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A STUDY OF LIQUIDS BY NEUTRON SCATTERING

by

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SUMMARY

A radial density-distribution function for liquids was derived with the aid of the assumption that there existed vibrational modes in the liquid. The distribution function was obtained only for elastically scattered neutrons.

A study of $s_i(s)$ curves as a function of temperature, as well as the energy distribution of the scattered neutrons, affords a way in which to construct a model describing the motion of atoms in a liquid.

I. Introduction

Interference effects observed in liquids by the scattering of X rays^(1,2) and low-energy neutrons⁽³⁻⁵⁾ indicate a regularity in the arrangement of the scatterers extending over several atomic distances from some origin atom.

In the case of X-ray scattering, Zernicke and Prins⁽⁶⁾ and Debye and Menke⁽⁷⁾ were the first to derive an expression for the radial density distribution from a differential scattering cross section based on the model of a solid composed of infinitely heavy scatterers; that is, temperature effects were neglected, since the change in the energy of the photon is negligible compared with the incident energy.

When temperature effects and energy transfer are not negligible, the motion of the scatterers must be considered. In liquids this motion is not well understood at present. Van Hove⁽⁸⁾ has presented a general theory of scattering for arbitrary particles in which a space- and time-dependent distribution function is introduced as a Fourier transform of the differential cross section. For liquids, when the assumption is made that atoms undergo a diffusive type of motion, in the limit of zero time displacement (static approximation) the formalism gives the ordinary radial distribution as derived by Zernicke and Prins.

Vineyard⁽⁹⁾ and Brockhouse,⁽¹⁰⁾ using the classical model of diffuse motion for liquids, have calculated the energy spread of the scattered neutrons. Recent experiments^(11,12) with water do not seem to verify this

broadening. Further, if the motion were purely diffuse, as proposed by Vineyard, the energy distribution of the scattered neutrons should be rather smooth. However, Hughes *et al.*⁽¹²⁾ and Larsson *et al.*,⁽¹³⁾ in contrast to Brockhouse, report the observation of definite peaks, which indicate quantum effects similar to those used in describing the motion of atoms in a solid.

The hypothesis that the liquid state may be considered as a polycrystalline material has been used fairly successfully by many investigators in studying specific heats as well as other thermodynamic quantities of liquids.⁽¹⁴⁾

Perhaps the diffuse motion of the atoms in a system may be considered as a cooperative diffusive motion, giving the effect of an oscillatory motion at some position of equilibrium.

The purpose of this and subsequent papers is to study the structure of fluids by means of a "quasi-polycrystalline" model for liquids. In other words, it is assumed that the atoms are in "vibrational motion" for a time long compared with the time associated with diffuse motion. Any diffusive motion of the atoms (molecules or aggregates of atoms) is considered as having a negligible effect on the scattering.

The differential scattering cross section for a system of oscillating atoms has been explicitly calculated by Zemach and Glauber⁽¹⁵⁾ and by others.⁽¹⁶⁾ The time- and space-dependent distribution function for this model has been considered by Van Hove and leads to the above result. In the present paper, however, the development of the radial density-distribution function starts with the explicit form of the differential cross section and is Fourier transformed later in the analysis. This procedure is essentially that of Zernicke and Prins.

Aside from starting with a differential cross section which involves temperature effects, the radial distribution obtained in this paper includes an angle-dependent scattering factor. This factor is usually neglected at small-angle scattering since it involves the size of the scattering sample, which is assumed to be large. The derivation in this paper leads to the small-angle effect which involves the size of the "crystallites" in a polycrystalline material rather than the sample size. This means that the factor manifests its effect at larger angles than is usually expected. Experimentally, this small-angle scattering is avoided and the intensity is then extrapolated to zero as the scattering angle approaches zero. The present analysis suggests that experiments include intensity measurements at smaller angles than those currently used.

The present paper is concerned with the preliminary analysis and will involve only zero phonon processes: elastic collisions in which no energy or momentum has been transferred to the system of scatterers. The effect of multi-phonon processes, elastic and inelastic, will be treated in a subsequent paper.

The differential cross section σ for a neutron with incident energy E_0 (wave vector $\underline{K}_0$) being scattered by a monoatomic system of vibrating nuclei about equilibrium positions $\underline{R}$, into the elemental solid angle $d\Omega$ about Ω and into the energy interval dE about E (wave vector $\underline{K}$) may be written as

$$\frac{d^2\sigma}{d\Omega dE} = \frac{d^2\sigma_{\text{inc}}}{d\Omega dE} + \frac{d^2\sigma_{\text{coh}}}{d\Omega dE} ,$$

where

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega dE} = \frac{K}{K_0} \frac{m_0^2}{\mu^2} N a_{\text{inc}}^2 e^{-2W} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\alpha e^{-i\alpha \Delta E} \times$$

$$\sum_{\ell=0}^{\infty} \frac{1}{\ell!} \left[\sum_{s=1}^{3N} \frac{\hbar [(\underline{K}_0 - \underline{K}) \cdot \underline{e}_s]^2}{4mN\omega_s \sinh(\hbar\omega_s/2k_0T)} \sum_{\eta_s=\pm 1} e^{-\frac{\eta_s \hbar \omega_s}{2k_0T}} e^{-i\alpha \eta_s \hbar \omega_s} \right]^{\ell} ,$$

and

$$\frac{d^2\sigma_{\text{coh}}}{d\Omega dE} = \frac{K}{K_0} \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} \sum_j \sum_k e^{i(\underline{K}_0 - \underline{K}) \cdot (\underline{R}_j - \underline{R}_k)} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\alpha e^{-i\alpha \Delta E} \times$$

$$\sum_{\ell=0}^{\infty} \frac{1}{\ell!} \left[\sum_{s=1}^{3N} \frac{\hbar [(\underline{K}_0 - \underline{K}) \cdot \underline{e}_s]^2}{4mN\omega_s \sinh(\hbar\omega_s/2k_0T)} e^{i\underline{q}_s \cdot (\underline{R}_j - \underline{R}_k)} \sum_{\eta_s=\pm 1} \times \right.$$

$$\left. e^{-\frac{\eta_s \hbar \omega_s}{2k_0T}} e^{-i\alpha \eta_s \hbar \omega_s} \right]^{\ell} .$$

These cross sections are readily obtained from Zemach and Glauber's⁽¹⁵⁾ Eqs. (3.19) and (3.20) by substituting the generating function for the summation term in Eq. (3.19), expanding the resulting exponential in a Taylor Series (phonon expansion), and using

$$x = \frac{\hbar [(\underline{K}_0 - \underline{K}) \cdot \underline{e}_s]^2}{2\omega_s mN \sinh(\hbar\omega_s/2k_0T)} e^{i\underline{q}_s \cdot (\underline{R}_j - \underline{R}_k)}$$

as the argument of the Bessel function.

The cross sections describe the process in which ℓ phonons (vibrational energy of frequencies ω_s and wave vectors $\underline{q}_s$) are emitted ($\eta_s = -1$) or absorbed ($\eta_s = +1$) by the system of N scatterers after interacting with a neutron whose energy has been changed by $\Delta E = E - E_0$.

The definitions of the remaining terms are as follows: m_0 is the neutron mass, m is the mass of the nuclei, μ is the reduced mass of the system, $\underline{e}_s$ is the unit polarization vector of the propagation vector $\underline{q}_s$, $\hbar$ is Planck's constant divided by 2π , k_0 is Boltzmann's constant, and T is the temperature of the scattering system.

The incoherent scattering length a_{inc} , due to isotope and spin effects, and the coherent scattering length a_{coh} are defined as⁽¹⁷⁾

$$a_{coh}^2 = \left[\sum_i p_i (w_+ a_{i+} + w_- a_{i-}) \right]^2$$

$$a_{inc}^2 = \sum_i p_i (w_+ a_{i+}^2 + w_- a_{i-}^2) - a_{coh}^2 \quad ,$$

where p_i is the abundance of the i^{th} isotope with scattering amplitude a_{i+} for total spin $I + \frac{1}{2}$, and a_{i-} for total spin $I - \frac{1}{2}$. The weighting factors w_+ and w_- are

$$w_+ = \frac{I+1}{2I+1} \quad ; \quad w_- = \frac{I}{2I+1} \quad .$$

The exponent in the Debye-Waller intensity factor e^{-2W} is given by

$$2W = \sum_{s=1}^{3N} \frac{\hbar [(\underline{K}_0 - \underline{K}) \cdot \underline{e}_s]^2}{2mN\omega_s} \coth \frac{\hbar\omega_s}{2k_0T} \quad ,$$

and with the approximation

$$\left[(\underline{K}_0 - \underline{K}) \cdot \underline{e}_s \right]_{avg}^2 = \frac{(\underline{K}_0 - \underline{K})^2}{3} \quad ,$$

becomes

$$2W = (\underline{K}_0 - \underline{K})^2 \sum_{s=1}^{3N} \frac{\hbar}{6mN\omega_s} \coth \frac{\hbar\omega_s}{2k_0T} \quad .$$

The Debye-Waller factor is a measure of the reduction in the intensity of the scattered beam due to the thermal motion of the scatterers.

The main features in the analysis leading to the radial density-distribution function may be presented by considering the zero-phonon ($\ell = 0$) process. This is essentially the first-order approximation for an elastic collision, $|\underline{K}| = |\underline{K}_0|$. The broadening in the angular distribution of the elastically scattered neutrons results from phonon processes of higher order ($\ell \geq 2$).

The incoherent and coherent cross sections for $\ell = 0$ reduce to

$$\frac{d\sigma_{\text{inc}}}{d\Omega} = \frac{m_0^2}{\mu^2} N a_{\text{inc}}^2 e^{-2W} ,$$

and

$$\frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} \sum_{j=1}^N \sum_{k=1}^N e^{i(\underline{K}_0 - \underline{K}) \cdot (\underline{R}_j - \underline{R}_k)} .$$

In a polycrystalline material, the geometric order of the atoms, which maximizes the coherent cross section, extends only over the crystallite if the crystallites are randomly oriented. If it be assumed that the crystallites in the sample to have an effective radius r_1 , the summations in the coherent cross section is performed over the atoms within this distance and then summed over all the crystallite. If N_C is the number of crystallites in the scattering sample and N_A is the number of atoms in the crystallite of size r_1 , then the cross section may be written as

$$\frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} \sum_{n=1}^{N_C} \left[\sum_{j=1}^{N_A} \sum_{k=1}^{N_A} e^{i|\underline{K}_0 - \underline{K}| |\underline{R}_j - \underline{R}_k| \cos \alpha_n} \right] ,$$

where α_n is the angle between the vectors $(\underline{K}_0 - \underline{K})$ and $(\underline{R}_j - \underline{R}_k)$, and defines the orientation of the n^{th} crystallite. The effect of random orientation is taken into account by averaging over $\cos \alpha_n$. Taking the co-ordinate axis to coincide with the direction $(\underline{K}_0 - \underline{K})$ and with the elemental solid angle $d\Omega_n = 2\pi \sin \alpha_n d\alpha_n$, the average of the exponential is

$$\frac{1}{4\pi} \int_{\Omega_n} d\Omega_n e^{isr_{jk} \cos \alpha_n} = \frac{\sin sr_{jk}}{sr_{jk}} ,$$

where $s = |\underline{K}_0 - \underline{K}|$ and $r_{jk} = |\underline{R}_j - \underline{R}_k|$. The averaged coherent differential cross section becomes

$$\frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} N_C \left[\sum_{j=1}^{N_A} \sum_{k=1}^{N_A} \frac{\sin sr_{jk}}{sr_{jk}} \right] .$$

Summing over j and k , the double summation is written as

$$\sum_{j=1}^{N_A} \sum_{k=1}^{N_A} \frac{\sin sr_{jk}}{sr_{jk}} = N_A + N_A \sum_{k=1}^{N_A-1} \frac{\sin sr_{jk}}{sr_{jk}} ,$$

where the first term is for $j = k$ and the second term is the sum over the distances r_{jk} of the k^{th} atoms from an origin atom j . Since the total number of atoms in the scattering sample is $N = N_A N_C$, the coherent cross section is then

$$\frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} N \left[1 + \sum_{k=1}^{N_A-1} \frac{\sin sr_{jk}}{sr_{jk}} \right] .$$

This expression is analogous to that derived by Debye and Menke for X rays. The relation for X rays with isotope and spin effects neglected may be obtained by replacing $(m_0^2/\mu^2) a_{\text{coh}}^2$ with the atomic structure factor which is dependent on the scattering angle. Aside from the Debye-Waller intensity factor, another difference is that the above relation is summed over the interatomic distances within the crystallite rather than over the whole of the sample.

The summation term is transformed into an integral form by introducing a radial density-distribution function $4\pi\rho(r)$ and integrating over the range of r from zero to the effective radius of the crystallite r_1 . For the case of liquids, this effective radius is the extent to which atoms may be considered as being arranged in some order from an origin atom. The distribution function is normalized so that

$$\int_{r_a}^{r_b} dr 4\pi r^2 \rho(r) = N_{a,b}$$

is the number of atoms about an origin atom contained within the spherical shell having an outer radius r_b and an inner radius r_a . The coherent cross section in terms of the radial density-distribution function then becomes

$$\frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} N \left[1 + \int_0^{r_1} dr 4\pi r^2 \rho(r) \frac{\sin sr}{sr} \right] .$$

The upper limit of integration may be extended beyond r_1 by introducing the average radial density ρ_0 , defined as

$$\rho_0 = \frac{\int_0^R dr 4\pi r^2 \rho(r)}{\int_0^R dr 4\pi r^2}$$

for large R , or

$$\rho_0 = \rho(r) \text{ for } r \geq r_1 \quad .$$

By adding and subtracting the term

$$\int_0^{r_1} dr 4\pi r^2 \rho_0 \frac{\sin sr}{sr} \quad ,$$

then, from the above condition the cross section is written as

$$\begin{aligned} \frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} N \left[1 + \int_0^\infty dr 4\pi r^2 \left\{ \rho(r) - \rho_0 \right\} \frac{\sin sr}{sr} \right. \\ \left. + \int_0^{r_1} dr 4\pi r^2 \rho_0 \frac{\sin sr}{sr} \right] . \end{aligned}$$

With the last term evaluated as

$$\int_0^{r_1} dr 4\pi r^2 \rho_0 \frac{\sin sr}{sr} = \frac{4\pi r_1^3}{3} \rho_0 \Phi(sr_1) = N \left(\frac{r_1}{R} \right)^3 \Phi(sr_1) \quad ,$$

where

$$\Phi(sr_1) = 3 \left\{ \frac{\sin sr_1 - sr_1 \cos sr_1}{(sr_1)^3} \right\} \quad ,$$

$$s = |\underline{K}_0 - \underline{K}| = 2k_0 \sin \frac{1}{2} \theta \quad ,$$

and θ being the scattering angle, the coherent cross section becomes

$$\begin{aligned} \frac{d\sigma_{\text{coh}}}{d\Omega} = \frac{m_0^2}{\mu^2} a_{\text{coh}}^2 e^{-2W} N \left[1 + \int_0^\infty dr 4\pi r^2 \left\{ \rho(r) - \rho_0 \right\} \frac{\sin sr}{sr} \right. \\ \left. + N \left(\frac{r_1}{R} \right)^3 \Phi(sr_1) \right] . \end{aligned}$$

The last term is the small-angle-scattering correction term, at $s \approx 0$, $\Phi(sr_1) \approx 1$. At large scattering angle, $\Phi(sr_1)$ approaches zero and, depending on the magnitude of r_1 , the term may be neglected.

The observed differential cross section is the sum of the coherent and incoherent parts:

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{obs}} = \frac{d\sigma_{\text{coh}}}{d\Omega} + \frac{d\sigma_{\text{inc}}}{d\Omega} ,$$

so that

$$\begin{aligned} \frac{\mu^2}{m_0^2} \frac{e^{2W}}{N a_{\text{coh}}^2} \left(\frac{d\sigma}{d\Omega}\right)_{\text{obs}} &= \frac{a_{\text{coh}}^2 + a_{\text{inc}}^2}{a_{\text{coh}}^2} + N \left(\frac{r_1}{R}\right)^3 \Phi(sr_1) \\ &+ \int_0^\infty dr \, 4\pi r^2 \left\{ \rho(r) - \rho_0 \right\} \frac{\sin sr}{sr} . \end{aligned}$$

Defining

$$\sigma = 4\pi \left\{ a_{\text{coh}}^2 + a_{\text{inc}}^2 \right\} = \sigma_{\text{coh}} + \sigma_{\text{inc}}$$

as the total scattering cross section for a bound isolated nucleus, and letting

$$i(s) = \left\{ \frac{\mu^2}{m_0^2} \frac{e^{2W}}{N \sigma_{\text{coh}}} \left(\frac{d\sigma}{d\Omega}\right)_{\text{obs}} - \frac{\sigma}{\sigma_{\text{coh}}} - N \left(\frac{r_1}{R}\right)^3 \Phi(sr_1) \right\} ,$$

the radial density relation becomes

$$si(s) = \int_0^\infty dr \, 4\pi r \left\{ \rho(r) - \rho_0 \right\} \sin sr .$$

From the Rieman-Lebesque lemmas,⁽¹⁸⁾ if

$$\int_0^\infty dr \, 4\pi r \left\{ \rho(r) - \rho_0 \right\}$$

exists and converges absolutely, and further if $4\pi r \left\{ \rho(r) - \rho_0 \right\}$ has limited total fluctuation in the range $(0, \infty)$ then, as $s \rightarrow \infty$,

$$si(s) \rightarrow 0 \, (1/s) .$$

With this condition in $si(s)$, the Fourier transform of the $si(s)$ relation yields the radial density distribution

$$4\pi r^2 \left\{ \rho(r) - \rho_0 \right\} = \frac{2r}{\pi} \int_0^\infty ds \, si(s) \sin sr \quad .$$

Measurements are made only over finite values of s . To satisfy the condition on $si(s)$, the usual procedure is to normalize the experimental data such that at large s , $i(s) = 0$, or

$$\left(\frac{d\sigma}{d\Omega} \right)_{\text{obs}} = \frac{m_0^2}{\mu^2} e^{-2W} \frac{N\sigma_{\text{coh}}}{4\pi} \left[\frac{\sigma}{\sigma_{\text{coh}}} + N \left(\frac{r_1}{R} \right)^3 \Phi(sr_1) \right] \quad .$$

The only unknown quantity in this expression is r_1 . In practice, for an experimentally realizable $s \sim 12\text{\AA}^{-1}$ and $r_1 > 10\text{\AA}$, the last term may be dropped.

However, the effective size of the crystallites may be obtained from the data at small-angle ($s \approx 0$) scattering. Taking the limit of $i(s)$ as $s \rightarrow 0$, then one gets

$$\lim_{s \rightarrow 0} i(s) = \lim_{s \rightarrow 0} \int_0^\infty dr \, 4\pi r^2 [\rho(r) - \rho_0] \frac{\sin sr}{sr} = 0 \quad ,$$

since

$$\int_0^\infty dr \, 4\pi r^2 [\rho(r) - \rho_0] = 0 \quad .$$

The above relation for the differential cross section is then valid for very small angles. In this limit, the Debye-Waller factor (which is a function of s^2) and $\Phi(sr_1)$ approach unity so that the differential cross section becomes

$$\left(\frac{d\sigma}{d\Omega} \right)_{\text{obs}} = \frac{m_0^2}{\mu^2} \frac{N\sigma_{\text{coh}}}{4\pi} \left[\frac{\sigma}{\sigma_{\text{coh}}} + N \left(\frac{r_1}{R} \right)^3 \right] \quad .$$

If the latter term predominates, the N^2 intensity behavior is modified by the crystallite and sample volume ratio.

II. Discussion

The above analysis gives a radial density-distribution relation which differs from previously and currently used expressions in 2 respects. The first difference is the inclusion of the Debye-Waller intensity factor, which results from the assumed vibrational motion of the atoms (temperature effect). The second difference, independent of the model used in describing the motion of atoms, is the addition of the term which involves the effects of small-angle scattering.

An experimental check on the existence or absence of vibrational modes in liquids is to study the temperature effect on the intensity through the Debye-Waller factor.

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