Crossover between hydrodynamic and kinetic modes in binary liquid alloys

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Inelastic x-ray scattering measurements of the dynamic structure factor in liquid $Na_{57}K_{43}$, which is sensitive to the atomic-scale coarse graining, reveal a sound velocity value exceeding the long wavelength continuum value and indicate the coexistence of two phononlike modes. By applying generalized collective mode analysis scheme, we show that the positive dispersion of the sound velocity occurs in a wavelength region below the crossover from hydrodynamic to atom-type excitations and, therefore, it cannot be explained as sound propagation over the light species (Na) network. The present result experimentally proves the existence of positive dispersion in a binary mixture due to a relaxation process, as opposed to fast sound phenomena.

I. INTRODUCTION

Alkali metals in their liquid state are outstanding examples of *simple liquids*, as they encompass most of the physical properties of complex fluids without system specific complications. For this reason, in the past 50 years, they have been the subject of intensive investigations, both experimentally and by means of computer simulations, aiming at an understanding of the mechanisms underlying their collective dynamics at a microscopic length scale.[1,2] Interest in the collective excitations and interdiffusion processes in liquid binary mixtures was stimulated by the pioneering molecular dynamics (MD) study on the dynamics of the liquid Na-K alloy by Jacucci and Mc Donald.[3] Disparate masses metallic alloys soon became the subject of intensive studies: The existence of a new high frequency mode with phase velocity exceeding the hydrodynamic value was reported by MD simulations[4–6] and experimentally proved by inelastic neutron scattering (INS) in Li_4Pb,[7–9] $NaCs$,[10] and $Li_{30}Bi_{70}$.[11] The very nature of this additional mode, traditionally named as *fast sound*, has been in focus of a lively debate in the past three decades. It was either interpreted as an atom-type acoustic mode, supported by the light (Li) ions only, which would merge at low Q with the corresponding low frequency mode (Pb) into a single hydrodynamic velocity, or like an opticlike excitation arising above a certain threshold Q value. The existence of a crossover from hydrodynamic into atom-type excitations can be theoretically rationalized within a concept of kinetic (nonhydrodynamic) modes in the framework of generalized collective modes (GCM) approach.[12,13] It has been shown that at low Q, two modes exist and can be associated with hydrodynamics density and concentration fluctuations (collective region) while above a certain characteristic wave number (dependent on mass ratio), each of the two excitation reflects the dynamics of a single atomic species (atom-type excitations). The experimental identification of the crossover, however, has been heavily debated. This was mostly due to the inherent difficulty of disentangling the coherent contribution from the total INS scattering signal, which is paired by the kinematic limitation restricting the accessible energy-momentum region. In He-Ne mixtures, for instance, the upper limit of the hydrodynamic region has

been questioned, which is also in view of the possible definitions of the excitation frequency in terms of the maxima of the dynamic structure factor (DSF), $S(Q,\omega)$, rather that those of the longitudinal current autocorrelation function.[14–17]

$$C^L(Q,\omega) = \frac{\omega^2}{Q^2} S(Q,\omega). \tag{1}$$

Similar controversies appeared in the case of water,[18–22] where for long time the existence of fast sound was debated until the viscoelastic origin of such phenomenon was clarified.[23–25] In fact, the advent of the new radiation sources opened the possibility to perform inelastic x-ray scattering (IXS), overcoming the previously mentioned limitations of INS. The purely coherent x-ray atomic cross section, on the one hand, and the relative high energy of the probing x rays, on the other hand, allowed us to study the purely coherent dynamics of disordered systems over a wide energy-momentum range covering the region across the first pseudo-Brillouin region (i.e., momentum transfers up to the first maximum of the static structure factor Q_M). As a consequence, the past decade saw a renewed interest in the investigation of high frequency dynamics in liquids and glasses.

It is worth emphasing in this context the lack of a unique and reliable methodology to analyze scattering experiments performed on binary liquids. In majority of experimental studies, one makes use of either single damped harmonic oscillator (DHO) model[26] or even a memory-function ansatz—designed for one-component liquids—to estimate the dispersion law of collective excitations in binary melts.[27] Within this kind of approaches, one necessarily ignores the existence of nonacoustic high frequency excitations in binary liquids, which beyond hydrodynamic region contribute to the shape of partial dynamical structure factors. In this study, we show how one can apply more sophisticated theoretical schemes to the analysis of experimental data in a molten metallic alloy, namely, a theoretical GCM approach, which consistently treats contributions from hydrodynamic and kinetic modes to dynamical processes in liquids.

We report here on an IXS study of the microscopic dynamics in the molten alkali alloy $Na_{57}K_{43}$ which allowed us to (i) identify the presence of two phononlike modes, i.e., an

high frequency and a low frequency one (ii) point out, by applying GCM theory to a binary mixture, the origin of these two modes and how they contribute to the measured IXS spectra, i.e., essentially to the mass density fluctuations spectra [specifically, we show the existence of a crossover from a hydrodynamic regime, where only one of the two modes is active, to an atom-type regime, where two excitations appear in the $S(Q,\omega)$]; and (iii) assign the sound velocity excess over the hydrodynamic value to a relaxation process similar to that observed in simple,[2] molecular,[28] and hydrogen bonding liquids,[23] as opposed to the idea of acoustic excitation propagating over the network supported by the light species or to the effect of opticlike excitations.

II. EXPERIMENT

The experiment reported in this work was carried out at the high resolution beam line ID16 of the European Synchrotron Radiation Facility (Grenoble, France). The backscattering monochromator and analyzer crystals, operating at the (11, 11, 11) silicon reflections, gave a total energy resolution of 1.5 meV, while energy scans were performed by varying the temperature of the monochromator with respect to that of the analyzer crystals. A five analyzers bench allowed us to simultaneously collect the spectra at five different values of constant momentum transfer, covering a Q region below the position of the main diffraction peak ($Q_M \approx 18$ nm^{-1}). The sample consisted of an NaK alloy at $T = 300$ K with sodium concentration $C_{\text{Na}} = 57$ at. % and was hold in a silica capillary of 2 mm inner diameter and 10 μm wall thickness, sealed, and kept in an inert atmosphere. Energy scans in the range of $-40 < E < 40$ meV where repeated up to a total integration time of 300 s/point. Energy scans of the empty cell were collected in the same Q-E range as for the sample, prior to filling the capillary, and were subtracted to the final IXS spectra.

The signal measured in an IXS experiment is related to the double differential cross-section depending on the exchanged momentum Q and energy $E = \hbar\omega$, and it is basically the convolution of the instrumental resolution, $R(\omega)$, with the classical x-ray weighted DSF $S_{IXS}(Q,\omega)$,

$$I(Q,\omega) = \int d\omega' R(\omega - \omega') S_{IXS}(Q,\omega') \frac{\hbar\omega'/KT}{1 - e^{-\hbar\omega'/KT}}, \quad (2)$$

in which the last term accounts for the detailed balance condition. The outcome of the IXS experiment (open circles) is reported in Fig. 1 for selected values of the exchanged momentum Q.

In a binary mixture, $S_{IXS}(Q,\omega)$ can be represented as a linear combination of the partial dynamic structure factors,

$$S_{ij}(Q,\omega) = \int dt e^{-i\omega t} \langle n_i(Q,0) n_j^*(Q,t) \rangle \quad (i,j = \text{Na}, \text{K}),$$

$$(3)$$

with weights depending on the Na and K atomic form factors as well as on the concentration.[29] In the previous relation, $n_i(Q,t)$ is the number density of species i. Alternative repre-
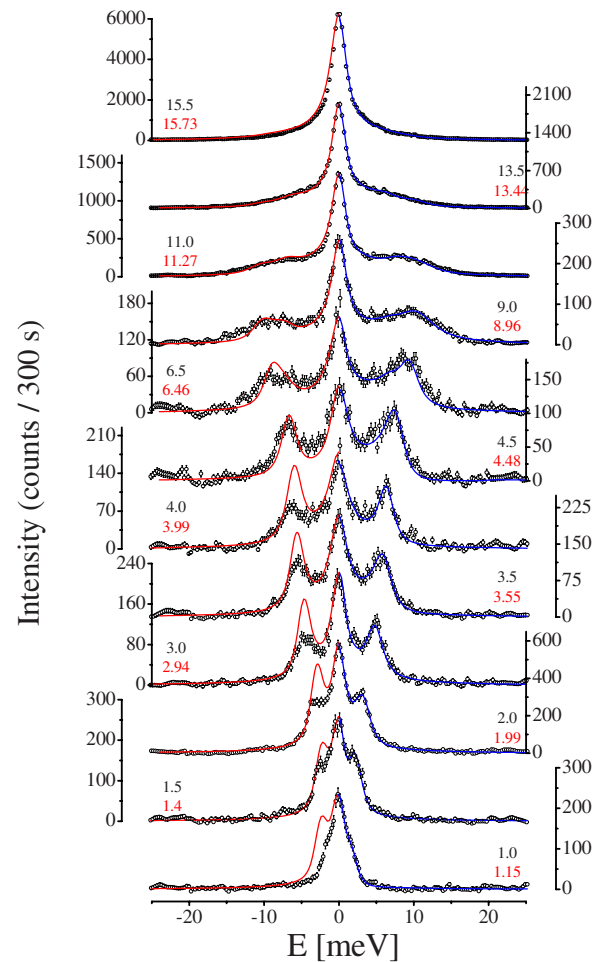

FIG. 1. (Color online) Selection of IXS spectra from the NaK liquid alloy ($C_{\text{Na}} = 57$ at. %, $T = 300$ K) measured for fixed Q values (nm^{-1}). The continuous line on the anti-Stokes side displays the outcome of GCM theory, while on the Stokes side shows Eq. (6), in which the eigenmodes are obtained as best fit to the data, as explained in the text.

sentations of $S_{IXS}(Q,\omega)$ can be given in terms of dynamical variables derived from the n_i as independent linear combinations. The most used options, in this respect, are the Bathia–Thornton[29] number concentration or the March mass-concentration variable.[30]

III. THEORETICAL MODEL

The usual approach to describe the DSF in a liquid is based on the solution of the Langevin equation for an appropriate set of dynamical variables. This can be done, for instance, by modeling a suitable memory kernel. At least two relaxation times must be accounted for in the second order memory function to describe the density fluctuations in a monatomic liquid,[2] and a nontrivial generalization for the case of binary liquids is required, along the line of the scheme proposed in Ref. 5. For this reason, we have chosen the GCM approach, which additionally treats coupling with thermal fluctuations and permits to solve the generalized

Langevin equation (GLE) in terms of dynamical eigenmodes.[13]

In such an approach, the GLE is solved in a Markovian approximation, while correlations faster than the hydrodynamic ones are accounted for by an appropriate choice for the set of dynamical variables. On this ground, the solution of the GLE is straightforwardly given in terms of eigenvalues and eigenvectors of the hydrodynamic matrix,

$$\mathbf{T}(Q) = \mathbf{F}(Q, t = 0) \cdot [\tilde{\mathbf{F}}(Q, z = 0)]^{-1}, \tag{4}$$

where $\mathbf{F}(Q, t)$ is the matrix of time correlation functions of all the variables belonging to the set and $\tilde{\mathbf{F}}(Q, z)$ is its Laplace transform.[12,13] The partial DSFs $S_{ij}(Q, \omega)$ are then obtained by means of a time Fourier transform of the corresponding components $F_{ij}(Q, t)$ related, for example, to the partial number densities n_i and n_j. As we will show below, the GCM approach captures all the essential features of the reported IXS experiment.

A suitable extended set of hydrodynamic variables for a binary mixture is provided by the set[12]

$$A^{(8)} = \{n_t(Q, t), n_x(Q, t), J_t(Q, t), J_x(Q, t),$$

$$\varepsilon(Q, t), \dot{J}_t(Q, t), \dot{J}_x(Q, t), \dot{\varepsilon}(Q, t)\}, \tag{5}$$

where the hydrodynamic variables reflecting the slowest fluctuations in the binary liquid are the total number density $n_t(Q, t)$, mass concentration density $n_x(Q, t)$, total longitudinal mass current $J_t(Q, t)$, and energy density $\varepsilon(Q, t)$. The variables with overdots correspond to the first time derivatives of the relevant fluctuations and are introduced to give a better representation of those processes that are faster than the hydrodynamic ones. It is worth to emphasize here that since total density and mass-concentration density can be easily represented as linear combinations of partial densities, one can represent set (5) via partial dynamical variables. Hence, the generalized hydrodynamic matrix $\mathbf{T}(Q)$ constructed on t-x dynamical variables [Eq. (5)] or on partial dynamical variables will have identical eigenvalues because both sets of dynamical variables are connected by linear transformation.

Within this scheme, the IXS weighted DSF can be expressed as

$$S_{IXS}(Q, \omega) = \frac{1}{\pi} \mathrm{Re} \left[\sum_1^8 \frac{G_\alpha^{IXS}(Q)}{z + z_\alpha(Q)} \right]_{z = i\omega}. \tag{6}$$

Both the eigenvalues [$z_\alpha(Q)$, which are complex or real in the case of propagating or diffusive modes, respectively] and the corresponding eigenvectors [$G_\alpha^{IXS}(Q)$, which are the x-ray weighted eigenvectors, i.e., are determined as the appropriate linear combination of partial Na and K densities, accounting for form factors and atomic concentration] can be evaluated within GCM theory once the Q-dependent elements of the hydrodynamic matrix are known. This requires the knowledge of initial values of time correlation functions of the variables set, as well as the correlation times of the kind

$$\tau_{mn}(Q) = \frac{1}{F_{mn}(Q, 0)} \int_0^\infty F_{mn}(Q, t) dt, \tag{7}$$

with $m, n = n_t, n_x, \varepsilon$.[13] To evaluate these latter, we performed molecular dynamics simulations on a system of 8000 particles interacting via effective two-body potentials[31] in a microcanonical ensemble at $T = 298$ K. The production run was performed over 3×10^5 time steps, and all the static and time correlation functions needed for GCM analysis were directly calculated in MD. The minimal wave number reached was 0.81 nm^{-1}. The resulting spectrum consists of eight dynamical eigenmodes: Complex-conjugated pairs of eigenvalues correspond to phononlike collective excitations $z_\alpha(Q) = \sigma_\alpha(Q) \pm i\omega_\alpha(Q)$, with $\omega_\alpha(Q)$ and $\sigma_\alpha(Q)$ as the frequency and damping of αth excitation, respectively, while real eigenvalues represent purely diffusive relaxation processes.

The prediction of parameter-free GCM theory (red lines) are reported in Fig. 1 along with the line shape of Eq. (6), in which the eigenmodes are obtained through a best fit to the experimental data (blue lines). In this latter case we used as initial values those from GCM and kept fixed all the frequencies of the propagating eigenvalues (imaginary part of the complex eigenvalues) and the (real) eigenvalues of the diffusive modes (diffusion coefficient). The weight coefficients $G_\alpha^{IXS}(Q)$ and the damping coefficients (real parts of propagating complex eigenvalues) were instead let as free parameters. Sum rules where used as constraints to further reduce the number of free parameters.[39] The stage of fitting procedure is necessary since the GCM analysis is based on MD simulations, which use effective two-body potentials. Namely, the use of effective potentials can introduce some extra error because it is well known that effective two-body potentials very rarely can yield the correct melting point, therefore underestimating or overestimating the contributions from relaxation processes to the shape of dynamical structure factors. In the anti-Stokes side of Fig. 1, one can see that the parameter-free GCM approach correctly predicts the frequency of collective excitations observed in the scattering experiments, only relative weight of relaxation processes (central peak) and propagating modes is not well reproduced. Namely, at that point, the aforementioned fitting procedure was applied, and later we will show that the fitting procedure does not alter the main results of GCM analysis.

Figure 2 shows the imaginary part of the two complex eigenvalues of the eight-variable GCM treatment (circles and dots). In order to ascertain the nature of these phononlike modes, we performed additional GCM analysis using four projected out onto processes of different origin three-variable eigenvalue problems using the following basis sets:

$$A^{(3t)} = \{n_t(Q, t), J_t(Q, t), \dot{J}_t(Q, t)\},$$

$$A^{(3x)} = \{n_x(Q, t), J_x(Q, t), \dot{J}_x(Q, t)\},$$

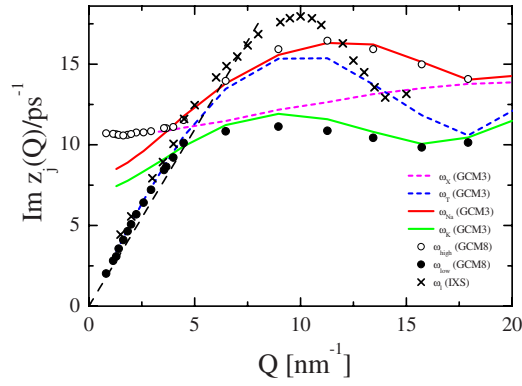$$A^{(3Na)} = \{n_{Na}(Q, t), J_{Na}(Q, t), \dot{J}_{Na}(Q, t)\},$$

FIG. 2. (Color online) Experimental dispersion relation obtained from the maxima of $C_{IXS}(Q, \omega)$ ($\times$), along with the hydrodynamic dispersion derived by ultrasound measurements (Ref. 32) (- - -). The two modes predicted by complete eight-variable GCM theory (high frequency, $\bigcirc$; low frequency, $\bullet$), and the outcome of partial three-variable GCM analysis with total (blue, - - -), concentration (pink, - - -), Na (red,——), and K (green,——) variables are also reported.

$$A^{(3K)} = \{n_K(Q,t), J_K(Q,t), \dot{J}_K(Q,t)\}. \qquad (8)$$

This approach allows us to ascertain the origin of each branch of collective excitations in the spectrum and processes responsible for them in different Q regions.[12,33,34] The result is reported in Fig. 2 and clearly shows how the atom-type excitations (full red and green lines for the Na and K subsets, respectively) correspond to the two eigenvalues of the eight-variable treatment in the high Q limit, while at low Q limit, the same eigenvalues are reproduced by the total density and concentration subsets. This is a clear indication of the existence of two dynamical regimes, a low Q, collective region, and a high Q, atom-type region, in agreement
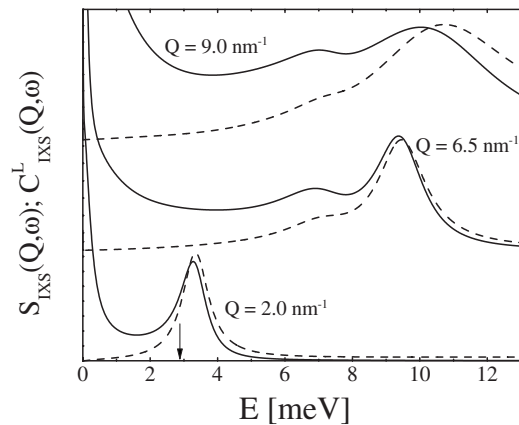

FIG. 3. Classical resolution deconvoluted $S(Q, \omega)$ (continuous line) and $C^L(Q, \omega)$ (dashed lines) obtained from the IXS measurement. The presence of a single mode exceeding the adiabatic frequency (marked by the arrow) is clearly visible at $Q=2$ nm^{-1}, i.e., well below the crossover to atom-type dynamics. For $Q=6.5$ nm^{-1}, two excitations appears, with the high frequency one dominating at $Q=9.0$ nm^{-1}.
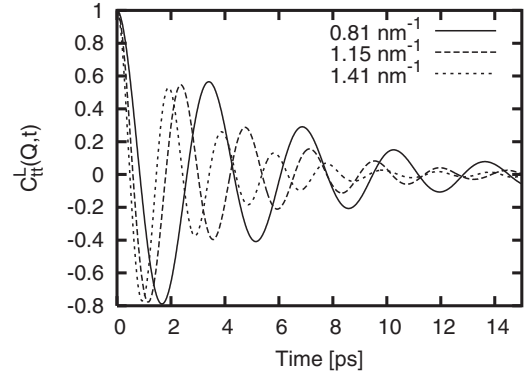

FIG. 4. Normalized total mass current autocorrelation function at the three lowest Q values investigated in MD.

with recent observation of opticlike modes above ≈ 2 nm^{-1} in MD simulations on Li alloys.[5] Interestingly, the experimental sound velocity $c_l(Q) = \omega_l(Q)/Q$ determined by the maximum ω_l of the raw current autocorrelation function $\omega^2 S_{IXS}(Q, \omega)$ (black crosses) turns out to be mainly determined by the low energy mode at low Q and by the high energy mode at high Q. In addition to that, it does not reach, even at the lowest explored Q, the hydrodynamic (adiabatic) value,[32] while exceeds it systematically.[40]

This is clearly shown in Fig. 3, in which we report the DSF, $S(Q, \omega)$, and longitudinal current spectra, $C^L(Q, \omega)$, from Eq. (6), adjusting the weight of the high frequency and low frequency modes to the IXS spectra. At the lowest Q values, a single mode dominates the spectrum, with an energy clearly exceeding the one expected from hydrodynamic dispersion $\omega = c_0 Q$. Around the first crossover ($Q = 6.5$ nm^{-1}) both the high frequency and the low frequency modes do contribute to the spectra, while at $Q=9$ nm^{-1}, most of the IXS signal is due to the high frequency Na-like excitation.

This picture is substantiated by direct inspection of the MD data. Different time correlation functions derived in MD simulations and analyzed by the GCM approach can reveal different types of collective excitations existing in binary liquids. In Figs. 4 and 5, we show the MD-derived longitudinal total and mass-concentration autocorrelation function at
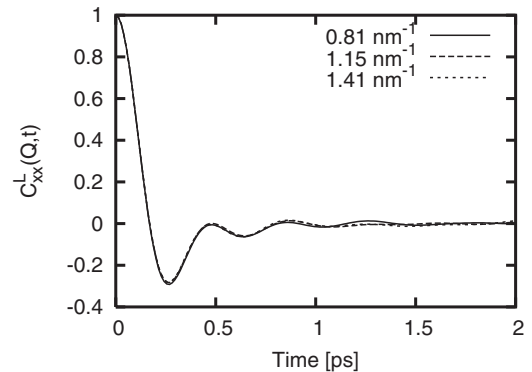

FIG. 5. Normalized mass-concentration current autocorrelation function at the three lowest Q values investigated in MD.
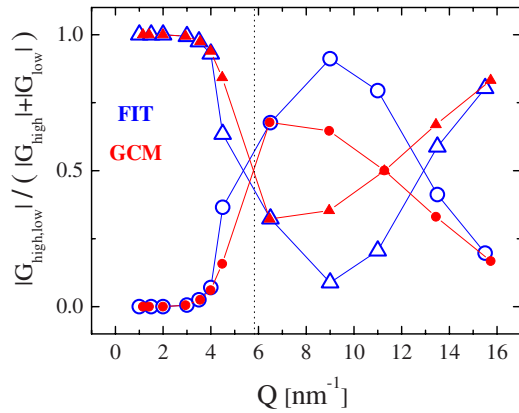
FIG. 6. (Color online) High (circle) and low (triangle) frequency modes from GCM (red) and from fitting procedure (blue). The crossover at $Q \approx 6$ nm^{-1} can be observed.

three lowest wave numbers sampled in simulations. It is clearly seen that the mass-concentration current autocorrelation functions $C_{xx}^L(Q,t)$ functions reflect propagating excitations with much smaller time scale than the sound excitations, which as in the case of single-component liquids contribute to the shape of $C_{tt}^L(Q,t)$ functions. Furthermore, almost identical shape of $C_{xx}^L(Q,t)$ functions at small wave numbers implies that the short-time excitations have almost the same frequency and damping in the long-wavelength limit, which is usually the evidence of kinetic opticlike excitations.

The presence of the two phononlike modes in the IXS spectra at intermediate Q values can be conveniently quantified by looking at the relative weights reported in Fig. 6.

The low frequency mode is clearly dominant below the sharp crossover occurring at $Q \approx 6$ nm^{-1}. Around this value, both modes contribute to the IXS spectra, while above the high frequency mode dominates up to a new crossover at $Q \approx 13$ nm^{-1}. The IXS cross section is indeed roughly proportional to the total density autocorrelation function, and hence, below the crossover it reflects the collective longitudinal excitation and not the opticlike mode. As previously mentioned, the amplitude values from GCM and from fitting procedure are quite different, but the oscillating behavior with the presence of sharp crossovers is captured by both the methods. The fact that positive dispersion observed from the IXS data occurs well below the first crossover, when the spectral frequency is solely reproduced by the low frequency mode, allows us to rule out a "fast sound" mechanism, i.e., the transition from the hydrodynamic regime to a dynamical regime in which the sound propagates over a network formed by the light component of the mixture, as it happens in disparate masses alloys. On the contrary, the origin of such an effect has to be traced back to a relaxation process of viscoelastic origin, similar to that invoked in simple[2] and molecular[28] liquids, glasses[35,36] and in the controversial case of water and hydrogen bonding systems.[23–25,37]

Along the same line, the inspection of the MD derived spectral functions $C_{NaNa}^L(Q,\omega)$ and $C_{KK}^L(Q,\omega)$ presented in Fig. 7 for a $Q=2$ nm^{-1}, i.e., well below the crossover, is particularly enlightening. The fingerprint of fast sound would
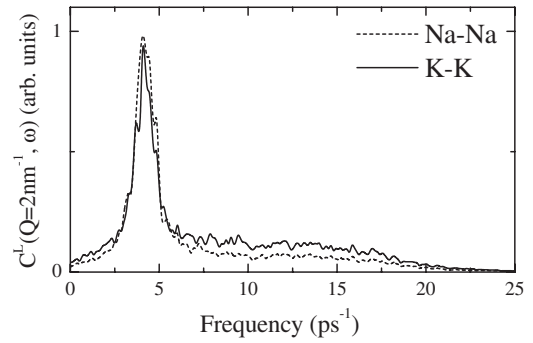

FIG. 7. MD calculated $C_{NaNa}^L(Q,\omega)$ (- - -) and $C_{KK}^L(Q,\omega)$ (—) for $Q=2$ nm^{-1}. The same acoustic mode turns out to dominate in both species, and no sign of fast soundlike phenomenology, in which the light component is dominated by the high frequency mode, is recovered at Q lower than the crossover value ($Q \sim 5$ nm^{-1}, see Fig. 8) from MD.

be here the presence of an extra high frequency mode in the light component (Na) spectra which, in turn, is absent in the corresponding heavy component (K) spectra. As can be observed in Fig. 7, this is certainly not the present case. Conversely, the same acoustic mode dominates in both the species, accompanied by a common weak (Q^2 vanishing) high frequency feature. As shown in Fig. 8, this scenario holds at any Q's below the crossover, while at high Q's, the partial spectra are dominated by distinct opticlike modes which can be identified with the high and the low frequency GCM modes.

IV. CONCLUSIONS

In conclusion, we reported the results of an experimental IXS investigation of high frequency dynamics in the liquid binary mixture Na$_{57}$K$_{43}$, which suggest the presence of two distinct phononlike excitations beside diffusive modes. A quantitative analysis based on GCM theory allows us to
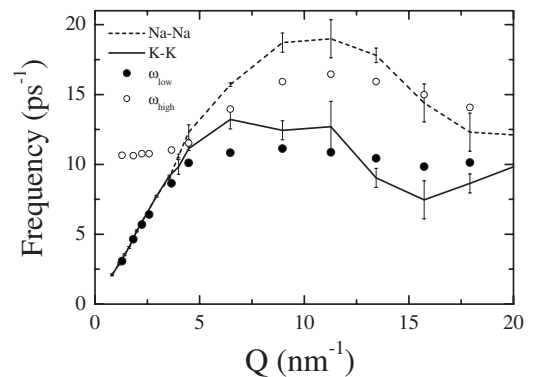

FIG. 8. MD calculated dispersion relation for Na (- - -) and K (—); the high (○) and low (●) frequency GCM modes are also reported. At Q's below the crossover, the same scenario depicted in Fig. 7 holds for the partial $C_{NaNa}^L(Q,\omega)$ and $C_{KK}^L(Q,\omega)$ currents, which are dominated by what can be addressed to as the GCM low frequency mode.

identify the nature of these excitations. The relative weight of the phononlike modes shows an oscillatory behavior with Q, ruled by sharp crossovers defining collective and atom-type dynamic regions, in which the global density and/or concentration fluctuation and the partial Na/K density fluctuations dominate, respectively. The observed positive dispersion of the acoustic branch, i.e., a sound velocity value exceeding the long wavelength limit, occurs well within the collective region. This indicates for this process an origin due to the existence of a microscopic relaxation, ubiquitous in monatomic liquids, as opposed to the fast sound phenomena observed in disparate masses mixtures, where enhanced sound velocity has been suggested to be either due to the propagation of sound over the light component network or to the presence of an optic branch.

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[40] Alternative definitions of the sound velocity have been recently discussed (Ref. 38). In this context, we just stress that both definitions lead to the same conclusion, i.e., higher than hydrodynamic values.