Collective excitations in liquid bismuth: the origin of kinetic relaxing modes

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Abstract
The origin of relaxing collective modes in liquid semimetallic Bi is studied within the generalized collective modes approach. It is found that the lowest kinetic relaxing mode strongly affects the shape of the density–density time correlation functions beyond the hydrodynamic region and determines specific de Gennes features slowing the density fluctuations near the main peak of the structure factor. It is shown that this kinetic relaxing mode appears due to structural relaxation in a liquid. Contributions of different collective modes to the shape of the density–density time correlation functions are studied in detail.

1. Introduction

In our recent study of collective excitations in liquid semimetallic Bi [1] (we will refer to this paper as paper I) at $n = 0.0289$ Å^{-3} and $T = 578$ K it was shown that

(i) the density–density time correlation functions (TCFs) obtained in our MD simulations were reproduced perfectly over a wide range of wavenumbers k within the nine-variable approximation of the generalized collective modes (GCM) approach;

(ii) there are three branches of propagating collective excitations in the spectrum for wavenumbers k up to 3 Å^{-1}.

The many-variable GCM approach allows one to take into account short-time processes of structural and thermal origin of higher order than hydrodynamic ones. Two branches among the generalized propagating modes were associated [1] with the viscoelastic properties of liquid Bi (generalized sound modes and high-frequency kinetic ones), and one branch has been identified as high-frequency heat waves. We have also found three generalized relaxing collective modes with purely real eigenvalues, the origin of which, however, was not completely established. It was only accepted that the lowest relaxing mode, which behaved as $D_T k^2$ in the hydrodynamic region with D_T being the thermodiffusion coefficient [2,3], could be considered throughout the whole k-range studied as the generalized thermodiffusive mode. In paper I it has also been established that the relative contribution from this lowest relaxing mode to the density–density time correlation function increases and even becomes dominant for large

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enough wavenumbers, so the question arises of what kinds of physical process beyond the hydrodynamic region result from the purely relaxing modes obtained.

Another interesting point to investigate is the k-dependence of the amplitudes of the mode contributions to the density–density TCFs beyond the hydrodynamic region. For pure liquids only the hydrodynamic behaviour of the amplitudes for thermodiffusive and propagating sound modes is well known [4]. Thus, in the case of a nine-variable treatment a study of separate mode contributions from generalized hydrodynamic and kinetic modes to TCFs would be of great interest.

The method of GCM and the scheme of our analysis presented in this report are quite general and can be applied to any pure or complex liquid. The results obtained in this study are similar to those obtained recently by us for liquid Pb [5], liquid Cs [6] and Lennard-Jones liquids [7]. The main focus in this report is not the specific features of liquid bismuth, but the origin of the relaxational behaviour in a pure liquid beyond the hydrodynamic region. More general comparison of relaxational behaviours in pure metallic, semimetallic and Lennard-Jones liquids as well as binary mixtures will be presented elsewhere.

The goals of this report are:

(i) to complete the study of generalized collective modes in semimetallic Bi by establishing the origins of all of the relaxing modes found in paper I;

(ii) to study the amplitudes of separate mode contributions to the density–density TCF and on this basis to estimate the most important mode contributions for different ranges of wavenumbers k.

2. Results and discussion

2.1. The origin of relaxational behaviour in liquid bismuth beyond the hydrodynamic region

For the details of the theoretical method used in our calculations on liquid Bi, we refer the reader to paper I. We simply state that for the study of long- and short-time processes in the liquid we use a basis set of nine dynamical variables [8, 9]:

$$A^{(9)}(k, t) = \left\{ n(k, t), J_l(k, t), e(k, t), \dot{J}_l(k, t), \dot{e}(k, t), \ddot{J}_l(k, t), \ddot{e}(k, t), \dddot{J}_l(k, t), \dddot{e}(k, t) \right\} \quad (1)$$

where the microscopic operators $n(k, t)$, $J_l(k, t)$ and $e(k, t)$ are the hydrodynamic densities of particle number, momentum and energy, respectively. In (1) the dots denote the orders of the time derivatives for the relevant operator. The basis set (1) is applied to generate the 9×9 eigenvalue problem for the generalized hydrodynamic matrix $\mathbf{T}(k)$ [10]. In the case of liquid Bi [1] we found within the nine-variable set (1) three pairs of complex-conjugate eigenvalues (propagating modes) and three purely real ones (relaxing modes) in the k-region $k < 3$ Å^{-1} (see figure 5 in paper I). To distinguish between the propagating and relaxing collective modes we will identify the purely real eigenvalues as $d_i(k)$ with the italic subscript $i = 1, 2, 3$ and the complex eigenvalues as

$$z_\alpha(k) = \pm i\omega_\alpha(k) + \sigma_\alpha(k) \quad (2)$$

with the Greek subscript $\alpha = 1, 2, 3$, where $\omega_\alpha(k)$ and $\sigma_\alpha(k)$ denote the dispersions and damping coefficients of propagating excitations, respectively.

In figure 1 the results for all of the generalized relaxing modes, obtained within the nine-variable treatment (1), are shown. In the hydrodynamic range of wavenumbers the thermodiffusive mode $d_1(k)$ (see figure 1) is proportional to k^2, in complete agreement with the predictions of the hydrodynamic theory [2, 3]. The other two relaxing modes are kinetic ones and tend to nonzero values when $k \to 0$. Therefore, they describe mainly
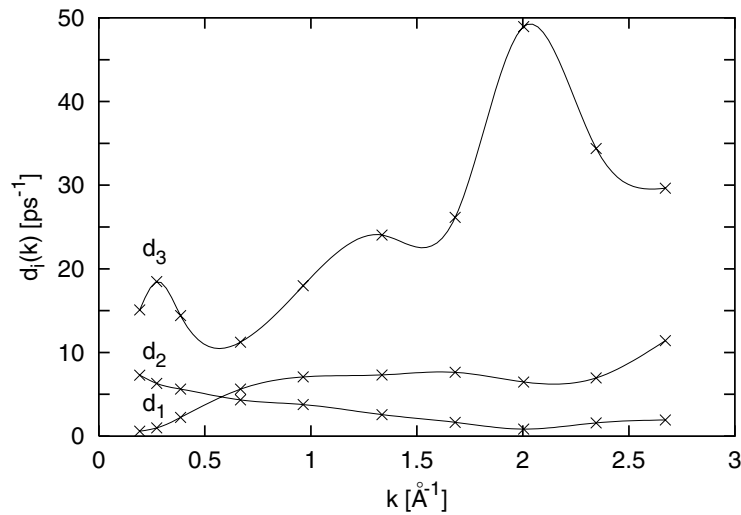
Figure 1. Three relaxing collective modes $d_i(k)$ obtained for the nine-variable basis set (1). As a guide to the eye, solid lines show the spline interpolation.

the short-time behaviour and cannot contribute substantially to dynamical processes in the hydrodynamic region, so the leading contribution from the relaxing modes is expected to be from the generalized thermodiffusive mode $d_1(k)$ when k is small. It is seen in figure 1 that appropriate classification of the eigenvalues obtained is a rather complicated problem (especially at intermediate values of k, where the two lowest relaxing modes overlap) and needs an additional analysis. Only in such a way can the correct k-dependence (and *the correct physical understanding*) of the corresponding damping coefficients be established. We note in this context that in paper I the relaxing mode identified as z_1^R was associated with the lowest purely real eigenvalue for the whole k-region.

To classify the purely real eigenvalues obtained in a correct way and to establish the physical origin of each branch, we apply the same scheme as was used in paper I for the study of generalized propagating modes. The main idea is the following: we split the set of dynamical variables (1) into two separate subsets:

$$A^{(4h)}(k, t) = \left\{ h(k, t), \dot{h}(k, t), \ddot{h}(k, t), \dddot{h}(k, t) \right\} \tag{3}$$

and

$$A^{(5)}(k, t) = \left\{ n(k, t), J_l(k, t), \dot{J}_l(k, t), \ddot{J}_l(k, t), \dddot{J}_l(k, t) \right\} \tag{4}$$

which describe by definition the dynamic properties arising from thermal and viscoelastic processes, respectively. Note that the dynamical variable $h(k, t)$ in (3) is the heat-density operator defined in paper I. Thus, the collective mode spectra can be recalculated for the separated subsets $A^{(4h)}$ and $A^{(5)}$, providing us with additional information about the 'bare' (or 'uncoupled') collective modes. This also allows one to assess the role and strength of coupling, by comparing the spectra obtained for the total set $A^{(9)}$ and the separate subsets (3) and (4). Such analysis was used previously in our study of the transverse dynamics in binary liquids [11, 12].

In figure 2 we compare the results for the generalized thermodiffusive mode $d_1(k)$ obtained for two different sets of dynamical variables: the basic nine-variable set $A^{(9)}(k, t)$, which takes
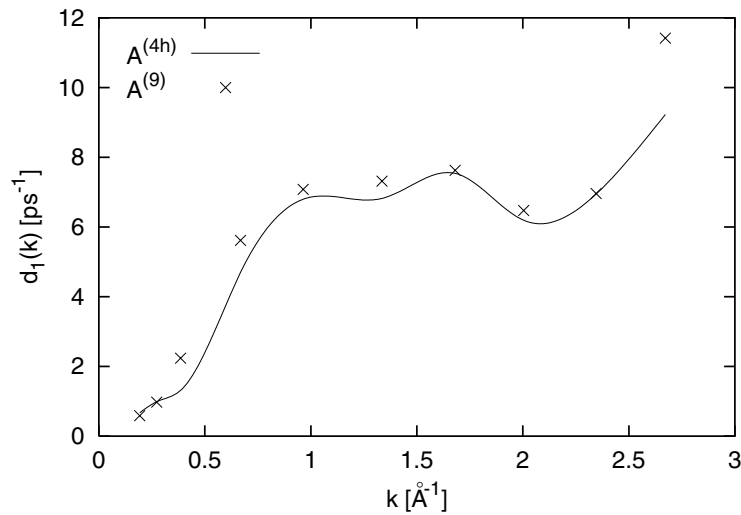
Figure 2. The generalized thermodiffusive collective mode $d_1(k)$ obtained for two sets of dynamical variables: the 'coupled' nine-variable set (1) (crosses); and the four-variable subset (3) (spline-interpolated solid line).

into account coupling between thermal and viscous processes (crosses in figure 2); and the four-variable subset $A^{(4h)}(k, t)$. One can see rather good agreement between the lowest thermal relaxing mode (solid line), obtained for the subset $A^{(4h)}(k, t)$, and the purely real eigenvalue $d_1(k)$ (crosses) found within the nine-variable treatment (1). Note that for intermediate and large wavenumbers, $k > 0.55$ Å^{-1}, the eigenvalue $d_1(k)$ is no longer (see figure 1) the lowest relaxing mode in the nine-mode spectrum.

Another purely real eigenvalue, obtained for the separated subset $A^{(4h)}(k, t)$, correlates well with the generalized kinetic relaxing mode $d_3(k)$ shown in figure 1. Thus, two relaxing collective modes $d_1(k)$ and $d_3(k)$, obtained for the 'coupled' nine-variable set $A^{(9)}(k, t)$, are caused mainly by thermal processes. Due to strong damping, the relaxing collective mode $d_3(k)$ can contribute only marginally to the dynamical properties of a liquid considered. Thus, the relaxational behaviour of time correlation functions is mainly determined by the generalized thermodiffusive mode $d_1(k)$ as well as the generalized kinetic relaxing excitation $d_2(k)$.

In figure 3 the purely real eigenvalues $d_2(k)$ of the nine-mode spectrum (crosses) are plotted together with the only relaxing mode (solid line) obtained for the separated 'viscoelastic' five-variable subset $A^{(5)}(k, t)$. It is seen that for $k > 1$ Å^{-1} the kinetic mode $d_2(k)$ is perfectly reproduced within the viscoelastic treatment. It is worth mentioning that the generalized mode approach used for the subset $A^{(5)}(k, t)$ does indeed give the higher-order version of viscoelastic theory [2, 3, 13, 14]. Note that no adjustable parameters are used in this version.

One can conclude from figure 3 that the viscoelastic approximation in the case of Bi does not work well in the range of small wavenumbers, for $k < 1$ Å^{-1}, where the coupling with the thermal processes becomes very important. When the thermal fluctuations are taken into account the coupling results in a pushing upwards of the eigenvalue $d_2(k)$ (crosses in figure 3) from the 'bare' relaxing mode, obtained in the viscoelastic approximation (solid line in figure 3).

In order to clarify further the physical meaning of the kinetic relaxing mode $d_2(k)$, we consider also the k-dependence of an eigenvalue:

$$d_0(k) = \tau_{nn}^{-1}(k) \tag{5}$$
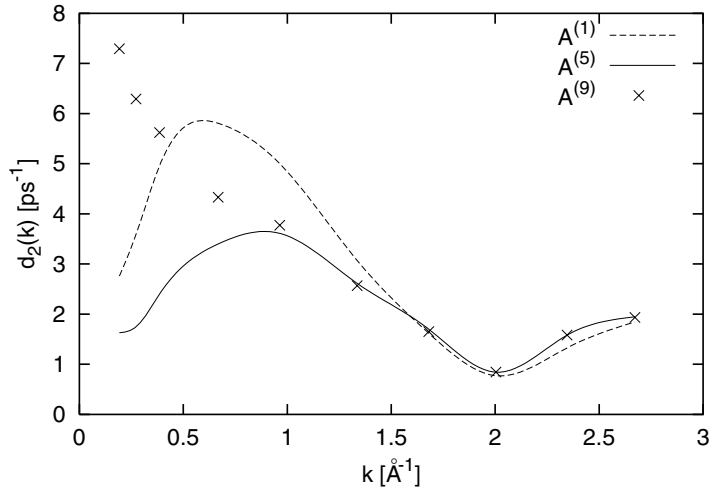
Figure 3. The kinetic relaxing collective mode $d_2(k)$ obtained for three sets of dynamical variables: the 'coupled' nine-variable set (1) (crosses); the five-variable subset (4) (spline-interpolated solid line); and the single-variable set $A^{(1)}$ (spline-interpolated dashed line).

which can be found within the simplest one-variable GCM theory for the density operator alone:

$$A^{(1)}(k, t) = \{n(k, t)\}. \tag{6}$$

The eigenvalue $d_0(k)$ gives additional information about the slowest density fluctuations in a liquid. In equation (5) the function $\tau_{nn}(k)$ is the generalized correlation time associated with the density–density TCF (see paper I). Qualitatively, the relaxing eigenvalues obtained for the five-variable subset $A^{(5)}(k, t)$ and for the single-variable treatment (dashed line in figure 3) show behaviours that are nearly the same. In the region of wavenumbers $1.5 \, \text{Å}^{-1} < k < 3 \, \text{Å}^{-1}$ the single-variable mode is very close to the kinetic relaxing mode $d_2(k)$. This implies that in the region of the main peak of the static structure factor $k \sim Q_p$ there is rather good separation of long- and short-time dynamical processes, represented by the relaxing mode $d_2(k)$ and propagating acoustic excitations, respectively.

The origin of the kinetic relaxing mode can be clearly established by taking into account that within the one-variable treatment the method of GCM leads to a single-exponential form for the density–density time correlation function:

$$F_{nn}^1(k, t) = G_{nn}^1(k) \exp\{-d_0(k)t\}$$

which follows from equation (10) of paper I for the case of basis set $A^{(1)}(k, t)$. Making use of equation (5) and the lowest sum rule, one gets

$$F_{nn}^1(k, t) = S(k) \exp\{-t/\tau_{nn}(k)\}. \tag{7}$$

One can see from (7) that within such a one-variable treatment, $\tau_{nn}(k)$ is a specific relaxation time and, in particular, for $k = Q_p$ it has the meaning of the lifetime of a particle in a cage of nearest neighbours. This implies that the kinetic relaxing mode $d_2(k)$ is connected with structural relaxation in a liquid, and, in particular, with the effect of cage diffusion [15].

2.2. Mode contributions to the density–density TCF

In the hydrodynamic region, three generalized hydrodynamic modes, $z_1^\pm$ and $d_1(k)$ (see figure 5 in paper I), are well separated from the kinetic ones. In particular, this explains why the

hydrodynamic result [4] for the density–density TCF works perfectly for small k. In order to see how the hydrodynamic picture of mode contributions [4] changes on increasing k and to study in more detail the crossover to relaxational behaviour of the TCF at large k-values, let us consider now the separate mode contributions to the shape of the density–density TCF in the nine-variable theory. In this case the density–density TCF can be expressed as a weighted sum of nine terms [11]:

$$F_{nn}^9(k, t) = \sum_{i=1}^{3} A_{nn}^i(k)\mathrm{e}^{-d_i(k)t} + \sum_{\alpha=1}^{3} \left\{ B_{nn}^\alpha(k) \cos[\omega_\alpha(k)t] + C_{nn}^\alpha(k) \sin[\omega_\alpha(k)t] \right\} \mathrm{e}^{-\sigma_\alpha(k)t} \quad (8)$$

where the amplitudes A_{nn}^i, B_{nn}^α and C_{nn}^α are functions of k and are defined via eigenvectors associated with relevant eigenvalues. The first three terms on the right-hand side of equation (8) describe the contributions from the relaxing modes $d_i(k)$, and the last six terms are connected with the generalized propagating modes (2). We will call terms with amplitudes B_{nn}^α and C_{nn}^α, caused by the propagating modes, symmetric and asymmetric ones, respectively. Obviously, for $t = 0$ one has

$$F_{nn}^9(k, 0) = \sum_{i=1}^{3} A_{nn}^i(k) + \sum_{\alpha=1}^{3} B_{nn}^\alpha(k) \equiv S(k). \quad (9)$$

The dynamical structure factor can be calculated in an analytical form by taking the Fourier transform of (8). This results in: three central Lorentzians at $\omega = 0$ (contributions from the relaxing modes); six Lorentzians (symmetric contributions) located at nonzero frequencies $\pm\omega_\alpha(k)$, determined by the dispersions of propagating modes (see (2)); and six non-Lorentzian corrections originating from the asymmetric contributions in (8). The latter reduce in the hydrodynamic limit to two anti-Stokes components, known from the standard hydrodynamic theory [2, 3].

We plot in figure 4, according to equation (9), two main contributions to the static structure factor $S(k)$ of liquid Bi. In this figure the static structure factor $S(k)$, obtained via Fourier
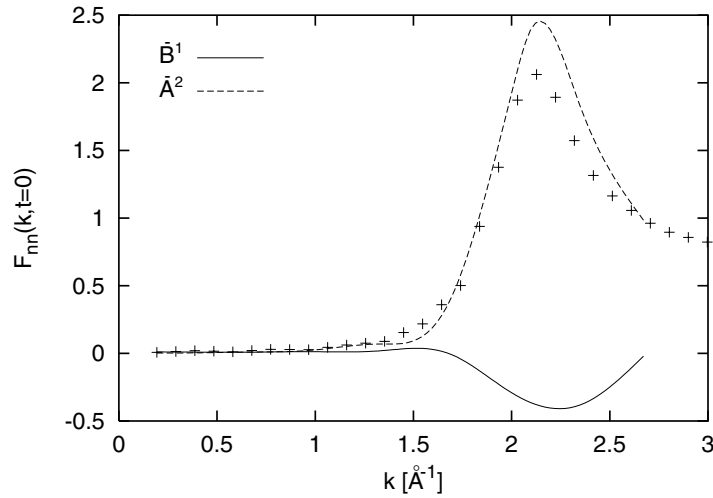
Figure 4. Contributions from the relaxing collective mode $d_2(k)$ (spline-interpolated dashed line) and generalized sound excitations (spline-interpolated solid line) to the zero-order frequency moment of $S(k, \omega)$, found in MD. The static structure factor $S(k)$ obtained via the MD-derived pair correlation function is shown by crosses.

transformation of the MD-derived pair correlation function, is also shown by crosses. One can see that in the range $k \sim Q_p$ two kinds of process mainly determine the shape of $S(k)$ (all of the other contributions are negligible on the scale used). They are formed by the long-time decay from the structural relaxation term (the collective mode $d_2(k)$, dashed line) and the shorter-timescale density fluctuations related to the acoustic-like excitations (solid line). Note that the relative contribution from $d_2(k)$ is almost six times bigger than the one from generalized acoustic excitations.

More precise information about the relative mode contributions can be obtained by considering the k-dependence of the normalized amplitudes, defined by the function $F_{nn}(k, t)/S(k)$. In this case, equation (8) can be rewritten in the form

$$\sum_{i=1}^{3} \bar{A}_{nn}^{i}(k) + \sum_{\alpha=1}^{3} \bar{B}_{nn}^{\alpha}(k) = 1$$

where the normalized amplitudes are defined as follows:

$$\bar{M}_{nn}^{g}(k) = M_{nn}^{g}(k)/S(k) \qquad M = \{A, B, C\} \qquad g = \{i, \alpha\}.$$

The results of our calculations for the normalized amplitudes are presented in figure 5.
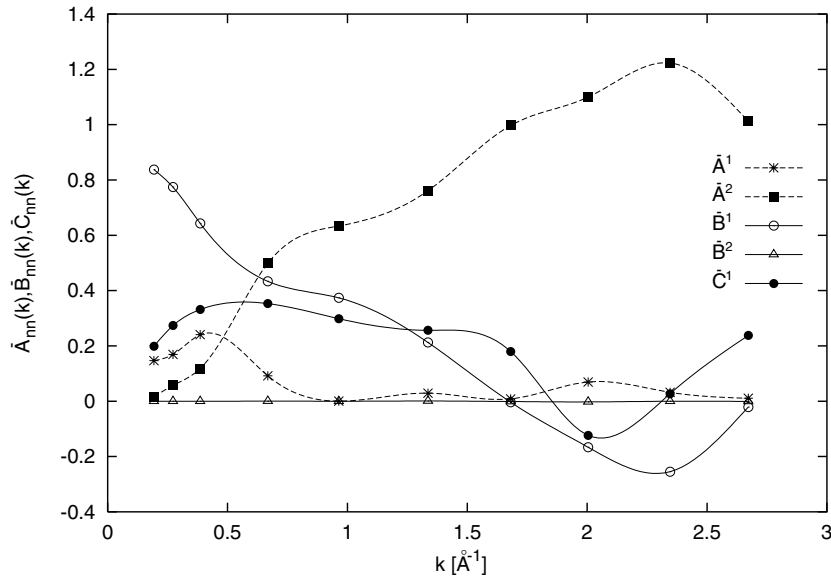
Figure 5. Normalized amplitudes of mode contributions to the 'density–density' time correlation function: relaxing modes d_1 (asterisks) and d_2 (closed boxes), generalized acoustic excitations z_1 (symmetric term: open circles; asymmetric one: closed circles), heat waves z_2 (symmetric amplitudes: open triangles). As a guide to the eye, the normalized amplitudes of relaxing and propagating modes are connected by spline-interpolated dashed and solid lines, respectively.

One can see in figure 5 that for small wavenumbers ($k < 0.5\,\text{Å}^{-1}$) the leading contributions to the function $F_{nn}(k, t)/S(k)$ are connected with the generalized hydrodynamic collective modes. When $k \to 0$, there remain only two finite contributions to the density–density TCF:

(i) the normalized symmetric amplitude $\bar{B}_{nn}^{1}(k)$ of the generalized sound excitations (open circles) tends to the value $1/\gamma$ with $\gamma \approx 1.12$, as estimated in paper I;
(ii) the normalized amplitude $\bar{A}_{nn}^{1}(k)$ of the generalized thermodiffusive mode (asterisks) tends to $1 - 1/\gamma$.

Such a behaviour is in complete agreement with the predictions of standard hydrodynamics [2, 3, 16]. It is also known from the literature [2–4, 16] that the asymmetric contribution $\bar{C}_{nn}^1(k)$ from the generalized sound excitations has to be a linear function of k and vanish when k goes to zero. The k-dependence of $\bar{C}_{nn}^1(k)$ is shown in figure 5 by closed circles, and it is seen that this function has an evident tendency to linear behaviour in the small-k region ($k < 0.3$ Å^{-1}). Note that according to (9) the asymmetric contributions are irrelevant for the zero-frequency moment of the dynamical structure factor $S(k, \omega)$, but they are very important for the correct reproduction of higher-order frequency moments. For small wavenumbers k the contribution $\bar{C}_{nn}^1(k)$ results in asymmetry of the Brillouin peaks.

It is seen in figure 5 that the generalized thermodiffusive mode $d_1(k)$ contributes substantially only when k is small enough. For intermediate and large wavenumbers ($k > 0.6$ Å^{-1}) this mode has no effect on the density fluctuations. Instead, the role of the relaxing kinetic mode $d_2(k)$ increases when k becomes larger, and this mode, together with the generalized sound excitations, in fact determines the shape of the function $F_{nn}(k, t)$ for the intermediate and large k-values considered. So, the approximate formula

$$\bar{A}_{nn}^2(k) \approx 1 - \bar{B}_{nn}^1(k)$$

for the mode contributions may be used herein (see figure 5). In particular, this explains why such a good agreement between the results obtained for the 'coupled' set (1) and the five-variable viscoelastic set (4) was found (see figure 3) in that region. In fact, for $k > 0.6$ Å^{-1} the viscoelastic theory must work fairly well because the coupling with the heat fluctuations is negligible. This conclusion is supported by the numerical results found for the contribution $\bar{B}_{nn}^2(k)$ of the heat waves (triangles in figure 5), which in the cases of solids [17] and liquid He [18] are also called the second-sound excitations, to the function $F_{nn}(k, t)/S(k)$. For the case of liquid Bi this contribution of heat waves is negligible, but for some liquids it may play a significant role by manifesting itself as a fast-sound-like phenomenon [19, 20].

3. Conclusions

Let us summarize the results obtained in this study for the generalized relaxing kinetic mode $d_2(k)$. We found that:

(i) The relaxing behaviour of $F_{nn}(k, t)$ in the region of the main peak of $S(k)$ is mainly determined by a single-mode contribution associated with $d_2(k)$, and this does indeed give the main mechanism of a dramatic slowing down of the decay of the density fluctuations, which results in so-called 'de Gennes narrowing' in the dynamical structure factor [21].

(ii) The kinetic relaxing mode $d_2(k)$ in the region of the main peak of $S(k)$ is determined entirely by viscoelastic processes, and the thermal processes have no effects therein. Moreover, the eigenvalue $d_2(k)$ is well reproduced for $k \sim Q_p$ even within the simplest one-mode approximation (6).

(iii) In the hydrodynamic range the damping coefficient of the kinetic mode $d_2(k)$ tends to a nonzero value, and its contribution to the density–density time correlation function $F_{nn}(k, t)$ becomes negligible (proportional to k^2; see the closed squares in figure 5). However, we point out that the role of the relaxing kinetic mode $d_2(k)$ increases substantially when k becomes larger, and the sharp increasing of the amplitude $\bar{A}_{nn}^2(k)$ as well as the rapid decreasing of $\bar{A}_{nn}^1(k)$ in the range $k \sim 0.6$ Å^{-1} can be considered as manifestations of the dynamic crossover from purely viscous to purely elastic behaviour [2] in liquid Bi.

Our recent results [5, 6], obtained for the liquid metals Cs and Pb, show obviously that beyond the small-k region the relaxing mode $d_2(k)$ is the lowest one and makes the main

contribution to the shape of the density–density TCF for intermediate and large wavenumbers. This allows us to assume that for highly viscous fluids the contribution from the generalized relaxing kinetic mode $d_2(k)$ must be observed in the dynamical structure factor even for rather small wavenumbers. This assumption needs further investigation and will be the subject of our next study, but we would like to stress that slightly beyond the hydrodynamic region the contribution from the relaxing mode $d_2(k)$ to the dynamical structure factor gives in fact a Mountain-like peak caused by the structural relaxation effects [2], which are typical for highly viscous fluids. Taking into account all of the facts collected above, we conclude that the relaxing kinetic mode $d_2(k)$ is connected with the decay of an instantaneous cage of neighbours and describes the main features of structural relaxation in a liquid. Note that in the one-variable treatment, $1/d_2(Q_p) \sim \tau_{nn}(Q_p)$ has the meaning of a particle's lifetime in the cage of nearest neighbours. Therefore, one can identify the kinetic eigenvalue $d_2(k)$ as the *generalized structural relaxation* mode. We note also that the results obtained allow us to state that the main distinction as regards the collective dynamics between semimetallic liquid Bi and the previously studied liquid metals [2, 16], with well-defined sound excitations visible in the dynamical structure factor up to $k \sim 0.7Q_p$, is not the strong damping of sound modes (as was concluded, e.g., in [22]) but rather their small relative contribution in comparison with that from the generalized structural relaxation mode $d_2(k)$. In particular, this conclusion is supported by our results found for the collective mode spectrum (see figure 5 in paper I) as well as our calculations of the normalized amplitudes for separate mode contributions, shown in figure 5 of the present paper. For instance, at $k \simeq 0.58$ Å^{-1} one gets $\sigma_1/\omega_1 \simeq 0.3$, but $\bar{A}_{nn}^2/\bar{B}_{nn}^1 \simeq 1.1$ and $d_2/\sigma_1 \simeq 1.4$. This means that at $k \geqslant 0.58$ Å^{-1} the sound modes are still well defined ($\sigma_1/\omega_1 \simeq 0.3 < 1$); however, they could be visible in the dynamical structure factor only as *shoulders* and not as well-separated side peaks.

In conclusion, the main results of this study are the following:

(i) Two relaxing collective modes $d_1(k)$ and $d_2(k)$, in addition to the propagating excitations, make the main contributions to the shape of the 'density–density' TCFs of liquid Bi. As was expected from the standard hydrodynamic treatment, for small wavenumbers k the contribution from the thermodiffusive mode $d_1(k)$ is dominant. However, for larger k ($k > 0.6$ Å^{-1}) another relaxing mode, $d_2(k)$, mainly determines the relaxing properties of the density–density TCF $F_{nn}(k, t)$. In this region the mode $d_2(k)$ has the smallest damping coefficient and contributes to $F_{nn}(k, t)$ with the largest amplitude.

(ii) The relaxing kinetic mode $d_2(k)$, which is irrelevant in the hydrodynamic region, makes the most significant contribution to the zero-order frequency moment of the dynamical structure factor in the range where the main peak of the static structure factor $S(k)$ is located. A relatively small (six times smaller in magnitude) and negative contribution of generalized sound excitations is also found in this range of k.

(iii) The kinetic relaxing collective mode $d_2(k)$ is closely connected by its origin with the structural relaxation in a liquid. In the region of the main peak of $S(k)$ the eigenvalue $d_2(k)$ can be used as an estimate of the average inverse lifetime of a particle in a cage of nearest neighbours. For intermediate values of k ($k \sim 0.6$ Å^{-1}), this mode causes the appearance of a Mountain-like peak in the dynamical structure factor.

(iv) The method used for the study of dynamical properties of liquid Bi allows us without any additional assumptions or adjustable parameters to describe correctly the crossover from purely hydrodynamic (or purely viscous) behaviour to the purely elastic response of the system and to explain the specific features of de Gennes narrowing at $k \sim Q_p$, where the main maximum of $S(k)$ is located.

As regards the future outlook, we might mention that the present work provides an example of how within a rigorous statistical treatment the problem of collective excitations in liquids can be understood on a microscopic level. Therefore we are interested in applying the method developed here in investigations of other liquids, the collective dynamics of which cannot be solely understood in terms of the hydrodynamic modes (see, e.g., [23]). A more specific problem is connected with the study of highly viscous and supercooled liquids, where one can expect to observe even for small wavenumbers a significant contribution from the generalized structural relaxation mode to the dynamical structure factor.

Acknowledgments

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