of the temperature and density. That this is indeed the case is shown by the following example. For  $\rho = 2$ ,  $T = 100$ , a direct Monte Carlo computation agrees quite well with that of Fickett and

Wood: A value of 9.2 is obtained for  $p/\rho kT$  when Fickett and Wood give the value 9.5. This is in strong disagreement with the results obtained from Barker and Henderson's theory.

Laboratoire associ5 au Centre National de la Recherche Scientifique, Orsay, France.

 $1$ J. A. Barker and D. Henderson, J. Chem. Phys. 47, 2856, 4714 (1967}.

 ${}^{2}$ R. W. Zwanzig, J. Chem. Phys. 22, 1420 (1954).

 $^3$ E. B. Smith and B. J. Alder, J. Chem. Phys. 30,

1190 (1959); H. Frish, J. L. Katz, E. Praestgaard,

and J. L. Lebowitz, ibid. 70, 2016 (1966); D. A.

McQuarrie and J. L. Katz, ibid. 44, <sup>2393</sup> (1966).

 ${}^{4}P$ . C. Hemmer, J. Math. Phys. 5, 75 (1964).

<sup>5</sup>J. L. Lebowitz, G. Stell, and S. Baer, J. Math. Phys. 5, 75 (1964).

 ${}^{6}$ M. Coopersmith and R. Brout, Phys. Rev.  $130$ , 2539 (1963).

<sup>7</sup>We have received a preprint by J. A. Barker, D. Henderson, and W. R. Smith where the same conclusion is reached.

 ${}^{8}$ F. H. Ree and W. G. Hoover, J. Chem. Phys.  $46$ , 4181 (1967}.

 ${}^{9}$ B. J. Alder and T. E. Wainwright, J. Chem. Phys. 33, 143S (1960).

 $\overline{10}$ L. Verlet, Phys. Rev. 165, 201 (1968).

 $11$ N. W. Ashkroft and J. Lekner, Phys. Rev.  $145$ , 83 (1966).

 $^{12}$ M. S. Wertheim, Phys. Rev. Letters 10, 321 (1963);

E.J. Thiele, J. Chem. Phys. 38, 1959 (1963).

 $^{13}$ D. Schiff and L. Verlet, to be published.

 $^{14}$ L. Verlet and D. Levesque, Physica  $36$ , 254 (1967).

<sup>15</sup>L. Verlet, Phys. Rev. 159, 98 (1967).

 $^{16}$ J. L. Lebowitz, J. K. Percus, and L. Verlet, Phys. Rev. 153, 250 (1967).

 $^{17}$ J. M. H. Levelt, Physica  $26$ , 361 (1960).

 $18$ W. Van Witzenburg and J. C. Stryland, Can. J. Phys. 46, 811 (1968).

 $\overline{^{19}}$ R. K. Crawford and W. B. Daniels, Phys. Rev. Letters 21, 367 (1968).

 $^{20}$ D. R. Lovejoy, Nature 197, 353 (1963).

 $21$ A. Michels, H. Wijker, and H. K. Wijker, Physica 15, 62V (1949).

 $^{22}E$ . Whalley and W. G. Schneider, J. Chem. Phys. 33, 633 (1965).

 $\overline{23}$ J. H. Dymond and B. J. Alder, J. Chem. Phys.  $48$ , 1 (1968).

 $^{24}$ J. K. Percus and G. Stell, report of work prior to publication.

 $^{25}$ J. P. Hansen and L. Verlet, to be published.

 $^{26}$ M. C. Donald and K. Singer, J. Chem. Phys.  $47$ , 4766 (1967).

 $27W$ . W. Wood, "The Physics of Simple Liquids" (to be published), Chap. 5.

 $^{28}$ W. W. Wood and F. R. Parker, J. Chem. Phys. 27, V20 (195V).

 $29W$ . Fickett and W. W. Wood, Phys. Fluids 3, 204  $(1960)$ .

#### PHYSICAL REVIEW VOLUME 182, NUMBER 1 5 JUNE 1969

# Correlation Length of Density Fluctuations in Liquids

Erik W. Aslaksen Bell Telephone Laboratories, Holmdel, New Jersey 07733 (Received 26 March 1969)

It is shown that in a liquid whose bulk modulus relaxes with relaxation time  $\tau'$ , there exist density fluctuations whose correlation length is approximately  $v\tau'/\pi$ , where v is the sound velocity at  $\Omega_0 = (\tau')^{-1}$ .

It has been suggested that one might study the correlation function of density fluctuations in a liquid by measuring the correlation function of the scattered light.<sup>1</sup> For this to be possible it is of course necessary that the correlation length in the liquid be comparable to the wavelength of the incident light. In the case of Hayleigh scattering near the critical point it is well known that the

correlation length does become very large, but in the case of Brillouin scattering the correlation length is not so clearly defined. Gori and Sette' assert that the correlation length is the distance traveled by the thermal acoustic waves before they are reabsorbed, however, this length is frequency dependent, so it would not define the correlation length uniquely, and also this definition would give correlation lengths which are in disagreement with experimental results.<sup>2</sup> We therefore present a short discussion of the correlation length involved in Brillouin scattering.

Let  $\delta \epsilon(\vec{r}, t)$  be the isotropic part of the fluctuations in the dielectric constant of the liquid. We can decompose  $\delta \epsilon(\vec{r}, t)$  as follows

$$
\delta \epsilon(\vec{\mathbf{r}},t) = \delta \epsilon_{\mathbf{c}}(\vec{\mathbf{r}},t) + \delta \epsilon_{\mathbf{D}}(\vec{\mathbf{r}},t) ,
$$

i.e. , into adiabatic and isobaric fluctuations. These are independent.

$$
\langle \delta \epsilon_{\mathbf{S}}(\vec{\mathbf{r}},t) \delta \epsilon_{\mathbf{D}}(\vec{\mathbf{r}}',t') \rangle = 0,
$$

so we can define a correlation function for the adiabatic fluctuations alone,

$$
\xi_{\mathcal{S}}(\Delta r, \Delta t) \equiv \langle \delta \epsilon_{\mathcal{S}}(\vec{r}, t) \delta \epsilon_{\mathcal{S}}(\vec{r} + \Delta \vec{r}, t + \Delta t) \rangle.
$$

The intensity of the polarized part of the scattered light is proportional to  $\langle |\delta \epsilon (\vec{q}, \Omega)|^2 \rangle$ ,<sup>3</sup> and the intensity in the Brillouin lines is then proportional to  $\langle |\delta \epsilon_S(\vec{q}, \Omega)|^2 \rangle$ . But with

$$
\delta \epsilon_S(\vec{q}, \Omega) = (2\pi)^{-2} \iint \delta \epsilon_S(\vec{r}, t) e^{-i(\vec{q} \cdot \vec{r} - \Omega t)} d^3r dt,
$$
  
we have

$$
S^{(\Delta r, \Delta t) = \pi^{-1} \int \int \langle |\delta \epsilon_S(q, \Omega)|^2 \rangle} \times e^{-i\Omega \Delta t} \left[ (\sin q \Delta r) / q \Delta r \right] q^2 dq d\Omega,
$$

which then effectively gives us a relation between the integrated intensity in the Brillouin lines and the correlation function of the adiabatic density fluctuations.

For the  $q$  dependence of the integrated intensity we may, to a first approximation, take the expression'

$$
I_B(q) \sim \frac{1}{K_{\infty}} + \frac{2(K_{\infty} - K_0)}{K_0^2} \frac{s}{1+s} , \qquad (1)
$$

where  $s = \rho_0 / \gamma K_0 q^2 \tau^{\prime 2}$ . We have assumed that the liquid is adequately characterized by a single relaxation time  $\tau'$ , and that the complex compression (longitudinal) modulus  $\overline{K}$  is given by

$$
\overline{K} = \frac{i\Omega K_{\infty} + K_0 / \tau'}{i\Omega + 1 / \tau'}.
$$

The intensity function of Eq. (1) is shown in Fig. 1, together with the approximation we shall use, namely that there are two contributions to  $I(q)$ : one is constant in the range  $0 < q < q_0$  and vanishes for  $q > q_0$ , the other is constant in the range  $0 < q$  $\langle q_{\rm max}$  and vanishes for  $q > q_{\rm max}$ . The parame ter  $q_{\text{max}}$  has been introduced for obvious physical reasons, and its magnitude is determined essentially by the molecular diameter.

One such rectangular intensity function will re-

such that

\n
$$
\xi_S(\Delta \gamma, 0) \sim \frac{a^3}{(a \Delta \gamma)^3} \left[ \sin(a \Delta \gamma) - a \Delta \gamma \cos(a \Delta \gamma) \right],
$$

where a is either  $q_0$  or  $q_{\text{max}}$ . This function is shown in Fig. 2, and it is reasonable to speak of a correlation range  $D=2.5a^{-1}$ . Of course, the function in Fig. 2 is not exactly like the actual correlation function due to our approximation, but at least in general outline it conforms to the experimental results.<sup>2</sup>

Thus we see that a relaxation of the bulk modulus, resulting in a decrease of the intensity at  $q = q_0$ , demands that there be fluctuations in the density with a correlation length of about  $2.5q_0^{-1}$ , but the mean amplitude of these fluctuations is much smaller than the equilibrium density. If we normalize the complete correlation function to unity, and define  $\alpha \equiv q_0/q_{\rm max}^{\phantom{\dagger}}$  , then the ampli tude of the long-range correlation is always less than  $\alpha$  times that of the short-range correlation



FIG. 1. Intensity of scattered light as a function of wave vector of the density fluctuations.



FIG. 2. One term of the spatial correlation function of the density fluctuations.

function. However, the frequency along the  $\Delta r$ axis of the former is only  $\alpha$  times that of the latter, thus the long-range correlation contributes significantly to the intensity integral. Also, since we are discussing radial correlation functions, the measure is not  $d(\Delta r)$ , but  $4\pi(\Delta r)^2d(\Delta r)$ , which measure is not  $u(\Delta \gamma)$ , but  $4\pi(\Delta \gamma) u(\Delta \gamma)$ , which<br>compensates for the  $({\Delta \gamma})^{-2}$  falloff of the correla tion functions.

<sup>1</sup>F. Gori and D. Sette, Phys. Rev. Letters 17, 361 (1966).

 $2^2$ For example, for liquid argon the characteristic decay length for thermal acoustic waves at 2 GHz is about  $1 \mu$  [P. A. Fleury and J. P. Boon, Bull. Am. Phys. Soc. 12, 1054 (1967)], whereas the correlation length is on the order of several  $\AA$  [A. Eisenstein and N. S. Gingrich, Phys. Rev. 58, 307 (1940); 62, 261 (1942)].

 ${}^{3}$ S. M. Rytov, Zh. Eksperim. i Teor. Fiz. 33, 514 (1957) [English transl.: Soviet Phys.  $-$  JETP 6, 401 (1958)].

 ${}^{4}$ L. van Hove, Phys. Rev. 95, 249 (1954); L. I. Komarov and I. Z. Fisher, Zh. Eksperim. i Teor. Fiz. 43, 1927  $(1962)$  [English transl.: Soviet Phys. - JETP 16, 1358] (1963)].

## PHYSICAL REVIEW VOLUME 182, NUMBER 1 5 JUNE 1969

## Magnetic Susceptibility of Isotopic Mixtures of Solid Helium\*

M. G. Richards and J. M. Homer

School of Mathematical and Physical Sciences, University of Sussex, Falmer, Brighton, Sussex, England (Received 20 November 1968)

Nuclear magnetic susceptibility measurements are reported for the  $He<sup>3</sup>$  spin system in isotopic mixtures of solid helium. The spin-ordering temperature  $\theta$  is derived from a plot of inverse susceptibility against temperatures between 0.3 and 1.2'K. For a wide range of density and He<sup>4</sup> concentration, it is found that  $\theta = 0 \pm 0.005$ °K. These results seem to conflict with the only other comparable published data.

### I. INTRODUCTION

It is well known that the atoms in solid helium have a zero-point motion whose amplitude is a sizable fraction (about a third) of the interatomic separation. This leads to exchange effects in solid He<sup>3</sup> several orders of magnitude greater than in any other known nuclear spin system. These exchange effects determine the nonequilibrium behavior of the spin system below about 1°K and affect appreciably the equilibrium behavior of the whole solid at temperatures below  $0.1\,^{\circ}\text{K}$ . The exchange rate  $J$ , as defined by writing the exchange Hamiltonian

$$
\mathcal{K}_{\mathbf{ex}} = -\frac{1}{2}\hbar J \sum_{i \neq j} \mathbf{I}_i \cdot \mathbf{I}_j,
$$

has been measured<sup>1-3</sup> by both types of behavio and  $J/2\pi$  is found to vary from about 1 Mc/sec at

molar volume 20 cm' to about 25 Mc/sec at molar volume 24 cm'. These frequencies correspond to spin-ordering temperatures of about  $0.06 \times 10^{-3}$  °K spin-ordering temperatures or about  $0.00 \times 10^{-4}$  and  $1.5 \times 10^{-3}$  °K, respectively. These result are in fairly good agreement with the theoretical predictions of Nosanow and co-workers, <sup>4</sup> who are also able to give the sign of J as favoring antiferromagnetism. Experimentally this point can only be clarified by susceptibility measurements as a function of temperature.

The presence of He<sup>4</sup> impurity greatly complicates the behavior of the He<sup>3</sup> nuclear spin system. Richards *et al.*<sup>2</sup> found that in a restricted temperature range hcp solid  $\text{He}^3$  containing 0.5%  $\text{He}^4$ behaved as though the impurity atoms were locally enhancing the exchange rate by a factor of about 40. At lower temperatures the nuclear spin system did not seem to be relaxing by the same sort of processes as are used to describe the pure solid. Another systematic investigation<sup>5</sup> showed that