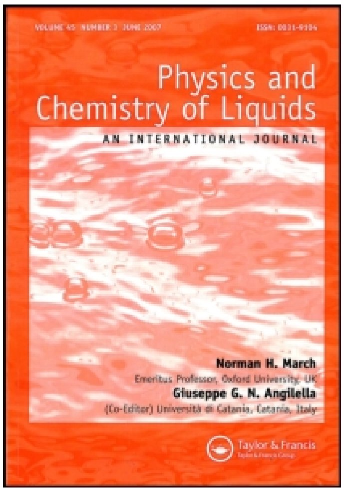

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# Dynamic Density Fluctuations in Molten Salts: Diagonalized-Relaxation Theory for Liquid RbBr

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# Dynamic Density Fluctuations in Molten Salts: Diagonalized-Relaxation Theory for Liquid RbBr†

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Autocorrelation spectra for ion-density fluctuations in molten RbBr are evaluated over a wide range of wave number (0.3 Å$^{-1}$ $\leqslant q \leqslant$ 7 Å$^{-1}$) in the microscopic region of wave number and frequency, using a memory-function theory which neglects thermal fluctuation effects. The results are critically compared with data obtained by the Argonne group through neutron inelastic scattering experiments and computer simulation. Agreement is good in an intermediate range of wave number (1 Å$^{-1}$ $\lesssim q \lesssim$ 4 Å$^{-1}$) but progressively worsens outside this range. In particular, the need for a more complete generalized-hydrodynamic theory at long wavelengths, inclusive of couplings to entropy fluctuations, is underlined.

## 1 INTRODUCTION

In the framework of a systematic investigation of the dynamical behaviour of simple liquids by the combined tools of neutron inelastic scattering and computer simulation, the Argonne group has recently reported[1,2] a detailed study of molten RbBr, as a first example of a molten salt. The aim of this paper is to assess the present status of the memory function theory[3,4] for the dynamic density fluctuations in these systems, as developed in a previous paper,[5] by comparison with these data.

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Autocorrelations of two density-fluctuation variables, as well as their cross correlations, are of interest in a two-component molten salt. As previously discussed,[5] a proper choice of the fluctuation variables greatly simplifies the description of the system. A hydrodynamic analysis[6,7] shows that the mass density and the charge density are independent fluctuation variables in the extreme hydrodynamic region of wave number and frequency, where their dynamics reflects itself in a Brillouin–Rayleigh spectrum and in a central peak due to a charge-current relaxation mode, respectively. These hydrodynamic modes become coupled with increasing wave number both directly through electrostrictive effects and indirectly through thermoelectric and thermoelastic effects.

At frequencies much larger than those relevant to hydrodynamics, computer simulations on rigid-ion fluids[2,8,9] show a prominent finite-frequency peak in the charge fluctuation spectrum. These modes are of course the remnants in the liquid of the longitudinal optical modes in the solid, as predicted by high-frequency theories,[10,11] and to establish their presence in the real ionic fluid is a problem of the greatest interest. Unfortunately in the case of molten RbBr no information on "optic" modes can be obtained from the neutron scattering experiment of Price and Copley,[1] owing to the close similarity of the scattering amplitudes of the two component ions. Alternative experimental information on optic modes, limited to wave numbers very near to zero, can be obtained from the infrared reflectivity spectrum,[12] but these data seem to be lacking for molten RbBr.

In the microscopic region of wave number and frequency of present interest, one would expect that the mass and charge fluctuation variables are approximately independent at "long" wavelengths (that is, at wavelengths of the order of a few mean interionic distances), especially in a fluid of nearly equi-sized ions such as RbBr and provided that indirect couplings via thermal fluctuations are small. At shorter wavelengths, instead, the mass and charge fluctuations strongly interact and mix, and suitable linear combinations which maintain approximate diagonalization of the matrix of correlation spectra are more convenient[5] for the extension of the memory-function theory to the two-component fluid. Inversion of this linear transformation is then needed for contact with the observed spectra. In the following, we present a detailed quantitative discussion of the calculated neutron spectrum and the (practically coincident) number-density spectrum of molten RbBr. More weight should be given to the simulation data for a test of the theory, since both are based on a rigid-ion model which neglects electronic polarization effects present in the real fluid. This model Hamiltonian tends to overemphasize structure in the wavelength dependence of spectral properties[2] and to overestimate the frequency of spectral peaks.[13] A comparison of the calculated spectrum of long-wavelength charge fluctuations with the computer simulation results is also presented and underlines the need for improvements in the theoretical description of the damping mechanisms.

## 2 THEORY

In this section we present a brief summary of the theory developed in Ref. 5 (hereafter referred to as I) and reconsider the estimation of the relaxation times, with a view to extending its applicability to longer wavelengths (wave number $q \lesssim 1$ Å$^{-1}$).

As discussed in detail in I, simplification of the structure of the density response functions for a two-component fluid in a Mori continued-fraction expansion[3] and development of reliable approximations hinge on a suitable choice of the dynamic density fluctuation variables. These are taken as linear combinations of the number density fluctuations of the two components, $n_+(\mathbf{q}, t)$ and $n_-(\mathbf{q}, t)$, of the form

$$A_1(\mathbf{q}, t) = m_+^{1/2} \sin \vartheta_q n_+(\mathbf{q}, t) + m_-^{1/2} \cos \vartheta_q n_-(\mathbf{q}, t) \tag{1}$$

and

$$A_2(\mathbf{q}, t) = m_+^{1/2} \cos \vartheta_q n_+(\mathbf{q}, t) - m_-^{1/2} \sin \vartheta_q n_-(\mathbf{q}, t) \tag{2}$$

where $m_+$ and $m_-$ are the ionic masses, with the requirements that these should reduce to the mass and charge density fluctuations at long wavelengths and to independent density fluctuations of the two components at short wavelengths. These limiting behaviors are automatically satisfied by determining the mixing angle $\vartheta_q$ as a function of wave number through the condition

$$\omega_{12}^{\infty}(q) = \omega_{12}^{0}(q) \tag{3}$$

where $\omega_{12}^{0}(q)$ and $\omega_{12}^{\infty}(q)$, as defined in Eqs. (9) and (10) below, are related to the zeroth and fourth moments of the van Hove functions for the dynamical variables $A_\alpha(\mathbf{q}, t)$,

$$S_{\alpha\beta}(q, \omega) = \frac{1}{4n} \int_{-\infty}^{\infty} \frac{dt}{2\pi} \exp(-i\omega t) < A_\alpha(-\mathbf{q}, 0) A_\beta(\mathbf{q}, t) + A_\beta(-\mathbf{q}, 0) A_\alpha(\mathbf{q}, t) \rangle. \tag{4}$$

In practice the condition (3), which in the lattice vibration problem would be equivalent to the diagonalization of the dynamical matrix for longitudinal modes, very nearly diagonalizes the matrix of van Hove functions (4), i.e. yields

$$\left| \frac{S_{12}(q, \omega)}{S_{\alpha\alpha}(q, \omega)} \right| \ll 1 \tag{5}$$

for all frequencies and wave numbers of interest.

The approximation developed in I for the evaluation of the correlation functions (4) involves the replacement of the matrix of second-order memory functions by a matrix of wavenumber-dependent inverse relaxation times, thus allowing an exact account of the zeroth, second and fourth spectral moments as in previous work[14] on liquid argon. This matrix is required to be diagonal for the particular choice of dynamical variables indicated above, and the final expressions for the van Hove functions (4) are

$$S_{\alpha\alpha}(q,\omega) = \frac{nk_B Tq^2}{2\pi\omega}\,\mathrm{Im}\left[\frac{\omega^2 - i\omega\Gamma^{(1)}_{\bar{\alpha}\bar{\alpha}}(q,\omega) - \omega^0_{\bar{\alpha}\bar{\alpha}}(q)}{D(q,\omega)}\right] \tag{6}$$

and

$$S_{12}(q,\omega) = -\frac{nk_B Tq^2}{2\pi\omega}\,\omega^0_{12}(q)\mathrm{Im}[D^{-1}(q,\omega)]. \tag{7}$$

Here, $\bar{\alpha}$ denotes the component different from $\alpha$, and $\Gamma^{(1)}_{\alpha\alpha}(q,\omega)$ are the first-order memory functions,

$$\Gamma^{(1)}_{\alpha\alpha}(q,\omega) = \frac{\omega^\infty_{\alpha\alpha}(q) - \omega^0_{\alpha\alpha}(q)}{i\omega + \tau_\alpha^{-1}(q)}, \tag{8}$$

with

$$\omega^0_{\alpha\beta}(q) = (-1)^{\alpha+\beta} nk_B Tq^2 S_{\bar{\beta}\bar{\alpha}}(q)[S_{11}(q)S_{22}(q) - S^2_{12}(q)]^{-1} \tag{9}$$

and

$$\omega^\infty_{\alpha\beta}(q) = (nk_B Tq^2)^{-1}\int_{-\infty}^{\infty} d\omega\,\omega^4 S_{\alpha\beta}(q,\omega). \tag{10}$$

Furthermore, $S_{\alpha\beta}(q)$ are the static structure factors for the dynamical variables $A_\alpha(\mathbf{q},t)$, $\tau_\alpha(q)$ are the relaxation times, and

$$D(q,\omega) = [\omega^2 - i\omega\Gamma^{(1)}_{11}(q,\omega) - \omega^0_{11}(q)][\omega^2 - i\omega\Gamma^{(1)}_{22}(q,\omega) - \omega^0_{22}(q)] - [\omega^0_{12}(q)]^2. \tag{11}$$

Clearly, contact with measured spectral functions is then achieved by inverting the linear transformation (1) and (2). In particular, we shall consider in the subsequent discussion the correlation spectra for the total number density fluctuations

$$S_{NN}(q,\omega) = (2n)^{-1}\int_{-\infty}^{\infty}\frac{dt}{2\pi}\exp(-i\omega t)\sum_{i,j=1}^{2}\langle n_i(-\mathbf{q},0)n_j(\mathbf{q},t)\rangle \tag{12}$$

and for the charge density fluctuations

$$S_{QQ}(q,\omega) = (2n)^{-1}\int_{-\infty}^{\infty}\frac{dt}{2\pi}\exp(-i\omega t)\sum_{i,j=1}^{2}(-1)^{i+j}\langle n_i(-\mathbf{q},0)n_j(\mathbf{q},t)\rangle, \tag{13}$$

as well as the symmetrized neutron scattering spectrum

$$\tilde{S}(q,\omega) = (2n)^{-1}(a_+ + a_-)^{-2}\exp\left(\frac{\hbar\omega}{2k_BT}\right) \times \int_{-\infty}^{\infty}\frac{dt}{2\pi}\exp(-i\omega t)\sum_{i,j=1}^{2} a_i a_j\langle n_i(-\mathbf{q},0)n_j(\mathbf{q},t)\rangle. \tag{14}$$

where $a_+$ and $a_-$ are the scattering lengths. The expressions of these spectra as linear combinations of the spectra (6) and (7) are easily obtained through the use of Eqs. (1) and (2).

The choice of the relaxation times entering Eq. (8) remains to be discussed. Previous work on liquid argon[14,15] provides some guidance in this delicate question, by suggesting that they should be connected with the elastic scattering functions $S_{\alpha\beta}(q, 0)$ and with the transport coefficients. The former requirement is useful at short wavelengths, where the elastic scattering functions can be evaluated by an independent particle model to yield

$$\tau_\alpha^{-1}(q) = 2\pi^{-1/2}[\omega_{\alpha\alpha}^{\infty}(q) - \omega_{\alpha\alpha}^{0}(q)]^{1/2} \qquad (qa \gg 1) \tag{15}$$

To establish a connexion with transport coefficients at long wavelength, we first recall that the $A_\alpha(\mathbf{q}, t)$ reduce in this limit to the mass density fluctuations $M(\mathbf{q}, t)$ and to the charge density fluctuations $Q(\mathbf{q}, t)$, aside from trivial factors. Using the long-wavelength forms of the corresponding static structure factors,[7] it is indeed easily seen that $D(q, \omega)$ in Eq. (11) factorizes for $q \to 0$, so that $S_{11}(q, \omega)$ and $S_{22}(q, \omega)$ describe in this limit two independent modes of collective mass and charge density fluctuations. The relaxation times can then be related through Kubo relations[7] to the corresponding transport coefficients, which are the sound-wave viscosity, $\frac{4}{3}\eta + \zeta$, and the electric conductivity, $\sigma$. Precisely, we find

$$\tau_1^{-1}(q) = n(m_+ + m_-)(\tfrac{4}{3}\eta + \zeta)^{-1}\frac{\omega_{11}^{\infty}(q) - \omega_{11}^{0}(q)}{q^2} \qquad (qa \ll 1) \tag{16}$$

and

$$\tau_2^{-1}(q) = \frac{m_+ m_- \sigma}{ne^2(m_+ + m_-)}[\omega_{22}^{\infty}(q) - \omega_{22}^{0}(q)] \qquad (qa \ll 1) \tag{17}$$

It should be noted for later reference that this connexion with hydrodynamic transport explicitly uses the inequality $\omega_{\alpha\alpha}^{\infty}(q) > \omega_{\alpha\alpha}^{0}(q)$.

In the numerical calculations reported below, we have used a simple interpolation between the expressions (15)–(17) for the inverse relaxation times, of the form[15]

$$\tau_\alpha^{-1}(q) = \tau_{\alpha\infty}^{-1}(q) + \left(1 + \frac{q^2}{q_0^2}\right)^{-1}[\tau_{\alpha 0}^{-1}(q) - \tau_{\alpha\infty}^{-1}(q)] \tag{18}$$

where $q_0$ is a parameter that we expect to be of the order of 1 Å$^{-1}$. In fact, the results do not depend very sensitively on the precise value of this parameter.

## 3 NUMERICAL RESULTS

The numerical values for the transport coefficients of molten RbBr near its freezing point were taken from Janz[16] and are $\eta = 1.38 \times 10^{-2}$ gr/(cm sec), $\zeta = 13.2 \times 10^{-2}$ gr/(cm sec) and $\sigma = 1.17$ (ohm cm)$^{-1}$. The other quantities needed for the calculation are the instantaneous structure factors $S_{\alpha\beta}(q)$ and the fourth moments $\omega^{\infty}_{\alpha\beta}(q)$, which obviously are linear combinations of the corresponding quantities for the density fluctuations for the two kinds of ions. The latter were provided by Copley and Rahman[2] from the results of their computer simulation study. These data need to be known with rather high accuracy for wave numbers $q \lesssim 1$ Å$^{-1}$, where the structure factors are very small and the fourth moments are varying rapidly, in order to obtain meaningful values for the quantities $\omega^{0}_{\alpha\beta}(q)$ and $\omega^{\infty}_{\alpha\beta}(q)$. In this range of wave number, it was found that the structure factors obtained from the simulation of the pair structure of the liquid were not sufficiently accurate to yield meaningful values of $\omega^{0}_{\alpha\beta}(q)$, and recourse had to be made to the values of the structure factors obtained from the simulation of the intermediate scattering functions in the zero-time limit. This, in turn, made it necessary to use interpolated values for the fourth moments. In particular, the difference $\omega^{\infty}_{22}(q) - \omega^{0}_{22}(q)$ is believed not to be known with sufficient quantitative accuracy, although it is quite clear from the numerical data that this quantity is small for $q \lesssim 0.8$ A$^{-1}$. If we take it to be zero, it is easily seen that the theory will yield a delta-function peak in the charge fluctuation spectrum. This behavior for RbBr is at variance from that reported[8] by simulation of other ionic fluids.

The main features of the calculated spectra over a wide range of wave number are compared in Figures 1–3 with the neutron scattering data of Price and Copley[1] and with the computer simulation data of Copley and Rahman.[2] These features are the elastic scattering function $S_{NN}(q, 0)$ (Figure 1), the full width at half maximum of $S_{NN}(q, \omega)$ (Figure 2), and the peak frequencies in the spectral functions for number-density currents, $\omega^2 S_{NN}(q, \omega)$ and for charge-density currents, $\omega^2 S_{QQ}(q, \omega)$ (Figure 3). The overall agreement with both sets of data is good, and it is particularly rewarding that the theory reproduces rather closely the various structures that are present in the computer simulation results. As discussed by Copley and Rahman,[2] less structure is present in the neutron scattering data, a likely reason for this discrepancy being the presence of electronic polarization effects in

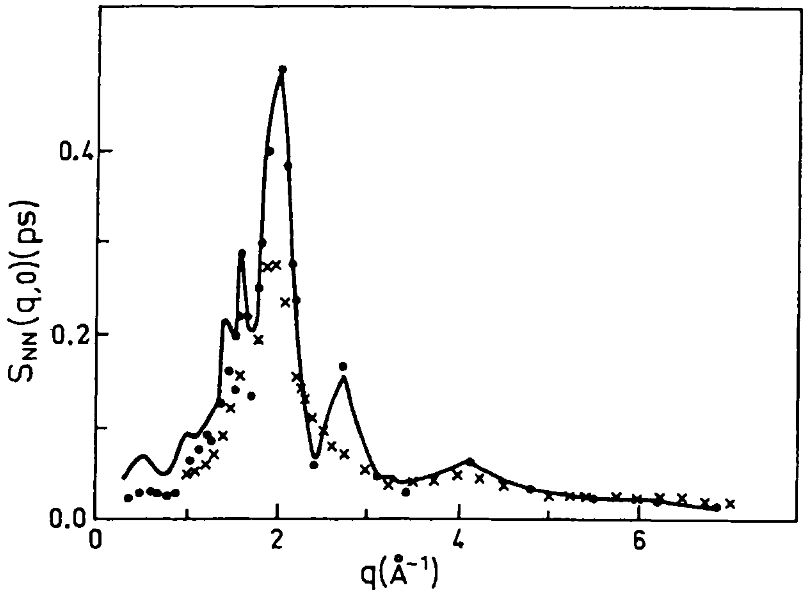

FIGURE 1 The elastic scattering function $S_{NN}(q, 0)$ as a function of wave number. Full curve: present theory, with $q_0 = 1.5\ \mathring{A}^{-1}$; dots, simulation results of Copley and Rahman;[2] crosses, neutron scattering results of Price and Copley.[1]

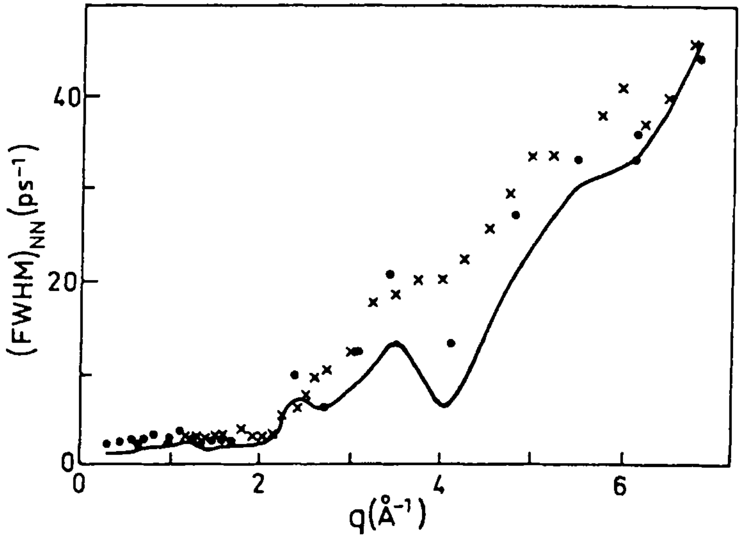

FIGURE 2 Full width at half maximum of the spectral function $S_{NN}(q, \omega)$ as a function of wave number. Symbols are as in Figure 1.

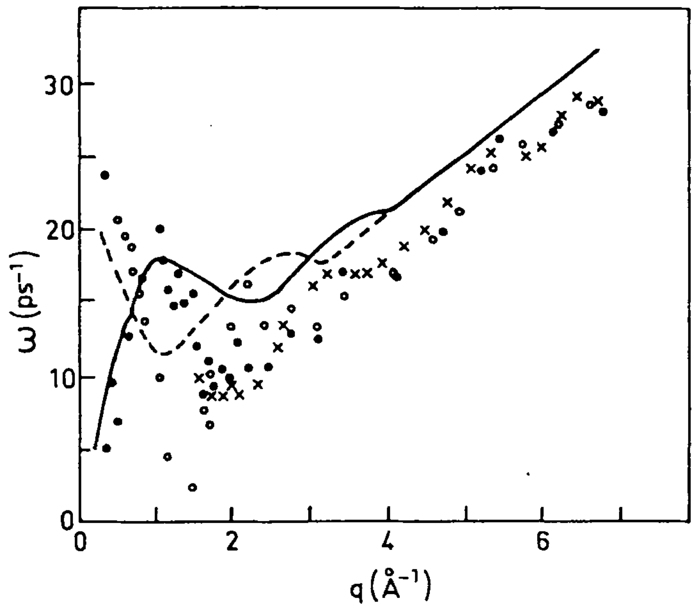

FIGURE 3 Peak positions in the current autocorrelation spectra for number-density $(\omega^2 S_{NN}(q, \omega))$ and charge-density $(\omega^2 S_{QQ}(q, \omega))$ fluctuations, as functions of wave number. Number-density peak: full curve, present theory with $q_0 = 1.5$ Å$^{-1}$; dots, simulation results;[2] crosses, neutron scattering results.[1] Charge-density peak: broken curve, present theory; circles, simulation results.[2]

the real fluid. A closer examination of Figures 1 and 2 shows that the theory tends to overestimate the height of the elastic scattering function at long wavelengths and to underestimate the width of the total-density fluctuation spectrum. The theoretical "dispersion curves" of Figure 3 have the same qualitative behaviours as the data, although the amplitudes of their oscillations with $q$ are less pronounced. Such discrepancies of detail also affect viscoelastic theories of the much simpler argon liquid.

Figures 4–6 present detailed illustrations of the spectral shapes. In Figures 4 and 5, the symmetrized neutron scattering function $\tilde{S}(q, \omega)$ is compared with the experimental neutron scattering spectra reported by Price and Copley[1] over the range of wave number 0.8 Å$^{-1}$ $\leq q \leq$ 7 Å$^{-1}$. The overall agreement is again quite reasonable, also on account of the discrepancies between experiment on one side, and simulation and theory on the other, for the elastic scattering function in Figure 1. A distortion of the spectral shape appears in the theoretical results for $q \gtrsim 4$ Å$^{-1}$, which is seen in Figure 5 to worsen with increasing wave number. At these wave numbers the scattering function should already be approaching the free-particle limit (as also indicated by the results reported in Figure 3) but the present

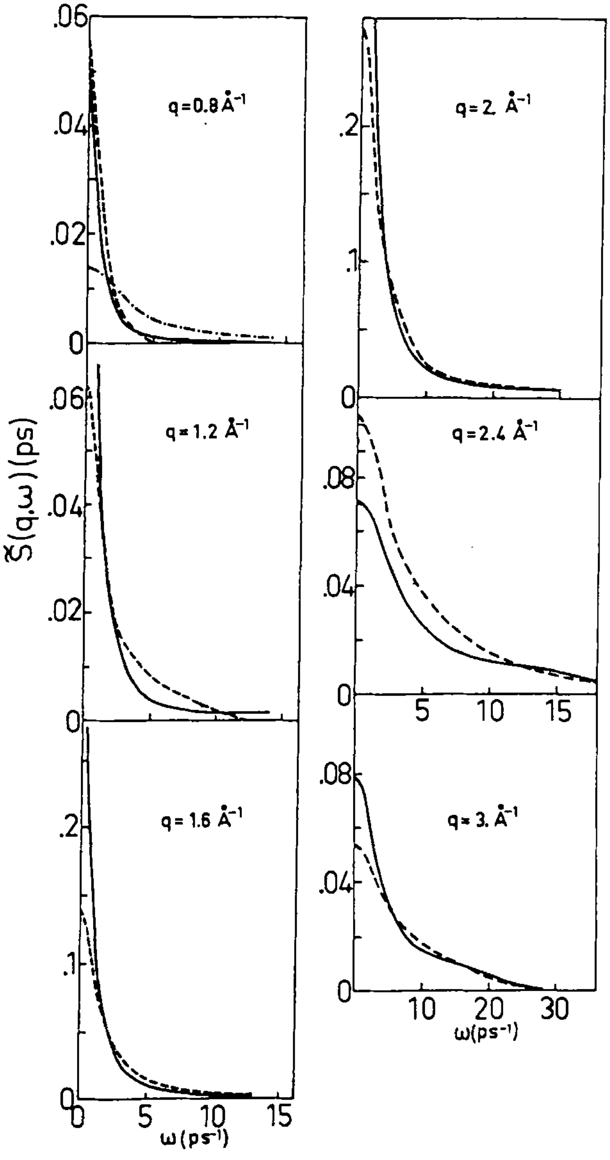

FIGURE 4 The symmetrized neutron scattering spectrum $\tilde{S}(q, \omega)$ for a set of values of the wave number. Full curves: present theory, with $q_0 = 1.5$ Å$^{-1}$; broken curves: neutron scattering data.[1] The dash-dot curve for $q = 0.8$ Å$^{-1}$ reports the theoretical results for the case $q_0 = 0$.

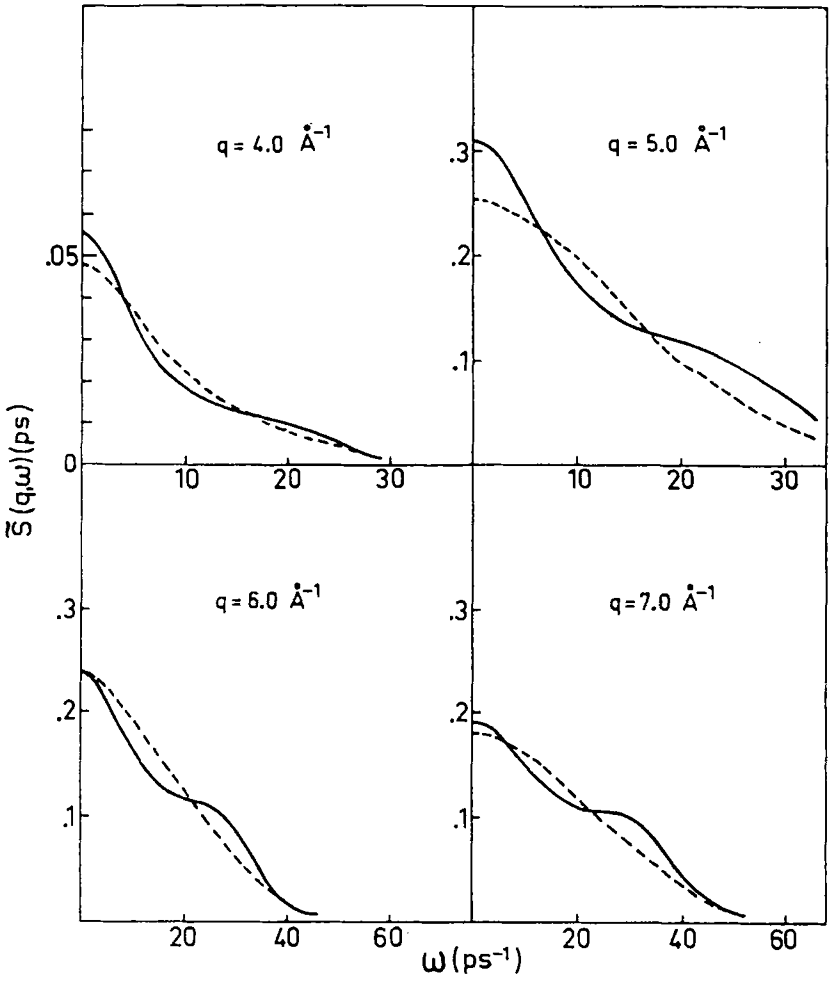

FIGURE 5 Same as Figure 4, for other values of the wave number.

simple form for the memory function in a truncated continued-fraction expansion does not allow for the correct Gaussian shape of the spectrum in this limit. Similar spectral distortions have been reported for magnetic systems[17] and can be mended by a more refined choice of the memory function.

Comparison with the neutron data in the particularly interesting range of long wavelengths ($q \lesssim 1$ Å$^{-1}$) is possible only at $q = 0.8$ A$^{-1}$, and is reported in Figure 4. A firm indication which emerges from the calculation is that contact with the hydrodynamic behavior of the fluid is already

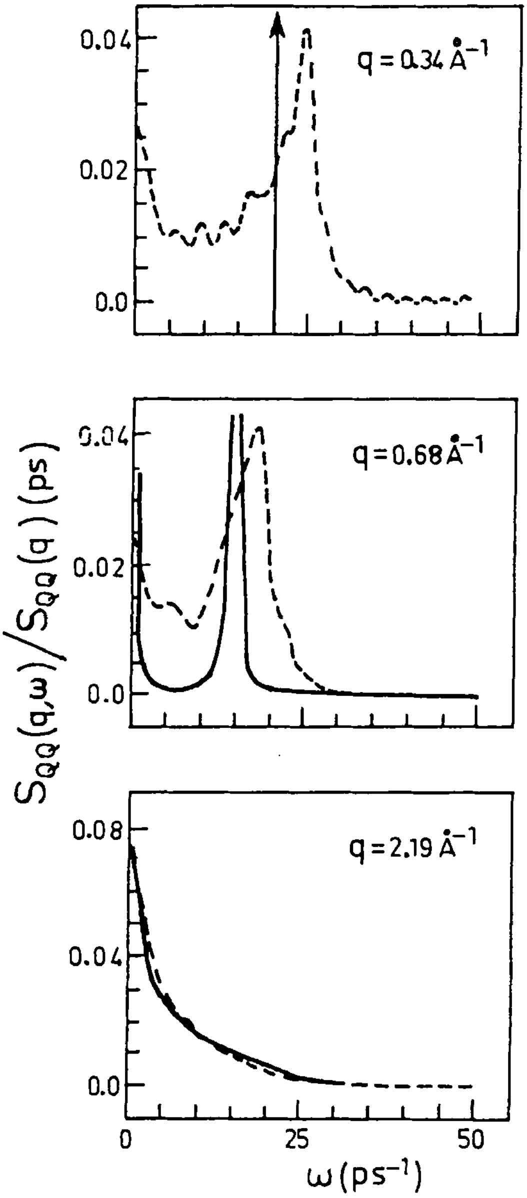

FIGURE 6 Spectral function $S_{QQ}(q, \omega)$ for charge density fluctuations, for three values of the wave number. Full curves, present theory with $q_0 = 1.5$ Å$^{-1}$; broken curves, computer simulation results.[2] For $q = 0.34$ Å$^{-1}$, the theoretical spectrum consists of a very narrow peak at $\omega = 19.5$ ps$^{-1}$ (shown by the arrow) and a very narrow peak at $\omega = 0$ (not shown). For $q = 0.68$ Å$^{-1}$, the data yield a slightly negative value for $\omega_{22}^{\infty} - \omega_{22}^{0}$, even though this quantity should be positive by definition:[3] the theoretical curve reported (with $\tau_{2\alpha}^{-1} = 0$) should thus be in effect equivalent to a delta function.

important at this wave number, as illustrated by reporting the numerical results obtained with the values $q_0 = 1.5$ Å$^{-1}$ (full curve) and $q_0 = 0$ (dash-dot curve) for the parameter entering the expression (18) of the relaxation times. The latter choice omits all contact with hydrodynamics and yields significantly poorer results at this wave number, while even at $q = 1.2$ Å$^{-1}$ the discrepancies between these two choices for $q_0$ are much smaller and rapidly disappear at larger wave number. The comparison with the experimental curve at $q = 0.8$ Å$^{-1}$ (and also at 1.2 Å$^{-1}$) is not significant in the high frequency tail, where experimental data are lacking and one has only a fitted curve which falls off too rapidly, as signalled[1] by large violations of the second-moment sum rule.

Finally, in Figure 6 we report a comparison of the spectral function for the charge density fluctuations with the computer simulation data[2] at three values of the wave number. Agreement is only qualitative at long wavelengths, where the theory badly underestimates the width of the charge fluctuation peak, as a consequence of the very small values attained by $\omega^{\infty}_{22}(q) - \omega^{0}_{22}(q)$. It would be useful to have spectral data on mass fluctuations in the same range of wave number.

## 4 CONCLUDING REMARKS

While the overall agreement of the theory with the available data on molten RbBr is felt to be good, some discrepancies have been underlined by our detailed comparisons. We briefly summarize them here with a view to future work on molten salt systems.

The discrepancies that emerge at large wave number, $q \gtrsim 4$ Å$^{-1}$, are in a sense trivial as they likely arise from the well known difficulty of describing a nearly free particle regime with a generalized hydrodynamic theory. Of greater interest is a more accurate theory for the low wave number range ($q \lesssim 1$ Å$^{-1}$) where generalized-hydrodynamic effects are seen to play a relevant role. In particular, the theory underestimates the damping of the charge fluctuations, and inclusion of thermal effects should therefore be important in this range in providing both a direct coupling between charge and entropy fluctuations and an indirect coupling between charge and mass fluctuations. We should stress here again the need for very accurate structural and moments data in this range.

The other major step forward that seems needed for a realistic description of the dynamics of real molten salts is the inclusion of electronic polarization effects. Present evidence suggests that these play a role in determining detailed features of the total-density fluctuation spectrum and an important role in the charge fluctuation spectrum, which is especially prominent at long wavelengths.

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