

Structure of Liquid Nitrogen, Oxygen, and Argon by Neutron Diffraction

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(Received August 26, 1953)

The structures of liquid oxygen, nitrogen, and argon have been investigated by measuring the scattering of neutrons of wavelength 1.08Å over the angular range 4° to 78° . The scattering curves have been transformed to give the time average of the atomic density as a function of the distance from an atom taken as center. The deviation from the mean density is an oscillating function with an amplitude which decreases more rapidly than the inverse square of the distance from the central atom. In nitrogen a single neighbor is found at a spacing of 1.1Å from each atom, corresponding to a diatomic molecule in the liquid. In oxygen 1.5 neighbors are found at approximately 1.25Å from each atom, suggesting that the degree of association is greater than diatomic. The results are not inconsistent with the suggested existence of the O_4 molecule. In argon the results are in reasonable agreement with published x-ray determinations.

INTRODUCTION

AS part of a program to investigate the scattering of slow neutrons by liquids, the angular distributions of neutrons scattered by liquid nitrogen, liquid oxygen, and liquid argon have been measured. Information about the atomic arrangements of the liquids can be obtained from such distributions. In particular, if a semiclassical model of neutron scattering is assumed¹⁻³ a transformation, well known in x-ray investigations, may be used to obtain from the measurements the radial variation of density about an atom in the liquid.^{4,5} Gingrich⁶ has reviewed x-ray investigations of liquid elements including the three discussed here. Previous measurements of the scattering of neutrons by liquids have been made by Chamberlain⁷ who investigated sulfur, lead, and bismuth, and by Sharrah and Smith⁸ who investigated lead and bismuth. In the interpretation of their measurements these workers assumed that the scattering pattern was the sum of a coherent diffraction pattern, to which the transformation is applicable, and an isotropic incoherent contribution.

The experimental results reported in this paper have been similarly transformed to give the radial density for each of the three liquids. The effects of variations in the treatment of the experimental results on the predicted structure are discussed.

APPARATUS

The angular distribution of scattered neutrons was measured by an automatic spectrometer with the monochromating crystal adjusted to provide neutrons of wavelength 1.08Å. Since the earlier description of this instrument⁹ some minor modifications have been made;

the monoenergetic neutrons were selected by diffraction at a sodium chloride crystal in reflection rather than transmission, the motor drive provided a wider choice of angular steps, and the neutron monitor was improved. The new monitor was a "thin" BF_3 counter interposed in the diffracted beam; this eliminated errors due to changes in the spectrum of neutrons leaving the reactor.

The liquids were held in a cryostat mounted on the specimen table of the spectrometer. A horizontal section, at the level of the neutron beam, is shown in Fig. 1. The cryostat was designed to hold liquid helium¹⁰ which has low scattering power and thus requires a large scattering chamber (12.7-cm diameter with 0.025-cm wall thickness). This chamber was also suitable for argon which has a small scattering cross section. For nitrogen and oxygen, which have larger cross sections, a small cylindrical aluminum cassette (1-cm diameter with 0.01-cm wall thickness) was mounted in the middle of the cryostat.

The scattered neutrons were detected by an enriched BF_3 proportional counter mounted on an arm rotatable about the axis of the cryostat. With nitrogen and oxygen the time to count 3200 neutrons was recorded for each of 44 equally-spaced positions of the arm in the angular

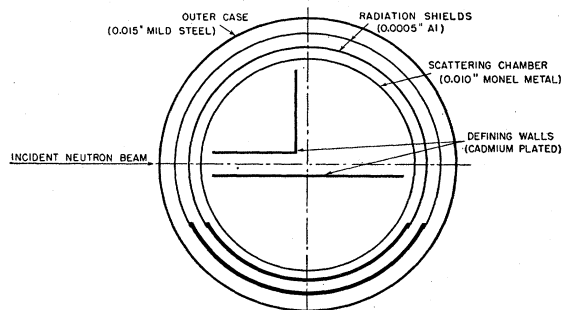


FIG. 1. A horizontal section of the argon scattering chamber at the level of the neutron beam. The cadmium-coated plates define the scattering volume and reduce the error caused by multiply-scattered neutrons. The radiation shields and liquid nitrogen baths to which the shields were connected formed an enclosure at liquid nitrogen temperature for the scattering chamber.

¹⁰ D. G. Henshaw and D. G. Hurst, *Phys. Rev.* **91**, 1222 (1953).

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¹ G. Placzek, *Phys. Rev.* **86**, 377 (1952).

² L. Goldstein, *Phys. Rev.* **84**, 466 (1951).

³ N. Z. Alcock and D. G. Hurst, *Phys. Rev.* **83**, 1100 (1951);

N. K. Pope, *Can. J. Phys.* **30**, 597 (1952).

⁴ P. Debye, *Ann. Physik* **46**, 809 (1915).

⁵ F. Zernike and J. Prins, *Z. Physik* **41**, 184 (1927).

⁶ N. S. Gingrich, *Revs. Modern Phys.* **15**, 90 (1943).

⁷ O. Chamberlain, *Phys. Rev.* **77**, 305 (1950).

⁸ P. C. Sharrah and G. P. Smith, *J. Chem. Phys.* **21**, 228 (1953).

⁹ D. G. Hurst and N. Z. Alcock, *Can. J. Phys.* **29**, 36 (1951).

range 4° to 77°. With argon the time to count 800 neutrons was recorded. The temperature of each sample was controlled and recorded by means of its vapor pressure using a commercial pressure controller.

EXPERIMENTAL RESULTS

The scattered intensities, normalized by means of the monitor to the same number of neutrons incident on the specimen, were corrected for background, resolution, double scattering, and in the case of argon, for the change in effective scattering volume with angle. The

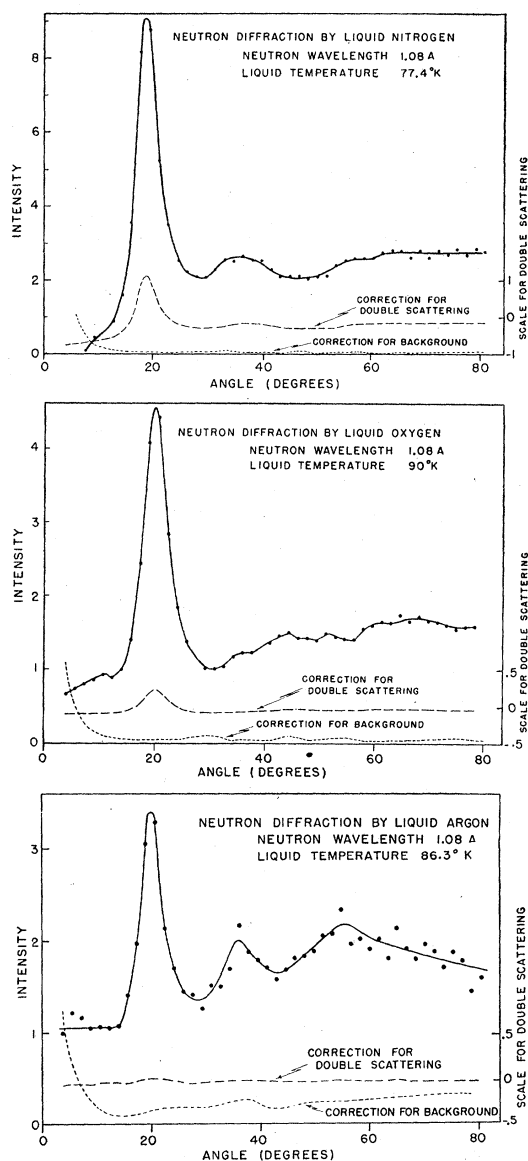


FIG. 2. The corrected intensities (in arbitrary units) for liquid oxygen at 90°K, nitrogen at 77.4°K and argon at 86.3°K as a function of the scattering angle. Also shown are the corrections applied for background and double scattering, while the correction for counter resolution, being small, has been omitted. A neutron wavelength of 1.08Å was used.

background was measured with the specimen holder empty. At small angles, where the background was large, it was assumed that the background neutrons in excess of those at large angles came predominantly from the region on the source side of the scattering volume. Accordingly, this excess was reduced by the transmission loss through the liquid before subtracting from the measurements. For nitrogen and oxygen the background correction at large angles was about 5 percent of the intensity and somewhat greater in the case of argon. The resolution of the instrument was determined by measuring the (200) line of powdered potassium bromide contained in the holder used for nitrogen and oxygen. This (200) line occurs at nearly the same angle as the main peak in nitrogen. The full width at half-height was found to be 1.67 degrees, and was applied as a correction to the measured pattern by a method due to Eckart.¹¹

Corrections for double scattering were made to the results for oxygen and nitrogen with the aid of the theorem that the average path length for particles originating with uniform source strength in a long cylinder of radius a is $(4/3)a$. For argon this same theorem was used for second scattering close to the position of first scattering; the contribution due to second scattering remote from the position of first scattering was calculated by numerical integration over the relevant portions of the scattering chamber. It was assumed, and this assumption was supported by calculations for special cases, that the corrections for multiple scattering could be calculated as if the scattering were isotropic. The attenuation of the neutron beam in its passage through the liquid was taken into account.

The effective scattering volume of argon depended upon the angle of scattering. The scattered intensities were normalized to unit scattering volume neglecting the angular divergences of the incident neutron beam and of the counter acceptance. The corrected results together with the corrections applied for background, counter resolution, and double scattering are plotted in Fig. 2 for nitrogen, oxygen, and argon, respectively.

INTERPRETATION

The formula of Debye⁴ and Zernike and Prins⁵ connects the coherently scattered intensity with the variation of atomic density measured radially from an atom in the liquid. When applied to neutron scattering without spin and isotope incoherence, it can be written as

$$4\pi r^2[\rho(r) - \rho_0] = (2r/\pi) \int_0^\infty si(s) \sin rs ds, \quad (1)$$

where r = distance from the atom chosen as center, $\rho(r)$ = atomic density at a distance r , ρ_0 = mean atomic density of the liquid, $s = (4\pi/\lambda) \sin(\varphi/2)$, λ = neutron wavelength, φ = angle of scattering, $(is) = [I(s) - I_\infty]/I_\infty$,

¹¹ C. Eckart, Phys. Rev. 51, 735 (1937).

$I(s)$ = intensity of coherent scattering (proportional to differential cross section) for the value s of the variable, and I_∞ = value of $I(s)$ for $s = \infty$ (see, however, later discussions).

In the derivation of Eq. (1) the quantity $I(s)$ is assumed to arise from coherent scattering proportional to the density of scattering material. Neutron scattering is considerably more complex than this simple process and it is necessary to inquire into the validity of the formula. The derivation treats the coherent scattering from the liquid as elastic. A rough estimate of the elastic neutron scattering may be obtained by comparing the liquid with an Einstein solid having an appropriate characteristic temperature. The elastic scattering from such a solid¹² is very small beyond the first peak in the angular distribution from the liquid. In practice it is impossible to separate the elastic from the total scattering, and it would not be feasible to restrict $I(s)$ to the elastic component. However, provided the energy transfers between liquid and neutron are small compared to the neutron energy, these transfers may be ignored, and the use of all or nearly all the scattering for $I(s)$ in Eq. (1) may lead to the correct result.¹⁻³

Spin and isotope incoherence, which often occur in neutron scattering, are known to be small for the three elements considered in the present paper.

Previous workers^{7,8} assumed that the incoherent scattering was isotropic and given by the small-angle scattering. They subtracted from the measured intensity the intensity observed at small angles and assumed that the resultant intensity was the coherent portion to which Eq. (1) applies.

In the present work nitrogen gave very little small-angle scattering but oxygen and argon gave relatively much greater amounts which were larger than those reported for lead, bismuth, and sulfur.^{7,8} In view of possible magnetic scattering by oxygen, of the possible existence of other small-angle scattering, and of the large experimental errors at small angle owing to background, it is not certain that the residual scattering should be subtracted. For most of the analysis of the present results $I(s)$ has been taken to be the scattering plotted in Fig. 2. The effect caused by subtracting a constant from the values of $I(s)$ has been studied.

The largest value of s for which measurements were made is 7.4\AA^{-1} . Unless some form for $i(s)$ is assumed in the range $7.4 < s < \infty$ the integration of Eq. (1) can be carried out only to this value of s . The extrapolation of $i(s)$ beyond the experimental limit of s is arbitrary except that $i(s)$ is almost certainly small. In x-ray and electron diffraction $i(s)$ is often assumed to be zero throughout this region,⁶ or to approximate to this condition an artificial temperature factor is sometimes introduced.¹³ The lack of direct knowledge of $I(s)$ as $s \rightarrow \infty$ permits the use of I_∞ as a parameter. For example I_∞ may be chosen to ensure⁷ that $\rho(r) \rightarrow 0$ as $r \rightarrow 0$. This

¹² B. N. Brockhouse and D. G. Hurst, Phys. Rev. 88, 542 (1952).

¹³ C. Degard, Bull. soc. roy. sci. Liège 7, 115 (1938).

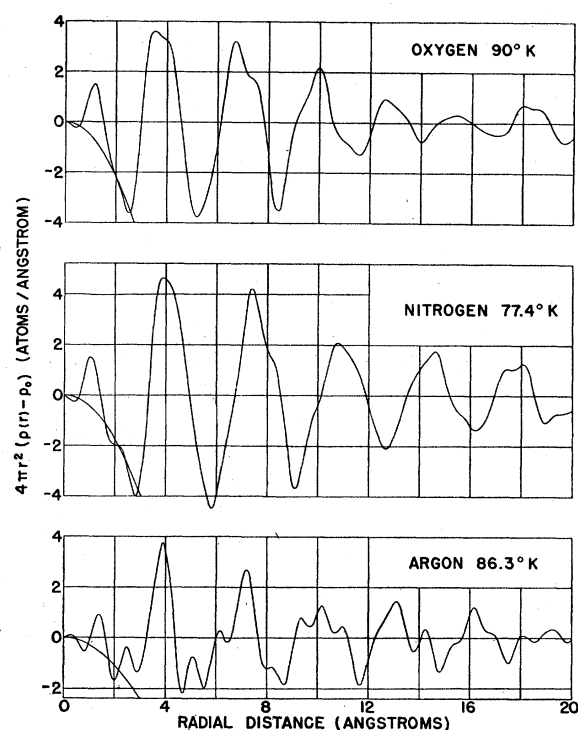


FIG. 3. The transforms $4\pi r^2[\rho(r) - \rho_0]$ for liquid oxygen, nitrogen, and argon calculated from the corresponding intensity curves of Fig. 2. The smooth curve with each transform is $-4\pi r^2 \rho_0$, where ρ_0 is the average density. ρ_0 for oxygen = 4.30×10^{-2} ; for nitrogen = 3.48×10^{-2} and for argon = 2.13×10^{-2} atoms per cubic angstrom.

condition requires that

$$-2\pi^2 \rho_0 = \int_0^\infty s^2 i(s) ds, \quad (2)$$

where again the upper limit of integration is experimentally unattainable. Extrapolation of $i(s)$ and the value of I_∞ are of course interrelated.

In the present work, transforms were computed on an I.B.M. Calculating Punch No. 602A at the Chalk River Laboratory and on the Ferranti Electronic Computer (FERUT) at the University of Toronto. Simpson's rule was used for the integrations. Calculations were carried out for several methods of terminating the integration and computing I_∞ . In the majority of cases I_∞ was calculated by Eq. (2). An extrapolation $i(s) \propto s^{-4}$ was tried. The effects of these variations were small, being mainly to add ripples with a wavelength $[2\pi/\text{cut-off value of } s]$ along the r axis; in the case of a sharp density peak these appear as ghosts on either side.^{14,15a,15b}

The transform $4\pi r^2[\rho(r) - \rho_0]$ given by Eq. (1) indicates the degree of order in the liquid in that it gives

¹⁴ C. Finbak, Acta Chem. Scand. 3, 1279 and 1293 (1949).

^{15a} T. Sugawara, Sci. Repts. Research Insts. Tohoku Univ. A3, 39 (1951).

^{15b} (added in proof) J. Waser and V. Schomacher, Revs. Modern Phys. 25, 671 (1953).

the deviation from the mean of the number of atoms per unit radial distance. This transform is plotted for the three liquids in Fig. 3. The values for $0 < r < 3\text{\AA}$ merit special attention for it is in this region that information regarding molecular association is obtained. The curves of Fig. 3 show that the transforms for nitrogen and oxygen have peaks at about 1.0\AA and 1.2\AA, respectively, while in argon there is a peak at 1.4\AA. For nitrogen and oxygen the transforms do not go appreciably below $-4\pi r^2 \rho_0$ and are equal to it for r in the range $1.8 < r < 2.3\text{\AA}$. In the case of argon, however, there are regions where the transform goes considerably below $-4\pi r^2 \rho_0$ and in fact seems to oscillate about it. Where the transform is less than $-4\pi r^2 \rho_0$, negative values of the density are implied. These physically unreal values are probably due to experimental inadequacies. The wide scatter of the experimental points for argon at large values of φ (Fig. 2) permits considerable latitude in the choice of $i(s)$. If a function $i(s)$ is assumed which will yield $\rho(r) = 0$ out to $r = 3\text{\AA}$, deviations from this function for $s > 6.3\text{\AA}^{-1}$, consistent with experimental error, will produce a peak similar to the peak at 1.4\AA. This peak is probably spurious and the present results cannot be considered as inconsistent with the monatomic nature of liquid argon. It is in-

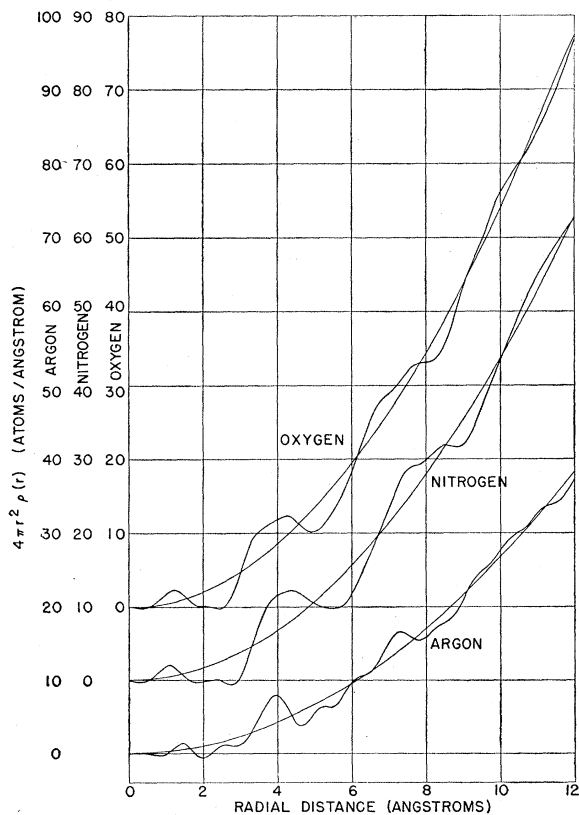


FIG. 4. The radial distribution functions $4\pi r^2 \rho(r)$ deduced from the transforms of Fig. 3 for liquid oxygen, nitrogen and argon. In each case $4\pi r^2 \rho_0$, the corresponding function for uniform atomic density, is also shown.

tended at a later date to repeat the measurements with greater accuracy.

The effect of varying the treatment of the experimental results is well illustrated by a comparison of three transforms for argon calculated (a) with the values of $I(s)$ plotted in Fig. 2 and I_∞ chosen to satisfy Eq. (2); (b) I_∞ chosen equal to $I(7.4)$ and (c) with the same values of $I(s)$ reduced by a constant amount equal to $0.62 I(7.4)$, i.e., by an amount equal to the value of the small angle scattering, and I_∞ satisfying Eq. (2).

The integrations were terminated at $s = 7.4\text{\AA}^{-1}$, equivalent to the assumption $i(s) = 0$ for $s > 7.4\text{\AA}^{-1}$.

In Case (b) the amplitude of the short wavelength ripples was increased. The curve obtained in (c) was very nearly the same form as in (a) but of approximately twice the magnitude. Of the three curves, (a) gave the smallest excursions into the region of negative density and is therefore more physically realistic. Similar behavior was found for oxygen: the subtraction of a constant as in (c) produced negative densities in the region $1.8\text{\AA} < r < 2.3\text{\AA}$ where previously the density was nearly zero. It is felt that these results support the decision to use the total scattered intensity in Eq. (1).

The radial distribution function $4\pi r^2 \rho(r)$ is such that $4\pi r^2 \rho(r) dr$ gives the number of atoms between r and $r + dr$ and is plotted in Fig. 4 for each of the liquids. The radial distribution function is greater than $4\pi r^2 \rho_0$ at regions where the density is greater than average. The existence of oscillations in this function indicates a tendency for the atoms to arrange themselves in groups of neighbors. Except for the first neighbors in nitrogen and oxygen, where the density is zero on either side of the peak, the groups are not clearly separated. As r increases the distinct character of the groups is rapidly lost, as can be seen by comparing the magnitude of the oscillations in Fig. 3 with the value of $4\pi r^2 \rho_0$. The finite resolution of the experiment contributes to the lack of distinctness.

For small values of r , it is usual to divide atoms into groups associated with the peaks. Some freedom exists in the division of atoms between the peaks. The method used in the present investigation is to write

$$T_n = 2(N - T_{n-1} - T_{n-2} \cdots T_1), \quad (3)$$

where T_n = number of atoms assigned to n th peak, and N = total number of atoms out to the position of the n th peak. N was obtained by integration of the radial distribution function.

Equation (3) insures that the atoms assigned to a peak are evenly divided between the two sides of the peak.

In Table I the positions and the number of atoms per peak have been listed for each of the three liquids. From the number of atoms under the first peak in nitrogen and oxygen the average number of atoms per molecule has been deduced, and this has been used to calculate the number of molecules under each peak.

TABLE I. Positions and magnitudes of atomic concentrations.

Liquid nitrogen 77.4°K			Liquid oxygen 90°K			Liquid argon 86.3°K	
Radius A	Number of atoms	Number of molecules (2 atoms each)	Radius A	Number of atoms	Number of molecular groupings (2.5 atoms each)	Radius A	Number of atoms
1.1	1.0		1.25	1.50		1.45	0.5 ^a
4.4	23.6	11.8	4.2	26.0	10.4	2.6	0.6 ^a
						3.9	8.2
						5.2	6.4 ^a
						6.2	11.0 ^a
						7.3	18.6

^a As discussed in the text, these probably result from the limitation on the experimental range of s and experimental error near this limit.

A study of Table I shows that nitrogen has 1.0 atom centered around a maximum at 1.1A.¹⁶ The existence of this 1 atom supports earlier evidence that nitrogen is diatomic in the liquid state. The second peak occurs at 4.4A and the number of neighbors at this spacing is 23.6 atoms or 11.8 diatomic molecules. Within experimental error this is equal to 12 molecules corresponding to a close packed structure. The spacing of 1.1A is in agreement with the value of 1.09A of the diatomic molecule. X-ray measurements on the liquid give 1.3A. In the x-ray analysis, in addition to the peak at 1.3A, peaks were found at 2.6A, 4.0A and 4.8A. It would appear that the peaks at 4.0A and 4.8A in the x-ray analysis have combined to give a peak at 4.4A in the neutron analysis. In the neutron analysis no peak is found at 2.6A, possibly owing to improved information at the larger values of s .

The results for liquid oxygen show 1.5 atoms centered around $r=1.25A$. The existence of more than one neighbor at $r=1.25A$ indicates that oxygen is not solely diatomic in the liquid state. The existence of O_4 has been suggested from the results of both spectroscopic and magnetic measurements on mixtures of liquid oxygen and nitrogen.¹⁷ From the magnetic susceptibilities of such mixtures, Lewis¹⁸ calculated that liquid oxygen at 77.4°K should contain 40 percent by weight of O_4 . Assuming that the molecule O_4 is tetrahedral with a spacing between atoms equal to that of the O_2 molecule, a simple calculation on the basis of 1.5 nearest neighbors would indicate the existence of 25 percent by weight of O_4 at 90°K. The agreement between the value of 40 percent by weight as determined from the magnetic measurements for oxygen at 77.4°K and the value of 25 percent by weight determined here for oxygen at 90°K is not unreasonable considering the experimental error which might be expected. The peak found at 4.2A has an area which corresponds to the existence of 26 atoms. On the basis of the 1.5 molecular neighbors, the 26 atoms would correspond to 10.4 molecular

¹⁶ The positions of the peaks will now be given at the maxima in the function $4\pi r^2\rho(r)$. The values quoted earlier are the positions of the peaks on the function $4\pi r^2[\rho(r) - \rho_0]$.

¹⁷ M. and B. Reuhmann, *Low Temperature Physics* (Cambridge University Press, London, 1937), p. 228.

¹⁸ G. N. Lewis, *J. Am. Chem. Soc.* **46**, 2027 (1924).

neighbors, which is not significantly different from the 12 neighbors required for close packing. X-ray measurements⁶ have given peaks at 1.3A, 2.2A, 3.4A, and 4.2A. The appearance in the x-ray analysis of 1.08 atoms at the 1.3A spacing together with the peak at 2.2A has been interpreted as arising from the existence of molecules of O_3 . The positions of the peaks at 1.3A and 4.2A correspond well with the neutron determination, but in the neutron determination no peak was found to correspond with the x-ray peak at 2.2A. Sugawara¹⁵ has concluded that this peak is spurious. The 3.4A peak appears to have become part of the broad peak at 4.2A.

The results for liquid argon are more difficult to interpret. An examination of the correlation function (Fig. 3) shows that the curves for liquid oxygen and nitrogen vary smoothly with r , while there appears to be high-frequency oscillations superimposed on the main pattern of argon. As has been pointed out earlier, the validity of the peak at 1.45A is in doubt. From Table I it may be seen that other peaks occur at 2.6A, 3.9A, 5.2A, 6.2A, and 7.3A. The main peaks have been italicized. There is a reasonable agreement between the present results and those obtained from the comprehensive work of Eisenstein and Gingrich with x-rays^{19,20} where peaks were found at 3.79A, 5.3A, and 7.3A. The number of atoms under the first main peak at 3.9A was 8.2, if the area under the subsidiary peaks is not included. This is considerably less than the 12 neighbors required for a close packed structure. In solid argon an atom has 12 nearest neighbors. The number of neighbors at the spacing of 3.8A in the liquid has been found by x-rays at three temperatures.^{19,20} The results have been plotted in Fig. 5, together with the value 8.2 from the neutron measurement. There appears to be reasonable agreement between the results of the x-ray

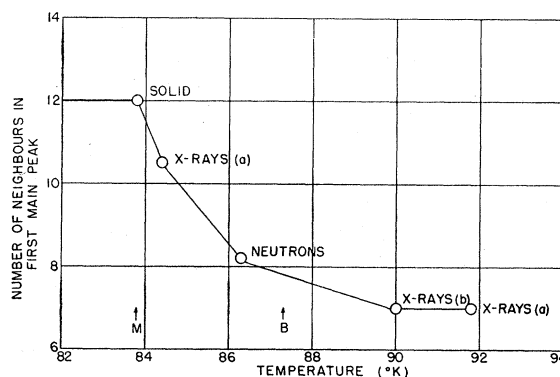


Fig. 5. The number of neighbors in the first main peak at 3.8A for liquid argon as a function of temperature. The x-ray results (a) and (b) were taken from the papers by Eisenstein and Gingrich (see references 19, 20) while the neutron value is from the present investigation. Point M represents the melting point (83.8°K and 512 mm Hg), while B represents the normal boiling point (87.5°K).

¹⁹ A. Eisenstein and N. S. Gingrich, *Phys. Rev.* **58**, 307 (1940).

²⁰ A. Eisenstein and N. S. Gingrich, *Phys. Rev.* **62**, 261 (1942).

determinations and the present neutron determination. An examination of the curve (Fig. 5) shows that the number of neighbors under the peak at 3.9A falls rapidly from 12 at 84°K to about 7 at 90°K, beyond which the curve would appear to fall more slowly. This means that the structure of the liquid changes rapidly for temperatures just above the melting point. Since the number of atoms per unit volume is approximately constant, a decrease in the number of atoms at a given spacing requires that these atoms appear elsewhere. It is intended to find the effect of temperature on the posi-

tions and heights of the peaks and to check the present results when experimental work can be resumed.

ACKNOWLEDGMENT

The authors wish to thank Dr. H. Gellman of the Theoretical Physics Branch for calculations of the Fourier transforms on the FERUT Computer and Miss B. Sears for assistance in similar calculations. Thanks are also due to Mr. W. McAlpin for the mechanical design of the cryostat, and to Mr. W. Woytowich for technical assistance.

Concentration of Uranium and Lead and the Isotopic Composition of Lead in Meteoritic Material

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(Received July 27, 1953)

The cosmic abundance of lead and uranium have been determined by studying the lead and uranium contents of meteoritic materials. Lead is found to be present to 8×10^{-3} atom/10 000 atoms of silicon, and uranium to 1×10^{-4} atom/10 000 atom of silicon. The new value for lead removes the hump in the cosmic abundance curve in the 206–208 mass region. The relative primordial abundances of lead isotopes of mass 204, 206, 207, and 208 are found to be 1:9.4:10.3:29.2, respectively.

THE concentrations of uranium in some stone meteorites and troilite from an iron meteorite were determined by the isotope dilution method.^{1,2} The concentrations of lead in a stone and iron meteorite and in troilite from an iron meteorite were determined colorimetrically, using Pb²¹⁰ for yield corrections.³ The lead samples so isolated were analyzed in a mass spectrometer, using the surface ionization method.² The effect of contamination upon the composition of the lead in the sample from the stone meteorite is as yet uncertain and, therefore, only the composition of the

troilite and the iron meteorite lead is reported here. The results are given in Table I.

Of the uranium concentrations in stone meteorites that have been previously reported,^{4–10} only the radon determinations of G. Davis agree with the values reported here. His values are given in Table II. It is believed that the higher values obtained by the early investigators resulted from a failure to exercise proper precautions in excluding moisture from electroscopes during radon measurements. Evans, Kip, and Moberg¹¹ recognized the critical nature of this phenomenon when they obtained anomalously high values for the radon content of sea water until the most stringent precautions were taken to dry the gases in the electroscop chamber. Other possible sources of error might be contamination

TABLE I. Uranium and lead in meteoritic material.

Sample	U concentration (ppm)	Pb concentration (ppm)	Pb composition (atomic ratios)		
			206/204	207/204	208/204
Canyon diablo troilite	0.009 ± 0.003	18 ± 1	9.41	10.27	29.16
Canyon diablo metal phase	—	(0.37 ± 0.05) ^a	(9.7) ^a	(10.5) ^a	(29.3) ^a
Modoc (total)	0.011 ± 0.002	0.9 ± 0.1	—	—	—
Norton county (total)	0.010 ± 0.003	—	—	—	—

^a Corrected for terrestrial lead contamination.

¹ G. Tilton, Atomic Energy Commission Report AECD-3182, 1951 (unpublished).

² G. Tilton *et al.*, Bull. Geol. Soc. Amer. (to be published).

³ C. Patterson, Atomic Energy Commission Report AECD-3180, 1951 (unpublished).

⁴ R. J. Strutt, Proc. Roy. Soc. (London) A77, 481 (1906).

⁵ T. Quirke and L. Finkelstein, Am. J. Sci. 44, 237 (1917).

⁶ G. Halledauer, Wien. Ber. 134 (IIa), 39 (1925).

⁷ A. Holmes, Bull. Natl. Research Council 81, 418 (1931).

⁸ F. Paneth and W. Koeck, Z. physik. Chem., Bodenstein-Festband, 145 (1931).

⁹ J. R. Horan, Oak Ridge Institute of Nuclear Studies, Oak Ridge, Tennessee reports a uranium concentration greater than 3 ppm in a carbonaceous chondrite from Murray, Callaway County, Kentucky (to be published in a new journal, Meteoritics). Mr. W. Nichiporuk of the California Institute of Technology has checked this meteorite by alpha counting and finds less than about 0.4 ppm of uranium plus thorium (the limit of the method).

¹⁰ G. L. Davis, Am. J. Sci. 243, 107 (1950).

¹¹ Evans, Kip, and Moberg, Am. J. Sci. 36, 241 (1938).