THE UNIVERSITY OF MICHIGAN

COLLEGE OF ENGINEERING
Department of Nuclear Engineering

Technical Report

FLUCTUATION ANALYSIS IN SIMPLE FLUIDS

A. Z. Akcasu
E. Daniels

ORA Project 08034

supported by:

NATIONAL SCIENCE FOUNDATION
GRANT NO. DK-1009
WASHINGTON, D.C.

administered through:

OFFICE OF RESEARCH ADMINISTRATION ANN ARBOR

July 1969

This report was prepared as a paper for submission to The Physical Review.

TABLE OF CONTENTS

	Page
ABSTRACT	1
1. INTRODUCTION	2
2. THE GENERALIZED LANGEVIN EQUATION	5
3. TRANSVERSE CURRENT CORRELATIONS	8
4. A GENERALIZED HYDRODYNAMIC DESCRIPTION	16
Transverse Equations	29
Longitudinal Equations	30
Conclusions	36
ACKNOWLEDGMENTS	37
REFERENCES	38
FIGURE CAPTIONS	40

ABSTRACT

Generalized Langevin Theory is applied to the analysis of fluctuations in simple fluids. Formulas for the current-current and density-density correlation functions are developed. More importantly, they are developed for a region of frequencies ($\omega \sim 10^{13}\text{sec}^{-1}$) and wave vectors ($k \sim 10^{8}\text{cm}^{-1}$) which are explored in typical slow neutron scattering measurements. Where applicable, comparisons are made with the results of the numerical calculations carried out by Rahman, and good agreement is generally found.

1. INTRODUCTION

The calculation of correlation functions in classical simple fluids in terms of microscopic quantities plays an important role in statistical mechanics, both for the interpretation of scattering experiments and the evaluation of the frequency and wavelength-dependent transport coefficients. Among these, the density-density (or Van Hove) correlation function has received the most attention because of its direct relation to the differential scattering cross-sections. Other correlation functions, such as the transverse current-current correlation functions which are not readily accessible in experiments, have been subject to quantitative investigations only recently following the publication of Rahman's[1] computer calculations in argon-like liquids. Computer studies of correlations[2] using molecular dynamical calculations provide a stringent test of the validity of the various classical theories introduced in correlation analysis because they only assume a known model interparticle potential, and involve no quantum effects.

The classical analysis of correlations is usually based on either a kinetic or hydrodynamic description of fluids. The kinetic description developed extensively by Nelkin[3] and his co-workers has been justified theoretically for dilute gases,[4] and used successfully to interpret Brillouin scattering from gases.[5,6] It has also been extended to dense fluids[7] and applied to Rahman's molecular dynamics calculations for liquid argon[2] with poor quantitative agreement.[8]

The hydrodynamic description of fluids has long been in use in the fluctua-

tion analysis in arbitrary continuous media as a phenomenological theory. In this approach, the conventional hydrodynamic equations are used to describe the linear response of the fluid, and the correlations functions are then related to the linear response by means of the fluctuation-dissipation theorem.[3] Using the formulation described by Landau and Lifschitz,[10] Rytov[11] applied the fluctuation-dissipation theorem to distributed parameter systems and calculated, among others, density-density correlation functions in an arbitrary continuous medium. This hydrodynamic approach has been used to interpret light scattering from liquids successfully by Mountain.[13] A systematic and general hydrodynamic description of fluids for the calculation of correlations and transport coefficients has been developed by Kadonoff and Martin.[12] The latter approach has been applied by Chung and Yip[14] to Rahman's calculations of current-current correlations.

The objective of this paper is to present a classical analysis of correlations in simple fluids, based on the generalized Langevin equation developed by Zwanzig,[15] and Mori[16,17] and to interpret quantitatively the current-current correlations computed by Rahman[2] for liquid argon. This approach has several appealing features. First by choosing the dynamical variable in the description of the fluid as the microscopic phase density function one obtains[18] an exact kinetic equation for the correlation function $\Gamma(\underline{v},\underline{v}';\underline{x},\underline{x}',t)$, which reduces by approximation to the kinetic equation derived by Zwanzig[19] and Nelkin.[8] However, by making an alternative choice of the dynamical variables to be microscopic densities in configuration space (e.g., mass, current, and energy densities) one arrives at an exact hydrodynamic description of cor-

relation functions in terms of frequency and wavelength-dependent transport parameters. The choice of the appropriate set of dynamical variables is arbitrary. For any choice of these variables one obtains exact expressions for the correlation functions of the variables in the set. Different levels of approximations can be obtained for a particular correlation function by adding new variables to the set and using the same simplifying assumption (e.g. Markoff assumption) in each case. The continued fraction expansion of correlation functions by Mori,[17] for example, can be obtained by using an orthogonal extension of the set starting from a given dynamical variable. The separation of thermodynamic and transport parameters can be given a geometric interpretation in terms of projections of dynamical variables on appropriate orthogonal axes, and the extension of their definitions to short wavelength where the anisotropies become significant can be made in a systematic way.

In this paper we will use the configuration space (hydrodynamic) description of fluids to investigate the current-current correlation functions, and obtain approximate formulas for transverse and longitudinal current-current correlations in the frequency and wavelength regions encountered in neutron scattering using a Markov assumption. The results will then be compared to Rahman's computer data[2] for liquid argon. We will also obtain the wavelength and frequency dependence of shear and longitudinal viscosities explicitly and discuss the influence of thermal effects as a function of wavelength. The objective of this paper is similar to that by Chung and Yip;[4] however their approach is based on the correlation function formalism developed by Martin and Kadonoff,[13] rather than on the projection operator formalism by Zwanzig[15] and Mori.[17]

2. THE GENERALIZED LANGEVIN EQUATION

Extending the projection operator technique first introduced by Zwanzig,[15] Mori[16] proves that the equation of motion of a set of dynamical variables, $a_j(t)$, can be written in the form,

$$\frac{da(t)}{dt} - i\Omega\cdot a(t) + \int_0^t \varphi(t-u)\cdot a(u)du = f(t) , \quad t \geq 0 . \tag{2.1}$$

The state vector a(t) is defined such that it has no invariant contribution, e.g.,

$$a(t) = A(t) - \langle A(t)\rangle , \tag{2.2}$$

where $\langle \ldots \rangle$ denotes the thermal average of the vector A(t). Equation (2.1) is the generalized form the Langevin equation (10,20) in the stochastic theory of Brownian motion. The random force vector f(t) is given formally by

$$f(t) = e^{t(1-P)iL}(1-P)\dot{a} , \quad (\dot{a} = iLa) , \tag{2.3}$$

where L is the classical Liouville operator and P is a projection operator defined for any arbitrary phase function G(t) by

$$PG(t) = \langle G(t)a^{\dagger}\rangle\cdot\langle aa^{\dagger}\rangle^{-1}\cdot a . \tag{2.4}$$

Here, $a^{\dagger}$ denotes the row vector which is the hermitian conjugate of a. The $\langle aa^{\dagger}\rangle^{-1}$ is the inverse of the square matrix $[\langle a_i a_j^{\dagger}\rangle]$ which is the static correlation matrix. It should be noted that the evolution of the force vector f(t) is determined by the special propagator $e^{t(1-P)iL}$, whereas the evolution of the state vector a(t) is given by

$$a(t) = e^{tiL}a \ . \tag{2.5}$$

It is shown by Mori[16] that

$$\langle f(t)a^{\dagger}\rangle \equiv 0 \ , \quad t \geq 0 \ . \tag{2.6}$$

The square matrices $\varphi(t)$ (damping function) and Ω (frequency matrix) are defined by

$$\varphi(t) = \langle f(t)f^{\dagger}(o)\rangle \cdot \langle aa^{\dagger}\rangle^{-1} \ , \tag{2.7}$$

and

$$i\Omega = \langle \dot{a}a^{\dagger}\rangle \cdot \langle aa^{\dagger}\rangle^{-1} \ . \tag{2.8}$$

Multiplying (2.1) by $a \cdot \langle aa^{\dagger}\rangle^{-1}$ from the right, taking the thermal average of the resulting equation and using (2.6) yields

$$\frac{dR(t)}{dt} - i\Omega \cdot R(t) + \int_o^t \varphi(t-u) \cdot R(u)du = 0 \ , \quad t \geq 0 \ , \tag{2.9}$$

where $R(t)$, the normalized dynamic correlation matrix, is defined by

$$R(t) = \langle a(t)a^{\dagger}\rangle \cdot \langle aa^{\dagger}\rangle^{-1} \ . \tag{2.10}$$

The one-sided Fourier transform of $R(t)$ is obtained from (2.8) as

$$\bar{R}(i\omega) = [i\omega - i\Omega + \bar{\varphi}(i\omega)]^{-1} \ , \tag{2.11}$$

where

$$\bar{\varphi}(i\omega) = \lim_{\varepsilon \to o} \int_o^{\infty} e^{-(i\omega+\varepsilon)t}\varphi(t)dt \ . \tag{2.12}$$

The projection technique enables one to find a closed set of linear equations for the correlation matrix $R(t)$ when the state variables $a_j(t)$ are chosen

as fluctuations from thermal equilibrium. The theory, although formally exact, serves only to transform the calculation from the direct computation of $R(t)$ to the computation of Ω and $\varphi(t)$. However, the frequency matrix Ω is determined from static correlations which are generally much easier to compute than the time-dependent correlation functions, and we may use approximations to compute the damping matrix $\varphi(t)$. In particular we will consider representations in which we can make a Markov approximation on $\varphi(t)$, viz.,

$$\bar{\omega} = \lim_{\omega \to o} \bar{\varphi}(i\omega) , \tag{2.13}$$

to approximate the transform of the correlation matrix $\bar{R}(i\omega)$ by

$$\bar{R}(i\omega) \tilde{=} [i\omega - i\Omega + \bar{\omega}]^{-1} . \tag{2.14}$$

We shall follow this formal procedure to calculate the current-current correlation function and its transform by choosing the components of the state vector as the spatial Fourier transforms of the local densities of conserved variables.

3. TRANSVERSE CURRENT CORRELATIONS

We first calculate the cosine transform of the transverse current correlation function as a simple application of the generalized Langevin equation, and then compare it with Rahman's computations. For this purpose, we choose the components of the state vector as

$$a_1 = \underline{J}_1(\underline{k}) \tag{3.1a}$$

$$a_2 = \underline{\underline{\Pi}}_{31}(\underline{k}) \tag{3.1b}$$

where $\underline{J}(\underline{k})$ and $\underline{\underline{\Pi}}(\underline{k})$ are the mass current density and the stress tensor, respectively. They are defined by

$$J_j(\underline{k}) \equiv \sum_{\alpha=1}^{N} mv_j^{\alpha} \exp(i\underline{k}\cdot\underline{x}^{\alpha}) , \tag{3.2a}$$

$$\Pi_{ij}(\underline{k}) \equiv \sum_{\alpha=1}^{N} \left[mv_i^{\alpha}v_j^{\alpha} + \frac{1}{2} \sum_{\substack{\beta=1 \\ (\beta\neq\alpha)}}^{N} \frac{x_i^{\alpha\beta}x_j^{\alpha\beta}}{|\underline{x}^{\alpha\beta}|^2} P^{\alpha\beta}(\underline{k}) \right] e^{i\underline{k}\cdot\underline{x}^{\alpha}} \tag{3.2b}$$

where

$$\underline{x}^{\alpha\beta} = \underline{x}^{\alpha} - \underline{x}^{\beta} \tag{3.2c}$$

$$P^{\alpha\beta}(\underline{k}) = R \frac{dV(R)}{dR} \frac{1-e^{-i\underline{k}\cdot\underline{R}}}{i\underline{k}\cdot\underline{R}} \Bigg|_{\underline{R}=\underline{x}^{\alpha\beta}} \tag{3.2d}$$

In these definitions, $\underline{x}^{\alpha}$ and $\underline{v}^{\alpha}$ denote the position and velocity of the αth particle in the system, and the subscripts i and j refer to the Cartesian components in a coordinate system in which $\underline{k}$ is parallel to the z-axis. With this choice of variables the static correlation matrix is diagonal, viz.,

$$\langle \underline{a}\,\underline{a}^{*} \rangle = \begin{bmatrix} \langle a_1 a_1^{*} \rangle & 0 \\ 0 & \langle a_2 a_2^{*} \rangle \end{bmatrix} \tag{3.3}$$

since the variables a_1 and a_2 are respectively odd and even functions of the particle velocities so that $\langle a_2 a_1^{*} \rangle = 0$. Furthermore, a direct evaluation of the diagonal terms using (3.2) yields

$$\frac{\beta}{V} \langle a_1 a_1^{*} \rangle = \rho_o \tag{3.4}$$

$$\begin{aligned} \frac{\beta}{V} \langle a_2 a_2^{*} \rangle &= C_{44}(k) \\ &= \left[\frac{n}{\beta} + n^2 \int d^3R \, g(R) \, \frac{\partial^2 V}{\partial X_2} \, \frac{(1-\cos kZ)}{k^2} \right] \end{aligned} \tag{3.5}$$

where $\beta = (1/k_B T)$ (k_B = Boltzmann's constant), V is the volume of the system, and ρ_o is the equilibrium density. Here $C_{44}(k)$ is one of the elastic moduli calculated by Schofield.[21] The frequency matrix Ω is calculated using (3.4) and (3.5), and noting $a_1 = ika_2$ as follows:

$$\Omega = k \begin{bmatrix} 0 & 1 \\ C_{44}(k)/\rho_o & 0 \end{bmatrix} . \tag{3.6}$$

Since $(1-P)\dot{a}_1 = ik(1-P)a_2 = 0$, the random force component $f_1(t)$ is identically equal to zero and the damping matrix φ has only one non zero element,

$$\varphi(t) = \begin{bmatrix} 0 & 0 \\ 0 & \varphi_{22}(t) \end{bmatrix} . \tag{3.7a}$$

where

$$\varphi_{22}(t) \equiv \langle (1-P)\dot{a}_2^{*} \, e^{t(1-P)iL} (1-P)\dot{a}_2 \rangle \langle a_2 a_2^{*} \rangle^{-1} . \tag{3.7b}$$

The generalized Langevin equation for the set $(a_1(t),a_2(t))$ becomes

$$\frac{da_1(t)}{dt} - ika_2(t) = 0 \tag{3.8a}$$

$$\frac{da_2(t)}{dt} - ik\frac{C_{44}}{\rho_o}a_1(t) + \int_o^t \varphi_{22}(t-u)a_2(u)du = f_2(t) . \tag{3.8b}$$

The transverse current correlation function is defined by

$$R_1(k,t) = \frac{\langle a_1(t)a_1^*\rangle}{\langle a_1 a_1^*\rangle} . \tag{3.9}$$

Its cosine transform $\bar{R}_1(k,\omega)$ is the transverse current power spectral density. The latter can be obtained directly from (3.8) by multiplying it by a_1^*, taking the thermal averages, using $\langle f_2(t)a_1^*\rangle \equiv 0$, and

$$R_1(k,\omega) = Re[i\omega+(k^2/\rho_o)\eta_s(k,i\omega)]^{-1} \tag{3.10}$$

where $\eta_s(k,i\omega)$ may be identified as a k- and ω-dependent shear viscosity defined by

$$\eta_s(k,i\omega) \equiv \frac{\rho_o}{ik}\frac{\langle \bar{a}_2(i\omega)a_1\rangle}{\langle \bar{a}_1(i\omega)a_1\rangle} \tag{3.11a}$$

$$= \frac{C_{44}(k)}{i\omega+\bar{\varphi}_{22}(k,i\omega)} . \tag{3.11b}$$

Rahman[2] has computed $R_1(k,\omega)$ for various values k and ω. We have been able to obtain an exact expression for it in terms of the Laplace transform of the damping function, viz., $\bar{\varphi}_{22}(k,i\omega)$. However, the evaluation of the latter using (3.7b) is as difficult as solving the Liouville equation, although perturbative

techniques such as expansions in density or interparticle potential[18] may be used in dilute systems. For sufficiently small frequencies, we can approximate (3.10) by replacing $\bar{\varphi}_{22}(k,i\omega)$ by its zero frequency limit $\bar{\varphi}_{22}(k,o)$. This approximation corresponds to a Markov description of the fluid in terms of $a_1(t)$ and $a_2(t)$, in which the convolution integral in (3.8b) is replaced by

$$\int_o^t \varphi_{22}(t-u)a_2(u) \simeq a_2(t) \int_o^\infty du\, \varphi_{22}(u) . \tag{3.12a}$$

The frequency range in which the Markov assumption may be expected to be valid can be estimated by considering the next term in the expansion of $\bar{\varphi}_{22}(k,i\omega)$ in powers of $(i\omega)$:

$$\omega << \left| \bar{\varphi}_{22}(k,o) \Big/ \frac{d\bar{\varphi}_{22}(k,i\omega)}{d(i\omega)} \right|_{i\omega = o} \tag{3.12b}$$

It is clear that the Markov assumption ceases to be valid if $\bar{\varphi}_{22}(k,o)$, the leading term in this expansion vanishes for some values of k. In such cases it turns out to be more convenient to go to a more complete description of the fluid by introducing new variables.

With these remarks, we obtain the following approximate form for $\bar{R}_1(k,\omega)$:

$$R_1(k,\omega) \simeq \frac{\omega_s(k)k^2C_{44}(k)/\rho_o}{\omega^2\omega_s^2(k) + \left(\frac{k^2C_{44}(k)}{\rho_o} - \omega^2\right)^2} \tag{3.13}$$

where we have introduced

$$\omega_s(k) = \lim_{\omega \to o} \bar{\varphi}_{22}(k,i\omega) . \tag{3.14}$$

Calculation of $\omega_s(k)$ directly from (3.14) is still a formidable task in dense fluids, although it is simpler than calculating $\bar{\varphi}_{22}(k,i\omega)$. Therefore, we choose to try to guess its k-dependence by considering the asymptotic behavior of $R_{\perp}(k,\omega)$ in the large-k limit, and of $\eta_s(k,i\omega)$ in the small-k limit. First, it is known that

$$\lim_{k,\omega\to o} \eta_s(k,i\omega) = \frac{C_{44}(o)}{\omega_s(o)} \tag{3.15}$$

is the conventional shear viscosity, η_s. Hence, the small k limit of $\omega_s(k)$ is given by

$$\omega_s(o) = \frac{C_{44}(o)}{\eta_s} \tag{3.16}$$

(Note that $C_{44}(o)$ is G_∞ in Zwanzig's notation[22]).

The large-k behavior of $R_{\perp}(k,\omega)$ may be predicted from the transverse current spectrum of an ideal gas, viz.,

$$R_{\perp}^{IG}(k,\omega) = \left(\frac{\Pi\beta m}{2k^2}\right)^{1/2} e^{-\beta m\omega^2/2k^2}, \tag{3.17}$$

This function has a single maximum at $\omega = o$ for all k. On the other hand, (3.13) attains its maximum for a fixed k at a frequency

$$\omega_{sm}^2(k) = \frac{k^2 C_{44}(k)}{\rho_o} - \frac{\omega_s^2(k)}{2} \tag{3.18}$$

for $\omega_s^2(k) < 2k^2C_{44}(k)/\rho_o$, and at $\omega = o$ otherwise. (The function $\omega_{sm}(k)$ is often referred to as the "dispersion relation.")[2] The dispersion relation (3.18) is expected to approach that of an ideal gas as $k \to \infty$ because for large-k the par-

ticles behave as free particles. (This is more apparent in the case of longitudinal current correlation function because it is related directly to the neutron scattering cross section, there the large values of k correspond to large momentum transfer to the scattering medium.) Hence, we require $\omega_{sm}^2(k)$ to approach zero for large values of k, i.e.,

$$\omega_s^2(k) \to \frac{2k^2 C_{44}(k)}{\rho_o} \quad . \tag{3.19}$$

Thus, we obtain the asymptotic behavior of $\omega_s(k)$ for small and large k from (3.16) and (3.19). The k-dependence of $\omega_s(k)$ for the intermediate values of k can be obtained by interpolating it between the zero and large k limits by the following formula:

$$\omega_s^2(k) = \frac{2k^2 C_{44}(k)}{\rho_o} + \frac{\left[\omega_s^2(o) - 2k^2 (C_{44}(k) - \rho_o/\beta m)/\rho_o\right]}{[1+k^2/k_o^2]} \quad , \tag{3.20}$$

where k_o is an adjustable parameter whose choice will be discussed presently.

It is interesting to compare (3.13) using the above expression for $\omega_s(k)$ to the ideal gas result in (3.17) for zero frequency and large k, because the Markov approximation becomes exact at $\omega = o$. Noting that $C_{44}(k)/\rho_o \to (1/\beta m)$ (cf. Equation (3.5)) we obtain $\bar{R}_1(k,o)$ from (3.13) as $(2\beta m)^{1/2}/k$ whereas (3.17) yields $(\Pi\beta m/2)^{1/2}/k$. The ratio is $\sqrt{4/\Pi} \approx 1.12$. Thus (3.13) recaptures the ideal gas result in the limit of small ω and large k. It may be pointed out here that (3.10) can be approximated for large frequencies by replacing $\bar{\varphi}_{22}(k,i\omega)$ by $\varphi_{22}(k,o)/i\omega$ (short-time expansion of $\varphi_{22}(k,t)$). Since our inter-

est lies in the small frequency region we shall not dwell on this point further even though $\varphi_{22}(k,o)$ is calculable exactly.

With the aid of (3.20) we have been able to obtain an expression for the transverse current power spectral density (3.13), which contains only one adjustable parameter, k_o. The value of k_o determines the transition from the small to large k limits. It is expected to be in the vicinity of the main peak of the structure function S(k), which occurs at $k = 2\text{Å}^{-1}$ in Rahman's computer calculations[2] for liquid argon. We have chosen $k_o = 1.5\text{Å}^{-1}$ which yielded the best fit to the computed curves although the value of k_o is found to be not too critical. The other constant in (3.20) is $\omega_s^2(o)$ which is obtained from (3.15) as $\omega_s^2(o) = .1 \times 10^{26}$ sec^{-1}, corresponding to a value for the shear viscosity $\eta_s = 2.8 \times 10^{-3}$ poise at $\rho_o = 1.407$ g/cm^3 and T = 76°K for liquid argon. The values of $C_{44}(k)$ were computed according to Eq. (3.5) in which the interparticle potential is taken, following Rahman, as

$$V(R) = 4\varepsilon\left[\left(\frac{\sigma}{R}\right)^{12} - \left(\frac{\sigma}{R}\right)^{6}\right]$$

with $(\varepsilon/k_B) = 120°K$ (k_B = Boltzmann constant) and $\sigma = 3.9$ Å. The variation of $C_{44}(k)$ is plotted in Fig. 1.

Figures 2 and 3 show the variation of $\omega_s(k)$ and $\eta_s(k,o) = C_{44}(k)/\omega_s(k)$ with k. We observe that the k-dependent shear viscosity decreases very rapidly by a factor of 100 in the region of k from zero to 2 Å^{-1}, and approaches zero as (1/k). This k-dependence of $\eta_s(k,o)$ appears to be crucial to the behavior of $R_1(k,\omega)$ for the k-values in 1-4 Å^{-1}.

Figures 4 and 5 show comparison between the calculated curves and Rahman's

data.[2] It is noteworthy that the present model predicts well the cut-off wavelength in the dispersion curve, i.e., $\omega_{sm}(k)$. Other features are self-explanatory.

4. A GENERALIZED HYDRODYNAMIC DESCRIPTION

The previous application indicates that the correlation function associated with a dynamical variable $a_1(t)$ (e.g., $J_1(\underline{k},t)$) can be obtained by solving the appropriate generalized Langevin equation. If only the autocorrelation function is of interest, the one-component description of the system is sufficient in principle. The correlation function in this case is obtained by solving

$$\dot{R}_1(t) + \int_0^t du\, \varphi_1(t-u)R_1(u) = 0 \qquad (4.1)$$

(Note that the frequency matrix is always zero in one-dimensional description.) The damping function $\varphi_1(t)$ involves $f(t) = \exp[t(1-P)iL](1-P)\dot{a}_1(o)$ where P projects a phase function onto $a_1(o)$. Although (4.1) is exact, the calculation of $\varphi_1(t)$ is as difficult as calculating $\langle a_1(t)a_1^*\rangle$ directly. Crude approximations for φ_1, such as the Markov assumption, are generally not precise enough to include even the qualitative features of the correlation function, or the power spectral density associated with it, for large values of ω and k. By introducing instead, a multidimensional description of the system, one actually extracts a great deal of information about the collective motion of the system through the frequency matrix even though one may still be interested only in the autocorrelation function of a single variable. This information is contained in $\varphi_1(t)$ in one-dimensional description. A proper choice of the additional variables in a given system, can lead to a sufficiently precise expression for the correlation function in a wide range of ω,k even with crude

approximations on the multidimensional damping function. The variables $J_1(\underline{k})$ and $\Pi_{31}(\underline{k})$ introduced in Section 3 provide such a description for the transverse-current correlations. The description of the longitudinal current correlations requires a more detailed description of the fluid including thermal and viscosity effects as will be demonstrated in this section. Moreover, a multidimensional description also allows the computation of the various cross-correlations between the variables in the set in terms of the same thermodynamic and transport parameters.

The purpose of this section is then to present a 14-dimensional description of a simple liquid, and to compute specifically the transverse and longitudinal current correlations. This description includes thermal effects, and sheds light on the anisotropies in the fluid for large k values.

For state variables, choose

$$\underline{a} \equiv \mathrm{col}[\rho, \theta, \sigma_\mu, J_i, q_j] \ , \tag{4.2a}$$

where J_i and q_j are vectors with three components ($i,j = 1,2,3$) and σ_μ is a 6-component vector with $\mu = 1,\ldots 6$, viz.,

$$\sigma_\mu = \mathrm{col}[\sigma_{11}, \sigma_{22}, \sigma_{33}, \sigma_{13}, \sigma_{23}, \sigma_{13}] \ . \tag{4.2b}$$

The variables θ, σ_μ and q_j are defined by

$$\rho(k) = m \sum_{\alpha=1}^{N} e^{i\underline{k}\cdot\underline{x}^\alpha} \tag{4.3}$$

$$\theta(k) = \left[E(k) - \frac{\langle E(\underline{k}) \rho^*(\underline{k}) \rangle}{\langle \rho(\underline{k}) \rho^*(\underline{k}) \rangle} \rho(\underline{k}) \right] \tag{4.4}$$

$$\sigma_{ij}(\underline{k}) \equiv \Pi_{ij}(\underline{k}) - \frac{\langle \Pi_{ij}(\underline{k})\rho^*(\underline{k})\rangle}{\langle \rho(\underline{k})\rho^*(\underline{k})\rangle}\rho(\underline{k}) - \frac{\langle \Pi_{ij}(\underline{k})\theta^*(\underline{k})\rangle}{\langle \theta(\underline{k})\theta^*(\underline{k})\rangle}\theta(\underline{k}) \qquad (4.5)$$

$$q_j(\underline{k}) \equiv Q_j(\underline{k}) - \langle Q_j \underline{J}^*(\underline{k})\rangle \cdot \langle \underline{J}(\underline{k})\underline{J}^*(\underline{k})\rangle^{-1} \cdot \underline{J}(\underline{k}) \qquad (4.6)$$

The definitions of $\underline{J}(\underline{k})$ and $\Pi_{ij}(\underline{k})$ have already been given in (3.2a) and (3.2b) respectively. The quantities $E(\underline{k})$ and $Q_j(\underline{k})$ are the energy density, and the energy current density respectively. They are defined as

$$E(\underline{k}) = \sum_{\alpha=1}^{N} \left[\frac{1}{2} m\underline{v}^\alpha \cdot \underline{v}^\alpha + \frac{1}{2} \sum_{\substack{\beta=1 \\ (\beta \neq \alpha)}}^{N} V(|\underline{x}^\alpha - \underline{x}^\beta|) \right] e^{i\underline{k}\cdot\underline{x}^\alpha} \qquad (4.7)$$

$$Q_j(\underline{k}) = \sum_\alpha \underline{v}^\alpha \left[\frac{1}{2} m\underline{v}^\alpha \cdot \underline{v}^\alpha + \frac{1}{2} \sum_{\substack{\beta=1 \\ (\beta \neq \alpha)}}^{N} V(|\underline{x}^\alpha - \underline{x}^\beta|) \right] e^{i\underline{k}\cdot\underline{x}^\alpha}$$

$$+ \frac{1}{4} \sum_{\alpha,\beta}{}' (\underline{v}^\alpha + \underline{v}^\beta) \cdot \underline{x}^{\alpha\beta} \frac{x_j^{\alpha\beta}}{|\underline{x}^{\alpha\beta}|^2} P^{\alpha\beta}(\underline{k}) e^{i\underline{k}\cdot\underline{x}^\alpha} \qquad (4.8)$$

($\underline{x}^{\alpha\beta}$ and $P^{\alpha\beta}(\underline{k})$ were defined previously in (3.2c) and (3.2d)).

The following usual conservation laws prevail among the variables ρ, J_j, Π_{ij}, E, and Q_j:

$$\frac{\partial \rho}{\partial t} = i\underline{k}\cdot\underline{J} , \qquad (4.9a)$$

$$\frac{\partial \underline{J}}{\partial t} = i\underline{k}\cdot\underline{\underline{\Pi}} , \qquad (4.9b)$$

$$\frac{\partial E}{\partial t} = i\underline{k}\cdot\underline{Q} . \qquad (4.9c)$$

The tensors Π_{ij} and σ_{ij} are symmetric and have only six independent components as implied in (4.2b). The variables σ_{ij} and q_j denote the viscous stress tensor and the heat flux vector. The $\Theta(t)$, defined by (4.4), will be replaced later by

$$T(\underline{k}) \equiv \frac{\Theta(\underline{k})}{\rho_o C_v(k)} \tag{4.10}$$

whose average with respect to a perturbed distribution function yields the temperature in the conventional linearized hydrodynamic description of a fluid.[16] Such an identification, however, is not needed for the present. The quantity $C_v(k)$ will be defined later (it will be identified as the specific heat at constant volume).

The average values of $\rho(k)$, $E(k)$, and $\Pi_{ij}(k)$ are zero for $k \neq 0$. When $k = 0$, we have[21]

$$\frac{1}{V}\langle\rho(0)\rangle = \rho_o \tag{4.11a}$$

$$\frac{1}{V}\langle E(0)\rangle = \frac{3}{2}\frac{\rho_o}{m\beta} + \frac{1}{2}\frac{\rho_o^2}{m^2}\int d^3R)(V(R))(g(R)) \tag{4.11b}$$

$$\frac{1}{V}\langle\Pi_{ij}(0)\rangle = \delta_{ij} P_o = \delta_{ij}\left[\frac{\rho_o}{m\beta} - \frac{\rho_o^2}{6}\int d^3R\, R\, \frac{dV(R)}{dR}\, g(R)\right] . \tag{4.11c}$$

Here, P_o is the equilibrium pressure. We shall always assume that these averages are subtracted from ρ, E, and Π_{ij} whenever they are not zero, so that the state vector $\underline{a}$ will denote the fluctuations.

Eight of the 14 components of the state vector $\underline{a}$ are even, and the remain-

ing six are odd functions of particle velocities. Hence $\underline{a}$ can be decomposed into even and odd parts as

$$\underline{a} = \mathrm{col}[\underline{a}^{e}, 0] + \mathrm{col}[0, \underline{a}^{o}] \, .$$

Consequently, the static correlation matrix $\underline{\underline{\phi}} = \langle \underline{a}\, \underline{a}^{*} \rangle$ splits into two disjoint submatrices as

$$\underline{\underline{\phi}} = \begin{bmatrix} \underline{\underline{\phi}}^{e} & 0 \\ 0 & \underline{\underline{\phi}}^{o} \end{bmatrix} \tag{4.12}$$

where

$$\underline{\underline{\phi}}^{e} \equiv \langle \underline{a}^{e}\, \underline{a}^{e*} \rangle = \begin{bmatrix} \langle \rho\rho^{*} \rangle & 0 & 0 \\ 0 & \langle \theta\theta^{*} \rangle & 0 \\ 0 & 0 & \langle \underline{\sigma}\, \underline{\sigma}^{*} \rangle \end{bmatrix} , \tag{4.13a}$$

and

$$\underline{\underline{\phi}}^{o} \equiv \langle \underline{a}^{o}\, \underline{a}^{o*} \rangle = \begin{bmatrix} \langle \underline{J}\, \underline{J}^{*} \rangle & 0 \\ 0 & \langle \underline{q}\, \underline{q}^{*} \rangle \end{bmatrix} \, . \tag{4.13b}$$

The block-diagonality of $\underline{\underline{\phi}}^{e}$ and $\underline{\underline{\phi}}^{o}$ is a consequence of the choice of the state variables as in (4.4), (4.5), and (4.6), which imply the following orthogonality relations:

$$\langle \rho\theta^{*} \rangle = \langle \sigma_{ij} \rho^{*} \rangle = \langle \sigma_{ij} \theta^{*} \rangle = \langle q_i J_j^{*} \rangle = 0 \, . \tag{4.14}$$

The static correlation functions appearing in (4.13a) and (4.13b) will be discussed later.

The frequency matrix $\underline{\underline{\Omega}} = \langle \dot{\underline{a}}\, \underline{a}^{*} \rangle \cdot \underline{\underline{\phi}}^{-1}$ can be written as

$$i\underline{\Omega} = \begin{bmatrix} 0 & \langle \dot{\underline{a}}^{e} \underline{a}^{o*} \rangle \cdot \underline{\underline{\phi}}^{o^{-1}} \\ \langle \dot{\underline{a}}^{o} \underline{a}^{e*} \rangle \cdot \underline{\underline{\phi}}^{e^{-1}} & 0 \end{bmatrix} \tag{4.15}$$

As a consequence of the conservation relations (4.9), $\underline{f}(o) = (1-P)\dot{\underline{a}}$ has only nine nonzero components:

$$\underline{f}(o) = \mathrm{col}\left[0,0,f_{\mu}^{\sigma},0,0,0,f_{j}^{q}\right] . \tag{4.16}$$

Therefore, the damping matrix $\underline{\underline{\varphi}}(t)$ is of the following form:

$$\underline{\underline{\varphi}}(t) = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & \underline{\underline{\varphi}}^{\sigma\sigma}(t) & 0 & \underline{\underline{\varphi}}^{\sigma q}(t) \\ 0 & 0 & 0 & 0 \\ 0 & \underline{\underline{\varphi}}^{q\sigma}(t) & 0 & \underline{\underline{\varphi}}^{qq}(t) \end{bmatrix} , \tag{4.17}$$

where $\underline{\underline{\varphi}}^{\sigma\sigma}$ and $\underline{\underline{\varphi}}^{qq}$ are 6 x 6 and 3 x 3 square matrices. The off-diagonal matrices $\underline{\underline{\varphi}}^{q\sigma}(t)$ and $\underline{\underline{\varphi}}^{\sigma q}$ are 3 x 6 and 6 x 3.

Substituting (4.15) and (4.17) into the generalized Langevin equation discussed in Section 2, we obtain the following set of equations:

$$\dot{\rho}(t) = i\underline{k}\cdot\underline{J}(t) \tag{4.18}$$

$$\dot{\underline{J}}(t) - \frac{\langle \dot{\underline{J}}\rho^{*} \rangle}{\langle \rho\rho^{*} \rangle} \rho(t) - \frac{\langle \dot{\underline{J}}\theta^{*} \rangle}{\langle \theta\theta^{*} \rangle} \theta(t) = i\underline{k}\cdot\underline{\underline{\sigma}}(t) \tag{4.19}$$

$$\theta(t) + \langle \theta\dot{\underline{J}}^{*} \rangle \cdot \langle \underline{J}\,\underline{J}^{*} \rangle^{-1} \cdot \underline{J}(t) = i\underline{k}\cdot\underline{q}(t) \tag{4.20}$$

$$\dot{\underline{\sigma}}(t) - \langle \dot{\underline{\sigma}}\underline{q}^* \rangle \cdot \langle \underline{q}\underline{q}^* \rangle^{-1} \cdot \underline{q}(t) + \int_o^t \underline{\underline{\varphi}}^{\sigma q}(t-u) \cdot \underline{q}(u) du$$

$$- \langle \dot{\underline{\sigma}}\underline{J}^* \rangle \cdot \langle \underline{J}\underline{J}^* \rangle^{-1} \cdot \underline{J}(t) + \int_o^t \underline{\underline{\varphi}}^{\sigma\sigma}(t-u) \cdot \underline{\sigma}(u) du = \underline{f}^{\sigma}(t) , \qquad (4.21)$$

$$\dot{\underline{q}}(t) - \frac{\langle \dot{\underline{q}}\theta^* \rangle}{\langle \theta\theta^* \rangle} \theta(t) + \int_o^t \underline{\underline{\varphi}}^{qq}(t-u) \cdot \underline{q}(u)$$

$$- \langle \dot{\underline{q}}\underline{\sigma}^* \rangle \cdot \langle \underline{\sigma}\underline{\sigma}^* \rangle^{-1} \cdot \underline{\sigma}(t) + \int_o^t \underline{\underline{\varphi}}^{q\sigma}(t-u) \cdot \underline{\sigma}(u) du = \underline{f}^{q}(t) \qquad (4.22)$$

n writing (4.20) and (4.22) we have used the orthogonality properties of the variables, and the identity:

$$\langle \dot{\underline{q}}\rho^* \rangle = 0 \qquad (4.23)$$

These follow from the sum rule $\langle \dot{A}B^* \rangle = -\langle A\dot{B}^* \rangle$ where A and B are two arbitrary dynamical variables.[21] Notice that we recover the conservation laws (4.9) in Eqs. (4.18), (4.19), and (4.20). Equations (4.18) through (4.22) describe the time evolution of the state vector $\underline{a} = \mathrm{col}[\rho, \theta, \sigma_\mu, J_i, q_j]$ exactly. We shall approximate them by neglecting the coupling between the viscous stress tensor and heat flux vector and introducing the Markov assumption in (4.21) and (4.22). The first approximation is equivalent to setting $\langle \dot{\underline{\sigma}}\underline{q}^* \rangle \equiv 0$ and $\underline{\underline{\varphi}}^{\sigma q}(t) \equiv 0$ in (4.21) and (4.22), and the second to

$$\int_o^t \underline{\underline{\varphi}}^{\sigma\sigma}(t-u) \cdot \underline{\sigma}(u) du \simeq \underline{\underline{W}}^{\sigma}(\underline{k}) \cdot \underline{\sigma}(t) \qquad (4.24a)$$

$$\int_o^t \underline{\underline{\varphi}}^{qq}(t-u) \cdot \underline{q}(u) du \simeq \underline{\underline{W}}^{q}(\underline{k}) \cdot \underline{q}(t) \qquad (4.24b)$$

where

$$\underline{\underline{W}}^{\sigma}(\underline{k}) \equiv \int_0^{\infty} dt\, \underline{\underline{\varphi}}^{\sigma\sigma}(t) , \tag{4.25a}$$

$$\underline{\underline{W}}^{q}(\underline{k}) = \int_0^{\infty} dt\, \underline{\underline{\varphi}}^{qq}(t) . \tag{4.25b}$$

Furthermore, the following definitions and equalities will be introduced:

$$\frac{\langle \dot{\underline{J}}\rho^{*}\rangle}{\langle \rho\rho^{*}\rangle} \equiv C_L^2(k) i\underline{k} \tag{4.26a}$$

$$\frac{\langle \dot{\underline{J}}T^{*}\rangle}{\langle TT^{*}\rangle} \equiv C_L^2(k)\rho_o\beta_L(k) i\underline{k} \tag{4.26b}$$

$$\frac{1}{V}\langle J_i J_j^{*}\rangle \equiv \frac{\rho_o}{\beta}\delta_{ij} \tag{4.26c}$$

$$\frac{\langle \theta\theta^{*}\rangle}{\langle J_1 J_1^{*}\rangle} \equiv \frac{\beta}{\rho_o}\left[\langle EE^{*}\rangle - \frac{|\langle E\rho^{*}\rangle|^2}{\langle \rho\rho^{*}\rangle}\right] \equiv T_o C_v(k) \tag{4.26d}$$

$$\frac{\langle \dot{\underline{q}}\theta^{*}\rangle}{\langle \theta\theta^{*}\rangle} \equiv i\underline{k}\, K_L(k) \tag{4.26e}$$

$$\frac{\beta}{V}\langle \sigma_\mu \sigma_\nu^{*}\rangle \equiv \Sigma_{\mu\nu}(k) \tag{4.26f}$$

Then, we obtain the following approximate description:

$$\dot{\rho}(t) = ik_j J_j(t) \tag{4.27a}$$

$$\dot{J}_j(t) - C_L^2(k) ik_j[\rho(t)+\beta_L(k)\rho_o T(t)] = ik_m \sigma_{jm}(t) \tag{4.27b}$$

$$\dot{\theta}(t) - \beta_L(k)C_L^2(k)T_o\dot{\rho}(t) = ik_j q_j(t) \tag{4.27c}$$

$$\dot{\sigma}_\mu(t) - \sum_{\mu\nu}(k)\varepsilon_\nu(t) + W^{\sigma}_{\mu\nu}(k)\sigma_\nu(t) = f^{\sigma}_{\mu}(t) \tag{4.27d}$$

$$\dot{q}_j(t) - K_L(k)ik_j\theta(t) + W^{q}_{jm}q_m(t) = f^{q}_{j}(t) \tag{4.27e}$$

where ε_{mn} is the rate of strain tensor, i.e.,

$$\varepsilon_{mn}(k) = \frac{i}{2\rho_o}[k_m J_n + k_n J_m] \tag{4.28}$$

We shall discuss the physical implications of the various quantities appearing in this set.

The $C_L^2(k)$, introduced in (4.26a) can be defined also by

$$C_L^2(k) = \frac{\langle \Pi_{33}\rho^* \rangle}{\langle \rho\rho^* \rangle} \tag{4.29a}$$

$$= \frac{1}{\beta m S(k)} \tag{4.29b}$$

where S(k) is the structure factor defined by

$$S(k) = 1 + \frac{\rho_o}{m}\int d^3R e^{i\mathbf{k}\cdot\mathbf{R}}[g(R)-1], \tag{4.29c}$$

g(R) being the static pair correlation function. We will refer to $C_L(k)$ as the longitudinal isothermal speed of sound. Its relation to the conventional isothermal speed $C_o(k)$ can be established by considering the definition of the latter:[16][21]

$$\frac{1}{3}\frac{\langle (\mathrm{Tr}\ \Pi_{ij})\rho^* \rangle}{\langle \rho\rho^* \rangle} = \left(\frac{\partial P}{\partial \rho}\right)_T \equiv C_o^2(k) \tag{4.29d}$$

where P is the thermodynamic part of pressure[16,21] $p = (1/3)\mathrm{Tr}\Pi_{ij}$ (the non-thermodynamic part, by definition, has no projection on ρ and T). If we define a transverse isothermal speed as

$$\frac{\langle \Pi_{22}\rho^* \rangle}{\langle \rho\rho^* \rangle} = \frac{\langle \Pi_{11}\rho^* \rangle}{\langle \rho\rho^* \rangle} \equiv \frac{1}{2} C_L^2(k) \tag{4.30}$$

then, $C_o^2 = (1/3)(C_L^2+C_T^2)$. In the isotropic limit where $\underline{k}\cdot\underline{a} \ll 1$, a being the mean linear force range, then $C_o^2 = C_T^2 = (1/2)C_L^2$, and the distinction between longitudinal and transverse isothermal speeds becomes irrelevant. It is important to note that only longitudinal isothermal speed is related to the structure function S(k) as in (4.29b).

The quantity $\beta_L(k)$ in (4.26b), which we refer to as the longitudinal thermal expansion coefficient, may be equivalently defined by

$$\frac{\langle \Pi_{33}T^* \rangle}{\langle TT^* \rangle} \equiv C_L^2(k)\rho_o\beta_L(k) \ . \tag{4.31}$$

We may also define a transverse expansion coefficient $\beta_T(k)$, similar to (4.30), as $(\rho_o\beta_T C_T^2/2) = \langle \Pi_{11}T^* \rangle / \langle TT^* \rangle = \langle \Pi_{22}T^* \rangle / \langle TT^* \rangle$. The conventional definition of $\beta(k)$ as a derivative of the thermodynamic part of pressure is

$$C_o^2(k)\rho_o\beta_o(k) = \frac{1}{3}\frac{\langle \mathrm{Tr}\Pi_{ij}T^* \rangle}{\langle TT^* \rangle} \equiv \left(\frac{\partial P}{\partial T}\right)_\rho \ . \tag{4.32}$$

Then, β_o is related to β_L and β_T by

$$\beta_o = \frac{\beta_L C_L^2+\beta_T C_T^2}{C_L^2+C_T^2} \ . \tag{4.33}$$

In the isotropic limit, $\beta_o = \beta_L = \beta_T$.

Equation (4.26d) defines the specific heat at constant volume. This definition is identical to that by Schofield.[21]

The quantity K_L introduced in (5.26e) can be equivalently defined by

$$K_L(k) \equiv \frac{\langle q_3 q_3^* \rangle}{\langle \theta\theta^* \rangle} = \frac{1}{\langle \theta\theta^* \rangle}\left[\langle Q_3 Q_3^* \rangle - \frac{\beta}{\rho_o V}\left|\langle E\Pi_{33}^* \rangle\right|^2\right] . \qquad (4.34)$$

It will be related later to the components of the thermal diffusivity tensor (cf. 4.42).

The symmetric tensor $\Sigma_{\mu\nu}(k)$ defined by (4.26f) is related to the elastic moduli tensor $C_{\mu\nu}$ by

$$\Sigma_{\mu\nu} = C_{\mu\nu} - \frac{\beta}{V}\left[\frac{\langle \Pi_\mu \rho^* \rangle\langle \rho \Pi_\nu^* \rangle}{\langle \rho\rho^* \rangle} + \frac{\langle \Pi_\mu T^* \rangle\langle T \Pi_\nu^* \rangle}{\langle TT^* \rangle}\right] \qquad (4.35a)$$

where

$$C_{\mu\nu} \equiv \frac{\beta}{V}\langle \Pi_\mu \Pi_\nu \rangle . \qquad (4.35b)$$

We have already encountered some elements of $C_{\mu\nu}$. In (3.5) we have used $C_{44} = (\beta/V)\langle \Pi_{31}\Pi_{31}^* \rangle$. Since $\langle \Pi_{3j}\rho^* \rangle = \delta_{j3}\langle \Pi_{33}\rho^* \rangle$ and $\langle \Pi_{3j}T^* \rangle = \delta_{j3}\langle \Pi_{33}T^* \rangle$, the terms in parenthesis are zero. We then obtain

$$\Sigma_{44} = C_{44} = C_{55} = \Sigma_{55} \qquad (4.36)$$

we shall also need Σ_{33} and Σ_{32}, in terms of $C_{33} \equiv \beta/V \langle \Pi_3 \Pi_{33}^* \rangle$ and $C_{32} \equiv \beta/V \langle \Pi_3 \Pi_{22}^* \rangle$ (clearly $\Sigma_{32} = \Sigma_{31}$, and $C_{32} = C_{31}$). From (4.35a) we get

$$\Sigma_{33} = C_{33} - \rho_o \gamma(k) C_L^2(k) \qquad (4.37a)$$

where we have introduced

$$\gamma(k) \equiv \left[1 + \frac{\beta_L^2(k)C_L^2(k)}{C_v(k)} T_o\right] . \qquad (4.37b)$$

(a consistent notation would be γ_{33}. For simplicity we let $\gamma \equiv \gamma_{33}$). The quantity $\gamma(k)$ reduces to the conventional ratio $\gamma = (C_p/C_v)$ in the limit of $k \to 0$ (the product $\sqrt{\gamma}\, C_L(k)$ can be interpreted as the longitudinal adiabatic speed of sound. However, such identifications for large values of k become ambiguous, and in fact are not needed. The important point is that all the elements of $\Sigma_{\mu\nu}$ are expressible in terms of the integrals involving the two- and three-particle static correlation function and interpartical potential.)

For Σ_{32} we find from (4.35a)

$$\Sigma_{32} = C_{32} - \rho_o C_L^2(k)\gamma_{32} \qquad (4.38a)$$

where

$$\gamma_{32} = \left[\frac{\langle \rho \Pi_{22}^* \rangle}{\langle J_1 J_1^* \rangle} + \beta_L \frac{\langle T\Pi_{22}^* \rangle}{\langle J_1 J_1^* \rangle}\right] . \qquad (4.38b)$$

We note that γ_{32} is different from γ in (4.37b). They become identical in the isotropic limit mentioned above.

It is known that in a crystal with cubical symmetry, the elastic moduli tensor has only three independent components as discussed by Kittel:[23]

$$C_{\mu\nu} = \left[\begin{array}{ccc|ccc} C_{11} & C_{12} & C_{12} & & & \\ C_{12} & C_{11} & C_{12} & & 0 & \\ C_{12} & C_{12} & C_{11} & & & \\ \hline & & & C_{44} & & 0 \\ & & & & C_{44} & \\ & 0 & & 0 & & C_{44} \end{array}\right] \qquad (4.39)$$

As an approximation we shall assume that both $\Sigma_{\mu\nu}$ and $C_{\mu\nu}$ have this form. The numerical values of C_{11}, C_{12} and C_{44} are plotted in Fig. 1. The values of $C_L^2(k)$ are determined from the knowledge of S(k) (cf. 4.29b). Therefore, if we approximate γ and γ_{32} by their limit for small k, we can compute the elements Σ_{12}, Σ_{11} and Σ_{44}.

We now focus our attention upon (4.27d) and (4.27e) which are generalizations of Maxwell's model for relaxation of the viscous stress tensor and Fourier's law. They both contain the relaxation frequency matrices $W^{\sigma}_{\mu\nu}$ and W^{q}_{jm}. Using the former and the relation $\sigma_{ij} = \eta_{ij\ell m}\varepsilon_{\ell m}$ valid for small frequencies wave numbers, we may define a "viscosity" tensor $\eta_{\mu\zeta}$ as

$$\Sigma_{\mu\nu} = \eta_{\mu} W^{\sigma}_{\xi\nu} \tag{4.40}$$

It is consistent with (4.39) to assume the same symmetry for $\eta_{\mu\zeta}$ as $\Sigma_{\mu\nu}$. Then, the viscosity tensor also contains only three independent elements η_{11}, η_{12} and η_{44}. Since the inverse of a matrix of the form of (4.39) has also the same form we find that the frequency matrix $W^{\sigma}_{\zeta\nu}$ posses only three independent frequencies ω_{11}, ω_{12} and ω_{44}:

$$\omega_{11} = \frac{\eta_{11}\Sigma_{11}+\eta_{12}(\Sigma_{11}-2\Sigma_{12})}{(\eta_{11}-\eta_{12})(\eta_{11}+2\eta_{12})} \tag{4.41a}$$

$$\omega_{12} = \frac{\Sigma_{12}\eta_{11}-\Sigma_{11}\eta_{12}}{(\eta_{11}-\eta_{12})(\eta_{11}+2\eta_{12})} \tag{4.41b}$$

$$\omega_{44} = \frac{\Sigma_{44}}{\eta_{44}} . \tag{4.41c}$$

These frequencies as well as components of the viscosity tensor are functions of k, which have to be determined from either (4.25a), or from the asymptotic

behavior of the correlation functions as discussed below.

In the case of W^{q}_{ij}, we define a thermal diffusivity $\underline{\underline{D}}_T(k)$

$$\underline{\underline{D}}_T(k) = [\underline{\underline{W}}^{q}]^{-1}_{ij} K_L(k) \tag{4.42a}$$

On the basis of symmetry arguments it is reasonable to assume that W^{q}_{ij} in diagonal with elements $\omega^{q}_{11} = \omega^{q}_{22}$ and $\omega^{q}_{33} \equiv \omega_o$. Only ω_o appears in the calculation of the longitudinal current correlations. The corresponding component of $\underline{\underline{D}}_T$ is

$$D_T(k) \equiv \frac{K_L(k)}{\omega_o(k)} , \tag{4.42b}$$

which reduces to the conventional thermal diffusivity in the limit of $k \to 0$.

TRANSVERSE EQUATIONS

The projection of $\underline{J}$ on a fixed axis perpendicular to $\underline{k}$ satisfies the following equations obtained from (4.19) and (4.20)

$$\dot{J}_1(t) = ik\sigma_4(t) , \tag{4.43a}$$

$$\dot{\sigma}_4(t) - \frac{ik}{\rho_o} C_{44} J_1(t) + W^{\sigma}_{4\nu}\sigma_\nu(t) = f^{\sigma}_{\mu}(t) . \tag{4.43b}$$

The assumption of cubical symmetries removes the coupling between $\sigma_4(t)$ and other components of $\sigma_\nu(t)$, because it implies $W^{\sigma}_{4\nu} = \delta_{4\nu}\omega_{44}$. If we let $\omega_1 = \omega_s$ in (4.43), they become identical to (3.8) in the Markov limit, which was obtained by using a two-component description. Hence, we can use (3.14) to compute $\omega_{44}(k)$. Using (3.11b) and (4.41c) we also find that

$$\eta_{44}(k) = \eta_s(k,0) \tag{4.44}$$

which enables us to interpret $\eta_{44}(k)$ as the k-dependent shear viscosity. It is important to note that Eq. (3.8), corresponding to (4.43) in the hydrodynamic description, is exact; whereas (4.43) are approximate. It therefore appears that the damping term in (3.8b) automatically includes all the coupling effects which are ignored in (4.43b). The definition of ω_{44} obtained from (4.25a) as a component of the multidimensional damping matrix is not identical to (3.14) because they involve different projection operators.

LOGITUDINAL EQUATIONS

The compenent of $\underline{J}$ parallel to $\underline{k}$ satisfies the following set of equations:

$$\left.\begin{aligned}
&\dot{\rho}(t) = ikJ_3(t)\\
&\dot{J}_3(t) - ikC_L^2[\rho(t)+\beta_L\rho_o T(t)] = ik\sigma_3(t)\\
&\dot{\sigma}_3(t) - \frac{ik}{\rho_o}\Sigma_{11}J_3(t) + \omega_{12}[\sigma_1(t)+\sigma_2(t)] + \omega_{11}\sigma_3(t) = f_{\underline{3}}^{\sigma}(t)\\
&\dot{\sigma}_1(t) - \frac{ik}{\rho_o}\Sigma_{12}J_3(t) + \omega_{12}\sigma_1(t) + \omega_{11}\sigma_2(t) + \omega_{12}\sigma_3(t) = f_{\underline{1}}^{\sigma}(t)\\
&\dot{\sigma}_2(t) - \frac{ik}{\rho_o}\Sigma_{12}J_3(t) + \omega_{11}\sigma_1(t) + \omega_{12}\sigma_2(t) + \omega_{12}\sigma_3(t) = f_{\underline{2}}^{\sigma}(t)\\
&\dot{\theta}(t) - \beta_L C_L^2 T_o\dot{\rho}(t) = ikq_3(t)\\
&\dot{q}_3(t) - \omega_o[q_3(t)-ikD_T\theta(t)] = f_{\underline{3}}^{q}(t)
\end{aligned}\right\} \qquad (4.45)$$

where we have already introduced the cubical symmetry. Multiplying these equations by $J_3^*(o)$, taking ensemble averages, using $\langle f_{\underline{\mu}}^{\sigma}(t)J_3^*(o)\rangle = \langle f_{\underline{j}}^{q}(t)J_3^*(o)\rangle = 0$, and Laplace transforming with respect to time, we obtain the following expression

for the longitudinal current power spectral density

$$R_{11}(k,\omega) \equiv \int_o^\infty dt \cos\omega t \frac{\langle J_3(t) J_3^*(o)\rangle}{\langle J_3 J_3^*\rangle} \tag{4.46a}$$

as

$$R_{11}(k,\omega) = \left[i\left(\omega - \frac{k^2 C_L^2}{\omega}\right) + \frac{k^2}{\rho_o}\eta_L(k,i\omega) + C_L^2 k^2(\gamma-1)\frac{1}{i\omega + k^2\delta(k,i\omega)}\right]^{-1} . \tag{4.46b}$$

where

$$\eta_L(k,i\omega) \equiv \frac{\rho_o}{ik}\frac{\langle \bar{\sigma}_3(i\omega) J_3^*\rangle}{\langle \bar{J}_3(i\omega) J_3^*\rangle} \tag{4.47a}$$

$$= \eta_B(k,i\omega) + \frac{4}{3}\eta_s'(k,i\omega) \tag{4.47b}$$

$$\eta_B(k,i\omega) \equiv \frac{\rho_o}{ik}\frac{\langle 1/3\ \mathrm{Tr}\bar{\sigma}_{ij}(i\omega) J_3^*\rangle}{\langle \bar{J}_3(i\omega) J_3^*\rangle} = \frac{1}{3}\frac{\Sigma_{11}+2\Sigma_{12}}{i\omega+\omega_B} \tag{4.48a}$$

$$\omega_B \equiv \omega_{11} + 2\omega_{12} = \frac{\Sigma_{11}+2\Sigma_{12}}{\eta_{11}+2\eta_{12}} \tag{4.48b}$$

$$\eta_s'(k,i\omega) \equiv \frac{1}{2}\frac{\Sigma_{11}-\Sigma_{12}}{i\omega+\omega_s'} , \tag{4.49a}$$

$$\omega_s' \equiv \omega_{11} - \omega_{12} = \frac{\Sigma_{11}-\Sigma_{12}}{\eta_{11}-\eta_{12}} \tag{4.49b}$$

$$\delta(k,i\omega) \equiv \frac{\langle \bar{q}_3(i\omega) J_3^*\rangle}{ik\langle \bar{\theta}(i\omega) J_3^*\rangle} \tag{4.50a}$$

$$= \frac{D_T(k)}{1+i(\omega/\omega_o)} \tag{4.50b}$$

The variations of $R_{11}(k,\omega)$ with k and ω has also been computed by Rahman[2] for an argon-like simple liquid. We shall interpret his data using an approximate form of (4.46). It is important to note that (4.46) is an exact result if $\eta_L(k,i\omega)$ and $\delta(k,i\omega)$ can be evaluated exactly using their definitions in (4.47a) and (4.50a) in terms of correlation function. The approximations which we have introduced above (i.e., the neglect of the coupling between heat current and viscous stress tensor, the Markov assumption, and the assumption of cubical symmetry), were actually made to evaluate $\eta_L(k,i\omega)$ and $\delta(k,i\omega)$ explicitly. We shall discuss them in some detail. We observe in (4.47a) that $\eta_L(k,i\omega)$ is the sum of two terms. The first term, $\eta_B(k,i\omega)$ may be identified as a generalization of the bulk viscosity as its definition in (4.48a) indicates. The explicit form in (4.48) has been obtained from (4.45) considering $\mathrm{Tr}\sigma_{ij} = \sigma_1+\sigma_2+\sigma_3$. The second term $\eta_s'(k,i\omega)$ defined by (4.49a) is not the k- and ω-dependent shear viscosity $\eta_s(k,i\omega)$ (cf. 3.11) as one might have expected. For small k limit, however, this is indeed the case. In the limit of $k \to 0$, the components of the elastic moduli tensor satisfies[21,23]

$$C_{11} - C_{12} = 2C_{44} . \tag{4.51}$$

Since $\gamma = \gamma_{32}$ in this limit, we also have $\Sigma_{11}-\Sigma_{12} = 2\Sigma_{44} = 2C_{44}$. Furthermore, $\eta_{11}-\eta_{12} = 2\eta_{44}$ because $\eta_{\mu\nu}$ and $\Sigma_{\mu\nu}$ are assumed to have the same form. Then, (4.49) yields $\omega_{11}-\omega_{12} = \omega_{44}$, i.e., the relaxation frequencies for the viscous stress tensor are not independent. Substituting these in (4.49) we obtain

$$\eta_s'(k,i\omega) \to \eta_s(k,i\omega) \text{ (isotropic limit)} \tag{4.52a}$$

and

$$\eta_L(k,i\omega) \to \eta_B(k,i\omega) + (\frac{4}{3})\eta_s(k,i\omega) \qquad (4.52b)$$

If, as an additional approximation we assume $\omega_{11} \gg \omega_{12}$ then, $\omega_B \approx \omega_s = \omega_{11} = \omega_L$, and we have for all k

$$\eta_L(k,i\omega) \approx \frac{C_{11}(k)-\rho_o\gamma(k)C_L^2(k)}{i\omega+\omega_L(k)} \qquad (4.53)$$

Physically, this assumption is equivalent to ignoring the coupling between σ_3 and σ_1 and σ_2 in (4.45) (set $\omega_{12} = 0$). An other consequence of this approximation is that the transverse relaxation frequency $\omega_s = \omega_{44}$ approaches ω_L for small k because $\omega_{44} \to \omega_{11}-\omega_{12} \approx \omega_{11}$. These observations have been verified approximately using Rahman's computer results as will shortly be discussed.

As a result of the k^2 factor multiplying $\delta(k,i\omega)$ in (4.46b), the thermal effects become insignificant for large values of k (e.g., $k \geq 1\text{Å}^{-1}$ for argon), and $R_{11}(k,\omega)$ reduces to

$$R_{11}(k,\omega) \simeq \mathrm{Re}\left[i\left(\omega - \frac{k^2C_L^2}{\omega}\right) + \frac{k^2}{\rho_o}\eta_L(k,i\omega)\right]^{-1} \qquad (4.54)$$

We have used (4.54) and (4.53) to interpret Rahman's data (we have set $\gamma(k) \approx 1$ as a further numerical simplification) which may be called the isothermal approximation:

$$R_{11}(k,\omega) \simeq \frac{\omega^2\omega_L(k)k^2[C_{11}(k)-\rho_oC_L^2(k)]/\rho_o}{\omega_L^2(k)[\omega^2-k^2C_L^2(k)]^2+\omega^2(\omega^2-k^2C_{11}(k)/\rho_o)^2} \; . \qquad (4.55)$$

The only unknown in this expression is $\omega_L(k)$. Its value for $k \to 0$ was

estimated using

$$\omega_L(o) = \frac{C_{11}(o)-\rho_o C_L^2(o)}{\eta_B + \frac{4}{3}\eta_s} \tag{4.56}$$

where η_B and η_s are the conventional bulk and shear viscosities, respectively. We note that

$$\lim_{k\to\infty} C_{11}(k) = K_\infty + \frac{4}{3} G_\infty \tag{4.57}$$

where G_∞ was defined in Section 3, and K_∞ is the bulk modulus in Zwanzig's notation.[22] The value of $C_{11}(o)$ was read from Fig. 1, and that of $C_L^2(o)$ (the ordinary isothermal speed of sound) was calculated from (4.29b) (Rahman has also calculated S(k)).

To model the large k behavior of $\omega_L(k)$ we consider the ideal gas,

$$R_{11}^{IG}(k,\omega) = \beta m \frac{\omega^2}{k^2}\sqrt{\frac{\Pi\beta m}{2k^2}}\, e^{-\beta m\omega^2/2k^2}, \tag{4.58}$$

which has a maximum at $\omega_m^2(k) = 2k^2/\beta m$. Requiring that the frequency at which (4.55) attains a maximum for a fixed k approach $2k^2/\beta m$ for large k, and using the same interpolation formula as in (3.20) we obtain the following expression for $\omega_L(k)$:

$$\omega_L^2(k) = \frac{8}{3} k^2 (C_{11}(k)-\rho_o C_L^2(k)-\rho_o/\beta m)/\rho_o + \frac{\omega_L^2(o) - \frac{8}{3} k^2 (C_{11}(k)-\rho_o C_L^2(k)-2\rho_o/\beta m)/\rho_o}{[1+k^2/k_o^2]}, \tag{4.59}$$

where $\omega_L(o)$ is given by (4.56). The variation of $\omega_L(k)$ is shown inFig. 2

($k_o = 1.5\ \text{Å}^{-1}$ as in the case of transverse correlations).

It is interesting to compare again $R_{11}(k,\omega)/\omega^2$ from (4.13a) and $R_{11}^{IG}(k,\omega)/\omega^2$ from (4.18) for $k \to \infty$ and $\omega \to 0$, as in the case of transverse current power spectral density. Using $C_L^2(k) \to (1/\beta m)$ and $C_{11}(k) \to (3\rho_o/\beta m)$ as $k \to \infty$, we find $\bar{R}_{11}(k,o)/\omega^2 \to (\beta m)^{3/2}\sqrt{3/2}\,/k^3$ and $\bar{R}_{11}^{IG}(k,o)/\omega^2 \to (\beta m)^{3/2}\sqrt{\pi/2}\,/k^3$. Thus, the approximate formula for $\bar{R}_{11}(k,\omega)$ yields the ideal gas limit for small ω and large k within a factor $\sqrt{3/\pi} \approx 0.98$.

The curves in Figs. 7 and 8 are calculated using (4.55) and (4.59). The agreement between Rahman's computer results and the theoretical curves are remarkably good.

The thermal effects dominate for small values of k such as those involved in light scattering.[13] We then approximate (4.46b) as

$$R_{11}(k,\omega) = \text{Re}\left[i\left(\omega - \frac{k^2 C_L^2(o)}{\omega}\right) + \frac{k^2}{\rho_o}\,\eta_L(o,o) + \frac{C_L^2(o)k^2(\gamma-1)}{i\omega + k^2 D_T(o)}\right]^{-1} . \quad (4.60)$$

which is the result corresponding to the conventional hydrodynamic description with constant transport parameters. Since the variation of the thermal diffusivity $D_T(k)$, thermal relaxation frequency and $\gamma(k)$ are not readily available, we may use their limiting values as $k \to 0$ in (4.46b), and substitute $\eta_L(k,\omega)$ from (4.59) to find

$$R_{11}(k,\omega) = \text{Re}\left[i\left(\omega - \frac{k^2 C_L^2(k)}{\omega}\right) + \frac{k^2}{\rho_o}\,\frac{C_{11}(k) - \rho_o \gamma C_L^2(k)}{i\omega + \omega_L(k)} + k^2 C_L^2(k)(\gamma-1)\left(i\omega + \frac{k^2 D_T(o)}{1+(\omega/\omega_o)}\right)^{-1}\right]^{-1} \quad (4.61)$$

where $\omega_L(k)$ is to be taken from (4.59). All the functions of k appearing in this expression are calculable in terms of S(k) (or g(R)) and interparticle potential, and the constants γ, $D_T(o)$ $\eta_L(o,o)$ are experimentally available.

CONCLUSIONS

In this paper we have used the generalized Langevin equation to investigate fluctuations in simple liquids in the frame-work of a generalized hydrodynamic description. The various transport coefficients appearing in this description have been defined in terms of time-correlations of the dynamical variables, and computed numerically whenever possible for argon-like liquids. The results, such as the variation of viscosities with wavelength, may be applicable to other simple liquids. In view of the good quantitative agreement with Rahman's data for all values of k and ω encountered in neutron scattering, we may expect the final expression (4.61) to be applicable to the interpretation of coherent neutron scattering as well as light scattering from dense fluids. (Figure 8 represents $\omega^2 S(k,\omega) = R_{11}(k,\omega)$ as a function of k and ω. No attempts have been made to compare these to actual neutron scattering data because the discussion of the contribution of incoherent scattering which is important in natural argon[14] is beyond the scope of this paper.) The formalism developed in this paper enables one to calculate correlations between other pairs of hydrodynamic variables, e.g., $\langle E(t)E^* \rangle$ and $\langle \underline{Q}(t)\underline{Q}^* \rangle$, $\langle J_1(t)J_{11}^* \rangle$, etc. Computer results for these correlation functions in conjunction with the analytical calculations will shed light on the k- and ω-dependence of the thermal parameters $\gamma(k)$, $\omega_o(k)$ and $D_T(k)$ as we have demonstrated for the viscosities.

ACKNOWLEDGMENTS

We wish to thank Professors R. K. Osborn and J. Duderstadt for their helpful comments and reviewing and editing of this paper. Also we thank Professor S. Yip for discussions and providing his own related calculations prior to publication. The work described above was supported in part by the National Science Foundation.

REFERENCES

(1) Rahman, A., Phys. Rev., 136, A405 (1964).

Rahman, A., Phys. Rev. Letters, 19, 420 (1967).

(2) Rahman, A., in Neutron Inelastic Scattering (International Atomic Energy Agency, Vienna, 1968), Vol. I.

(3) Nelkin, M, VanLeeuwen, J.M.J., and Yip, S., Inelastic Scattering of Neutrons in Solids and Liquids, Vienna (1965), Vol. II, 35-57.

(4) VanLeeuwen, J.M.J. and Yip, S., Phys. Rev. 139, A1138 (1965).

(5) Nelkin, M. and Yip, S., Phys. Fluids, 9, 380 (1966).

(6) Greytak, J. J. and Benedek, G. B., Phys. Rev., Letters, 17, 179 (1966).

(7) Nelkin, M. and Ranganathan, S., Phys. Rev., 164, 222 (1967),

Ranganathan, S. and Nelkin, M., Journ. Chem. Phys., 47, 4056 (1967).

(8) Nelkin, M. and Ortoleva, P. J., IAEA Symp. Inelastic Neutron Scattering (Copenhagen, 1969).

(9) Callen H. B. and Welton, T. A., Phys. Rev., 83, 34 (1951),

Kubo, R., Tokyo Summer Lectures in Theoretical Physics, Part 1, Many-Body Theory (1966).

(10) Landaue, L. D. and Litschitz, E. M., Statistical Physics (Pergamon Press, London-Paris, 1959).

(11) Rytov, S. M., Soviet Phys. JETP, 6, 130 (1958).

(12) R. Mountain, Rev. Mod. Phys., 38, 205 (1966).

(13) Kadonoff L. P. and Martin, P. C., Annals of Physics, 24, 419 (1963).

(14) Chung, C. H. and Yip, S., to be published in Phys. Rev. (1969).

(15) Zwanzig, R., "Statistical mechanics of irreversibility," in Lectures in Theoretical Physics, ed. by W. E. Brittin, (John Wiley, New York, 1961), Vol. 3.

(16) Mori, H., Progress of Theoretical Physics, Vol. 33, No. 3, March 1965.

(17) Mori, H., Progress of Theoretical Physics, 34, 399 (1965).

(18) Akcasu, A. Z. and Duderstadt, J. J., to be published in Phys. Rev.

(19) Zwanzig, R., Phys. Rev., 144, 170 (1966).

(20) Wang, M. C. and Uhlenbeck, G. E., Rev. Mod. Phys., 17, 323 (1965); Chandrasekhar, S., Rev. Mod. Phys., 15, 1 (1943).

(21) Schofield, P., Proc. Phys. Soc., 88, 149 (1966).

(22) Zwanzig, R., and Mountain, R., J. Chem. Phys., 43, 4464 (1965).

(23) Kittel, C., Introduction to Solid State Physics (John Wiley, New York, 1953).

FIGURE CAPTIONS

Fig. 1. Variation of elastic moduli with wavelength.

Fig. 2. The variation of the transverse and longitudinal relaxation frequencies with wavelengths.

Fig. 3. The variation of shear viscosity with wavelength.

Fig. 4. The transverse current-current correlation, $R_{\perp}(k,\omega)$, vs. ω for various k.

Fig. 5.1. The maxima of a transverse current-current correlation function vs. wave number, k. Solid line due to present theory and points represent Rahman's data.

Fig. 5.2. The frequency at which the transverse current-current correlation function is maximum.

Fig. 5.3. The transverse current-current correlation at zero frequency.

Fig. 6. The longitudinal current-current correlation function vs. ω for various k. The points are from Rahman.

Fig. 7.1. The maxima of the longitudinal current-current correlation function vs. wave number, k. The points are from Rahman.

Fig. 7.2. Frequency at which the longitudinal current-current correlation function is maximum.

Fig. 8. The scattering function, $S(k,\omega)$, vs. ω for various k.

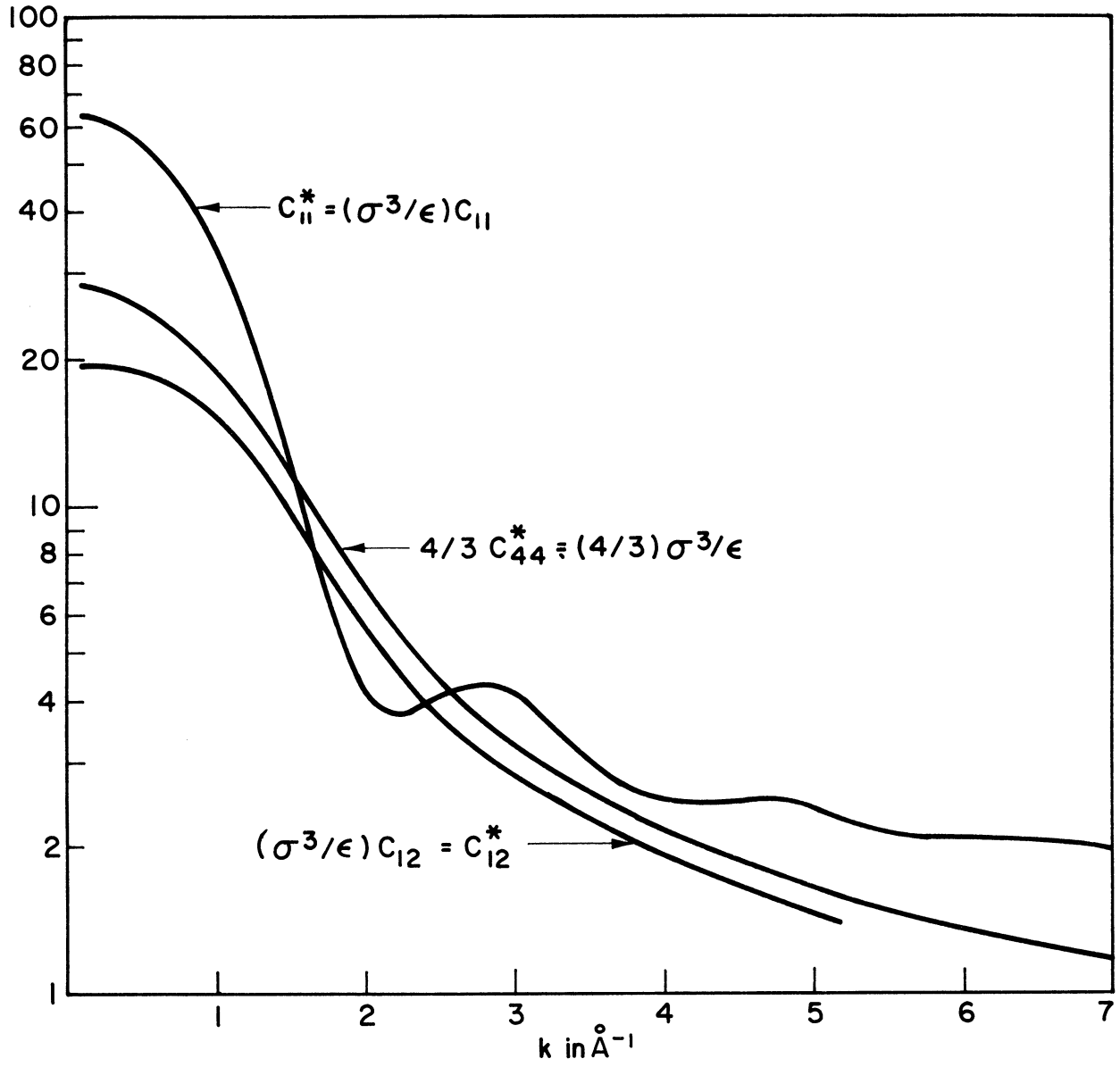
100
80
60
40
20
10
8
6
4
2
1
$C_{11}^* = (\sigma^3/\epsilon)C_{11}$
$4/3\ C_{44}^* = (4/3)\sigma^3/\epsilon$
$(\sigma^3/\epsilon)C_{12} = C_{12}^*$
1
2
3
4
5
6
7
k in Å^{-1}

Fig. 1

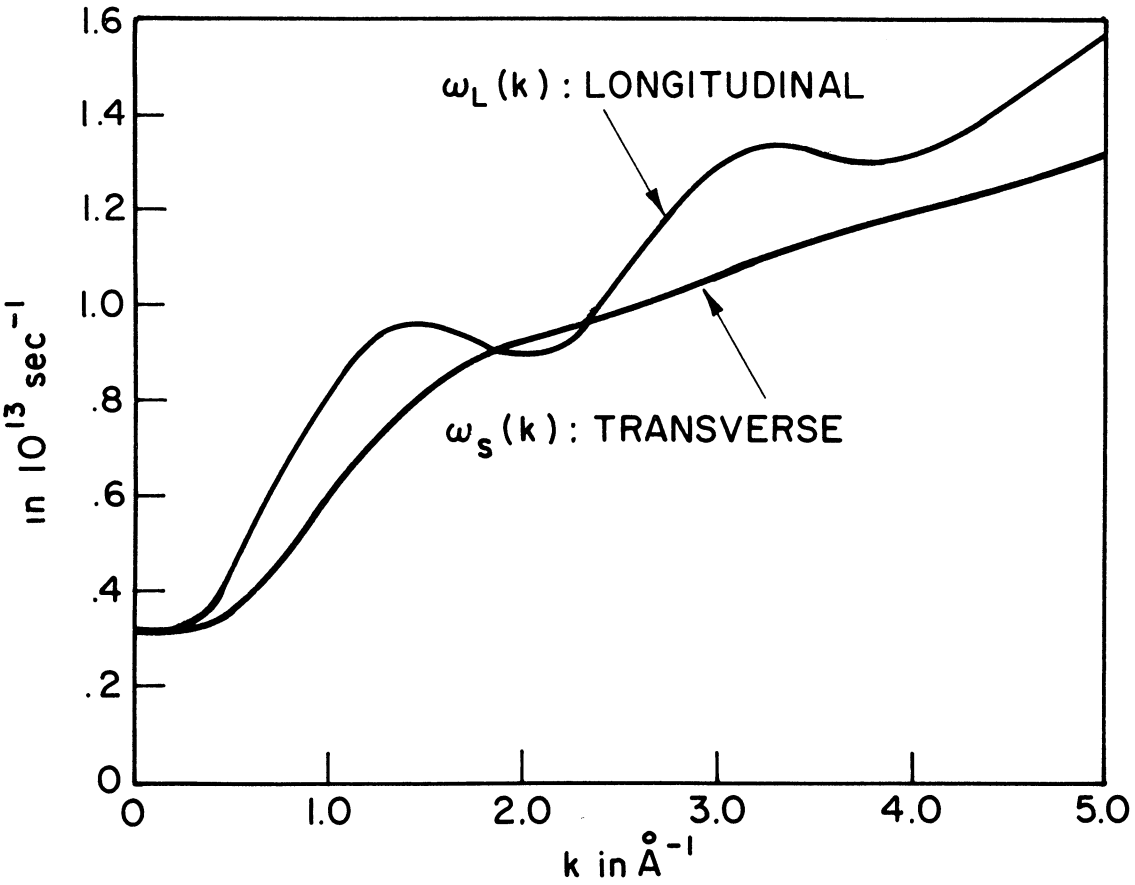
1.6
1.4
1.2
1.0
.8
.6
.4
.2
0
0
1.0
2.0
3.0
4.0
5.0
$\omega_L(k)$: LONGITUDINAL
$\omega_s(k)$: TRANSVERSE
in 10^{13} sec^{-1}
k in $Å^{-1}$

Fig. 2

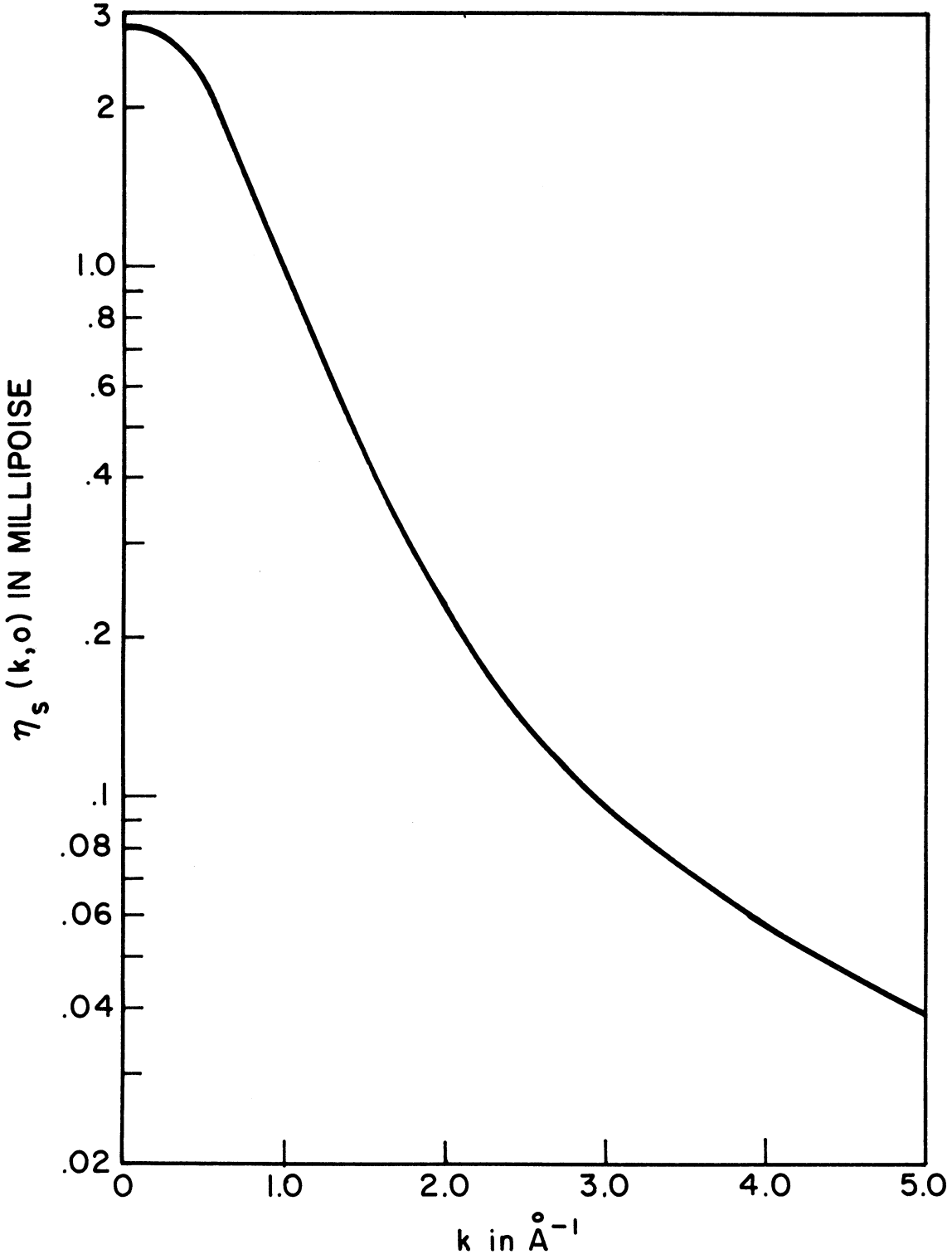
3
2
1.0
.8
.6
.4
.2
.1
.08
.06
.04
.02
η_s (k,o) IN MILLIPOISE
0
1.0
2.0
3.0
4.0
5.0
k in Å^{-1}

Fig. 3

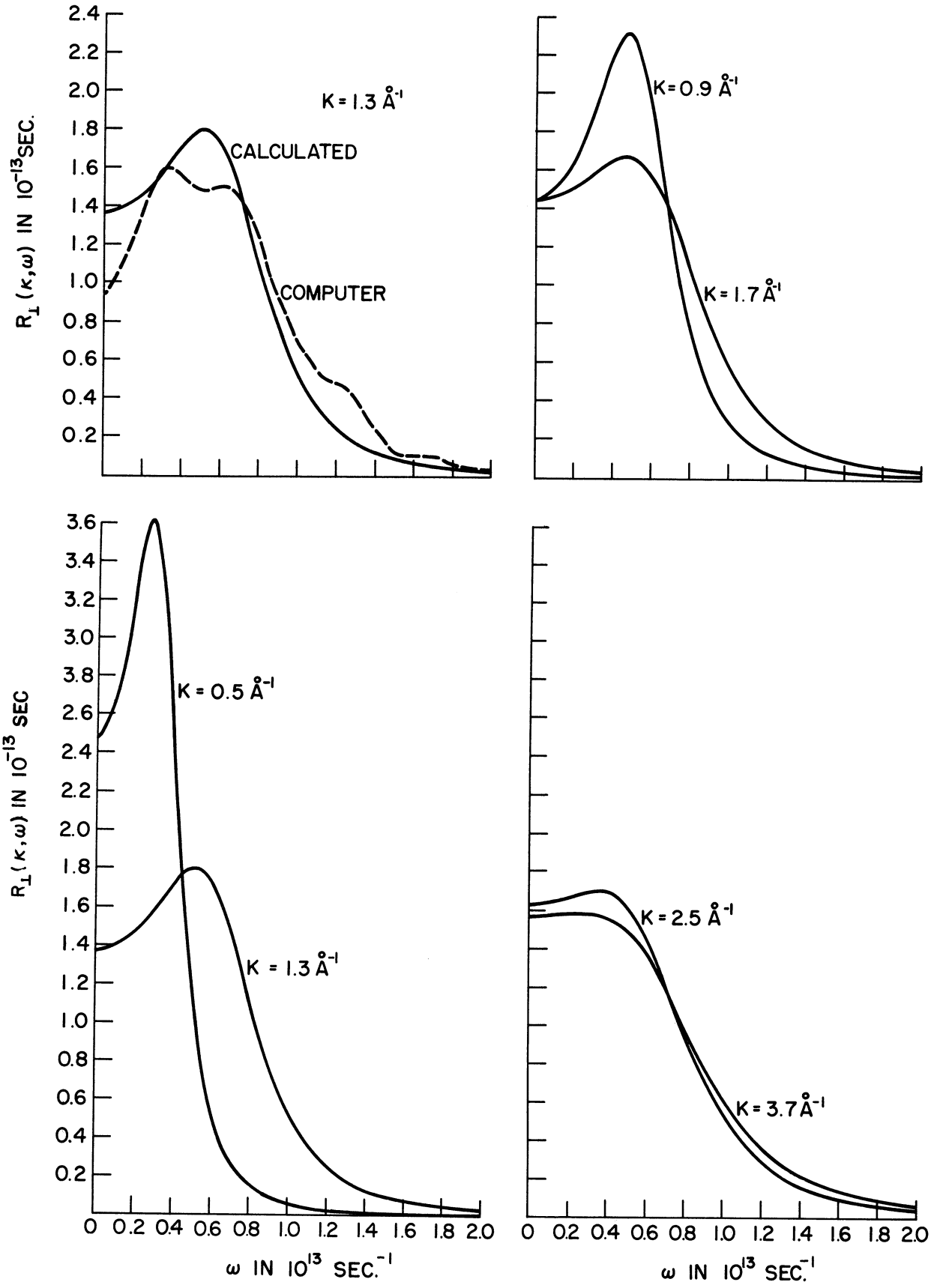
K = 1.3 Å⁻¹
CALCULATED
COMPUTER
K = 0.9 Å⁻¹
K = 1.7 Å⁻¹
K = 0.5 Å⁻¹
K = 1.3 Å⁻¹
K = 2.5 Å⁻¹
K = 3.7 Å⁻¹
R⊥(κ,ω) IN 10⁻¹³ SEC.
ω IN 10¹³ SEC.⁻¹

Fig. 4

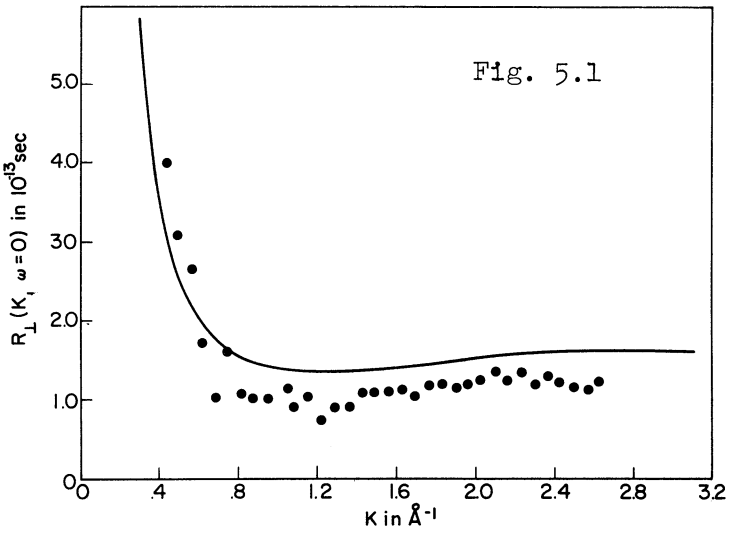
Fig. 5.1
$R_\perp(K, \omega=0)$ in 10^{13} sec
5.0
4.0
3.0
2.0
1.0
0
0
.4
.8
1.2
1.6
2.0
2.4
2.8
3.2
K in Å^{-1}

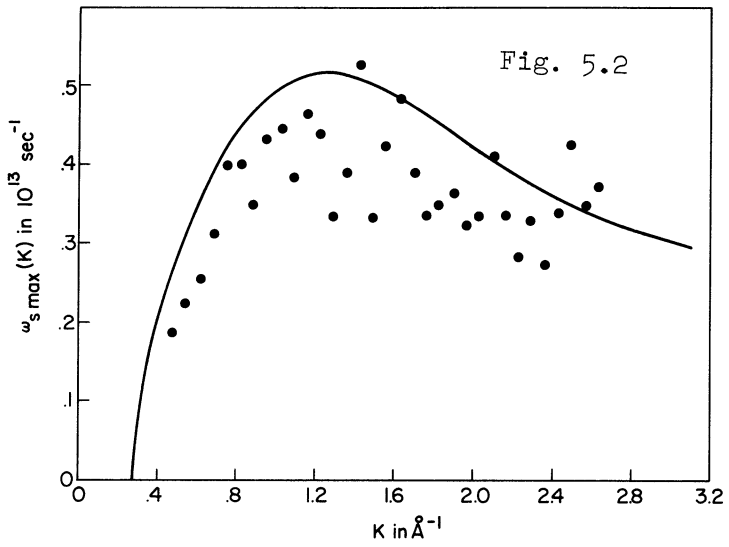
Fig. 5.2
$\omega_{s\,max}(K)$ in 10^{13} sec^{-1}
.5
.4
.3
.2
.1
0
0
.4
.8
1.2
1.6
2.0
2.4
2.8
3.2
K in Å^{-1}

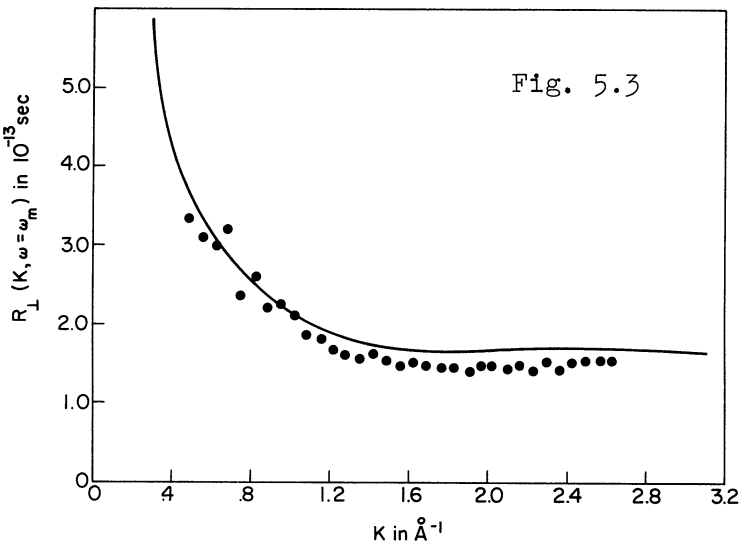
Fig. 5.3
$R_\perp(K, \omega=\omega_m)$ in 10^{-13} sec
5.0
4.0
3.0
2.0
1.0
0
0
.4
.8
1.2
1.6
2.0
2.4
2.8
3.2
K in Å^{-1}

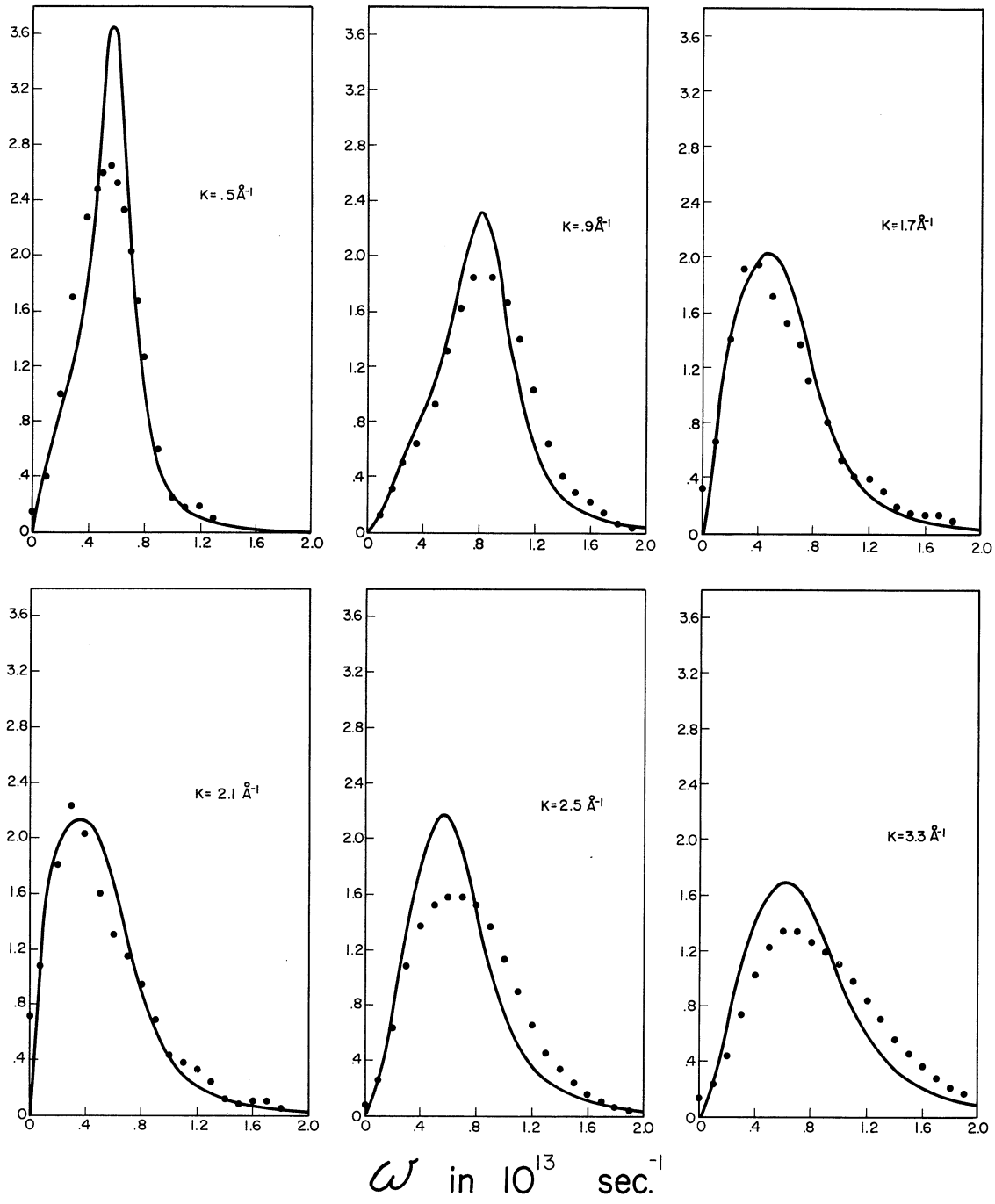

Fig. 6

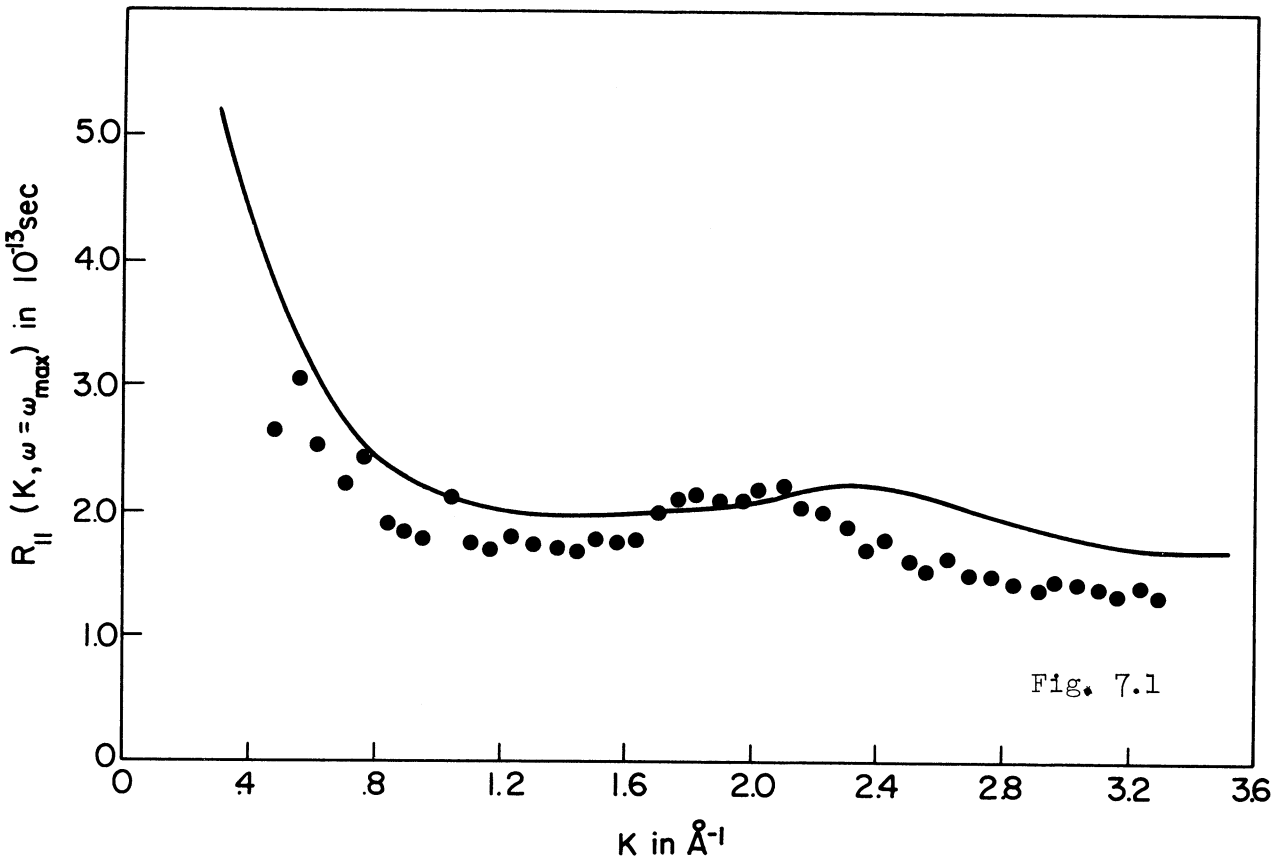
$R_{11}(K, \omega = \omega_{max})$ in 10^{-13}sec
5.0
4.0
3.0
2.0
1.0
0
0
.4
.8
1.2
1.6
2.0
2.4
2.8
3.2
3.6
K in Å^{-1}

Fig. 7.1

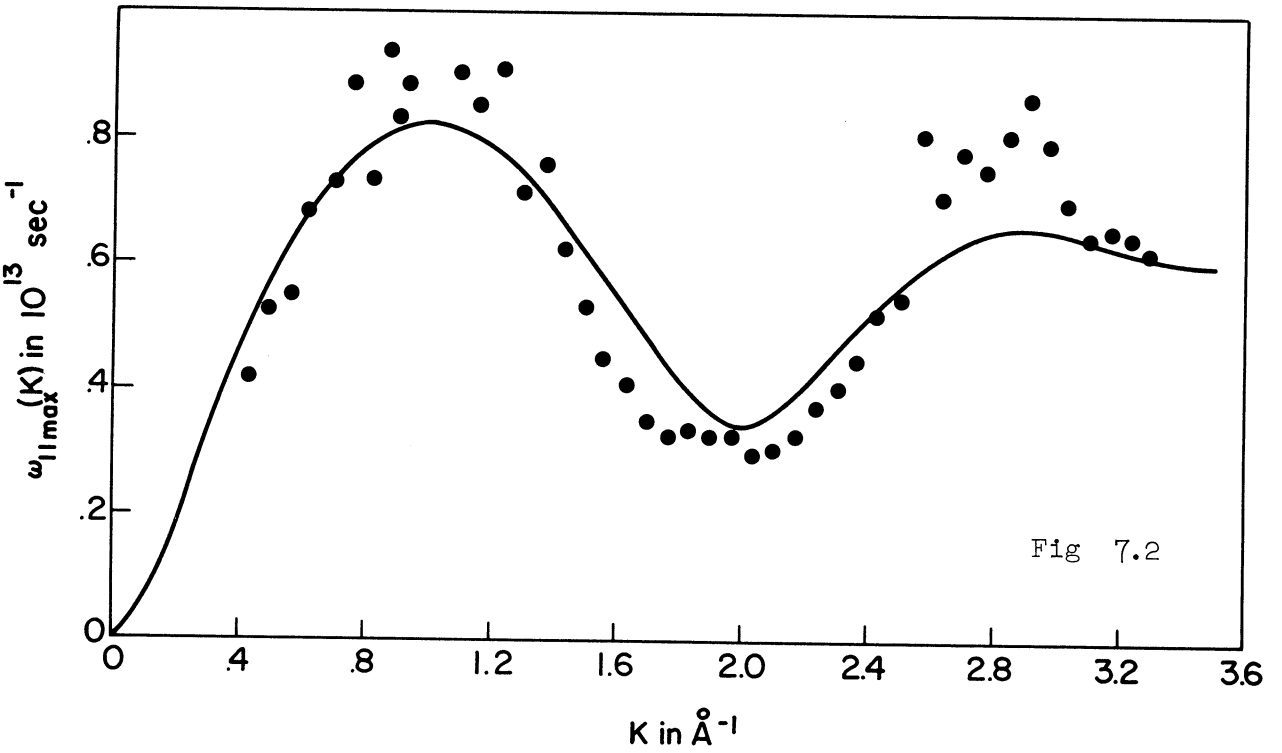
$\omega_{11max}(K)$ in 10^{13} sec^{-1}
.8
.6
.4
.2
0
0
.4
.8
1.2
1.6
2.0
2.4
2.8
3.2
3.6
K in Å^{-1}

Fig 7.2

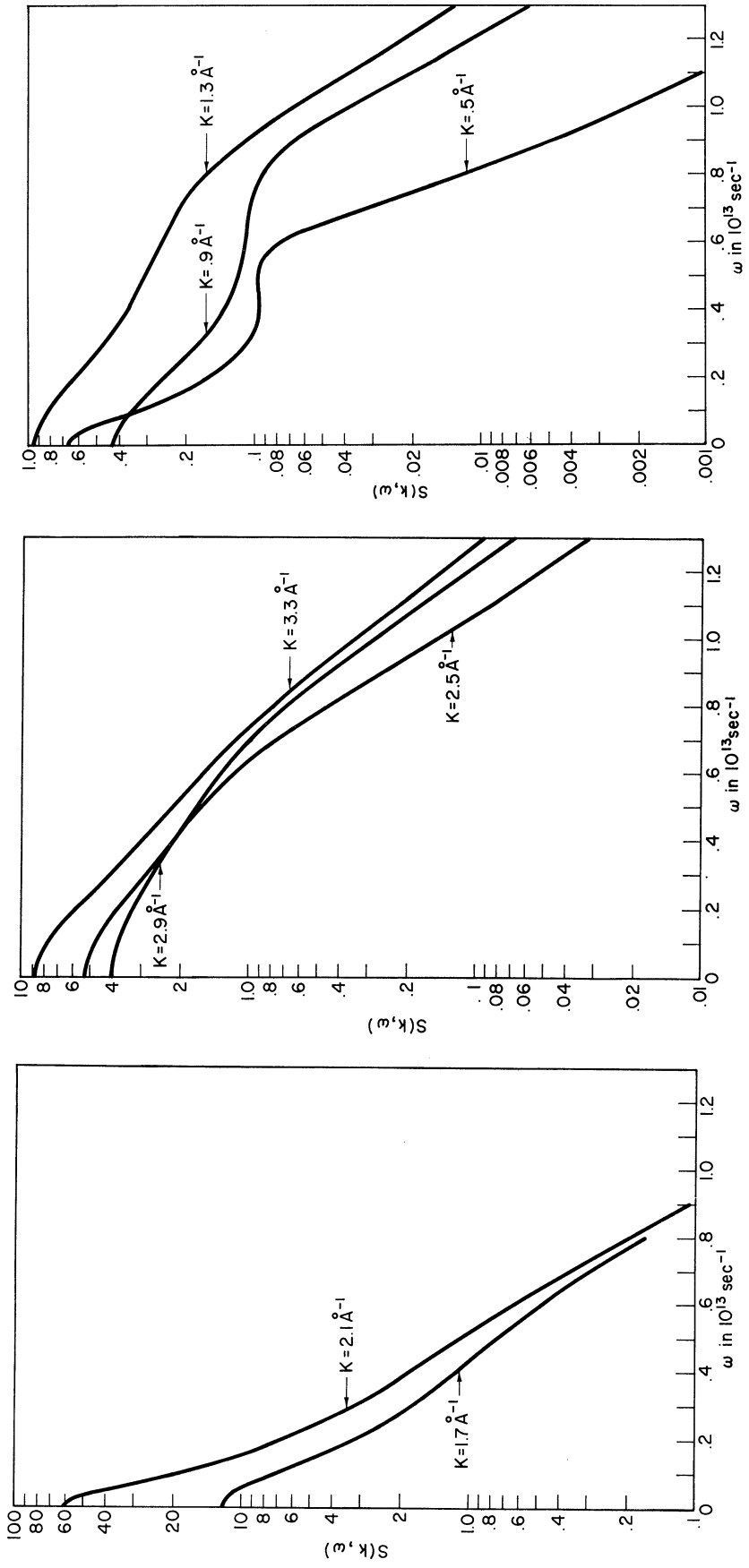
S(k,ε)
ω in 10^13 sec^-1
K=1.7Å^-1
K=2.1Å^-1
K=2.9Å^-1
K=3.3Å^-1
K=2.5Å^-1
K=.9Å^-1
K=1.3Å^-1
K=.5Å^-1

Fig. 8