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MASTER

SCATTERING LAW FOR LIQUID SODIUM AT
SMALL MOMENTUM TRANSFERS

P. D. Randolph

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AT SMALL MOMENTUM TRANSFERS

P. D. Randolph

IDAHO NUCLEAR CORPORATION

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ABSTRACT

The thermal neutron scattering law for liquid sodium at 102°C has been measured for the momentum transfer range $\hbar\kappa$ of $0.3 \lesssim \kappa \lesssim 1.5 \text{ \AA}^{-1}$ for energy transfers out to 0.032 eV. These measurements extend earlier scattering law measurements to the region of small momentum transfer where incoherent scattering predominates. The measurements were made at small scattering angles (3° to 18°) in an evacuated flight path at incident energies of 30 and 51 meV. The scattering law results are presented combined with the older measurements. Agreement is good except at small energy transfers where resolution effects are important. Analysis of the quasi-elastic incoherent peaks gives a Debye-Waller coefficient of $0.22 \text{ \AA}^2$. A width analysis of the peaks indicates that liquid sodium acts like a solid for times $\geq 1.5 \times 10^{-12}$ sec.

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SCATTERING LAW FOR LIQUID SODIUM

AT SMALL MOMENTUM TRANSFERS

I. INTRODUCTION

In studying the dynamics of microscopic atomic motion in simple liquids, the scattering law $S(\kappa, \omega)$ plays a central role. It contains information, on a short time scale, of self diffusive or single particle motion and information about the correlated motion of pairs of atoms, and it is important for obtaining the scattering kernels for reactor calculations. The methods of slow neutron inelastic scattering give the most complete information presently obtainable on $S(\kappa, \omega)$. The variables in the scattering law are the neutron momentum transfer $\hbar\kappa$ and the energy transfer $\hbar\omega$. The kind of motion sensed by the neutron is determined by the nuclear scattering cross section of the atoms involved. If the cross section is incoherent, the single particle motion, or self diffusion is observed. For coherent cross sections, the correlated motion of pairs of particles is measured. Most monatomic real liquids are purely coherent or a combination of both types of cross sections, and in the latter case the observed scattering cannot in general be separated into self diffusion and pair motion over a wide range of momentum transfer except by the use of theoretical models such as the convolution approximation⁽¹⁾ or one of its variations. Of the monatomic simple liquids, only sodium^(2,3), lead⁽⁴⁾, argon⁽⁵⁾ and aluminum⁽⁶⁾ have been extensively studied. Of these materials the measurements on liquid sodium⁽²⁾ span the largest range of momentum transfer ($1 \leq \kappa \leq 10 \text{ \AA}^{-1}$). The scattering in this range contains the interference effects of the correlated motion as well as the diffusive

contributions. The theoretical models available to separate these two types of contributions are not entirely adequate. In the region $\kappa \leq 1.5 \text{ \AA}^{-1}$, however the interference effects are much smaller, and the primary contributions to the scattering arise from the single particle motions.

In this report we present scattering law measurements for liquid sodium just above its melting point in the small momentum transfer region. This is an extension of the earlier measurements⁽²⁾ made at this laboratory. The new measurements span the momentum transfer range $.22 \leq \kappa \leq 1.5 \text{ \AA}^{-1}$ and energy transfer range of $0 \leq \hbar\omega \leq 32 \text{ meV}$. Besides presenting the data in the form of scattering law, an analysis of the quasi-elastic scattering is presented. This analysis shows that liquid sodium near its melting point acts much like a solid with a Debye-Waller coefficient of $0.22 \text{ \AA}^2$ and a Debye temperature for the diffusive motions of 103°K .

II. DESCRIPTION OF THE EXPERIMENT

The experiment was performed using the MTR phased chopper velocity selector. In order to obtain small values of κ with thermal neutrons, small scattering angles are necessary. A cadmium-lined evacuated flight path and scattering chamber was installed to remove air scattering. The flight path subtended scattering angles between 0 and 20 degrees, with a sample-to-detector flight path of 2.26 meters. Figure 1 shows the experimental geometry. The sample was completely surrounded by a cadmium shield except at the position of the entrance and exit beams. A short (19 cm) 5-slit cadmium collimator was installed between the last chopper and the cadmium shield which served to reduce undesirable time dependent background near the elastic peaks. The detectors were

in 12 sets of two one-inch diameter, 10 atmosphere helium-3 proportional counters. Each bank of two counters subtended 1.4 degrees at the sample. This gives a spread in κ of $\pm 0.06 \text{ \AA}^{-1}$ for elastic scattering. The scattering angles ranged from 3.3 to 18.3° .

The sample was in the form of a rectangular slab 11.4 cm high by 7.6 cm wide having a thickness of 0.635 cm. It was mounted with the long dimension perpendicular to the scattering plane and intercepted a beam of 10.1 cm by 3.2 cm. For the small scattering angles used, the sample orientation was normal to the beam. The sample container windows were of 0.0025 cm vanadium. In the κ region of interest, at the elastic peak, the vanadium windows contribute about 14 per cent of the total scattering observed, but this is removed on background subtraction. The measured transmission of the sample was 0.94 . The sodium was controlled at $375.8 \pm 2^\circ\text{K}$ (102°C) by means of copper-constantan thermocouples and temperature control was obtained using a chart recorder.

Data runs were made at incident energies of 30.5 and 51.0 meV at a chopper speed of $10,000$ rpm. The 12 angles spanned a κ range from 0.22 to $1.4 \text{ \AA}^{-1}$ for elastic scattering. Background runs were obtained by alternately cycling the filled sample holder and an empty holder into the beam at 10 minute intervals. The time-of-flight analyzer was used with 256 channels of $5 \text{ \mu sec}$ for each scattering angle. For the 30.5 meV data both the sample and the background were run for 187 hours, while for the 51 meV run the time was 155 hours each. The runs were made long because the inelastic scattering in liquid sodium in this κ range is small, and also because the presence of the Cd collimator just ahead of the sample reduced the incident beam by about 25 per cent. The velocity selector was calibrated for absolute cross section measurements

at 30 meV by comparison with a vanadium standard. The vanadium was in the form of rectangular bars 0.475 cm x 0.475 cm x 5 cm long. Each pair of bars was separated by 0.050 cm cadmium spacers to reduce multiple scattering. Background corrections for the calibration run were made with an identical sample holder without the vanadium but containing the cadmium spacers. Figure 2 shows an example of raw data for the 30 meV run at 11.53° and the scattering cross section. Even though background counting rates were low in the inelastic region, (~ 1.3 counts/channel hour), the signal-to-background ratio is only about 2:1. In order to smooth the inelastic data somewhat, a bipolar average was taken, i.e. the counts in a given channel were taken to be the average of the counts in the channel in either side of it ($N_j = 1/2(N_{j-1} + N_{j+1})$). This was used only in analyzing the inelastic data, no averaging was done for the quasi-elastic peaks. The measured energy resolution for elastic scattering as obtained from the shape of forward beam bursts was 1.25 meV for the 30 meV data and 2.4 meV for the 51 meV data.

III. RESULTS

In Figure 3 are shown representative plots of the inelastic partial differential cross section for the 30.5 eV ($1.638 \text{ \AA}$) data. As can be seen from Figure 2, the quasi-elastic cross section is ~ 50 times larger than the inelastic cross section. The inelastic data has some features of interest, which can be most easily observed on the short wavelength side of the elastic peaks. The main feature which appears for all scattering angles is a broad peak at ~ 1.35 to $1.4 \text{ \AA}$ which at the larger angles becomes a shoulder. Two other features appear in some of the angles. Thus there appears to be a small shoulder at $\sim 1.42 \text{ \AA}$ and at

Figure 1 - Plan view of the velocity selector geometry showing the evacuated small angle flight path.

Figure 2 - a) Raw data at 11.53 degrees. The elastic peak in the background is due to the vanadium windows of the sample holder. b) Scattering cross sections obtained from this data.

Figure 3 - Inelastic scattering cross sections for liquid sodium.
 ---- Estimated multiple and inelastic scattering. — Cross section
 for a Debye frequency spectrum including one and two phonon contributions.
 The vertical lines show the assumed boundary of the quasi-elastic peak.

1.54 Å in some of the angles. If these are real, the fact that they appear only in some angles indicates they could be due to coherent inelastic scattering, i.e. excitation of quasi-phonons in the liquid. The statistical fluctuations of the data in this region however are sufficiently large that these little peaks or shoulders may not be real.

A. SCATTERING LAW

In this section the new data are presented in the scattering law form together with the results obtained from the earlier experiment⁽²⁾. The form of the scattering law used is the reduced partial differential cross sections $S_R(\kappa, \omega)$. This is defined in terms of the energy and angle dependent scattering cross section by

$$\frac{d^2\sigma(E_o, E, \theta)}{dE d\Omega} = \left(\frac{E}{E_o}\right)^{1/2} e^{-\frac{\beta}{2}} S_R(\kappa, \beta)$$

$$\beta = \frac{E - E_o}{k_B T} = \frac{\hbar\omega}{k_B T}$$

$$\vec{\kappa} = \vec{k} - \vec{k}_o$$

where E_o , E and $\vec{k}_o$, $\vec{k}$ are the incident and scattered neutron energies and wave vectors, respectively. The scattering law $S_R(\kappa, \beta)$ has the dimensions of barns/[eV steradian atom] and is an even function of the neutron energy transfer $\hbar\omega$. Also since the sample is isotropic it depends on the neutron momentum transfer $\hbar\vec{\kappa}$ only through the magnitude of $\vec{\kappa}$.

This form of the scattering law is related to the dimensionless form⁽⁷⁾ $S(\alpha, \beta)$ by

$$S_R(\kappa, \beta) = \frac{\sigma_b}{4\pi k_B T} S(\alpha, \beta)$$

where

$$\alpha = \frac{\hbar^2 \kappa^2}{2M k_B T}.$$

The mass of the scattering atom is M , T is the absolute temperature and σ_b is the bound atom scattering cross section. Figures 4, 5, and 6 show results combining the data of this experiments (diamond shaped points) with the earlier data⁽²⁾. There has been no normalization of the new to the old data. For β values $\geq .08$ (2.57 meV) the agreement between the two experiments is quite good. As the energy transfer increases the scatter in the new data increases due to counting statistics. Beyond $\beta = .5$ only the energy gain points which have better statistics are plotted for the 30 meV data. For $\beta \leq .06$ the new data show a considerable amount of splitting. The energy loss points lie below the energy gain points. Since $S_R(\kappa, \beta)$ is even in β , the two sets of points should be on the same line. This region lies at the base of the sharp (in energy) quasi-elastic peak. The splitting is attributed mainly to a small contribution of multiple elastic scattering which tends to give a slight distortion to the quasi-elastic peak shape. The symmetric form of the scattering law is very sensitive to small distortions of this type. In addition to the splitting, there is a marked discrepancy in the two sets of data at $\beta = 0$ and 0.02. This is due to the difference in energy resolutions used in the two experiments. The newer experiment has an energy resolution of 1.25 meV while the resolution for the older data is 2.0 meV. The data with the better resolution should be, and is, higher in the quasi-elastic peak.

Figure 4 - Scattering law data for liquid sodium at 100°C for $0 \leq \beta \leq .15$.

Figure 5 - Scattering law data for liquid sodium at 100°C for $.20 \leq \beta \leq .70$.

Figure 6 - Scattering law data for liquid sodium at 100°C for $.80 \leq \beta \leq 1.5$.

In order to see how the scattering law looks as a whole, a hand drawn smooth curve scattering law is shown in Figure 7. Since resolution effects have not been removed for either experiment they are present in both sets of data. They are most pronounced for $\beta \leq .08$. For $\beta = 0$ and 0.02 resolution effects are sufficiently large that no curve could be drawn that fitted smoothly onto the older results.

B. QUASI-ELASTIC PEAKS

In liquid sodium the static structure factor $S(\kappa)$ which is due to coherent interference scattering has its main maximum at $\kappa = 2.02 \text{ \AA}^{-1}$. The quasi-elastic data of this experiment lie below this, between 0.2 and $1.5 \text{ \AA}^{-1}$, and arise predominantly from incoherent scattering. The structure factor, though small in this region, is not zero, so that the observed quasi-elastic peaks have some coherent scattering. In addition, there are expected to be some multiple scattering and inelastic scattering contributions in the peak.

The quasi-elastic peaks have been analyzed to obtain the Debye-Waller factor for the liquid, and quasi-elastic widths have been obtained. The Debye-Waller factor was obtained after correcting the data for estimated contributions for inelastic and multiple scattering at the base of the peaks, and further correcting for the known coherent contribution. In Figure 3 are shown (dotted lines) the estimated combined contributions of multiple scattering and inelastic single scattering. The quasi-elastic peak was assumed to be the region between the vertical lines in Figure 3. These were estimated by eye, but as will be shown, are reasonable. The correction for coherent contributions was calculated using the experimental structure factor $S(\kappa)$ given by Gingrich and Heaton⁽⁸⁾.

Figure 7 - Smooth curve scattering law from the data of Figures 4, 5 and 6.

$$\left. \frac{d\sigma}{d\Omega} \right)_{\text{coh}} = \frac{\sigma_{\text{coh}}}{4\pi} S(\kappa).$$

The value of σ_{coh} of 1.55 barns⁽⁹⁾ was used. The results are shown in Figure 8. Figure 8a shows the uncorrected values for the integrated quasi-elastic peaks. Also shown are the contributions from coherent scattering and the estimated values of the inelastic and multiple scattering cross sections. Note that the multiple and inelastic contribution is of the same order and has about the same shape as the coherent contribution. At small κ , the total correction amounts to about 4 per cent and gradually rises to about 30 per cent at $\kappa = 1.4 \text{ \AA}^{-1}$ and is not in general negligible. Figure 8b shows the corrected incoherent elastic cross sections plotted as $\ln \frac{d\sigma}{d\Omega}$ against κ^2 , and combines data from both the 30 and 51 meV runs. The data have been least squares fit to the form:

$$\left. \frac{d\sigma}{d\Omega} \right)_{\text{inc}} = \frac{\sigma_{\text{inc}}}{4\pi} e^{-a\kappa^2}.$$

This is the form of the cross section for the elastic scattering from a solid incoherent scatterer. The solid line in Figure 8b shows the fit. The value of σ_{inc} given by the intercept is 1.98 barns compared to the published value $\sigma_{\text{inc}} = \sigma_{\text{b}} - \sigma_{\text{coh}} = 1.85$ barns. The Debye-Waller coefficient obtained is $a = 0.221 \text{ \AA}^2$. The value of σ_{inc} from this fit is 7 per cent higher than the published one. That this discrepancy is small gives some confidence to the estimates of multiple scattering. If multiple elastic scattering were a relatively large effect in liquid sodium at these κ 's the measured elastic cross sections would be much higher than observed, due to elastic double scattering around the main maximum of $S(\kappa)$.

Figure 8 - Angular distribution of the quasi-elastic peaks. a) Uncorrected data. b) Corrected distribution.

Using the relation valid for $T > \theta$, $\alpha\kappa^2 = \frac{\kappa^2 R^2}{6} \approx \kappa^2 \left(\frac{6h^2 T}{2M\kappa_B \theta^2} \right)$ we obtain a value for the RMS displacement of $R = 1.15 \text{ \AA}$ and a Debye temperature of $\theta \approx 103^\circ\text{K}$.

Widths of the quasi-elastic peaks have been obtained from the 30.5 meV data which has the better energy resolution. These widths were corrected for energy resolution by assuming the time-of-flight distribution of the incident beam is Gaussian in shape, and that the natural line width is a Lorentzian in energy. The ratio of the Lorentzian width to the observed width changed from .15 to .33, showing that most of the observed line width is due to the finite resolution of the incident beam. Figure 9 shows the quasi-elastic widths plotted vs. κ^2 . Also shown is the simple diffusion model, and several curves for the jump diffusion model⁽¹⁰⁾ given by:

$$\Delta E = \frac{2\hbar}{\tau_0} \left(1 - \frac{e^{-\alpha\kappa^2}}{1 + D\kappa^2 \tau_0} \right) .$$

The width given by this expression is the limiting case for jump diffusion when the jump time for an atom is very much smaller than the characteristic time τ_0 during which an atom is assumed to oscillate about a fixed position. In evaluating this expression the value of $D = 4.3 \times 10^{-5} \text{ cm}^2/\text{sec}$ has been taken for the macroscopic coefficient of self diffusion⁽¹¹⁾, and the Debye-Waller coefficient of $a = 0.22 \text{ \AA}^2$ as obtained in this experiment. At larger κ^2 the widths are narrower than the simple diffusion model predicts. This is characteristic of liquids. Above $\kappa^2 \approx 0.3 \text{ \AA}^{-2}$ the jump diffusion model with a value of $\tau_0 = 1.5 \times 10^{-12} \text{ sec}$ gives the best fit. In the region below $\kappa^2 = 0.3 \text{ \AA}^{-2}$, the data lie higher than any of the curves. Since these widths should approach zero, we conclude that the

energy resolution is too great a fraction of the observed total width to allow a good determination of the natural line width for $\kappa^2 \leq 0.3 \text{ \AA}^{-2}$. In the cold neutron experiment of Cocking⁽³⁾ a value of $3.6 \times 10^{-12} \text{ sec}$ was obtained on the assumption of jump diffusion for data analyzed at four angles in the range $0.5 < \kappa \leq 1.5 \text{ \AA}^{-1}$. We are not able to explain this discrepancy.

C. INELASTIC SCATTERING

The inelastic shoulder at $\kappa \approx 1.35 \text{ \AA}$ appears in all the spectra (Figure 3), and increases in amplitude with scattering angle. Since the single-scattering inelastic events in this κ region arise almost completely from incoherent scattering, the shape is assumed to be mostly due to the frequency spectrum of the velocity auto-correlation function. It is expected however that there is a large contamination of multiple scattering. The amount of multiple scattering is of the same order or somewhat smaller than that involved in the elastic case. However in the inelastic region the relative amount is much larger since the inelastic cross section is very small compared to the elastic peak ($\sim 1/50$). Since we do not have an adequate way of calculating the multiple inelastic scattering contributions for both coherent and incoherent events, no quantitative analysis of the inelastic scattering was attempted. However some qualitative information can be obtained. The fact that a Debye-Waller factor can be obtained over a reasonable κ range together with the fit of the widths to a solid-like model shows that the assumption of solid-like behavior just above the melting point is a reasonable one. In this spirit the inelastic data has been analyzed in the one-phonon incoherent cubic approximation, i.e.:

$$\frac{d^2\sigma}{d\Omega d\epsilon} = \frac{\sigma_{inc}}{4\pi} \frac{\kappa}{\kappa_0} e^{-a\kappa^2} \frac{\hbar^2 \kappa^2}{4M} \frac{e^{-\frac{\epsilon}{2k_B T}} G(\epsilon)}{\epsilon [\sinh \frac{\epsilon}{2k_B T}]}$$

where $\epsilon = E - E_0$, and $G(\epsilon)$ is the "one-phonon" spectrum. The Debye-Waller coefficient used is $a = 0.22 \text{ \AA}^2$, the value obtained from the elastic peak analysis. Figure 10 shows a typical $G(\epsilon)$ for a scattering angle of 14.30° , together with error flags due to counting statistics. Positive values of the "phonon energy" indicate neutron energy loss, and the ordinate is in units of eV^{-1} . The spectrum has a single maximum for phonon energies in the region of ± 14 to 15 meV , and for energy transfers less than this has a shape similar to a Debye spectrum but rises faster. A value of 14 meV is a reasonable cutoff value for the frequency spectrum. The longitudinal phonon branch in solid sodium in the $[111]$ direction has a maximum at 14 meV indicating a singularity in the frequency spectrum at this value. Above 14 meV , the spectrum falls off rather slowly. Calculations of the two phonon scattering with a Debye spectrum shows that incoherent multiphonon contributions are very small in this κ range. In Figure 3 the wavelength plots show the calculated one plus two phonon contributions. Thus the extra contributions to the one-phonon spectrum of Figure 10 above 14 meV are presumably due largely to multiple inelastic scattering. Figure 10 also shows the Debye spectrum with an assumed cutoff at 14 meV for both energy gain and loss events. The shapes below 14 meV are in reasonable agreement.

IV. CONCLUSIONS

A slow neutron inelastic scattering experiment on liquid sodium in the momentum transfer range where the scattering is predominantly

Figure 9 - Widths of the quasi-elastic peaks for liquid sodium.

Figure 10 - Typical frequency spectrum of the velocity auto correlation function analyzed in the one phonon incoherent solid assumption. The error flags are due to counting statistics.

incoherent has been performed. Multiple scattering effects are relatively small in the quasi-elastic peaks, and large in the inelastic spectrum. The scattering law has been extended to low momentum transfers and agrees well with earlier data, except where resolution effects are important. From the quasi-elastic peaks a value of the Debye-Waller coefficient has been obtained and the quasi-elastic widths analyzed. A qualitative analysis of the inelastic spectrum shows a shape characteristic of a simple Debye type frequency spectrum for the velocity auto correlation function. The results are consistent with a solid-like model of jump diffusion, having a Debye temperature of 103°K, and a mean vibrational time of residence $\tau_0 = 1.5 \times 10^{-12}$ sec.

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