

## Collective Effects and the Hydrodynamic Approximation in Neutron Scattering from Fluids\*

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The scattering law is investigated for gases and liquids. It is shown that the convolution approximation ignores perturbations caused by the test particle for  $t > 0$ . For a dilute gas, a normal-mode expansion of the Boltzmann equation is made to estimate the size of the "collective effects" contribution to the scattering law. It is found that for small energy and momentum transfers the "collective-effects" contribution dominates the solution. The hydrodynamic and convolution approximations are compared to experimental results for liquid sodium.

### I. INTRODUCTION

THAT the neutron scattering cross section is a product of two terms, one dependent on the properties of the incident neutron and the other on the properties of the scattering system, has been shown by Van Hove.<sup>1</sup> The latter factor will be denoted by  $S(\mathbf{k}, \omega)$ , where  $\hbar\mathbf{k}$  is the momentum transfer and  $\hbar\omega$  the energy transfer. It is related to classical space-time correlation functions by<sup>2,3</sup>

$$S(\mathbf{k}, \omega) = \exp\left(\frac{\hbar\omega}{2kT} - \frac{\hbar k^2}{8mkT}\right) a^{-2} [a_{\text{inc}}^2 S_s^{CL}(\mathbf{k}, \omega) + a_{\text{coh}}^2 S^{CL}(\mathbf{k}, \omega)] + O(\hbar^2), \quad (1.1)$$

where  $S^{CL}(\mathbf{k}, \omega)$  is the space-time Fourier transform of  $G(\mathbf{r}, t)$ :

$$S^{CL}(\mathbf{k}, \omega) = \frac{1}{(2\pi)^3} \int d^3r \int dt \exp[i(\mathbf{k} \cdot \mathbf{r} - \omega t)] G(\mathbf{r}, t), \quad (1.2)$$

and  $S_s^{CL}(\mathbf{k}, \omega)$  is the similarly defined Fourier transform of the self (or "test" particle) portion of the correlation function  $G_s$ . One writes

$$G(\mathbf{r}, t) = G_s(\mathbf{r}, t) + G_d(\mathbf{r}, t), \quad (1.3)$$

where  $G_d(\mathbf{r}, t)$  represents the probability that there be a particle at  $(\mathbf{r}, t)$  different from the one that was at the origin at  $t=0$ . Relation (1.1) has been derived for the self-correlation function by Aamodt *et al.*,<sup>4</sup> but a derivation of the complete relation is still lacking.

In general, it is quite simple to formulate models for calculating  $G_s(\mathbf{r}, t)$ , and many such models have been

proposed.<sup>5-8</sup> Calculation of  $G_d(\mathbf{r}, t)$  is more difficult and considerably less progress has been made in developing a theory for obtaining this quantity. This paper is mainly devoted to a discussion of this problem.

Vineyard<sup>9</sup> wrote

$$G_d(\mathbf{r}, t) = \int g(\mathbf{r}') H(\mathbf{r}, \mathbf{r}', t) d^3r', \quad (1.4)$$

where  $g(\mathbf{r})$  is the ordinary static pair correlation function and  $H(\mathbf{r}, \mathbf{r}', t)$  is the conditional probability that, given a particle at the origin and a second particle at  $\mathbf{r}'$  at time  $t=0$ , the second particle will be found at  $\mathbf{r}$  at time  $t$ . Vineyard (and most other authors) took  $H(\mathbf{r}, \mathbf{r}', t)$  to be  $G_s(|\mathbf{r}-\mathbf{r}'|, t)$ . This is the well-known and widely used convolution approximation. That  $H(\mathbf{r}, \mathbf{r}', t)$ , the function describing the motion of the "field" particles, cannot be the same as  $G_s$  has been well appreciated, but it has not been stressed that the convolution approximation ignores perturbations caused by the test particle for  $t > 0$ .

The plan of this paper is as follows: For a dilute gas the equations for  $G(\mathbf{r}, t)$  and  $G_s(\mathbf{r}, t)$  can be found<sup>10,11</sup> and the properties of the functions discussed. For such gases,  $g(\mathbf{r}) = n$ , where  $n$  is the number density. Equation (1.4) would give  $G_d = n$  and hence  $G = G_s + n$ . A direct calculation of  $G$  shows that this is incorrect, and a simple physical reason for the discrepancy can be arrived at. This is done in Sec. II.

Since there has been considerable interest in the effect of collective phenomena on neutron scattering from fluids,<sup>12,13</sup> this subject is discussed for the case of dilute gases in Sec. III. The analysis is based on the normal-mode expansion of the Boltzmann equation.<sup>14</sup>

<sup>5</sup> A. Rahman, K. S. Singwi, and A. Sjölander, *Phys. Rev.* **126**, 986 (1962).

<sup>6</sup> K. S. Singwi and A. Sjölander, *Phys. Rev.* **120**, 1093 (1961).

<sup>7</sup> M. Nelkin and A. Ghatak, *Phys. Rev.* **135**, A4 (1964).

<sup>8</sup> A. G. Gibbs and J. H. Ferziger, *Phys. Rev.* **138**, A701 (1965).

<sup>9</sup> G. H. Vineyard, *Phys. Rev.* **110**, 999 (1958).

<sup>10</sup> J. M. J. VanLeeuwen and S. Yip, *Phys. Rev.* **139**, A1138 (1965).

<sup>11</sup> M. Nelkin, J. M. J. VanLeeuwen, and S. Yip, in *Inelastic Scattering of Neutrons* (International Atomic Energy Agency, Vienna, 1965), Vol. II.

<sup>12</sup> R. C. Desai and M. Nelkin, *Phys. Rev. Letters* **16**, 839 (1966).

<sup>13</sup> S. H. Chen *et al.*, *Phys. Letters* **19**, 269 (1965).

<sup>14</sup> J. K. Buckner and J. H. Ferziger, *Phys. Fluids* (to be published).

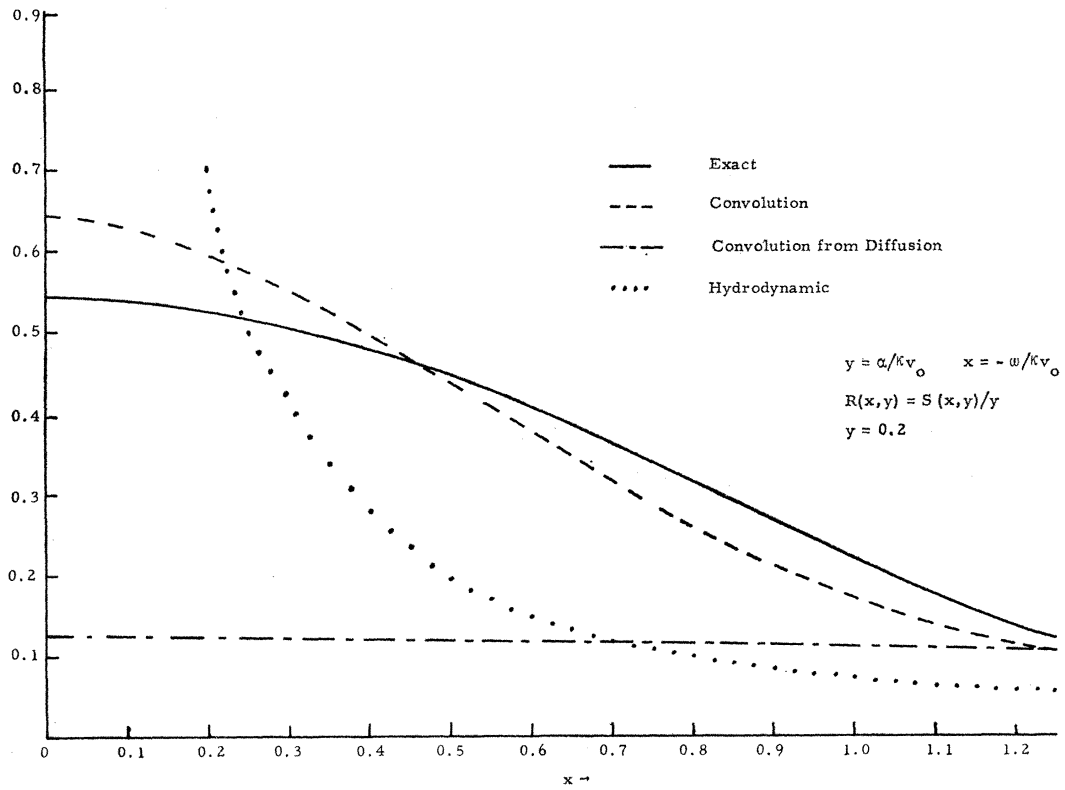
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<sup>1</sup> L. Van Hove, *Phys. Rev.* **95**, 249 (1954).

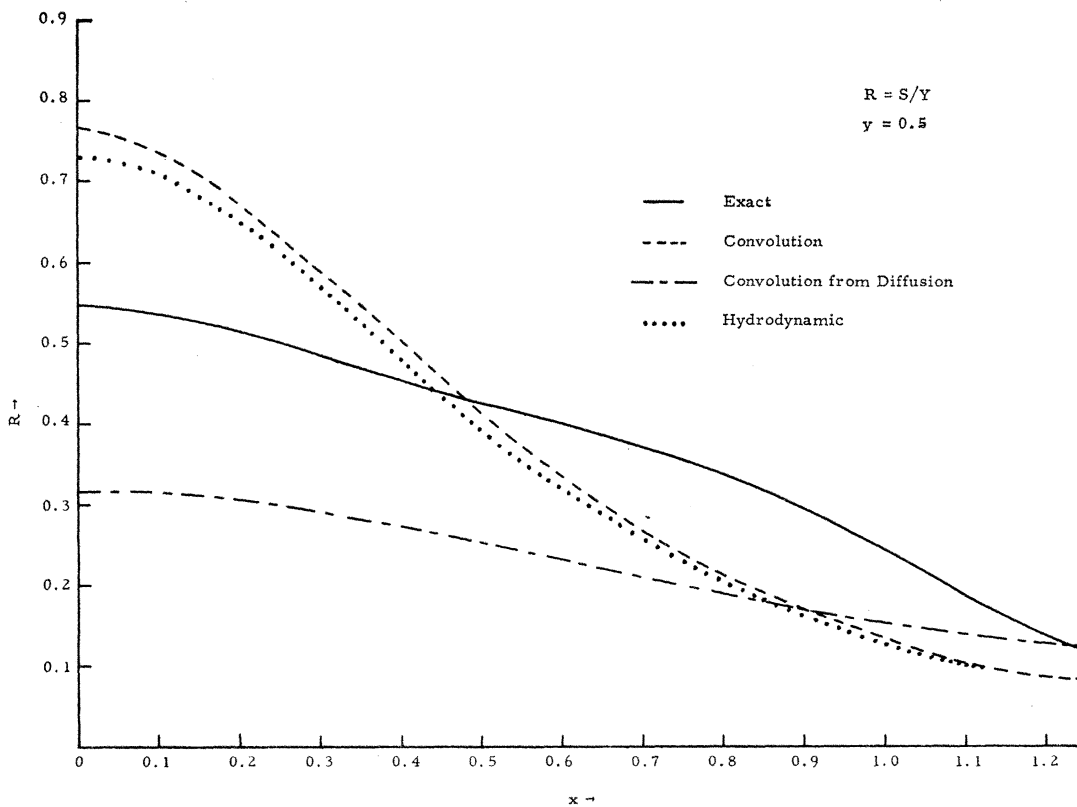
<sup>2</sup> P. Schofield, *Phys. Rev. Letters* **4**, 239 (1960).

<sup>3</sup> P. A. Egelstaff, in *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1961).

<sup>4</sup> R. Aamodt, K. M. Case, M. Rosenbaum, and P. F. Zweifel, *Phys. Rev.* **126**, 1165 (1962).



(a)



(b)

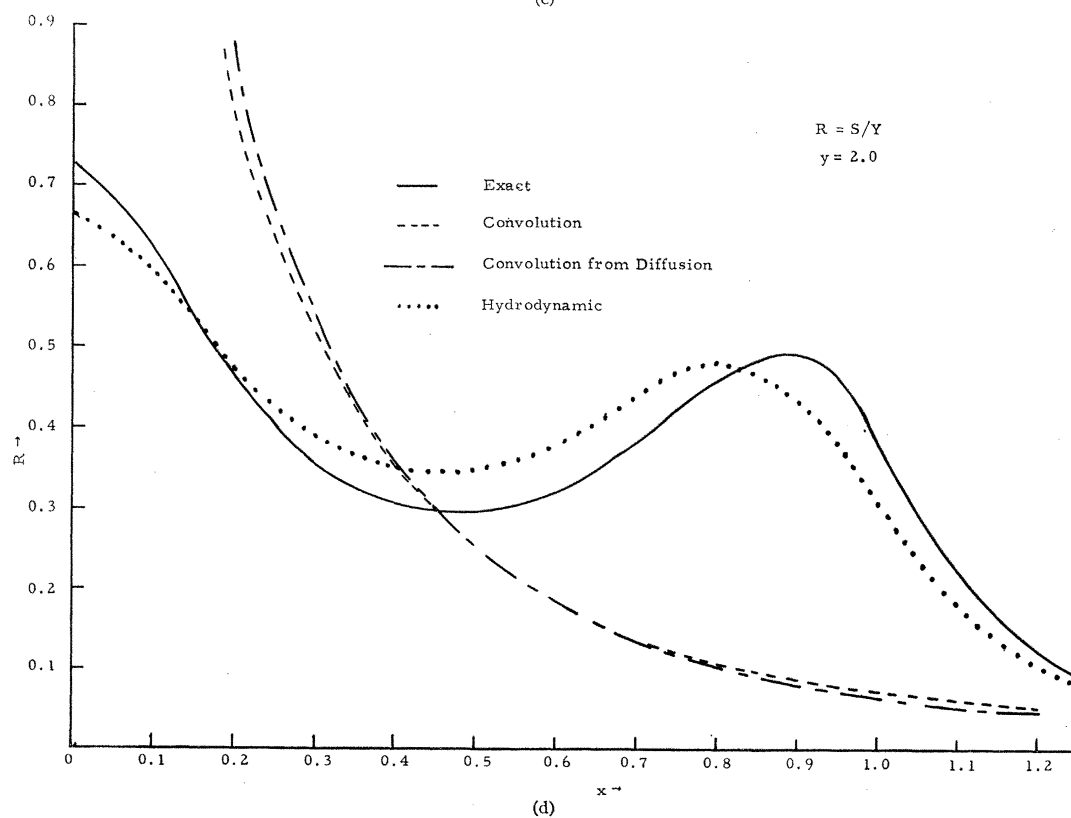
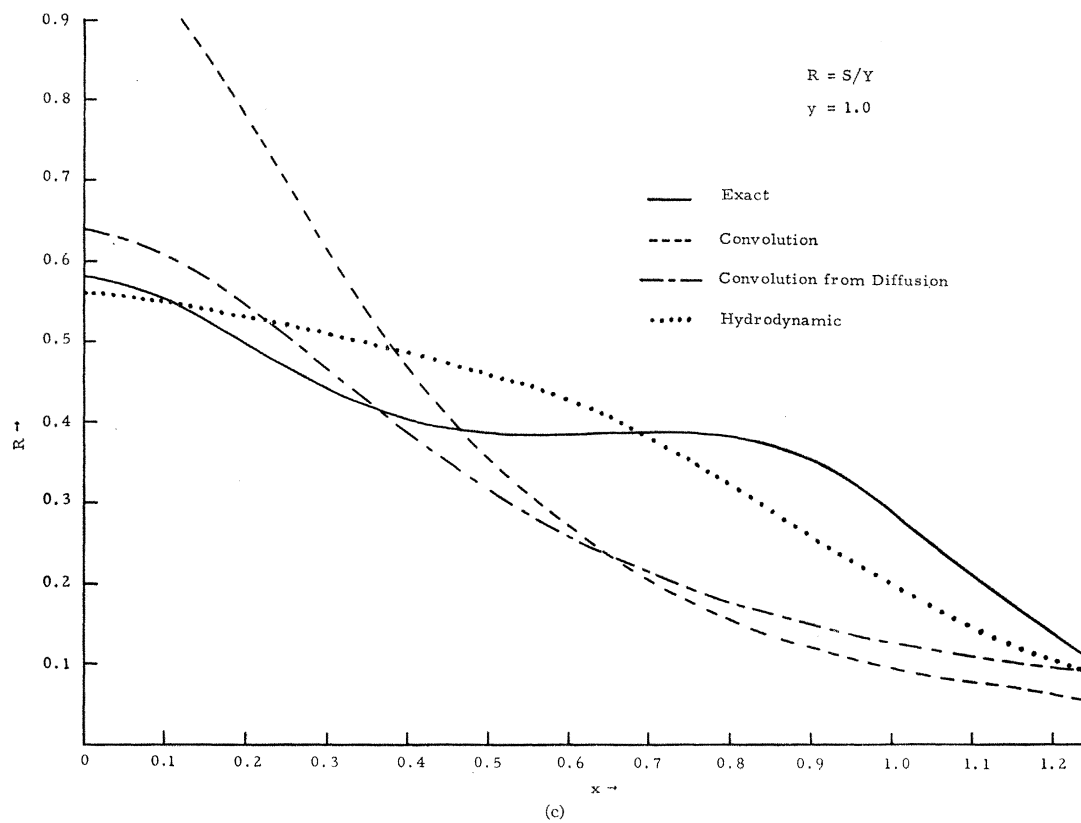
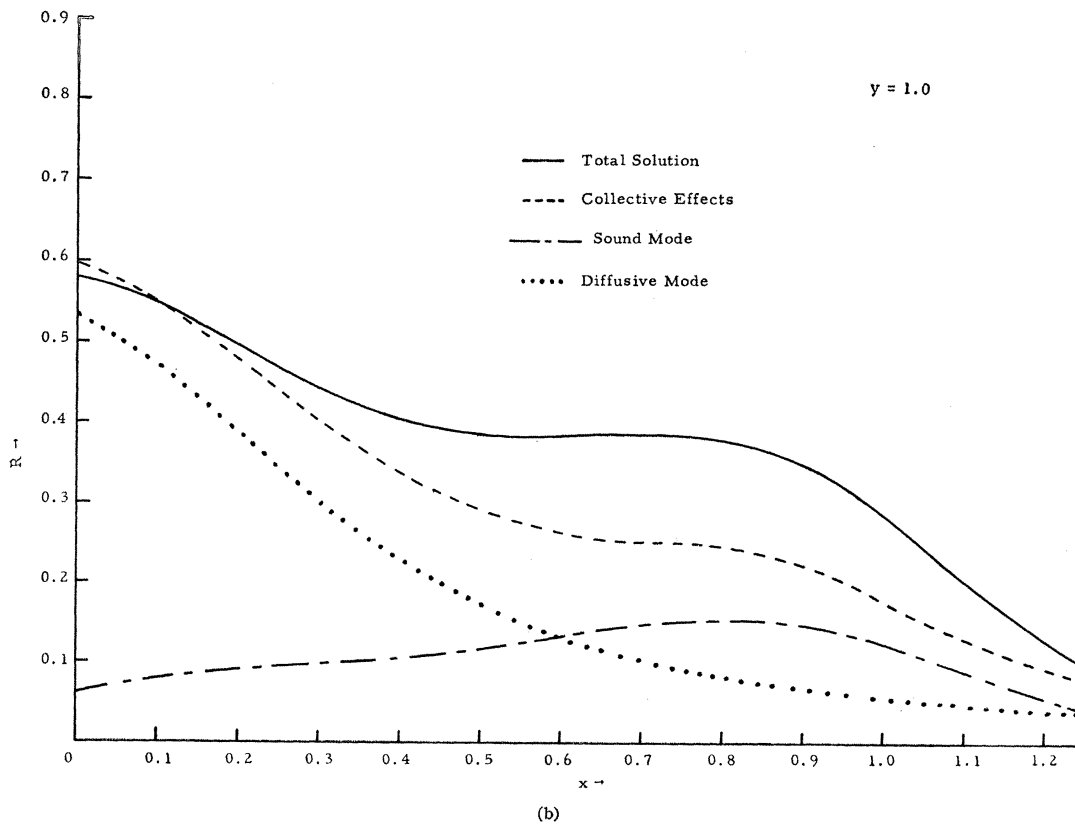
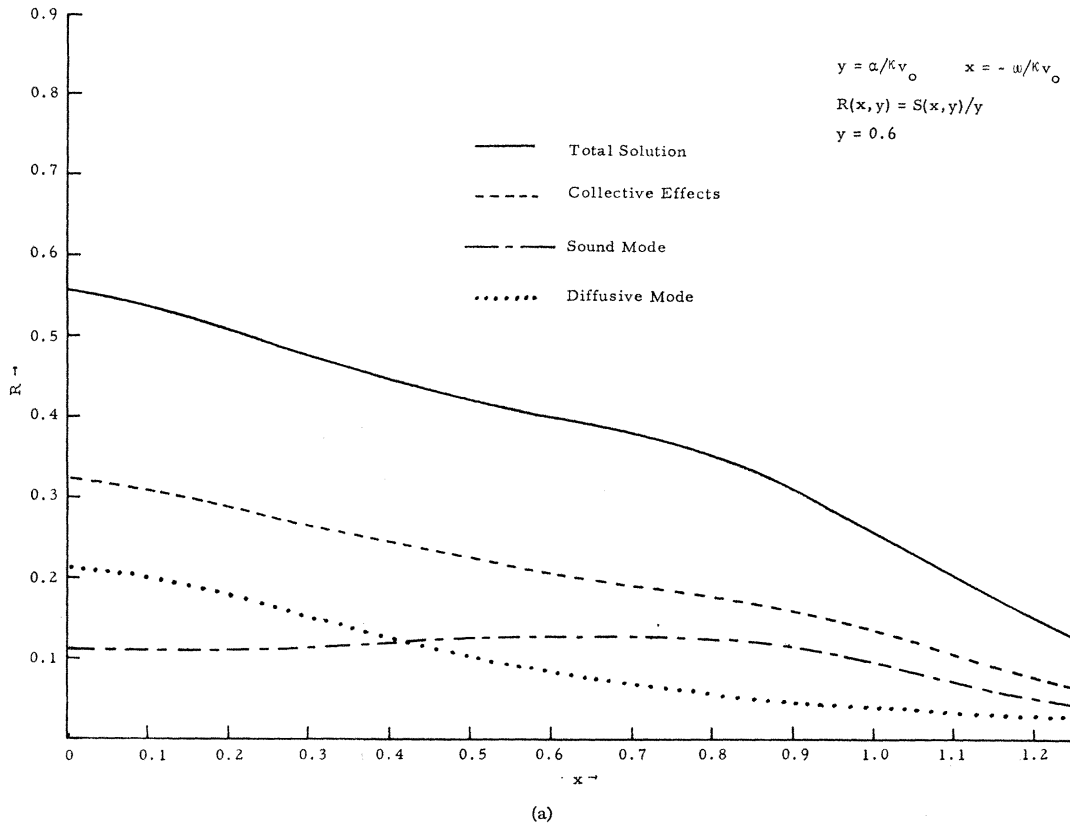


FIG. 1. Comparison of exact solution with Nelkin convolution, diffusion convolution, and hydrodynamic approximation for various  $y = \alpha/\kappa v_0$ ,  $x = -\omega/\kappa v_0$ ,  $R = (\kappa v_0/\pi)S(x, y)$ . Here,  $\alpha$  is the collision frequency and  $v_0$  is the thermal speed  $(2kT/m)^{1/2}$ .



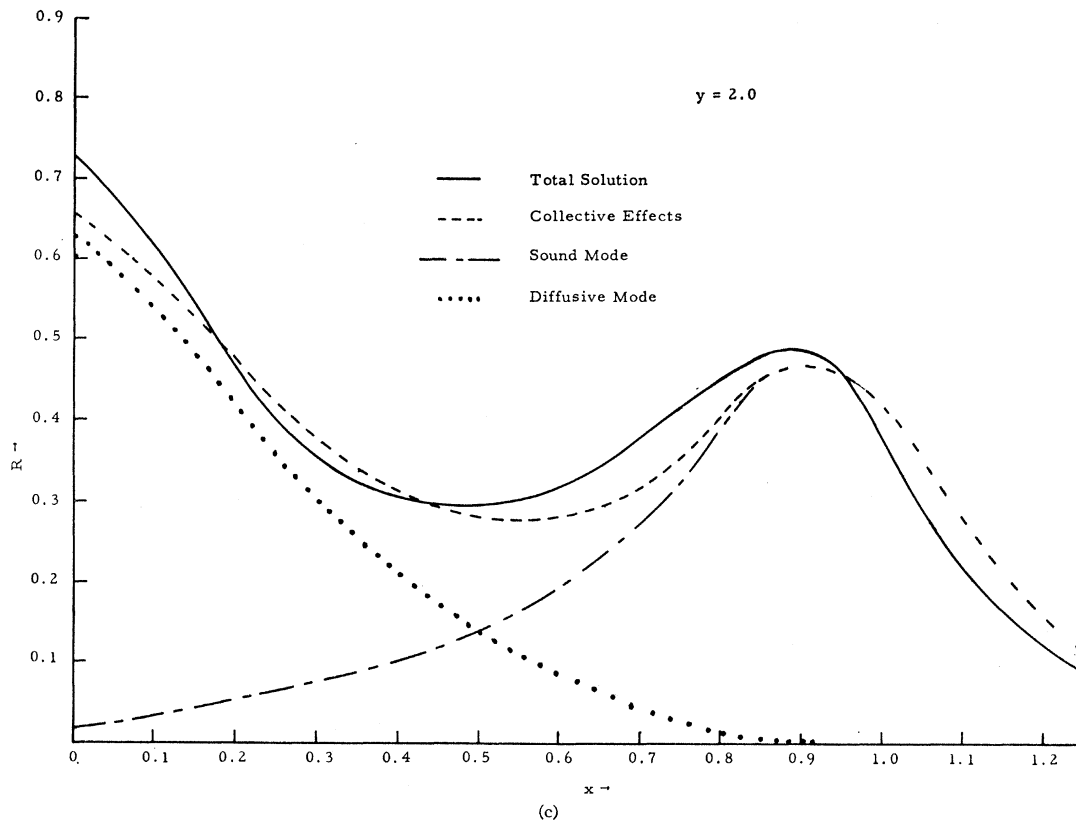


FIG. 2. Comparison of total solution with collective effects contribution for various  $y = \alpha/\kappa v_0$ ,  $x = -\omega/\kappa v_0$ ,  $R = (\kappa v_0/\pi)(Sx, y)$ .

Unfortunately, the results for dilute gases cannot be easily extended to dense gases or liquids. In order to apply some of the results of Secs. II and III to liquids (for which the bulk of experimental data is available) we study the small- $\kappa$ -small- $\omega$  regime in which the hydrodynamic approximation is valid. The use of hydrodynamics is not new<sup>15,16</sup>; however, we shall compare the calculations with existing experimental data. We also compare with the convolution approximation using several models for  $G_s$ .

## II. DILUTE GASES

For a dilute gas, it is convenient to work with the velocity-dependent correlation function  $F(\mathbf{r}, \mathbf{v}, t)$ , which is defined as the probability that there will be a particle with velocity  $\mathbf{v}$  at  $(\mathbf{r}, t)$ , given that there was a particle with an equilibrium velocity distribution at the origin at  $t=0$ . Yip and Van Leeuwen<sup>10,11</sup> have shown that for a dilute gas this function obeys the linearized Boltzmann equation

$$DF = LF, \quad (2.1)$$

with the initial condition

$$F(\mathbf{r}, \mathbf{v}, 0) = [\delta(\mathbf{r}) + g(\mathbf{r})]M(v), \quad (2.2)$$

<sup>15</sup> L. P. Kadanoff and P. C. Martin, *Ann. Phys. (N. Y.)* **24**, 419 (1963).

<sup>16</sup> R. D. Mountain, *Rev. Mod. Phys.* **39**, 205 (1966).

where  $M(v)$  is the Maxwell distribution function. For a dilute gas,  $g(\mathbf{r})$  becomes simply the number density  $n$ . In Eq. (2.1),  $D$  is the drift operator,

$$D = (\partial/\partial t) + \mathbf{v} \cdot \nabla, \quad (2.3)$$

and  $L$  is the linearized collision operator of kinetic theory.<sup>16</sup> For our purposes, the only important property of  $L$  is that it possesses a fivefold degenerate zero eigenvalue with the eigenfunctions corresponding to the five collisional invariants.

Yip and Van Leeuwen<sup>10</sup> have also shown that the self part of  $F(\mathbf{r}, \mathbf{v}, t)$  satisfies the linearized Boltzmann equation

$$DF_s = L_s F_s, \quad (2.4)$$

with the initial condition

$$F_s(\mathbf{r}, \mathbf{v}, 0) = \delta(\mathbf{r})M(v), \quad (2.5)$$

where  $L_s$  is a linear collision operator which possesses only a single zero eigenvalue.<sup>17</sup> This is the starting point of the analyses of several authors.<sup>7,17,18,19</sup>

One can readily solve these equations for the Fourier

<sup>17</sup> S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-Uniform Gases* (Cambridge University Press, New York, 1961).

<sup>18</sup> S. Yip and M. Nelkin, *Phys. Rev.* **135**, A1241 (1964).

<sup>19</sup> R. C. Desai, *J. Chem. Phys.* **44**, 77 (1966). R. C. Desai, Ph.D. thesis, Cornell University, 1966 (unpublished).

transforms of  $F$  and  $F_s$  if simple forms are taken for the collision operators. Then  $S^{CL}$  and  $S_s^{CL}$  are easily obtained by integration over velocity.<sup>7,17,18</sup>

For a dilute gas the convolution approximation gives  $G(\mathbf{r},t) = G_s(\mathbf{r},t) + n$ , which is clearly incorrect. To see why this is so, we first subtract Eq. (2.4) from Eq. (2.1) to obtain

$$DF_D = LF_D + (L - L_s)F_s = LF_D + f_s, \quad (2.6)$$

where  $F_D = F - F_s$ . Subtracting initial conditions, Eqs. (2.2) and (2.5), gives

$$F_D(\mathbf{r}, \mathbf{v}, 0) = g(\mathbf{r})M(\mathbf{v}). \quad (2.7)$$

Again,  $g(\mathbf{r}) = n$ , so the initial condition gives only the contribution  $nM(\mathbf{v})$  to  $F_D$ . This, in turn, gives no contribution to the cross section, and we shall ignore it from here on. Now, assuming that we have solved for  $F_s$ , we can formally solve Eq. (2.6) by

$$F_D(\mathbf{r}, \mathbf{v}, t) = \int \mathcal{G}(|\mathbf{r} - \mathbf{r}'|; \mathbf{v}, \mathbf{v}'; t - t') f_s(\mathbf{r}', \mathbf{v}', t') \times d^3\mathbf{r}' d^3\mathbf{v}' dt', \quad (2.8)$$

where  $\mathcal{G}$  is the Green's function of the operator  $D - L$ . The convolution approximation completely ignores the contribution represented by Eq. (2.8). Clearly, Eq. (2.8) represents the perturbation in the field particle distribution caused by the test particle for  $t > 0$ . The additional perturbation is given by  $f_s(\mathbf{r}', \mathbf{v}', t')$  and this propagates according to the Green's function  $\mathcal{G}(|\mathbf{r} - \mathbf{r}'|; \mathbf{v}, \mathbf{v}'; t - t')$ .

These results give some physical insight into the reasons for the inadequacy of the convolution approximation. Nossal,<sup>20</sup> among others, has put forth more formal arguments for this inadequacy.

### III. COLLECTIVE EFFECTS

Another approach to looking at the problem discussed in the previous section is to write  $G(\mathbf{r},t)$  in terms of the normal modes of the system. In this way it is possible to separate collective effects from purely single-particle effects. There has been much discussion of the role of collective effects in neutron scattering from fluids (e.g., see Refs. 12 and 13) and one can obtain some insight into the problem in this way. Also, as we shall see, hydrodynamics essentially gives the collective effects (although not exactly) and thus this procedure gives further information as to the usefulness of the hydrodynamic calculations.

A discussion of Eq. (2.1) with an arbitrary initial condition has been given by Buckner and Ferziger.<sup>14</sup> If the collision operator is modeled,<sup>21</sup> Eq. (2.1) has the form

$$\frac{\partial F}{\partial t} + c_z \frac{\partial F}{\partial z} + F = \sum_{i=1}^N (\lambda_i + 1) \psi_i \int e^{-c^2} \psi_i(\mathbf{c}) F(\mathbf{c}) d^3c, \quad (3.1)$$

<sup>20</sup> R. J. Nossal, Phys. Rev. 143, 74 (1966).

<sup>21</sup> L. Sirovich and J. K. Thurber, Brown University, Providence, Rhode Island, Division of Applied Mathematics, Technical Report No. 58, 1964 (unpublished).

where the  $\lambda_i$  and  $\psi_i$  are the eigenvalues and eigenfunctions of the linearized collision operator.

By considering solutions to Eq. (3.1) of the form

$$F(\mathbf{c}, z, t) = F_{\kappa\omega}(\mathbf{c}) e^{i(\omega-1)t - i\kappa z}, \quad (3.2)$$

Buckner and Ferziger show that the spatial Fourier transform of  $G(\mathbf{r},t)$ , denoted by  $\chi(\kappa,t)$ , has the form

$$\chi(\kappa,t) = \sum_{l=1}^N a_l(\kappa) e^{i\omega_l(\kappa)t} + \int_{-\infty}^{\infty} A(\lambda,\kappa) e^{i(\lambda-1)t} d\lambda, \quad (3.3)$$

where the  $\omega_l$  ( $l=1, 2, \dots, N$ ) are the (complex) discrete eigenvalues of Eq. (3.1) and  $a_l(\kappa)$  and  $A(\lambda,\kappa)$  are functions which are readily calculated from the initial condition (2.2). A similar approach applied to Eq. (2.4) with initial condition (2.5) gives a similar representation for  $\chi_s(\kappa,t)$ , the self part of  $\chi(\kappa,t)$ . The functions  $a_l(\kappa)$  and  $A(\lambda,\kappa)$  are, of course, different in this case and, more importantly, the number of discrete eigenfunctions is different. In particular, the "sound" modes which arise from the degeneracy of the zero eigenvalue of the scattering operator are absent.

The discrete term in Eq. (3.3) may be interpreted as the "collective" effects while the integral term represents "single-particle" effects.<sup>22</sup> Thus the representation (3.3) (which resembles the Lehmann representation for the many-fermion problem<sup>23</sup>) makes it particularly simple to discuss collective effects. It is also interesting to note that when Eq. (3.3) is Fourier transformed, the discrete terms give rise to "phonon" peaks, while the integral gives a continuous background; i.e.,

$$S^{CL}(\kappa,\omega) = \sum_l \frac{a_l(\kappa)}{\omega^2 - \omega_l^2(\kappa)} + \int_{-\infty}^{\infty} \frac{A(\lambda,\kappa) d\lambda}{(\omega - \lambda)^2 + 1}. \quad (3.4)$$

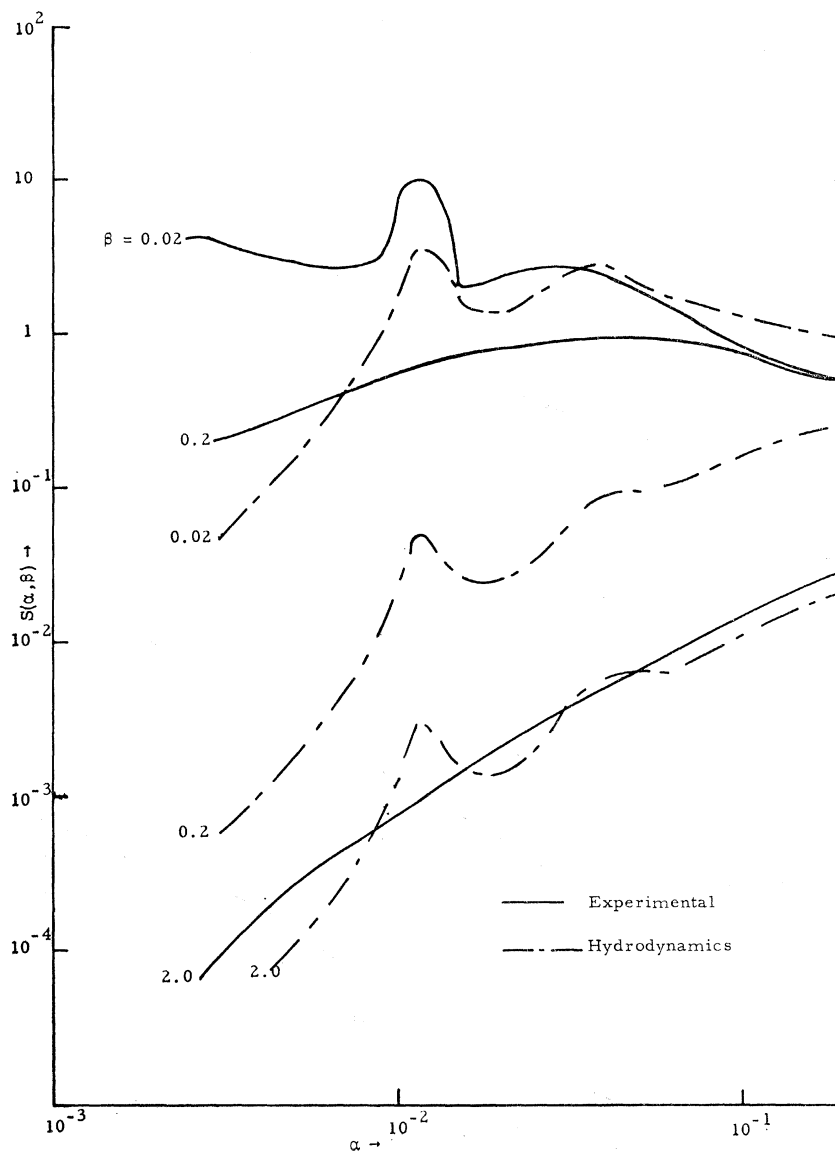
Note that because of the degenerate zero eigenvalue there are always at least three discrete terms at low  $\kappa$  (the spherical symmetry of the problem prevents the other two from contributing). One of these eigenvalues is pure imaginary and corresponds to a heat-diffusion mode and gives rise to a peak at  $\omega=0$ . The other two are incoming and outgoing damped sound waves and give rise to peaks at  $\omega = \pm \text{Re}(\omega_l)$ .

At low wave numbers  $\kappa$  the discrete terms in Eq. (3.4) completely dominate the integral term and the neutron scattering is thus due almost entirely to collective effects. Furthermore, the hydrodynamic approximation gives only the first three discrete terms and at small  $\kappa$  gives an excellent approximation to them. These points are illustrated in Figs. 1 and 2. In Fig. 1 the exact solution is taken from Yip and Nelkin's<sup>18</sup> Eq. (6) and  $F_s$  for the convolution from Nelkin and Ghatak's<sup>7</sup> Eq. (28).

<sup>22</sup> G. E. Uhlenbeck and G. Ford, *Lectures in Statistical Mechanics* (American Mathematical Society, Providence, Rhode Island, 1963).

<sup>23</sup> P. Nozières, *Theory of Interacting Fermi Systems* (W. A. Benjamin, Inc., New York, 1964).

FIG. 3. Comparison of scattering-law hydrodynamic approximation with Randolph's experimental results for sodium at 100°C.



As  $\kappa$  increases, the number of discrete modes decreases. In fact, there is a point [for the model of Bhatnager, Gross, and Krook (BGK),  $\gamma = \alpha/\kappa v_0 \sim 0.53$ ] at which all discrete modes cease to exist. As this cutoff is approached, the continuum contribution becomes increasingly more important, as is shown in Fig. 2. Finally, beyond the cutoff, all of the contribution to  $G(\mathbf{r}, t)$  comes from the continuum modes; collective effects no longer exist. Also, in this regime, the hydrodynamic approximation is completely invalid; again this is due to the divergence of the asymptotic approximation that the hydrodynamic equations represent. This is illustrated by Fig. 1.

It should also be remarked that, in this formalism, the central peak in the scattering function is due to a collective phenomenon: the heat-diffusion mode.

#### IV. LIQUIDS

Since a kinetic equation corresponding to the Boltzmann equation cannot be written for a liquid, the above discussion is not directly applicable to liquids. However, some of the qualitative features of the results for gases have implications for liquids. Thus we may expect that hydrodynamics will give a good approximation for small  $\kappa$  and/or  $\omega$  and that when these parameters are small the neutron scattering may be attributed to collective effects. A complete theory for scattering from liquids encompassing large values of  $\kappa$  and  $\omega$  will undoubtedly require the development of a more complete theory of liquids. We may surmise, however, that the complete expression for  $S(\kappa, \omega)$  will contain a sum of discrete terms representing collective effects and,

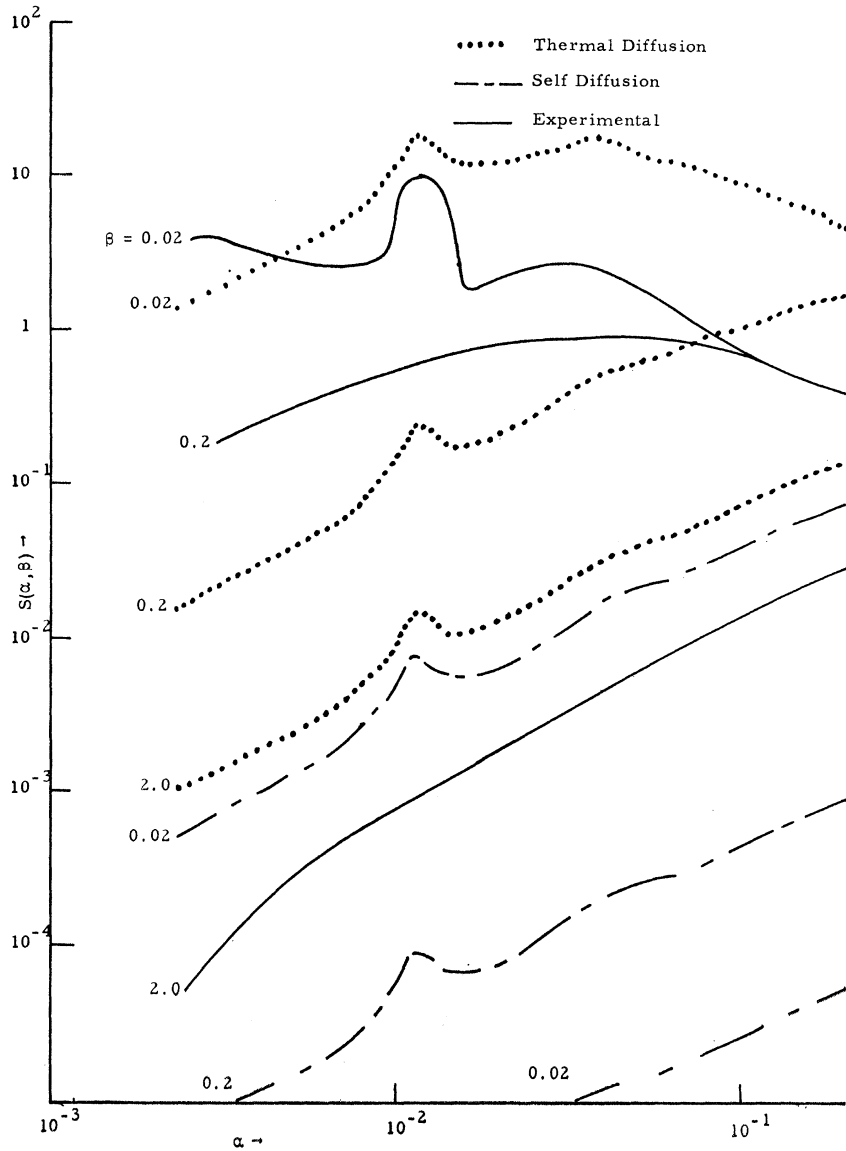


FIG. 4. Comparison of scattering-law convolution approximation using self diffusion and thermal diffusivity with Randolf's experimental results for sodium at 100°C.

most likely, an integral term that accounts for short-time behavior.

The application of hydrodynamics to the calculation of correlation functions is not new and has been discussed by Kadanoff and Martin,<sup>15</sup> Mountain,<sup>16</sup> and Yip and Nelkin.<sup>18</sup> To obtain  $G(\mathbf{r}, t)$  one solves the linearized Navier-Stokes equations with the initial conditions

$$\delta n(\mathbf{r}, 0) = \delta(\mathbf{r}) + [g(\mathbf{r}) - n], \quad (4.1)$$

$$\delta v(\mathbf{r}, 0) = \delta T(\mathbf{r}, 0) = 0. \quad (4.2)$$

Again, for a gas, this is just the hydrodynamic approximation to Eq. (2.1) and initial condition (2.2). Using the results of Ref. 16, we have

$$S^{CL}(\mathbf{k}, \omega) = (1/\pi)[1 + \Gamma(\mathbf{k})] \operatorname{Re}\{n(\mathbf{k}, \omega)\}, \quad (4.3)$$

with

$$n(\mathbf{k}, \omega) = \frac{-\omega^2 + i(a+b)\kappa^2\omega + ab\kappa^4 + c^2(1-1/\gamma)\kappa^2}{-i\omega^3 - (a+b)\kappa^2\omega^2 + i(c^2\kappa^2 + ab\kappa^4)\omega + ac^2\kappa^4/\gamma}, \quad (4.4)$$

where

$$a = \lambda_0/\rho_0 c_v, \quad b = (\frac{4}{3}\eta_0 + \xi_0)/\rho_0, \quad (4.5)$$

and  $\rho_0$  is the equilibrium density,  $\lambda_0$  the thermal conductivity,  $\eta_0$  and  $\xi_0$  the shear and bulk viscosities, respectively,  $c$  the sound speed,  $\gamma$  the ratio of specific heats, and  $c_v$  the specific heat at constant volume. For small  $\kappa$ , Eq. (4.4) can be factored into a sum of three terms, two of which represent the contribution of sound modes and the third a heat-diffusion mode. These, of course, are precisely the modes referred to earlier.



The contribution of the heat-diffusion mode has an energy width proportional to the thermal diffusivity  $D_T$ ; again, this is what one ought to expect. On the other hand, the hydrodynamic approximation to Eq. (2.4) yields the well-known result

$$S_s(\kappa, \omega) = \frac{2D\kappa^2}{(D\kappa^2)^2 + \omega^2}, \quad (4.6)$$

where  $D$  is the self-diffusion coefficient. As is well-known (see, for example, Ref. 11), for a gas  $D \approx D_T$ , but for liquids  $D$  is several orders of magnitude less than  $D_T$ . The reason is that in a liquid the dominant mechanism of energy transport is collisional transfer rather than transport by individual particles and the collisional mechanism gives a contribution to  $D_T$  but not to  $D$ . Thus, Vineyard's<sup>9</sup> replacement of  $H$  in Eq. (1.4) by the  $G_s$  obtained from the diffusion equation [i.e., the inversion of (4.6)] cannot be correct. Certainly, a better result would be obtained if  $D_T$  were used instead of  $D$ ; this, however, still ignores a contribution to  $G_d$  of the type given by (2.8) and is also equivalent to replacing  $H$  of Eqs. (1.4) or (2.8) solely by its heat-diffusion component. It therefore seems more appropriate to bypass the convolution approximation entirely and to calculate  $G(\mathbf{r}, t)$  from hydrodynamics and  $G_s(\mathbf{r}, t)$  from hydrodynamics or one of the improved models that have been suggested. Unfortunately, this means that it is unlikely that the models put forth for computing  $G_s$  can be applied to the calculation of  $G$  or  $G_d$  and that improved calculation of  $G$  will have to await a more fundamental approach.

Finally, we include a comparison of hydrodynamic calculations with experimental data for liquid sodium.<sup>24</sup>

<sup>24</sup> P. D. Randolph, Phys. Rev. (to be published).

TABLE I. Sodium parameters<sup>a</sup> at 100°C used in calculation of Figs. 3 and 4.

Specific heat	$c_p$	$1.39 \times 10^7$ erg/g
Isothermal compressibility	$k=1/B$	$19.16 \times 10^{-12}$ cm <sup>2</sup> /dyn
Shear viscosity	$\eta$	0.705 cP
Bulk viscosity	$\xi$	1.06 cP
Sound speed	$c$	$2.525 \times 10^5$ cm/sec
Volume expansivity	$\beta$	$2.445 \times 10^{-4}$ C
Thermal conductivity	$\lambda$	$0.8581 \times 10^7$ erg/sec cm
Self diffusion coefficient	$D$	$2.5 \times 10^{-4}$ cm <sup>2</sup> /sec
Density	$\rho$	0.916 g/cm <sup>3</sup>

<sup>a</sup> *International Critical Tables* (McGraw-Hill Book Company, Inc., New York, 1933); M. Sittig, *Sodium, Its Manufacture, Properties and Uses* (Reinhold Publishing Corporation, New York, 1956); *Liquid Metals Handbook, Sodium Supplement* (U. S. Government Printing Office, Washington, D. C., 1955).

These are given in Fig. 3 for a range of parameters for which hydrodynamics should be valid. The thermodynamic parameters for sodium are found in Table I. Since sodium contains both a coherent and incoherent contribution, Eq. (1.1) was used with  $S_g^{CL}$  calculated using Eq. (4.6) and  $S^{CL}$  calculated using Eq. (4.3). Except for  $\beta=0.2$ , the comparison is not bad. That the peaks in the theoretical curves are not reproduced in the experimental data may be due to the fact that for the values of parameters shown, the experiment is difficult to perform and the resolution is less than would be desirable. Figure 4 shows the convolution approximation using both the self-diffusion coefficient and the thermal diffusivity. The latter matches the experimental data about as well as the full hydrodynamic calculation. This is because, in the parameter range shown, the sound-mode contribution is small. Also this is apparently an indication that in liquids the perturbations caused by the particle at  $t>0$  are less important than the initial perturbation.