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DYNAMICAL STUDY OF LIQUID ALUMINIUM *

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ABSTRACT

Recent molecular dynamics data of Ebbsj8 et al. in liquid aluminium have been analysed through the memory function formalism. Two forms of the memory functions which have correct asymptotic limit at large wavenumbers but accounts for interatomic correlations in a different manner are considered. The results for $\omega^2 s(q, \omega)$ obtained from both models are compared with experimental data.

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I. INTRODUCTION

The continued fraction expansion of Mori (1965) have played an important role in the study of dynamical correlations in condensed systems. The correlation function is expressed in terms of relaxation kernels, also called memory function, whose form is not known a priori. An exact first principle calculation of the memory function is rather difficult, although several attempts are made in this direction. Recently with a mode coupling ansatz Munakata and Igarashi (1977, 1978) and Götze and co-workers (Götze and Lücke 1975, Bosse et al. 1978a, 1978b) have tried to calculate the memory function through the projection operator technique. Attempts have also been made to obtain it from the kinetic equation (Sjögren and Sjölander 1976, Sjögren 1978) of motion for phase-space distribution function. The above calculations, however, become quite tedious and need a self-consistent solution for the correlation functions. This involves a considerable amount of computer time for numerical evaluations. The simple method is to assume a functional form for the memory function which has given good results in liquid argon and rubidium (Kahol et al. 1976, Copley and Lovesey 1975).

In the past many authors made theoretical attempts to approximate memory function through a generalized hydrodynamic approach (Akcasu et al. 1970, Chung and Yip 1969) or assumed a functional form for it. Another approach is to start from the free-particle system and then build interatomic correlations necessary at intermediate wavenumbers. In this paper we use the latter approach and consider two memory functions which have correct free-particle limits at large wavevectors but incorporate the effect of particle interactions in a different fashion. In the first case we start with a memory function corresponding to a non-interacting gas and then introduce the effect of collisional damping through a renormalization parameter. In the second case, the memory function is expressed in terms of its kinetic part and an interaction part which takes into account the polarization process due to the mutual interaction between particles. From the mean-field approach, the former is equivalent to the Pathak-Singwi (PS) (1970) model, i.e. it gives a density response function whose polarization potential depends only on wavevector but the screened response is of renormalized free-particle one. The latter gives rise to a frequency and wavevector dependent polarization potential and a free-particle response function.

The paper is organized as follows. In Sec.II we first briefly give the basic definitions and then discuss models for memory function in Secs.IIA and IIB, respectively. Numerical results and their discussion is given in Sec.III.

II. FORMULATION

Let us consider a liquid metal composed of N ions of mass m to be perturbed by a weak external probe. The density fluctuation and the longitudinal current fluctuation are given by

$$\rho(q, t) = \sum_{i=1}^N e^{iq \cdot r_i(t)} \quad (1)$$

$$J_L(q, t) = \sum_{i=1}^N v_i^z(t) e^{iq \cdot r_i(t)}$$

where $r_i(t)$ and $v_i^z(t)$ are, respectively, the position and the velocity component of the i th ion along the direction of q . The density and the longitudinal current fluctuations are connected by a continuity equation which relates the longitudinal current correlation function $c(q, \omega)$ to the dynamic structure factor $s(q, \omega)$ as

$$c(q, \omega) = \frac{\omega^2}{q} s(q, \omega) \quad (2)$$

The Laplace transform of the density correlation function $G(q, z)$, whose real part is denoted as $s(q, \omega)$, is defined in terms of the density response function $\chi(q, z)$ via the relation

$$\chi(q, z) = \frac{n}{k_B T} [s(q) - z G(q, z)] \quad (3)$$

where $s(q)$ is the static structure factor, n mean ionic density, and k_B is the Boltzmann constant. In Mori's formalism $G(q, z)$ is expressed as a continued fraction expansion:

$$G(q, z) = \frac{s(q)}{z + M_1(q, z)} ; M_l(q, z) = \frac{\delta_l}{z + M_{l+1}(q, z)} \quad (4)$$

where $l = 1, 2, \dots$ and the $\delta_l = M_l(q, t=0)$ are related to the frequency moments of $S(q, \omega)$. $M_l(q, z)$ is called the l th order memory function and is the only unknown function. In the following we shall discuss two models to approximate this function.

A. Renormalized memory function

In the limit of large wavevectors $q > 6 \text{ \AA}^{-1}$, the collision time between successive collisions is much larger than the relaxation time of the density fluctuation produced by an external probe. In such a short interval of time the ions can be considered to be free, and $M_2(q, z)$ can be obtained exactly by taking the free-particle limit of the expression (4). Thus, in the free-particle limit, we get the second-order memory function as

$$M_{2f}(q, z) = [z - (z^2 + \omega_0^2)G_f(q, z)]/[z G_f(q, z) - 1] \quad (5)$$

where $\omega_0^2 = \frac{q^2 k_B T}{m}$ and

$$G_f(q, z) = -i(2\pi\omega_0^2)^{1/2} \int_{-\infty}^{\infty} dt e^{-t^2} / [t - (iz/\sqrt{2}\omega_0^2)] \quad (6)$$

However, in the region $0.5 < q < 6 \text{ \AA}^{-1}$, the collision time is of the same order as the relaxation time and has the physical effect of broadening the line shape of $S(q, \omega)$. Mathematically, we incorporate this effect by introducing a renormalization parameter $\gamma(q)$ in the energy width of $G(q, z)$. This leads to the following form for the memory function:

$$M_2(q, z) = [z^2 - (z^2 + \tilde{\omega}_0^2)\tilde{G}(q, z)]/[z \tilde{G}(q, z) - 1] \quad (7)$$

where $\tilde{\omega}_0^2 = [2q^2 k_B T + m\gamma(q)]/2m$, and $\tilde{G}(q, z)$ is the same as (6) except that ω_0^2 is now replaced by $\tilde{\omega}_0^2$. With the above choice of memory function, theory satisfies exactly the zeroth and second moment sum rules. The fourth moment can be satisfied by fixing $\gamma(q)$ through the relation

$$\gamma(q) = \frac{\langle \omega^4 \rangle}{\langle \omega^2 \rangle} - \langle \omega^2 \rangle [2 + 1/S(q)] \quad (8)$$

Here and in the following ω^{2n} ($n = 1, 2, \dots$) are defined as the frequency moments of $S(q, \omega)$. It can be seen that in the limit of large wavenumbers $\gamma(q)$ vanishes and Eq. (7) takes the form of (5).

It may be mentioned here that Eqs. (7), (4) and (3) give the same expression for the density response function as obtained by PS from the generalized random-phase approximation

$$\chi(q, z) = \frac{\chi_{sc}(q, z)}{1 - \psi(q) \chi_{sc}(q, z)} \quad (9)$$

with $\chi_{sc}(q, z)$ taken as the response of renormalized free-particles and the parameters $\psi(q)$ and $\gamma(q)$ determined from the zeroth and fourth frequency moments.

B. The model of Yoshida et al.

In order to understand collective excitations in liquids through Mori's formalism Yoshida (1977a, 1977b) divided the i^{th} -order memory function in terms of its free part and an interaction part as

$$M_i(q, t) = M_{if}(q, t) + M_{iI}(q, t) \quad (10)$$

where $M_{if}(q, t)$ is a kinetic part obtained by summing all the contributions associated with a free-particle system and $M_{iI}(q, t)$ is the remaining part which takes into account the effect of interaction between particles including the collective aspects. The kinetic part of the second-order memory function, $M_{2f}(q, z)$, is given by Eq.(5) and for $M_{2I}(q, z)$ we assume a Gaussian form

$$M_{2I}(q, t) = d(q) e^{-\alpha(q) t^2} \quad (11)$$

where $\alpha(q)$ is a decay constant and $d(q)$ is the normalization constant. Their values can be determined from the fourth and sixth frequency moments of $s(q, \omega)$ as

$$d(q) = \frac{\langle \omega^4 \rangle}{\langle \omega^2 \rangle} - \langle \omega^2 \rangle \left(2 + 1/S(q) \right) \quad (12)$$

$$\alpha(q) = \frac{1}{2 d(q)} \left[\frac{\langle \omega^6 \rangle}{\langle \omega^2 \rangle} - 6 \langle \omega^2 \rangle - \left(\frac{\langle \omega^4 \rangle}{\langle \omega^2 \rangle} \right)^2 \right] \quad (13)$$

Again one can see that $d(q)$ vanishes in the limit of large wavenumbers and therefore both the memory functions (10) and (7) tend to the same asymptotic form (5).

We now look at the above model of the memory function from a mean-field approach. Using Eqs.(10), (4) and (3) and after some manipulations, the density response function can be written as

$$\chi(q, z) = \frac{\chi_f(q, z)}{1 - V(q, z) \chi_f(q, z)} \quad (14)$$

where

$$V(q, z) = \frac{k_B T}{m} \left[\left(1 - 1/S(q) \right) - z M_{2I}(q, z) \right] \quad (15)$$

and $\chi_f(q, z)$ is the free-particle response function. A comparison of Eq.(14) with (9) reveals that the memory function (10) enables us to describe the system as if it interacts with a frequency-dependent effective potential, whereas a frequency-independent effective potential is given by the memory function (7). In the next section we shall compare results obtained from both models.

III. RESULTS AND DISCUSSION

The calculation of the dynamic structure factor from the models discussed in the previous section requires a knowledge of the static structure factor $S(q)$ and the frequency moments up to six. We use the results obtained in a previous paper (Dubey *et al.* 1979) with Duesbery-Taylor (1973) potential and for a number density of $0.522 \times 10^{-24} \text{ cm}^{-3}$ at the temperature of 1056 K. Theoretical results are compared with the molecular dynamics (MD) data for $S(q, \omega)$ obtained by Ebbsjö *et al.* (private communication) using the Duesbery-Taylor potential. Ebbsjö *et al.* have also performed MD calculations with two other potentials, namely the Ashcroft pseudopotential with Geldart-Vosko screening (AGV) and the Shaw model potential with Geldart-Taylor screening (SGT). However, the SGT potential which shows a sound mode peak only for q around 0.3 \AA^{-1} is less reliable than the AGV potential. This as well as Duesbery-Taylor potential clearly shows the existence of "sound waves" for $q < 1.2 \text{ \AA}^{-1}$ and a plateau in $S(q, \omega)$ for $1.5 \text{ \AA}^{-1} < q < 1.8 \text{ \AA}^{-1}$.

In Fig.1 we have plotted $\omega^2 S(q, \omega)$ for four values of q , namely 2.1, 2.672, 4.006 and 4.995 \AA^{-1} . For $q < 2 \text{ \AA}^{-1}$ our results for $S(q, \omega)$ are a monotonically decreasing function of ω and do not give any evidence of a plateau shown by MD data. The kinetic theory of Sjögren (1978) also does not give any resonance peak for $q > 0.5 \text{ \AA}^{-1}$. We are therefore not sure whether

the evidence of collective excitations shown by MD results in liquid aluminium is a real one. It may sometimes appear due to the inaccuracies in MD calculations. For $q > 2 \text{ \AA}^{-1}$ it can be seen from the figure that both the models of memory functions provide an overall good agreement with MD data. However, the peak heights of the PS model are in better agreement with MD for all q 's but the peak positions for $q = 2.1 \text{ \AA}^{-1}$ and 4.006 \AA^{-1} are slightly shifted to the smaller ω region. On the other hand, Yoshida's model seems to give better agreement at $q = 2.1 \text{ \AA}^{-1}$ and 4.006 \AA^{-1} but for other wavevectors its peak positions are shifted to the larger ω region. It may be mentioned that Yoshida's model satisfies sum rules up to sixth order and therefore incorporates information of pair as well as triplet correlations, whereas the PS model contains information of pair correlations only. We may thus conclude that at wavevectors $q = 2.1 \text{ \AA}^{-1}$ and 4.006 \AA^{-1} which correspond to first and second peaks in static structure factor, the triplet correlations are quite important.

To get an insight of the memory functions (11) and (7), in Fig.2 we have shown the real part, denoted as $M'(q, \omega)$, and the minus of the imaginary part, denoted as $-M''(q, \omega)$, of the memory function for some values of q . The dashed curves are the free-particle memory functions and solid lines are the results when the interaction part is also included in Yoshida's model. The dash-dot curves indicate the renormalized memory functions. It can be seen from the figure that with the introduction of the renormalized parameter $\gamma(q)$ the $M'(q, \omega)$ decreases much slower than the free-particle one. Also the values of renormalized $M'(q, \omega)$ are similar to the Yoshida model with the difference that the latter contains structure in it. At large wavevectors both the models approach asymptotically the free-particle value. The renormalized form of the memory function, however, has one advantage: it contains only one parameter, and therefore with equal knowledge of the sum rules one can incorporate higher poles into the continued fraction expansion of correlation functions.

In conclusion, we find that for wavenumbers corresponding to the peak positions in the static structure factor the effect of triplet correlations is quite important, whereas for other wavenumbers ($q > 2 \text{ \AA}^{-1}$) the binary collisions provide an appropriate mechanism for damping. For $q < 2 \text{ \AA}^{-1}$, the present analysis as well as the kinetic theory of Sjögren show no evidence of collective excitations. Therefore we believe that either the plateau in MD data is due to some inaccuracy in MD calculations or a second relaxation mechanism is needed in this region.

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FIGURE CAPTIONS

Fig.1 $\omega^2 S(q,\omega)$ versus ω for four selected values of q . Full curve (—) is the result of Yoshida's model and chain curve (-·-·-·-) is the result of renormalized memory function. Filled circles represent molecular dynamics data of Ebbsjo et al.

Fig.2 The frequency-dependence of real $M'(q,\omega)$ and the minus imaginary part $-M''(q,\omega)$ of the memory function. Dashed curve (---) is the free-particle memory function; solid line (—) when interaction part is also included in Yoshida's model and (-·-·-·-) is the renormalized memory function.

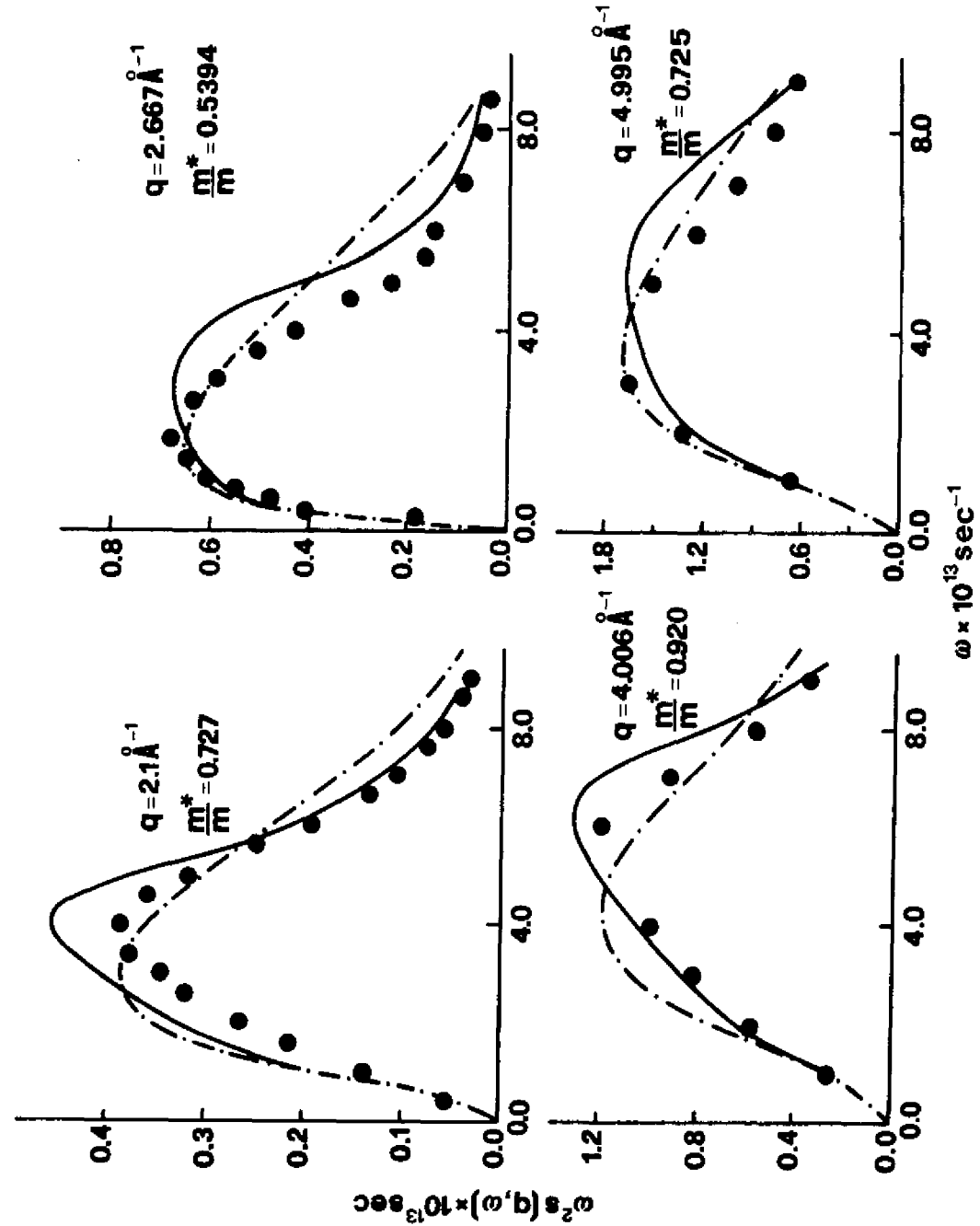


Fig.1

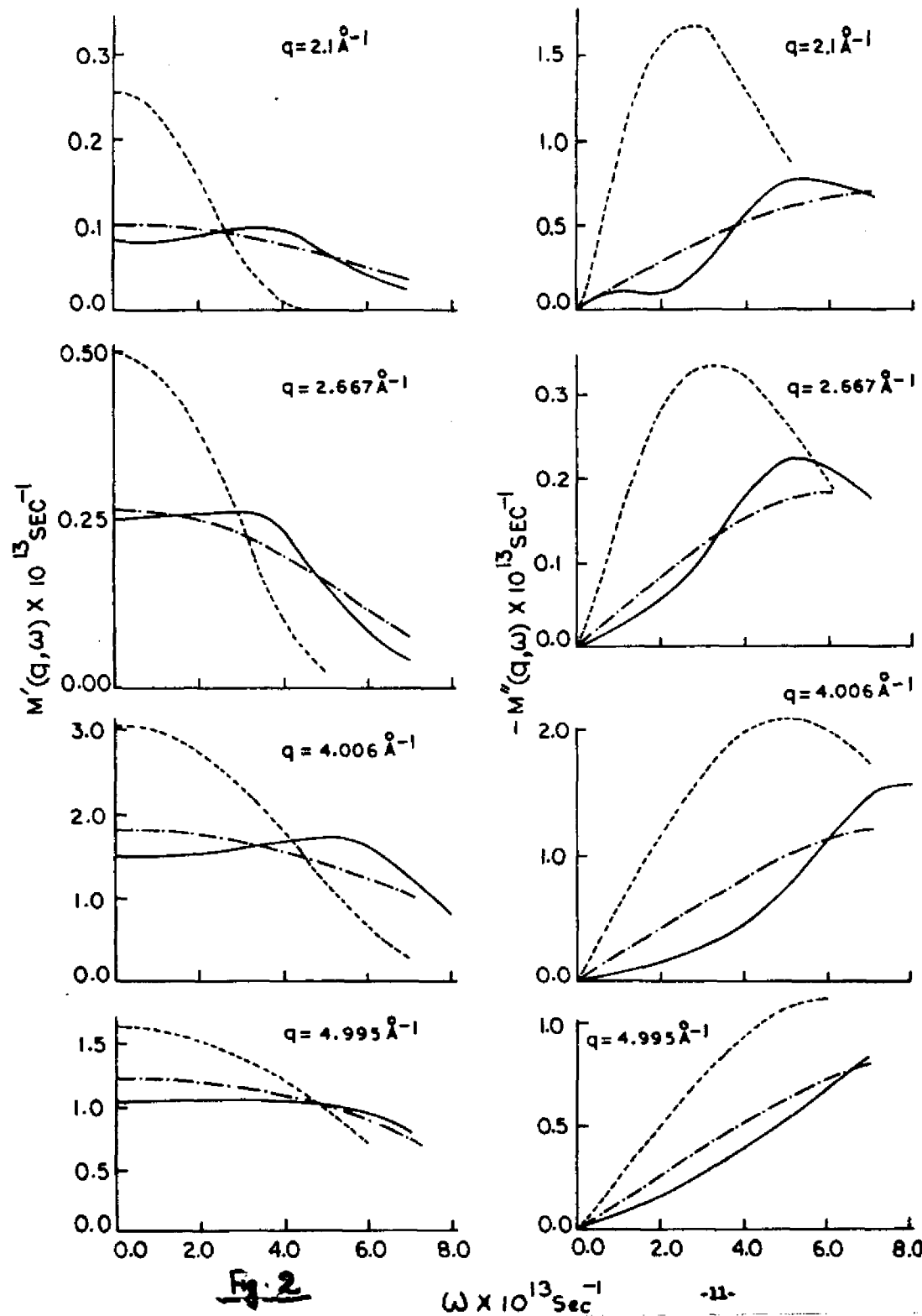


Fig. 2

 $\omega \times 10^{13} \text{Sec}^{-1}$

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