Fingerprints of order and disorder on the high-frequency dynamics of liquids

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It is largely accepted that liquids are characterized by a short-range order usually corresponding to that of the solid phase at the same density. It is less clear to what extent dynamic properties of liquids and crystals can be compared. In particular, high-frequency collective excitations reminiscent of phonons in solids exist as well in liquids. They are however traditionally discussed in terms of relaxation processes characteristic of the liquid phase. We report here on a quantitative comparison of the collective excitations in liquid and polycrystalline sodium. We show that liquid sodium exhibits acoustic excitations of both longitudinal and transverse polarization at frequencies strictly related to those of the corresponding crystal. The only relevant difference between the liquid and the polycrystal appears in the broadening of the excitations: An additional disorder-induced contribution comes into play in the case of the liquid, which we show to be related to the distribution of local structures around the average one. These results establish a direct connection between structural and dynamic properties of liquids, with short-range order and overall structural disorder leaving very specific fingerprints.

vibrational dynamics | X-ray scattering

The short-range order in liquids, characterized by parameters like, e.g., the first neighbors distance and number, is usually similar to that of the corresponding solids (1, 2). Ab initio calculations of liquid structures spanning a large density range do support this idea (3). For what concerns dynamic properties, the situation is more complex. Local vibrations appear in the liquid as in the corresponding solid, although they are usually much broader (1). This broadening is generally related to the distribution of local structures that is present in the liquid phase (4) and that is, e.g., characterized by the distribution of first neighbors distances. Collective excitations exist as well in liquids, and their broadening is usually attributed to relaxation processes (5–7). In fact, the response of a crystal to an external perturbation can be formally expressed in terms of phonons, the modes of lattice vibration that can be defined in presence of a long-range, periodic structure. In liquids this description is instead formally not appropriate, and the concept of relaxation is usually introduced with reference to a dynamic process that controls the reequilibration of the system after it has been perturbed (8). Several distinct relaxation processes can be introduced to describe the dynamics of liquids, each one characterized by a characteristic time, τ, that sets the timescale over which it is active and observable. Indeed, if the perturbation frequency, ω, is such that $\omega\tau \ll 1$, the relaxation is active, whereas when $\omega\tau \gg 1$ the relaxation process does not have the time to take place and is frozen. In the infinite frequency limit, where all relaxations are frozen, the liquid behaves like a solid: This limit corresponds to the elastic regime, where only the instantaneous atomic positions and the interatomic forces determine the response to a perturbation (9). But how high should the frequency be in order to reach this elastic regime? And once reached, how is the liquid dynamics related to one of the corresponding crystalline phase?

Well-defined high-frequency acoustic excitations, appearing as peaks in the spectrum of the density–density fluctuations $S(q,\omega)$, have been observed in liquids up to wavenumbers q in the Å^{-1} range (5). Their characteristic frequencies exhibit a solid-like sinusoidal dispersion curve, $\omega(q)$, with the average interatomic distance, a, marking the border of a pseudo-Brillouin zone at $q_0 \simeq \pm\pi/a$ (10). For q values in the 0.1 Å^{-1} range, they propagate with a sound velocity higher than the adiabatic one measured in the macroscopic limit (9) and close to that of the corresponding crystals (11–15). Liquid metals are characterized by especially long living collective excitations visible up to about two-thirds of the pseudo-Brillouin zone size (5, 16). This feature makes them ideal candidates to probe the role of relaxation processes at high frequency. For this reason, we decided to study the high-frequency acoustic excitations of the simplest liquid metal, sodium. Despite the several studies carried out on this prototypical system (17–19), a systematic comparison of the results for the liquid and the crystal is still missing.

Results and Discussion

Selected density–density fluctuations spectra for the liquid and polycrystalline phases, measured by inelastic X-ray scattering (20), are reported in Fig. 1. Concerning the polycrystal, two excitations with predominantly longitudinal and transverse polarization are well visible for $q > 0.3$ Å^{-1}. The spectra corresponding to the liquid phase are similar to previously published data (18, 19) and appear clearly broader than for the polycrystal. For what concerns the polycrystal, the presence of transverse excitations for q values well within the first Brillouin zone in a spectrum that corresponds to the dynamic structure factor $S(q,\omega)$ and that as such probes the longitudinal dynamics, might result at first sight surprising. It has to do with the fact that acoustic modes have a pure transverse or longitudinal polarization only along high symmetry directions: In all other directions the modes have a mixed polarization, so that the corresponding transverse modes (or, more precisely, quasi-transverse modes) have always a nonzero projection along the longitudinal direction. When dealing with a polycrystal, the intrinsic orientational average automatically adds to the measured spectrum the contribution from these quasi-transverse modes also in first Brillouin zone. In the case of polycrystalline sodium, a simple elasticity-based model (21) indeed confirms on a quantitative ground this interpretation. As a matter of fact, the larger the elastic anisotropy of the system, the more visible is the quasi-transverse component in the $S(q,\omega)$ spectrum. One of the reasons to choose sodium for this study was indeed that, among simple metals, it is one of those showing the largest elastic anisotropy.

In simple liquids, the most important relaxation process is called structural, and is responsible for viscoelasticity and ultimately for the glass transition in glass-forming liquids (8). It can be described qualitatively in terms of the well-known cage effect, its characteristic time τ_α being the time for a particle to escape the cage of the nearest neighbors. In simple liquids the

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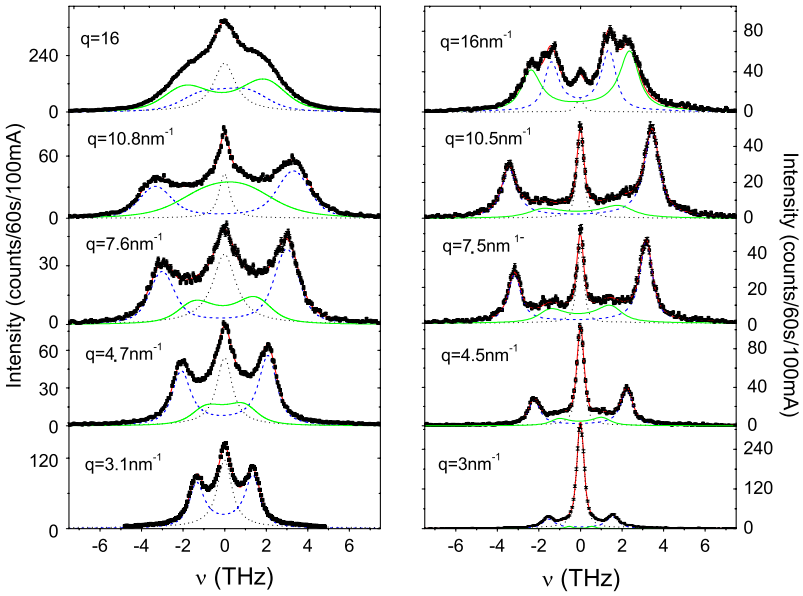

Fig. 1. Acoustic excitations in liquid and polycrystalline sodium. Inelastic X-ray scattering spectra of sodium at 393 K in the liquid phase (*Left*) and at room temperature in the (poly)crystalline phase (*Right*) at the indicated q values. The experimental data (squares) are reported together with the best fitting lineshape of model Eqs. **1–3** (full red line). The quasi-elastic (dotted black line) and inelastic transverse (dot-dashed green line) and longitudinal (dashed blue line) components of the fitting model are also reported after convolution to the instrumental function, which has an almost Lorentzian shape (FWHM = 0.34–0.36 THz). At the lowest investigated q the inelastic transverse component is negligible: Only for $q > 0.3$ Å^{-1} a fit based on two excitations is strictly required. The intensities are normalized to a 100 mA current in the storage ring and are reported as counts per 60 s.

structural relaxation time, τ_α, is of the order of 1 ps or less (5). In sodium at the melting point $\tau_\alpha \simeq 0.2$ ps (19). On a shorter timescale another relaxation process—dubbed microscopic—has been reported and ascribed to largely uncorrelated atomic collisional events (16). In sodium, its characteristic time, τ_μ, is$\simeq$ 0.04 ps (19). The difference between these two timescales is important here. In fact, the structural relaxation gives the main contribution to the quasi-elastic line intensity and width in the spectra of the liquid (Fig. 1), whereas it has almost no effect on the broadening of the acoustic excitations, as the condition $\omega\tau_\alpha >$ 1 is always fulfilled (5, 16). The microscopic relaxation, instead, comes out to be strongly coupled to the acoustic excitations (16, 19). It is important to emphasize that its existence places the elastic regime at frequencies as high as at least $1/\tau_\mu \simeq$ 10 THz so that, up to these frequencies, a truly liquid-specific theory in terms of relaxation processes seems to be required to correctly describe the dynamics of liquids. In contrast to this picture, the analysis presented below shows that no sign of relaxation processes faster than the structural relaxation can in fact be found in liquid sodium. This result clarifies that even at high frequency the structural relaxation is the only relevant liquid-specific dynamic feature, whereas on a timescale shorter than the structural relaxation one a truly elastic crystal-like description is appropriate.

In order to get to this point, we establish a direct connection between the spectra of the liquid and of the polycrystal, as shown in Fig. 2 (polycrystal, squares; liquid, circles) for two selected q values. The comparison is carried out within the quasi-harmonic approximation, i.e., assuming that frequencies simply scale with density, a scheme that works well for sodium (22, 23). Discarding the elastic line, obviously different due to the presence of the structural relaxation in the liquid, the spectra look similar, though those of the liquid are clearly broader. In particular, the shoulder at $\simeq$1.3 THz for the liquid nicely corresponds to the transverse excitations in the polycrystal, thus suggesting the existence of transverse excitations in the liquid too. The appealing hypothesis of a strict correspondence between the spectra of the two phases can be tested by broadening the spectra of the polycrystal convolving them with a function that for simplicity we choose to be Gaussian. This operation is done after rescaling the spectra of the polycrystal to the temperature of the liquid using the Bose distribution factor, $n(\omega)$, so to normalize the spectra to the same thermal population of the excitations (5, 9). The result is reported in the same figure (red line) and looks compelling: The spectral shape of the liquid is almost perfectly reproduced for what concerns the inelastic component, with reference not only to the position of the inelastic peaks but also to the intensity ratio between the longitudinal and transverse excitations.

With the aim of making this analysis quantitative, we fit the spectra of the liquid using as a model a Lorentzian function for the quasi-elastic line and two damped harmonic oscillator (DHO) functions for the longitudinal and transverse excitations convoluted with a Gaussian broadening function $G(\omega)$ with standard deviation σ. The model is then multiplied by$[n(\omega)+1]/k_B T$ in order to take into account the quantum nature of the measured spectra (5, 9) and finally is convoluted to the instrument function $R(\omega)$ prior to be fitted to the experimental data. We then write

$$S(q,\omega) = S_{el}\frac{\Gamma_0}{\omega^2+\Gamma_0^2} + S_{in}(q,\omega) \otimes G(\omega); \quad [1]$$

$$S_{in}(q,\omega) = I_L\frac{\Omega_L^2\Gamma_L}{(\omega^2-\Omega_L^2)^2+\omega^2\Gamma_L^2} + I_T\frac{\Omega_T^2\Gamma_T^2}{(\omega^2-\Omega_T^2)^2+\omega^2\Gamma_T^2}; \quad [2]$$

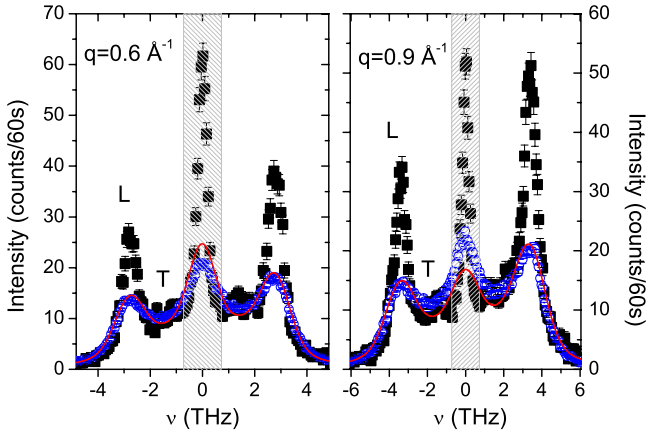

Fig. 2. Similarity of the acoustic excitations in the liquid to broadened acoustic excitations in the polycrystal. Inelastic X-ray scattering spectra of liquid (○) and polycrystalline (□) sodium at the indicated q values. The energy scale of the spectra for the liquid has been scaled by the density ratio between the polycrystal and the liquid. The red curve corresponds to the spectrum of the polycrystal after (*i*) correction for the Bose factor to account for the different temperature of the liquid and the solid; (*ii*) convolution to a Gaussian function; and (*iii*) scaling of the intensity to match the spectrum of the liquid. The elastic line is shadowed, because this comparison concerns only the inelastic peaks.

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$$I(q,\omega) = \left[A\frac{\omega(n(\omega)+1)}{k_B T}S(q,\omega)\right] \otimes R(\omega). \quad [3]$$

Here $\otimes$ represents the convolution operation; $\Omega_L(\Omega_T)$ is the frequency position of the longitudinal (transverse) excitations, and $\Gamma_L(\Gamma_T)$ the corresponding broadening (FWHM); $2\Gamma_0$ is the broadening of the quasi-elastic line; and A, S_{el}, S_{in}, I_L, and I_T are spectral weights. The same model has been used to describe the spectra of the polycrystal where, however, a delta function replaces in Eq. **1** both the Lorentzian contribution (elastic scattering) and the function $G(\omega)$ (no disorder-induced broadening). The spectra of both the liquid and the polycrystal can be well described using this model, as shown for the examples reported in Fig. 1.

In the following we discuss the fitting parameters derived from this analysis. Fig. 3 reports the dispersion curves for the longitudinal and transverse excitations for both the polycrystal (squares) and the liquid (circles). The phonon branches measured on a sodium single crystal at 90 K along the main symmetry directions (22) are shown as well. All the energy values are rescaled to the density of the polycrystal at room temperature and are reported in reduced zone units, q/q_0, where q_0 corresponds to the first diffraction peak for the liquid (2.02 Å^{-1}) and for the crystal ([110] Bragg peak) (24). The error bars in the figure correspond to $\pm\Gamma_L/2$ and $\pm\Gamma_T/2$, i.e., the half width at half maximum of the inelastic peaks obtained from the bare DHO model of Eq. **2**. In the same figure, we also report the theoretical elastic limit for the frequency of the longitudinal and transverse excitations in the liquid (9), calculated using the pair correlation function from literature diffraction data (25) and the pseudopotential developed by Price et al. (26), that was found to work well for liquid alkali metals. Fig. 3 shows that acoustic excitations with transverse polarization are present in the liquid for $q/q_0 \geq 0.2$. Qualitative similar observations have already been reported in other liquids (5, 7, 27). The striking result of Fig. 3 is the very close agreement between the results for the liquid and the polycrystal concerning both the frequency position and the broadening of the acoustic excitations and for both polarizations. It is clear as well that the frequency of the observed excitations corresponds to the orientational average of the modes of the single crystal, whereas the bare DHO width is closely related to their separation. Fig. 3 elucidates then that the spectra of the liquid are nothing but broadened versions of those of the polycrystal.

The existence of transverse excitations in sodium down to $q/q_0 \approx 0.2$ has far reaching implications. It means in fact that the complex spectral shape that was interpreted as the signature of the coupling of the longitudinal modes to a very fast microscopic relaxation process is instead largely the consequence of the presence of transverse modes in the liquid. This result implies that there is no need to invoke any microscopic relaxation related to the structural disorder to describe the high-frequency longitudinal dynamics of the liquid: At frequencies higher than the inverse structural relaxation time the liquid is well within the elastic limit, as it is confirmed by the nice agreement in Fig. 3 between the experimental data for the liquid and the theoretical elastic limit calculation over the whole first pseudo-Brillouin zone. Moreover, the nice correspondence between the acoustic branches of the liquid and the polycrystal reveals the real origin of the transverse excitations in the $S(q,\omega)$ of liquids: It has nothing to do with the structural disorder of the liquid (27) but rather with its short-range order, similar to (poly)crystals.

The key point is now the origin of the additional broadening of the acoustic excitations in the liquid with respect to the polycrystal. The standard deviation σ of the Gaussian describing it is reported in Fig. 4*A*: σ is negligible at low q, rapidly increases in the range $0.15 \leq q/q_0 \leq 0.3$, reaches then a plateau, and finally exhibits a pronounced minimum in correspondence to the position of the first diffraction peak. But what does σ represent? We have seen that the spectra of the liquid contain the same information—though somehow washed out—as those of the polycrystal in terms of acoustic branches. We know as well that, despite the fact that the average short-range order in the liquid is

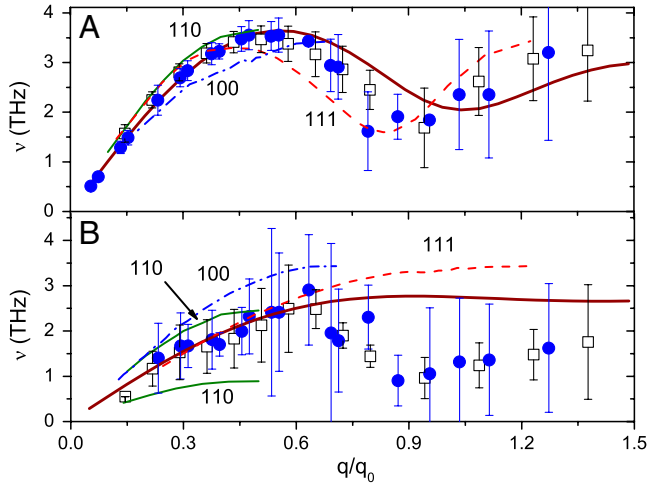

Fig. 3. Effect of the average short-range order on the acoustic dispersion curves of liquid sodium. The frequency position of the longitudinal A and transverse B excitations is reported as a function of q/q_0 for both the polycrystal (squares) and the liquid (bullets), q_0 being the position of the first diffraction peak in the liquid and in the polycrystal. The length of the error bars corresponds to the half width at half height of the bare DHO function of Eq. **2**. The results are compared with the single crystal phonon branches along the main symmetry directions (22) ([100], dot-dashed; [110], solid; [111], dashed lines). The theoretical prediction for the acoustic frequencies of the liquid in the infinite-frequency elastic limit is also reported (thick solid wine-colored line). All values are scaled to the polycrystal density.

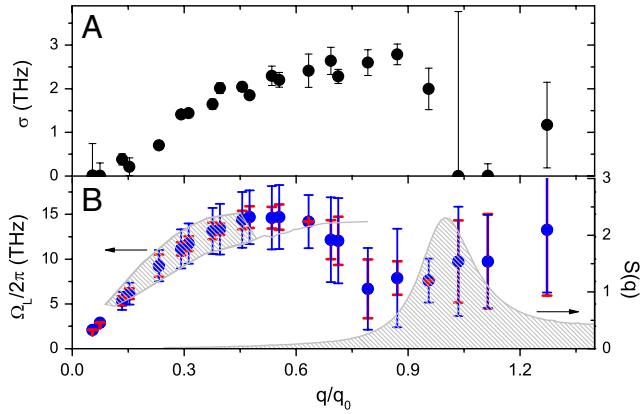

Fig. 4. Effect of structural disorder on the acoustic dispersion curves of liquid sodium. *A* shows the standard deviation of the Gaussian function describing the disorder-induced broadening of the acoustic excitations for the liquid, as a function of q/q_0. *B* shows the frequency position of the longitudinal excitations in liquid sodium (left axis). The larger error bars correspond to the entire broadening (coming from the combined DHO and Gaussian contributions; blue) whereas the smaller error bars correspond to the effect of the average short-range order only (coming from the DHO component; red). The distribution of local structures in the liquid can be described in terms of distribution of first neighbors distances using the FWHM, $\Delta = 0.45$ Å^{-1}, of the first diffraction peak in the static structure factor (shadowed gray line, right axis). In order to evaluate the effect of this distribution, the position of the highest frequency ([110]) and lowest frequency ([100]) acoustic branches for the single crystal are reported as a function of $q/(q_0 - \Delta)$ (upper gray line) and $q/(q_0 + \Delta)$ (lower gray line), respectively. These correspond indeed to the highest and lowest single crystal branches allowed by the considered distribution of first neighbors distances, and therefore of Brillouin zone sizes. The separation between these two branches (shadowed region) agrees well with the overall broadening of the longitudinal acoustic excitations.

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very similar to that in the polycrystal, the overall disorder brings in a distribution of local structures which can be characterized, e.g., by the distribution of first neighbors distances. As a consequence there is a distribution of local elastic moduli (28, 29), and then of acoustic frequencies. A rough but direct way to take into account this effect is to consider the distribution of pseudo-Brillouin zone sizes which follows from the distribution of nearest neighbors distances described in terms of the broadening of the first diffraction peak of the liquid, whose FWHM is $\Delta = 0.45$ Å^{-1}. Fig. 4*B* shows the implications of this simple idea for the longitudinal excitations (similar results hold for the transverse ones). Specifically, we report the position of the highest frequency ([1 1 0]) and lowest frequency ([1 0 0]) dispersion curves for the crystal as a function of $q/(q_0 - \Delta/2)$ and $q/(q_0 + \Delta/2)$, respectively. These correspond to the highest and lowest dispersion curves allowed by the considered distribution of nearest neighbors distances and then of Brillouin zone sizes: Their separation represents the broadening one should expect for the acoustic excitations in the liquid. Indeed we find such a correspondence when we consider the entire broadening (coming from the combined DHO and the Gaussian contribution) of the experimental longitudinal acoustic modes of the liquid, also reported in the figure as blue error bars. The take-home message here is that the broadening of the acoustic excitations in the liquid is well described by the combination of the effects coming from (*i*) the average short-range order, via the separation of the single crystal branches, and (*ii*) the distribution of local structures, described by the broadening of the first diffraction peak of the liquid. In the case studied here, the two contributions are almost equally important, the balance between them will however depend on the considered system.

In conclusion, the present work clarifies the connection between structural and high-frequency dynamic properties in liquids and in the corresponding solids. We have shown that the high-frequency (>0.5 THz) range can be properly treated within the elastic limit, and that no further liquid-specific relaxation process faster than the structural one is required to describe the experimental results. This result implies that at these frequencies a liquid is by all means indistinguishable from an amorphous solid. Moreover, we have shown that the short-range order and the overall structural disorder leave both very distinct fingerprints on the dynamic properties of liquids: The former dictates the acoustic dispersion curves, of both longitudinal and transverse polarization, which are indeed strictly related to those of the corresponding polycrystal, whereas the latter gives rise to the broadening of the acoustic excitations through the distribution of local structures, as it can be characterized by the distribution of nearest neighbors distances obtained from a simple analysis of the structural properties of the liquid. The present approach is thus strongly foretelling: The high-frequency dynamics of a liquid can be predicted once its structure and the dispersion curves of the corresponding crystal are known.

It is worth underlining that the approach followed here to describe the high-frequency dynamics of liquids rests on crystal-like concepts, differently from the liquid-like approach inherent in the traditionally utilized generalized hydrodynamics description (5). The latter formalism is a phenomenological modification of the continuum, simple hydrodynamics approach valid for the description of the macroscopic dynamics of simple liquids; it relies on the introduction of q dependence for the thermodynamic parameters and transport coefficients entering the simple hydrodynamics formalism in order to take care of the microscopic structure of the liquid. Clearly, the liquid- and crystal-like approaches are not necessarily mutually excluding: We actually believe that the combination of the two will strongly help to further advance in the understanding of the dynamics of liquids.

Materials and Methods

The high-frequency dynamics of sodium has been investigated analyzing the wavenumber and frequency dependence of the dynamic structure factor, $S(q,\omega)$, probed in a high-resolution inelastic X-ray scattering experiment carried out at beamline ID 16 (20) of the European Synchrotron Radiation Facility (ESRF). For the measurements on the liquid (solid), the monochromatic X-ray beam at the sample position had an energy of 23,725 (21,747) eV, a spot of 300×150 μm^2 horizontal × vertical sizes, and an intensity of $3 \cdot 10^9$ ($5 \cdot 10^9$) photons per second corresponding to 200 mA current in the storage ring. In our experiment, frequency spectra were measured in the −8 to 8 THz range at several q values between 0.1 and 2.6 Å^{-1} with a total integration time of about 600 (400) s per point. The instrumental resolution was q independent with a FWHM of 0.34 (0.36) THz, whereas the q resolution was fixed to 0.04 Å^{-1} FWHM. The sample cell was a windowless austenitic steel container mounted in a vacuum chamber with Kapton windows for the incoming and scattered (forward direction) X-ray beam. The length of the sample along the beam path was 7 mm. The sodium sample (99.5% purity, purchased from Aldrich) was loaded in the sample cell in an Ar-filled glove box. For the measurements in the liquid phase, a furnace was used to heat up the sample through its melting point $T_m = 371$ K to the temperature chosen for the experiment $T = 393$ K. The temperature stability has been 0.5 K during the whole experiment. The polycrystal was prepared from a sodium suspension in toluene by removing the solvent in vacuum. The resulting grain size was in the range of a few micrometers, which ensures a good orientational average. The measurements on the polycrystal were carried out at room temperature. The empty cell has been carefully checked to give a negligible contribution to the measured spectra.

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