

bubble is stable with $a_2(t)$ bounded by its initial value. Finally, at a critical value $R(0)/R_C \cong 1.063$, the Taylor instability due to the \dot{R} term sets in, as ω_2 approaches the subharmonic value $\omega_0/2$. Figure 3, curve *d* displays a beat frequency between ω_2 and the difference frequency $\omega_0 - \omega_2$ as both approach $\omega_0/2$. Above the critical value (Fig. 3, curve *e*) the bubble becomes unstable, with a_2 increasing without bound. This critical amplitude is very likely only a lower bound for the stability region, because stabilizing effects such as viscosity and terms nonlinear in a_2 have been neglected in this calculation.¹²

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Structural Rearrangements in Low-Temperature Heavy Water

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The isochoric temperature derivative of the neutron liquid structure factor is shown to be more sensitive to the variation of O-D and D-D distances than the measurable structure factor itself. Careful determinations of this quantity have been made, with measurements on either side of the density maximum at 11.2°C. These results are compared to the computer data of Impey, Klein, and McDonald; significant discrepancies are found which highlight the differences between the computer model and real water.

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In recent years there have been a number of attempts to predict the structure of liquid water, as contained in x ray and neutron diffraction patterns, from the results of computer molecular dynamics (CMD) simulations based on a (rigid) molecular pair potential. To a first approximation these attempts have been successful¹⁻³ and

consequently a more precise comparison of experimental data with the simulated results would be useful. In particular we have measured the isochoric temperature derivative (ITD) of the neutron structure factor [$S(q)$] and compared it to the predicted data. In the case of the CMD data we shall show that this function is more sen-

sitive to the OH bond distance, for example, than the structure factor itself. Because of the higher sensitivity significant discrepancies between measured and predicted data are revealed.

Lie, Clementi, and Yoshimine² and Impey, Klein, and McDonald³ have made CMD calculations using the Matsuoka-Clementi-Yoshimine potential,⁴ for several states of water at a density of $18 \text{ cm}^3 \text{ mol}^{-1}$. This density corresponds to the maximum density of heavy water, which occurs at 11.2°C . The three partial correlation functions (O-O, O-D, and D-D) were obtained from which the x-ray and neutron diffraction patterns may be predicted. (Throughout this Letter we follow the rigid-molecule approximation of the CMD work but will employ a flexible-molecule approximation in a subsequent paper.)

We have compared our calculations of the x-ray and neutron structure factors (using the r -space functions from Ref. 2) to the published results of Narten and Levy⁵ and to our neutron data.⁶ The peaks in the x-ray data, which depend mainly on the O-O correlations, are shifted relative to the experiment. Also the principal neutron peak, which depends mainly on the O-D correlations, is shifted slightly further. The significance of these discrepancies has not been understood.¹⁻³

In Fig. 1(a) we show in r space the observable neutron pair correlation function deduced from the CMD data of IKM.³ This function is given in

terms of the partial functions as

$$g_N(r) = 0.092g_{\text{OO}}(r) + 0.486g_{\text{DD}}(r) + 0.422g_{\text{OD}}(r) \dots \quad (1)$$

We have Fourier transformed $g_N(r)$, then truncated it at $q = 6.7 \text{ \AA}^{-1}$ and transformed back into r space. We chose this point since at about 6 \AA^{-1} the intramolecular scattering dominates the intermolecular scattering, and so experimental data are effectively truncated there even though measurements extend to higher q . The comparison between the original r -space data and the truncated and back-transformed data is given at Fig. 1(a). The loss of resolution is such that the O-D peak at 1.92 \AA is smoothed out, and the predicted function has the same appearance as the results of Gibson and Dore⁷: We conclude that experimentally it would be extremely difficult to observe the O-D peak this way. In Fig. 1(b) we show a similar comparison for the ITD $\Delta g/\Delta T$. For this quantity the smoothing does not eliminate the combined O-D and D-D peaks at 2 \AA , and so we might expect to see an experimental peak in the ITD. The reason is that this peak is more temperature sensitive than other parts of $g(r)$ and so stands out more prominently in $\Delta g/\Delta T$ than in the original data [for example, compare the behavior near 2 \AA in the $g(r)$ of Ref. 7 with our data in Fig. 2(b)].

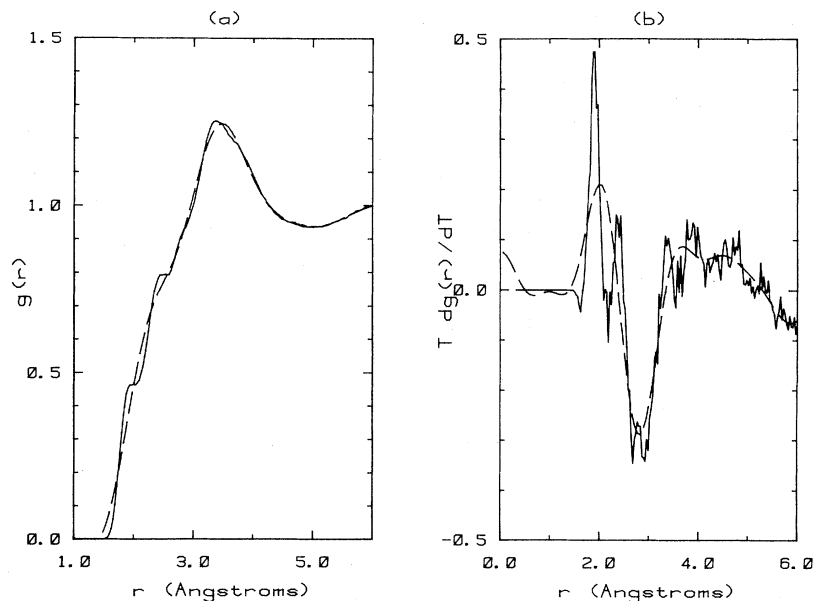


FIG. 1. A comparison of original (Ref. 3) data, full line, and q -space truncated data (after truncation at 6.7 \AA^{-1} and back transformation), dashed line. (a) $g_N(r)$; with use of an average of 9° and 49° data. (b) $T\Delta g_N/\Delta T$; with use of 9° minus 49° data.

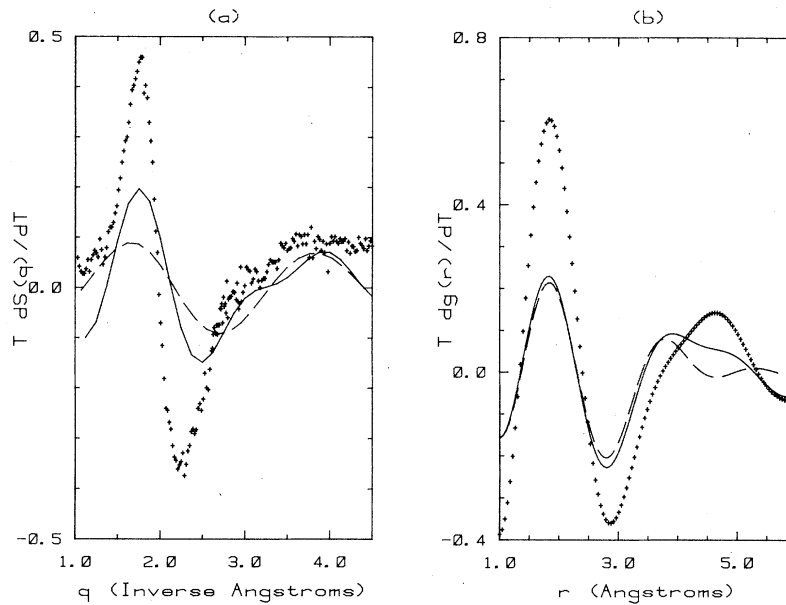


FIG. 2. A comparison of experimental and predicted values of the isochoric temperature derivative: Experimental 1.6° data minus 23.3°C data, crosses; predicted results with use of data from Ref. 3, by using 9° and 49° data, full line; and with use of -31° and 9° data, dashed line. (a) $T\Delta S(q)/\Delta T$; (b) $T\Delta g_N/\Delta T$; the predicted results in (a) were truncated at 4.5 \AA^{-1} in q space and transformed back to r space.

In the case of water, it is possible, without applying pressure, to obtain an ITD since the density as a function of temperature passes through a maximum at 11.2°C . Measurements taken on either side of this value may be chosen to have the same density but different temperatures. Data taken at 1 atm and other temperatures should be interpreted according to the equation

$$\left. \frac{\partial S(q)}{\partial T} \right|_P = \left. \frac{\partial S(q)}{\partial T} \right|_\rho - \alpha \rho \left. \frac{\partial S(q)}{\partial \rho} \right|_T \dots, \quad (2)$$

where α is the volume coefficient of thermal expansion. Walford and Dore⁸ have published measurements of $S(q)$ as a function of temperature, but they did not extend their data below 11.2° and their data did not have sufficient precision to give reliable isochoric temperature derivatives. Nevertheless they pointed out the many advantages of temperature-difference experiments and took temperature differences from a reference state (23°C). They were able to show that their results were roughly independent of temperature, which indicates that the second term on the right-hand side of Eq. (2) is less than their errors. But its effect is significant when higher precision is used. In our experiment the second term is eliminated.

Measurements were made on a high-purity heavy-water sample with use of the GWELFNEUD

II diffractometer at the NRU reaction Chalk River and a wavelength of 2.395 \AA . Data were taken at steps of $\Delta q \sim 0.02 \text{ \AA}^{-1}$ and $\sim 5 \times 10^5$ counts were recorded for each point. The temperature was varied from 23.5° to -1.7°C in steps of $\Delta T \sim 4^\circ\text{C}$. When these data are subtracted the usual corrections to such results cancel, apart from the temperature-dependent ones (e.g., the container background which is removed before subtraction). The inelasticity correction⁹ for free water molecules is negligible except at low angles, and we assume that this conclusion will hold in the real case.⁶

In Fig. 2(a) we plot the experimental data for $T\Delta S/\Delta T$ obtained from the 23.2° and 1.6°C data, together with the results we obtained from the IKM³ correlation functions. The latter results were truncated at the limit in this experiment ($q = 4.5 \text{ \AA}^{-1}$) and then back transformed; these data are shown in Fig. 2(b) in comparison to the transform of the experimental data. There is a marked difference in magnitude of the oscillations in both q space and r space, especially near the important O-D and D-D peaks at 2 \AA . It is clear that the temperature dependence of the intermolecular distances has not been predicted correctly by the Matsuoka-Clementi-Yoshimine potential simulations. These discrepancies are due to a bodily shift—larger than predicted—of the major

peak in the diffraction pattern to higher q as the temperature is raised at constant density. The interpretation of this effect in r space [see Fig. (2)] is a combination of small- and large- r changes in g_{OD} and g_{DD} which will be discussed in a later publication.

To summarize, we have proposed and experimentally demonstrated a sensitive method of studying the temperature variation of O-D and D-D distances, which uses the ITD. This quantity allows us to detect structural rearrangements as the temperature is altered, free from effects due to overall volume changes. It also avoids the difficult experimental problems of the inelasticity and multiple-scattering corrections. In addition a systematic use of CMD data allowed us to assess the loss of resolution in r -space data due to a necessary q -space truncation of $S(q)$, and to demonstrate that the ITD technique can partly overcome this disadvantage. Finally the ITD itself is a good test of the assumptions underlying the model used in the CMD calculations. A more detailed account of the experiment and its interpretation will be published elsewhere.

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Amplitude- and Temperature-Dependent Pinning and Unpinning of Dislocations in bcc and hcp ^3He

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Measurements of the temperature dependence of the longitudinal sound velocity and attenuation in solid ^3He show a frequency-dependent anomaly due to interactions between the ultrasonic waves and dislocations. By application of high-amplitude sound pulses at temperatures below about 0.4 K, the attenuation can be greatly reduced and the velocity changed. Upon warming, the velocity and attenuation return to their initial values by a thermally activated process whose activation energy appears to be that of mobile vacancies.

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For dielectric crystals, the adiabatic sound velocity is expected from energy considerations to have a temperature dependence of the form

$$v_a(T) = v_0 + aT^4 + bT^6 + \dots \quad (1)$$

Ultrasonic experiments in the megahertz re-

gion^{1,2} on pure hcp ^4He ($\leq 1 \text{ ppm } ^3\text{He}$) have shown that Eq. (1) describes the sound velocity only from the melting point to about 1 or 1.5 K. At lower temperatures, frequency-dependent deviations from Eq. (1) occur, accompanied by a large increase in sound attenuation with falling tempera-