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Neutron Scattering from a Liquid on a Jump Diffusion Model

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Abstract. The incoherent inelastic scattering cross section of slow neutrons from liquids is calculated using a simple model in which the liquid is assumed to have appreciable short range order in a quasi-crystalline form. Diffusive motion takes place in large discrete jumps, between which the atoms oscillate as in a solid. The model predicts a definite, easily calculable cross section which is not dominated by diffusion effects as when continuous diffusion is assumed, but shows a characteristic variation with angle which could be looked for experimentally. The related pair correlation functions are dominated at small r and t by vibrational effects. Although simple and extreme the model explains several aspects of the observations of Brockhouse and Pope in 1959 and others. A brief discussion of the coherent scattering cross sections for the model is given although explicit formulae are not obtained.

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§ 1

OBSERVATION of the inelastic scattering cross section of slow neutrons from liquids promises to be an important method in determining their basic properties, since these cross sections contain more information than any other type of measurement. Several preliminary results have been reported mainly on two liquids of rather different kinds, water (Brockhouse 1959, Hughes *et al.* 1959), and lead (Pelah, Whittemore and McReynolds 1959, Turberfield, to be published). More recently Brockhouse and Pope (1959) have reported much more detailed results on lead. These results have been transformed using the formalism of van Hove (1954) who showed that the inelastic cross sections were related to the Fourier transform of the correlation function $G(\mathbf{r}, t)$ between pairs of atoms. In the classical form $G(\mathbf{r}, t)$ is the probability of finding an atom at $\mathbf{r}$ at time t if there was an atom at the origin at $t = 0$. It is well known that in the scattering of energetic particles (e.g. x-rays) where the energy transfer is unimportant the differential scattering cross section is the Fourier transform of the instantaneous pair correlation function $G(\mathbf{r}, 0)$.

In fact

$$\frac{\partial^2 \sigma}{\partial \Omega \partial \epsilon} = \frac{a^2 k'}{\hbar k} \Gamma(\mathbf{x}, \omega) \qquad \dots\dots (1)$$

$$\Gamma(\mathbf{x}, \omega) = \int \int \exp i(\mathbf{x} \cdot \mathbf{r} - \omega t) G(\mathbf{r}, t) \, d\mathbf{r} \, dt \qquad \dots\dots (2)$$

where a is the appropriate scattering length, $\mathbf{k}$ and $\mathbf{k}'$ the wave vectors of the incident and outgoing neutrons. The momentum transfer is

$$\hbar\mathbf{x} = \hbar(\mathbf{k} - \mathbf{k}')$$

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and ϵ is the energy lost by the neutron on collision

$$\epsilon = \hbar\omega = \hbar^2(k^2 - k'^2)/2m_0.$$

For interpretation the experimental results are most conveniently displayed as $G(\mathbf{r}, t)$ and Brockhouse's measurement are the first sufficiently detailed to allow its evaluation. However, several Fourier transformations are required on data taken in a limited range of $\varkappa$ and ω, and the extrapolation of this data to cover the whole range may give rise to errors.

The present status of the theory of liquids is totally inadequate for the interpretation of such results, and at the present time the best method of approach appears to be to compare the results with simple models representing various extreme cases. Pioneer work of this kind was done by Vineyard (1958) who discussed, free, oscillatory and diffuse motion and evaluated $G(\mathbf{r}, t)$ in a classical approximation. Schofield (1960) has pointed out that one must proceed to the classical limit with care; crudely speaking because the energy transfers $\hbar\omega$ may all be lost if $\hbar \to 0$ is taken in $G(\mathbf{r}, t)$. He notes that it is safer to consider not $G(\mathbf{r}, t)$ but

$$\gamma'(\varkappa, t) = \gamma\left(\varkappa, t + \frac{i\hbar}{2kT}\right) \qquad \ldots\ldots(3)$$

where γ is defined as

$$\gamma(\varkappa, t) = \int \exp\left[i(\varkappa \cdot \mathbf{r})\right] G(\mathbf{r}, t)\, d\mathbf{r}. \qquad \ldots\ldots(4)$$

Thus for evaluation of the data it is convenient to have predictions of G, γ' and Γ for particular simple models. It has become clear from both the water and lead results (Brockhouse and Pope 1959) that a part of the diffusive motion in liquids takes place in comparatively large diffusive jumps. It therefore seemed of some interest to evaluate the predictions of a specific model of diffusion by large jumps. Brockhouse (1958) was the first to suggest that such a model might be applicable and to investigate some of its properties.

§ 2. Model

The model used is basically that proposed by Frenkel for liquids close to the melting point in which he assumes that the liquid has locally a lattice-like structure. It is known from x-ray scattering that $G(\mathbf{r}, 0)$ is peaked at about the expected atomic spacing. In the model the atomic motion is largely oscillatory as in the solid but because of the randomness left in the structure and the high density of vacancies compared with a solid an atom can occasionally jump into a neighbouring lattice site. If the observed coefficient of self-diffusion in lead $D = 2 \cdot 5 \times 10^{-5}\, \mathrm{cm^2\, sec^{-1}}$ (Hall and Rothman 1956) is due to large jumps of an interatomic spacing $l = 3 \cdot 5 \times 10^{-8}$ cm the time between jumps

$$\tau = l^2/6D \sim 7 \times 10^{-12}\ \mathrm{sec.}$$

Assuming a Debye model of a solid the average time of oscillation is the inverse of the mean frequency $\bar{\nu}$

$$\frac{1}{\bar{\nu}} = \frac{4h}{3k\theta} = 7 \times 10^{-13}\ \mathrm{sec}$$

assuming a Debye temperature of $80°$K. Thus the atom performs many oscillations between jumps and it seems that this quasi-crystalline model would be a good first approximation.

If we assume that during the jumping process the atom moves as a free particle the time in transit

$$\tau' = l \left(\frac{M}{2kT} \right)^{1/2} \sim 1 \cdot 5 \times 10^{-12} \text{ sec}$$

where M is the mass of the lead atom. This is much shorter than the time between jumps, and will be neglected in considering the diffusion process.

It is convenient to divide a discussion of $G(\mathbf{r}, t)$ into the self-correlation function $G_s(\mathbf{r}, t)$ which defines the probability of finding the atom which began at the origin at $\mathbf{r}$ at t, and $G_d(\mathbf{r}, t)$, the correlation function of different atoms. G_d is further conveniently subdivided into its mean value—the density ρ—and fluctuations about this mean which die away in a few atomic radii (G_d'). ρ remains constant and since its contribution to scattering occurs only at $\varkappa = 0$, $\omega = 0$, which cannot be observed, it may be neglected.

In substances where the nuclear scattering is incoherent because of isotope and spin effects only G_s is observable. Since protons have a very large incoherent cross section this is always the dominant effect in hydrogenous materials. In coherent scatterers like lead it is possible to effect an approximate separation of the effects of G_s and G_d (Brockhouse and Pope 1959, Egelstaff, unpublished). It is also very much more difficult to make a calculation of G_d than one of G_s. Accordingly we devote most of the rest of the paper to an evaluation of G_s on our model, giving only a brief discussion of G_d in a later section.

Taking the view that the atomic motion is oscillatory about well-defined points between which the atom moves by diffusion we write

$$G_s(\mathbf{r}, t) = \int G_s^D(\mathbf{R}, t) G_s^V(\mathbf{r} - \mathbf{R}, t) \, d\mathbf{R} \qquad \ldots \ldots (5)$$

where G^D describes the diffusive and G^V the vibrational motion. The Fourier transform of (5) is a simple product

$$\gamma_s(\varkappa, t) = \gamma_s^D(\varkappa, t) \gamma_s^V(\varkappa, t) \qquad \ldots \ldots (6)$$

while

$$\Gamma_s(\varkappa, \omega) = 2\pi \int \Gamma_s^D(\varkappa, \omega') \Gamma_s^V(\varkappa, \omega - \omega') \, d\omega'. \qquad \ldots \ldots (7)$$

In writing (5) it has been assumed that the oscillatory and diffusive motions are completely uncorrelated, and that the diffusion takes place very rapidly so that the oscillatory motion has gone on for the whole time interval t.

§ 3. THE DIFFUSIVE MOTION

The diffusion equation for discrete jumps in space is a difference equation. If $P(\mathbf{r})$ is the probability of finding a particle at $\mathbf{r}$, and it can jump to a distribution of sites $\mathbf{l}$ from $\mathbf{r}$, of which there are n

$$\frac{\partial P(\mathbf{r})}{\partial t} = \frac{1}{n\tau} \sum_{\mathbf{l}} [P(\mathbf{r} + \mathbf{l}) - P(\mathbf{r})]. \qquad \ldots \ldots (8)$$

Thus $G_s^D(\mathbf{r}, t)$ is a solution of this equation with the boundary condition

$$G_s^D(\mathbf{r}, 0) = \delta(\mathbf{r}). \qquad \ldots \ldots (9)$$

The equation is conveniently solved by considering the Fourier transform of G_s

$$G_s^D(\mathbf{r}, t) = (2\pi)^{-3} \int \exp(-i\varkappa \cdot \mathbf{r}) \exp(-f(\varkappa)t) \, d\varkappa \qquad \ldots \ldots (10)$$

where

$$f(\varkappa) = -\sum_{1} (\exp{(i\varkappa.1)} - 1)/n\tau. \qquad \ldots\ldots(11)$$

This may now be evaluated for various arrangements of sites. For example, if the diffusion is on a simple cubic lattice of spacing a

$$f(\varkappa) = (3 - \cos\kappa_x a - \cos\kappa_y a - \cos\kappa_z a)/3. \qquad \ldots\ldots(12)$$

The actual form of $G(\mathbf{r}, t)$ is more difficult to find although in this case it is possible to give the result in closed form. At lattice point $(\alpha a, \beta a, \gamma a)$

$$G(\mathbf{r}, t) = \exp{(-t/\tau)} I_\alpha(t/3\tau) I_\beta(t/3\tau) I_\gamma(t/3\tau) \qquad \ldots\ldots(13)$$

where I_n is the Bessel function of imaginary argument. It will be noticed that at the origin the decay is not given simply by $\exp{(-t/\tau)}$ but is modified by a probability that it will return.

For a close-packed lattice with atomic distance a

$$f(\varkappa) = [1 - \cos{(\kappa_x a/\sqrt{3})}\cos{(\kappa_y a/\sqrt{3})}\cos{(\kappa_z a/\sqrt{3})}]/\tau. \qquad \ldots\ldots(14)$$

Since it is not certain that the long range order implied by these lattices does not exist in a liquid an alternative model is to assume jumps of a fixed distance l but in random directions, although this probably overestimates the randomness of the motion. The sum in (11) is now replaced by an integral and

$$f(\varkappa) = \frac{1}{\tau}\left(1 - \frac{\sin\kappa l}{\kappa l}\right). \qquad \ldots\ldots(15)$$

If the jumps have a distribution $q(l)$

$$f(\varkappa) = \frac{1}{\tau}\int\left(1 - \frac{\sin\kappa l}{\kappa l}\right)q(l)\,dl \bigg/ \int q(l)\,dl. \qquad \ldots\ldots(16)$$

The case of jumps of fixed length is a well-known problem in the theory of random walks and is treated in detail by Chandrasekhar (1943). After one jump the distribution function has a contribution $\delta(\mathbf{r}-1)$. After two jumps $G_s^{(2)}$ has two steps:

$$G_s^{(2)} = 2Z \qquad 0 < r < 1,$$
$$= Z \qquad 1 < r < 2l,$$
$$= 0 \qquad r > 2l,$$

where Z is a constant. Expressions after three and four jumps are given in the reference cited, and after many jumps G_s takes on a Gaussian distribution

$$G_s(\mathbf{r}, t) = \left(\frac{1}{4\pi Dt}\right)^{3/2}\exp{\left(-\frac{r^2}{4Dt}\right)}. \qquad \ldots\ldots(17)$$

This formula is of course obtained at all times in the limit of small jumps when the difference equation (8) becomes a differential equation.

In the foregoing work the discrete nature of the jumping in space has been emphasized but it has been assumed that this takes place at a uniform rate. A better approximation was proposed by Vineyard (1958) and extended by Schofield (1960) using the Langevin equation as for Brownian motion. They demonstrate that these effects can be included by replacing t in the exponent of (10) by

$$\zeta = t + \frac{1}{\eta}(e^{-\eta t} - 1) \qquad \ldots\ldots(18)$$

where $\eta = kT/MD$.

The form of these results means that the usual methods of discussing the width of the distribution G_s as a function of time must be interpreted with care. At small times the jumping process produces peaks in G_s at the average value of l but does not broaden the central peak. After one or two jumps have taken place, however, all such bumps and discontinuities have been smoothed out, particularly if there is a distribution of jump lengths, and after those times the form of G_s is given by (17). Thus, if measurements are made on the central peak of G_s (as is usual in substances with coherent scattering since the outer tails are obscured by G_d, and is certainly the case with Brockhouse and Pope (1959)), the diffusive broadening is not observed until times of two or three times τ.

§ 4. The Vibrational Motion

The form of $G_s(\mathbf{r}, t)$ in crystals has been extensively discussed by van Hove (1954), Vineyard (1958) and others, so that we give only the relevant results. For a cubic crystal

$$G_s{}^V(\mathbf{r}, t) = \left(\frac{L}{2\pi}\right)^{3/2} \exp{(-Lr^2)} \qquad \ldots\ldots(19)$$

where

$$L^{-1} = 2(M(0) - M(t)),$$

and the quantum mechanical expression for $M(t)$ is

$$\frac{\hbar}{2M} \int \frac{g(\nu)}{\nu} \left[1 - \exp{\left(-\frac{\hbar\nu}{kT}\right)}\right]^{-1} \left[\exp{(i\nu t)} + \exp{\left\{-\nu\left(it + \frac{\hbar}{kT}\right)\right\}}\right] d\nu,$$

$$\ldots\ldots(20)$$

$g(\nu)$ is the density of modes per unit frequency range. (20) contains all the complications due to the phonon spectrum, and the cross sections computed from it are conveniently divided into processes where a fixed number of phonons are created or destroyed. The most important contributions to the cross section come from elastic no-phonon processes where $\Gamma_s \sim \delta(\omega)$, one-phonon creation processes where

$$\Gamma_s{}^{(1)} = e^{-2W}\kappa^2 g(\omega)/\omega(1 - \exp{(-\hbar\omega/kT)}) \qquad \ldots\ldots(21)$$

and a similar term with an appropriate temperature factor for destruction processes. The first term in (21) is the Debye–Waller factor and

$$W = \tfrac{1}{2}\kappa^2 M(0). \qquad \ldots\ldots(22)$$

In the limits of high temperatures or high incident neutron energies many-phonon processes are important and this detail is not observed. Under these conditions it is possible to approximate to the width function (20) of G_s. The classical result (Vineyard 1958) is

$$L^{-1} = \int \frac{g(\nu)2kT}{M\nu^2} (1 - \cos{\nu t}) d\nu \qquad \ldots\ldots(23)$$

and Schofield (1960) gives an improved form for the appropriate γ'. The oscillatory nature of this width is rapidly damped out and as demonstrated by Vineyard the width increases with time quadratically at first and saturates with minor oscillations after $t \sim 1/\bar{\nu}$, at a value corresponding to the Debye–Waller factor. For a Debye spectrum this value is $L^{-1} = 12\hbar^2 T/Mk\theta^2$ where θ is the Debye temperature.

§ 5. RESULTS

The predictions of the diffusive model can be displayed in various ways using formulae (5)–(7). The integration in (5) to form G_s is tedious but the general behaviour is clear. At short times G_s takes the form of a Gaussian whose width increases with time as described by (20) or (23) and saturates at a value L beyond $t \sim 1/\bar{\nu}$. The diffusion at first causes peaks to appear in the wings of G_s at times of about τ and only when $t \sim 2\tau$ does the central peak of G_s become broadened by the diffusion.
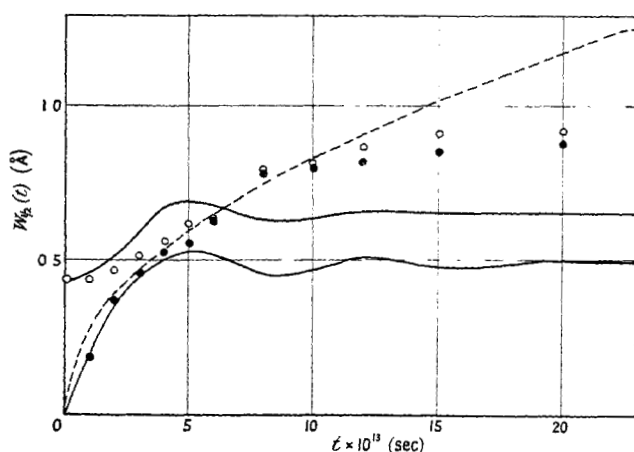

Fig. 1. The width at half height of G_s (full circles) and the first peak of G_d (open circles) in liquid Pb after Brockhouse and Pope (1959). The lower full curve is the width of G_s predicted using a Debye crystal model from Eqn (23). The upper full curve is obtained by adding the vibrational width to the original width of G_d at $t = 0$ using (28). The broken curve is that given by continuous diffusion.

A comparison with experiment is made most conveniently by comparing the widths of the central peak of G_s, which is essentially the same as the central peak of G in a coherent scatterer. This explicitly excludes the peaks which appear in the wings at short times. Thus the width is predicted to have a plateau between about $1/\bar{\nu}$ and 2τ. In Fig. 1 the results of Brockhouse and Pope are compared, the widths being predicted for Debye oscillations and continuous diffusion appropriate to his experimental conditions. This jump model predicts a width like the Debye model with some slight increase at large t and a very considerable increase at $t \sim 10^{-11}$ sec to raise the curve up to the diffusion value. The experimental results do show a rapid rise at short times followed by near saturation with a small slope. The magnitude of the saturation value is, however, almost twice as large as that predicted by the Debye theory. Thus, while the jump diffusion model is satisfactory in predicting a saturation of the width below the diffusion value in this time region, there appears to be a further source of broadening not treated in the model. One possibility arises from the fact that the nearest neighbours of one atom are found with a considerable spread about their mean distance. From the width of the first peak of $G_d(\mathbf{r}, 0)$ this is about $\delta_0 \sim 0.4$ Å in lead. An atom whose nearest neighbours are more distant than the average might be expected to move out more freely and so $G_s(\mathbf{r}, t)$ takes up a

width similar to that of the first peak of $G_d(\mathbf{r}, 0)$ at a rate appropriate to free motion. Such a width added to the vibrational width would certainly give reasonable agreement with experiment.

The cross section results transformed only once from their energy dependence to give $\gamma'(\mathbf{x}, t)$ are probably a more reliable source of comparison with theory. $\gamma_s'(\mathbf{x}, t)$ is given in a very convenient form for the above model

$$\gamma_s'(\mathbf{x}, t) = \exp\left[-\tfrac{1}{2}\kappa^2 L(t) - f(\mathbf{x})\zeta\right] \qquad \ldots\ldots(24)$$

using (11), (18) and (23) or its improvement given by Schofield. At small κ, $f(\mathbf{x})$ has in all cases the form $D\kappa^2$ which is appropriate to continuous diffusion. This is because small κ reflects the behaviour of G_s at large $\mathbf{r}$ where the discreteness of the jumping process is not noticeable. Conversely at large κ, $f(\mathbf{x})$ oscillates in the true lattices but allowing for disorder as in (15) or (16) it saturates, and leads to a term in Γ independent of κ. This reflects the fact that at small $\mathbf{r}$, G_s is given by the vibrational motions.

If the quasi-lattice model with phonons is applicable, the best mode of comparison is with $\Gamma(\mathbf{x}, \omega)$. The zero-phonon contribution is broadened in energy (sometimes called the quasi-elastic contribution) and has Lorentzian form

$$\Gamma_s^0(\mathbf{x}, \omega) = e^{-2W} \frac{2f(\mathbf{x})}{\omega^2 + f(\mathbf{x})^2}. \qquad \ldots\ldots(25)$$

One-phonon creation processes give

$$\Gamma_s^{+1}(\mathbf{x}, \omega) = \frac{h\kappa^2}{4\pi^2 M} e^{-2W} \int \frac{g(\nu)d\nu}{\nu[1 - \exp(-\hbar\nu/kT)]} \cdot \frac{f(\mathbf{x})}{(\omega + \nu)^2 + f(\mathbf{x})^2} \qquad \ldots\ldots(26)$$

and one-phonon destruction processes

$$\Gamma_s^{(-1)}(\mathbf{x}, \omega) = \frac{h\kappa^2}{4\pi^2 M} e^{-2W} \int \frac{g(\nu)d\nu}{\nu[\exp(\hbar\nu/kT) - 1]} \cdot \frac{f(\mathbf{x})}{(\omega - \nu)^2 + f(\mathbf{x})^2}. \qquad \ldots\ldots(27)$$

In the improved formulation of Schofield (1960), using (18) for the diffusive term in this product, the result is an infinite series of which the term in (25)–(27) is only the first. The effect of this series is to give a somewhat narrower function than the Lorentzian but having basically the same form.

The energy width of the quasi-elastic scattering is thus seen to depend on $\mathbf{x}$ like $f(\mathbf{x})$, i.e. it rises like $D\kappa^2$ at small κ and then saturates. The single-phonon contributions also have an extra broadening in energy because of the diffusion terms and the amount depends on the angle through $f(\mathbf{x})$. In a crystal the spread of these contributions is independent of angle, and because of the jump nature of the diffusion the extra broadening is a comparatively small effect, although it does lead to an increase in width with increasing angle. Fig. 2 shows a typical result actually evaluated for lead, although experimentally this would be difficult to separate from the G_d. In a hydrogenous substance the comparison with experiment is simpler. In his experiments with water Brockhouse (1959) notes that the energy width does increase with scattering angle but less rapidly than is expected for free or diffusional motion, which is the form of result predicted here. It would appear to be instructive to compare the inelastic cross sections with the crystal phonon form. Unfortunately the best results in this form by Hughes *et al.* (1959) on water show a considerable structure due to rotations. It would be interesting to have experiments on a more symmetrical hydrogenous (and hence incoherent) molecular scatterer like methane.
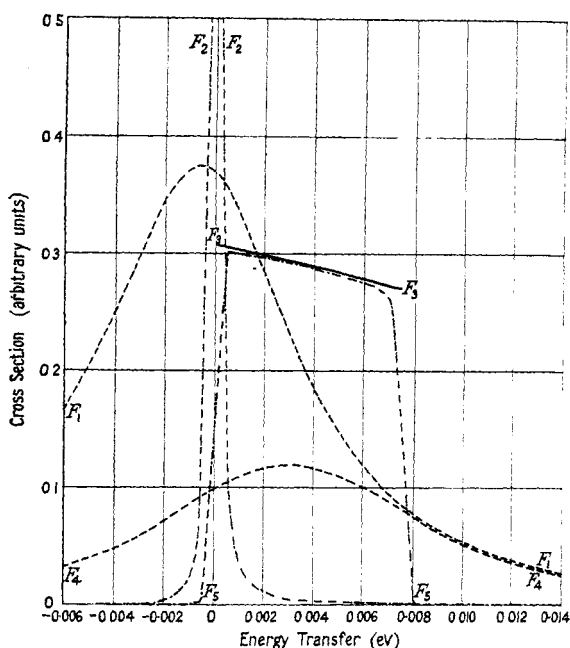
Fig. 2.   Neutron scattering cross sections as a function of energy transfer predicted by the model for the scattering of thermal energy neutrons at a scattering angle of 90° from liquid lead at a temperature of 600°K (the melting point) with a Debye temperature of 88°K.   The cross sections shown are:

F_1, no-phonon cross section on a continuous diffusion model;
F_2, no-phonon cross section on a jump diffusion model;
F_3, one-phonon gain cross section of lead as a Debye solid;
F_4, one-phonon gain cross section on the continuous diffusion model;
F_5, one-phonon gain cross section on the jump diffusion model.

The model of jump diffusion is therefore seen to lead to quite definite predictions about the nature of the neutron scattering in a liquid, which can be compared with the experimental results in various ways.   The data at present available seem to indicate that it does agree with many salient features, and it will be of some interest to see how far this crude model can explain the more detailed results which it is hoped will shortly be available.

§ 6. Discussion of G_d

In conclusion it seems appropriate to add a brief discussion of the behaviour of G_d on the jump diffusion model.   It is much more difficult to do this on a quantitative basis than it is for G_s unless one makes even more sweeping approximations—like the convolution approximation of Vineyard which is certainly bad at small distances where the crystalline structure is important.   It is possible to give some discussion of G_d on an *ad hoc* basis—and in particular of the first peak which is its most prominent feature.   At $t = 0$ the width of this peak is finite (δ_0) because of the randomness in the positions of the shell of first neighbours.   At short times the vibrations cause an increase in width

$$w = (\delta_0{}^2 + L^{-1})^{1/2}$$

where L is given by (25).   Results for a Debye model are shown in Fig. 1. ω increases slightly with time and then saturates in a manner similar to the experiment but at a low value.   The jump diffusion again does not influence the width much until $t \sim 2$ or 3τ.   At $t < \tau$ (the mean jump time) atoms in the first neighbour shell cannot jump to the origin because it is occupied, and rearrangements within the first shell will not affect the width of the first peak.   At $t > 2\tau$ all the peaks of G_d will be rapidly ironed out.

An alternative method of approach is through the inelastic cross sections on the basis of a lattice-like theory involving phonons.   The one-phonon processes in solids lead to $d^2\sigma/d\Omega d\epsilon$ which are a few δ-functions of energy which vary in their position with angle.   These lines will in a liquid be broadened in angle (i.e. in $\varkappa$) because of the smooth variation of $G_d(\mathbf{r})$.   They will be broadened in energy because of the diffusive motions.   If diffusion is by jumps the first form of broadening will be dominant except possibly in the case of very slow neutrons. A quasi-phonon picture of this type has been partly worked out by Butterworth and Marshall (1957).

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