

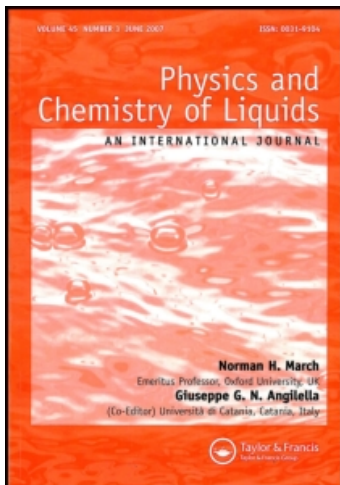
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Structure of Expanded Fluid Metals

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REVIEW

Structure of Expanded Fluid Metals

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A survey is given of neutron diffraction investigations of the static structure factor $S(Q)$ of liquid rubidium and cesium expanded by heating towards conditions close to their critical points. The data are used to derive the characteristic changes of the microscopic structure—such as the distance and number of nearest neighbours—as a function of density. After a brief discussion of recent measurements of the isothermal density derivative of $S(Q)$ of expanded liquid cesium, which is related to the triplet correlation function, we describe the theoretical attempts which have been undertaken so far to extract information from the structure data about the density dependent changes of the effective interaction potential as the metal-nonmetal transition is approached.

KEY WORDS: Liquid Metals, Neutron Scattering, High Temperature, High Pressure, Structure

1 INTRODUCTION

Over the last years, considerable effort has been put into investigating the structural, thermodynamic, electrical and magnetic properties of liquid alkali metals expanded by heating towards their liquid-vapour critical points¹⁻¹⁰. Much of this effort has focused on rubidium and cesium whose critical points occur at temperatures and pressures sufficiently low that the physical properties of these metals can be studied in the critical region with accurate static experimental methods (critical data of Cs: $T_c = 1924$ K, $P_c = 92.5$ bar, $d_c = 0.38$ gcm⁻³; of Rb: $T_c = 2017$ K, $P_c = 124.5$ bar, $d_c = 0.29$ gcm⁻³⁶). Experimental measurements such as those of the liquid-vapour coexistence curve⁶, the critical data⁶, the electrical conductivity¹, the static magnetic susceptibility³ and the Knight shift⁴ have indicated, that liquid rubidium and cesium undergo a metal-nonmetal transition near their liquid-vapour critical point.

The metal-nonmetal transition resembles that proposed by Mott in his pioneering studies of the metal-nonmetal transition in expanded monovalent crystals¹¹. Mott and Hubbard have shown that short range electron-electron interactions can lead to electron localization at low densities in systems with one electron per atom such as liquid alkali metals^{11,12}. Subsequent theoretical work¹³ showed that the electron gas on the metallic side of the metal-nonmetal transition should be highly correlated, having a low instantaneous fraction of doubly occupied states, which leads to an increase in the effective mass and, consequently, to an enhancement of the paramagnetic susceptibility. The latter prediction has been confirmed by static magnetic

susceptibility and nuclear magnetic resonance experiments on expanded liquid cesium^{3,4}.

The occurrence of the metal-nonmetal transition in expanded alkali metals implies that the interatomic forces must exhibit drastic changes as the density of the fluid is decreased, especially when the metal-nonmetal transition is approached. Useful information about these changes can be obtained from studies of a combination of equation of state data, electrical properties and structural data. For that reason, a series of neutron scattering experiments have been performed to determine the structure of liquid rubidium and cesium at conditions covering the whole liquid range from the melting point up to the critical point^{5,14}, where the metallic properties disappear. In addition the isothermal density dependence of the structure factor $S(Q)$ of expanded cesium has been measured over a wide range of temperatures, ranging from 340 K up to 1670 K³¹. Such measurements are of actual interest because recent theoretical work¹⁶⁻¹⁸ demonstrates that the isothermal density derivative of $S(Q)$ which is related to the three-body correlations¹⁵ will play an essential role in any effort to extract quantitative information on the effective pair potential from experimental diffraction data.

In the following we present recent experimental results together with theoretical attempts which have been undertaken so far to extract information from them about the state dependence of the effective interaction potential.

2 EXPERIMENTAL RESULTS AND DISCUSSION

As an example, Figure 1a shows a selection of measured structure factors $S(Q)$ of expanded liquid cesium for different temperatures and densities near the liquid-vapour coexistence curve, covering the whole liquid range from the melting point up to the critical point⁵. For $Q \leq 0.28 \text{ \AA}^{-1}$, the structure factors are extrapolated to $S(0) = nk_B T \chi_T$, with the number density n and isothermal compressibility χ_T taken from the most accurate PVT-data⁶. The neutron-diffraction experiments have been performed using the two axis diffractometer D4B at the high-flux reactor of the Institute Laue-Langevin in Grenoble. The high-pressure high-temperature autoclave necessary for performing these experiments and the data evaluation procedure for obtaining $S(Q)$ from the raw data are described in detail in Ref. 19.

Figure 1b displays the corresponding Fourier-transform of the $S(Q)$ -data, the pair correlation function $g(R)$. With decreasing density or increasing temperature the following changes in $S(Q)$ or $g(R)$ of the liquid metal are apparent:

The intensities of the first peaks of $S(Q)$ and $g(R)$ are strongly reduced and broadened, whereas the peak positions Q_1 and R_1 shift only slightly towards lower Q - or higher R - values, respectively. Compared with a simple nonmetallic liquid such as argon, the oscillations of $S(Q)$ for liquid cesium are drastically damped with increasing temperature. Only a broad first maximum is seen in $S(Q)$ of cesium near the critical point, whereas in the case of liquid argon three well defined maxima and minima are observable near the critical point²⁰. This difference in behaviour can be attributed to differences in the repulsive part of the effective interaction potential $\varnothing(R)$ for liquid

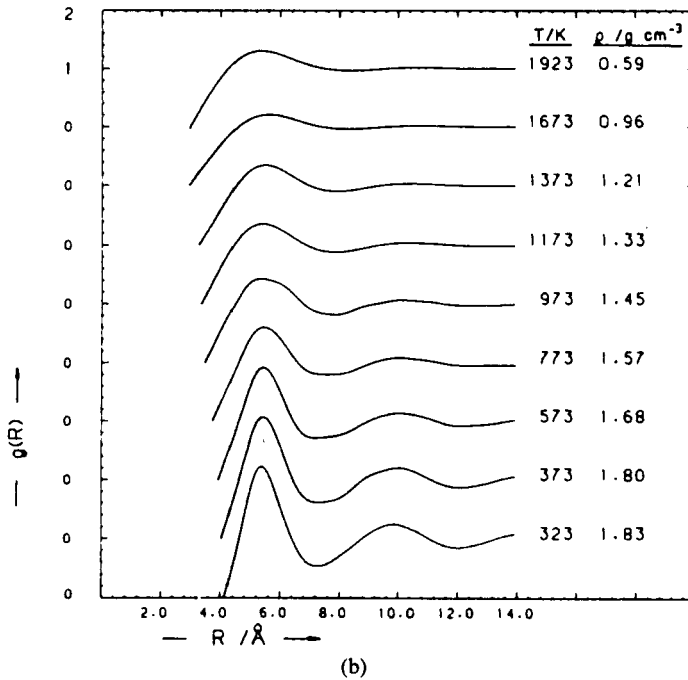
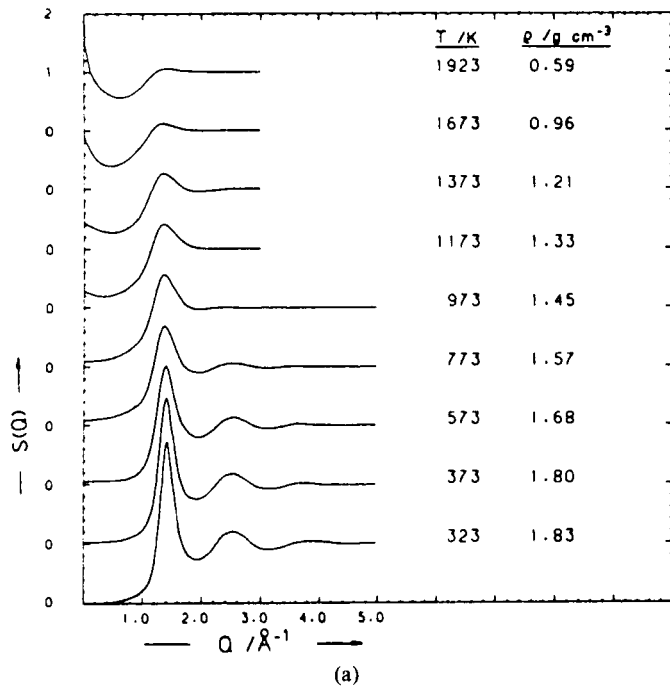


Figure 1 (a) The structure factor $S(Q)$ of expanded liquid cesium at conditions near the liquid-vapour coexistence curve. (b) The pair correlation function $g(R)$ of expanded liquid cesium.

alkali metals and argon²¹. The repulsive part is much softer in metals than in argon. This characteristic of $\varnothing(R)$ for the alkali metals is also qualitatively indicated by the change of $g(R)$ with increasing temperature (see Figure 1b). With increasing temperature, the first ascent of $g(R)$ becomes less steep and starts at a smaller R -value, i.e., a smaller hard-core radius. In the case of liquid argon, the position of the first peak and the steepness of the first ascent of $g(R)$ remain almost unchanged along the saturated vapour-pressure curve, only the height of the maximum decreases²⁰.

The average nearest-neighbour distance R_1 is given by the position of the first peak in $g(R)$, whereas the average coordination number N_1 can be obtained from the area under the first peak. The absolute value of N_1 depends sensitively on the method employed to define and integrate the first neighbour peak²². Depending on the model,

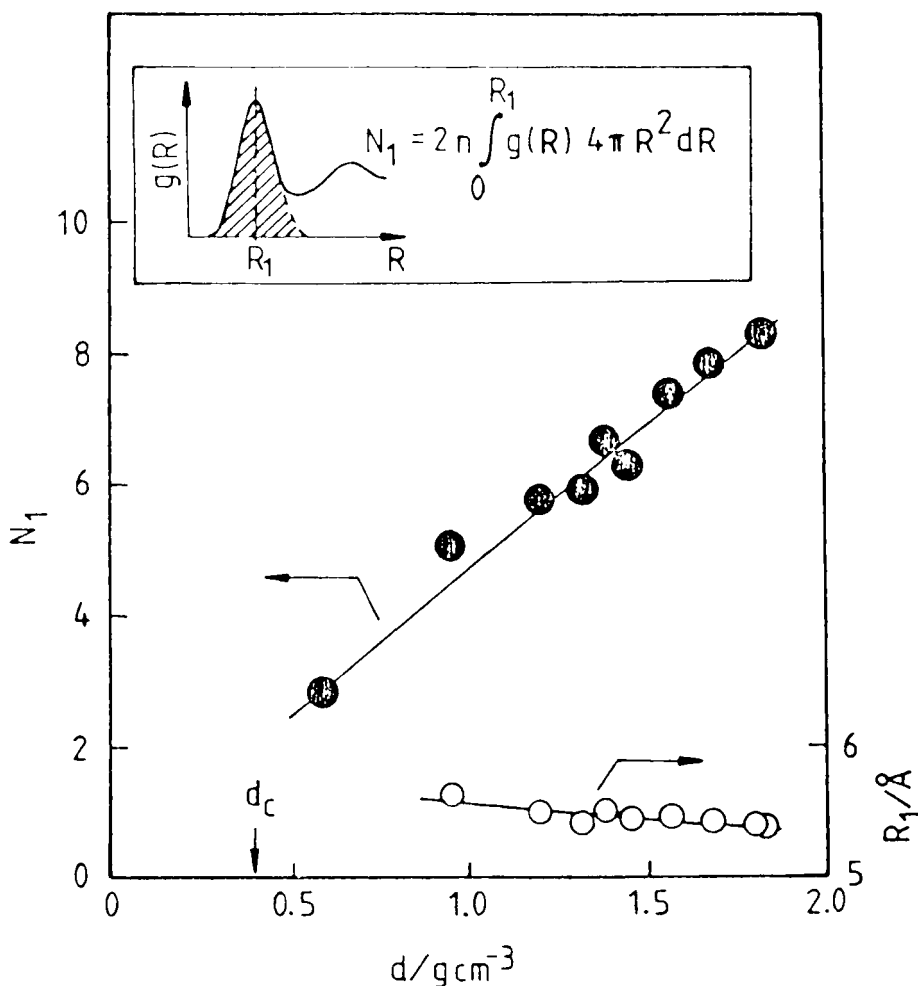


Figure 2 Average number N_1 and average distance R_1 of nearest neighbours for liquid cesium as a function of density (inset: method for calculating N_1).

the absolute value of N_1 can vary within $N_1 \pm 2$. We used the method of symmetrical main maximum (see inset of Figure 2).

The analysis of the data displayed in Figure 1b shows (Figure 2) that for cesium N_1 tends to decrease linearly with density from 9 close to the melting point to roughly 3 in the critical point region, whereas the position R_1 of the nearest neighbours remains essentially constant. These data clearly demonstrate that the density reduction by expanding liquid cesium develops mainly from a decrease in the average number of nearest neighbours rather than from an expansion to larger interatomic distances. A similar behaviour has been observed for expanded liquid rubidium and argon^{5,14,20}

Figure 3 displays the height of the first maximum of $S(Q)$, $S(Q_1)$, of a few liquid metals as a function of temperature, both quantities are reduced to their values at the melting point^{5,14,23,24}. Obviously, no reduced correlation between the alkali metals and the polyvalent metals is observed, indicating that reduced correlations are unlikely to hold for the liquid metals as a group. However, the alkali metals can be reduced in corresponding regions of the phase diagram, which indicates that the thermodynamic state dependence of the effective interaction potentials in metals may be very similar within groupings of metals like the group of the alkali metals. This indication is also supported by the existence of a law of corresponding states for the equation of state of the alkali metals rubidium and cesium^{2,6}.

As is well known, electrical transport measurements are directly correlated with the structure of the liquid metal, i.e. the structure factor $S(Q)$ ²¹. In the nearly-free

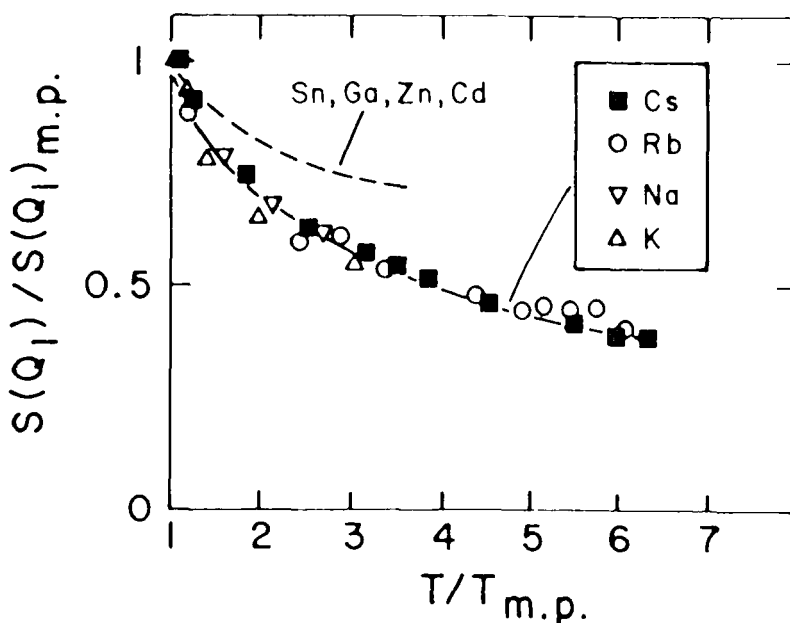


Figure 3 The height $S(Q_1)$ of the first peak of $S(Q)$ for a few liquid metals as a function of temperature (both values are reduced to their value at the melting point).

electron (NFE) approach, the electrical conductivity is described by the Faber-Ziman-formula²⁵

$$\sigma^{-1} = \frac{3\pi m^2 \Omega}{4\hbar^3 e^2 k_F^6} \int_0^{2k_F} S(Q) |V(Q)|^2 Q^3 dQ \quad (1)$$

where $V(Q)$ denotes the screened ion pseudopotential, k_F the wavenumber of the electrons at the Fermi surface and Ω is the atomic volume. For calculating $V(Q)$ we used the Ashcroft empty core potential²⁶, combined with a density dependent dielectric function $\epsilon(Q)$ which takes into account exchange and correlation effects^{5,27-29}. Figure 4 shows the results of these calculations compared with the experimental values of the dc-electric conductivity for expanded liquid cesium³⁰. The agreement is satisfactory for higher densities. The applied formalism starts to fail already at a density of about 1.4 g cm^{-3} , i.e., at about 3-4 times the critical density d_c . The region where the NFE-model breaks down is very close to the region where the onset of the magnetic susceptibility enhancement has been observed^{3,4}, indicating that electron-electron correlation effects become important. In addition, the electron mean

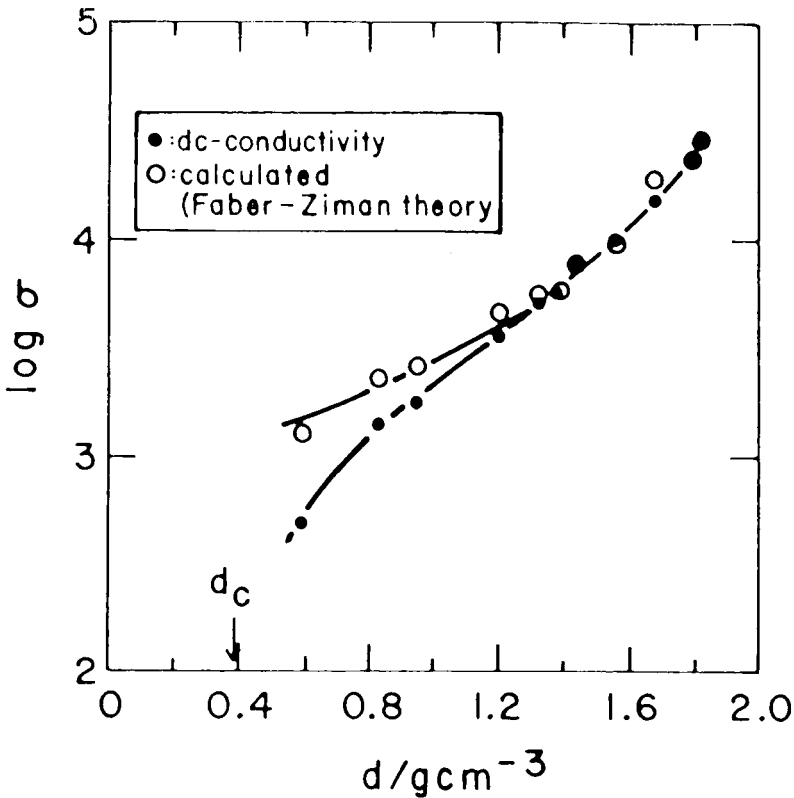


Figure 4 Measured and calculated (Faber-Ziman NFE-model) electrical conductivity of liquid cesium as a function of density.

free path becomes small in this region and approaches a value which is comparable to the mean interatomic separation of the metal atoms. This necessarily leads to the breakdown of the weak scattering Faber-Ziman approximation and marks the onset of the strong thermodynamic state dependence of the electronic structure as the metal-nonmetal transition is approached¹.

When the alkali metals are expanded at conditions near the coexistence curve, density and temperature are changed simultaneously. In order to understand the influence of each of these thermodynamic variables on the structural properties upon expansion in more detail, it is necessary to study separately the isobaric temperature and isothermic pressure dependence of $S(Q)$. The effect upon $S(Q)$ of increasing the temperature or pressure at constant pressure and temperature, respectively, is illustrated for one example in Figure 5. It can be seen that the temperature increase broadens the main maximum of $S(Q)$ and shifts its main peak position Q_1 to a smaller value, whereas the compression results in a slight shift of Q_1 to a higher value, while its peak height rises only by a small amount. This example clearly shows that the temperature and pressure effects on $S(Q_1)$ and Q_1 and therefore also on the distance of nearest neighbours, R_1 , have opposite signs and compensate each other to some extent during the expansion of the metal.

The isothermal pressure derivative of the structure factor $S(Q)$ is of particular interest, because it is related to three-body correlations and plays an important role in extracting quantitative information on the effective pair potential from experimental diffraction data¹⁵⁻¹⁸.

Figure 6 shows the experimental results for the pressure dependence of the structure factor $S(Q)$ of liquid cesium at different subcritical temperatures between 340 and 1700 K³¹. It can be seen that in the high density metallic region of cesium at 343 K the dominant pressure effect of $S(Q)$ (or equivalently on the pair correlation function $g(R)$) is a slight shift of the first peak position, reflecting a corresponding slight change in the mean interatomic distance R_1 , while otherwise little change is seen. In a theoretical paper of Bratkovsky *et al.*³², the pressure-dependence of $S(Q)$ of alkali metals has been studied using a model potential of the Animalu-Heine type in combination with the Percus-Yevick equation. Their calculated $S(Q, P)$ shows an increase in peak position and height of the main peak under compression. Their results are in good qualitative agreement with the observations for cesium and also with similar data obtained for liquid potassium³³ and liquid rubidium³⁴.

However, this behaviour of liquid alkali metals is quite unlike that of normal non-conducting liquids like the rare gases, for which pressure change mainly leads to a change in the number of nearest neighbours²⁰, but it is in good agreement with the predictions of the uniform fluid model (UFM) which has been successfully used by Egelstaff and his colleagues to explain the density dependence of $S(Q)$ of liquid rubidium near the melting point^{34,35}. The UFM can be characterized in the following way. At high densities, the alkali metal effective interaction potential $\phi(R)$ is assumed to have a soft core repulsion for small distances R and a weak attractive minimum at intermediate R . At larger distances, the potential oscillates according to $\phi(R) = \cos(2k_F R)/R^3$. Here, $k_F = (3n\pi^2)^{1/3}$ represents the Fermi wave vector and n the number density of the conduction electrons. The effect of density change is then

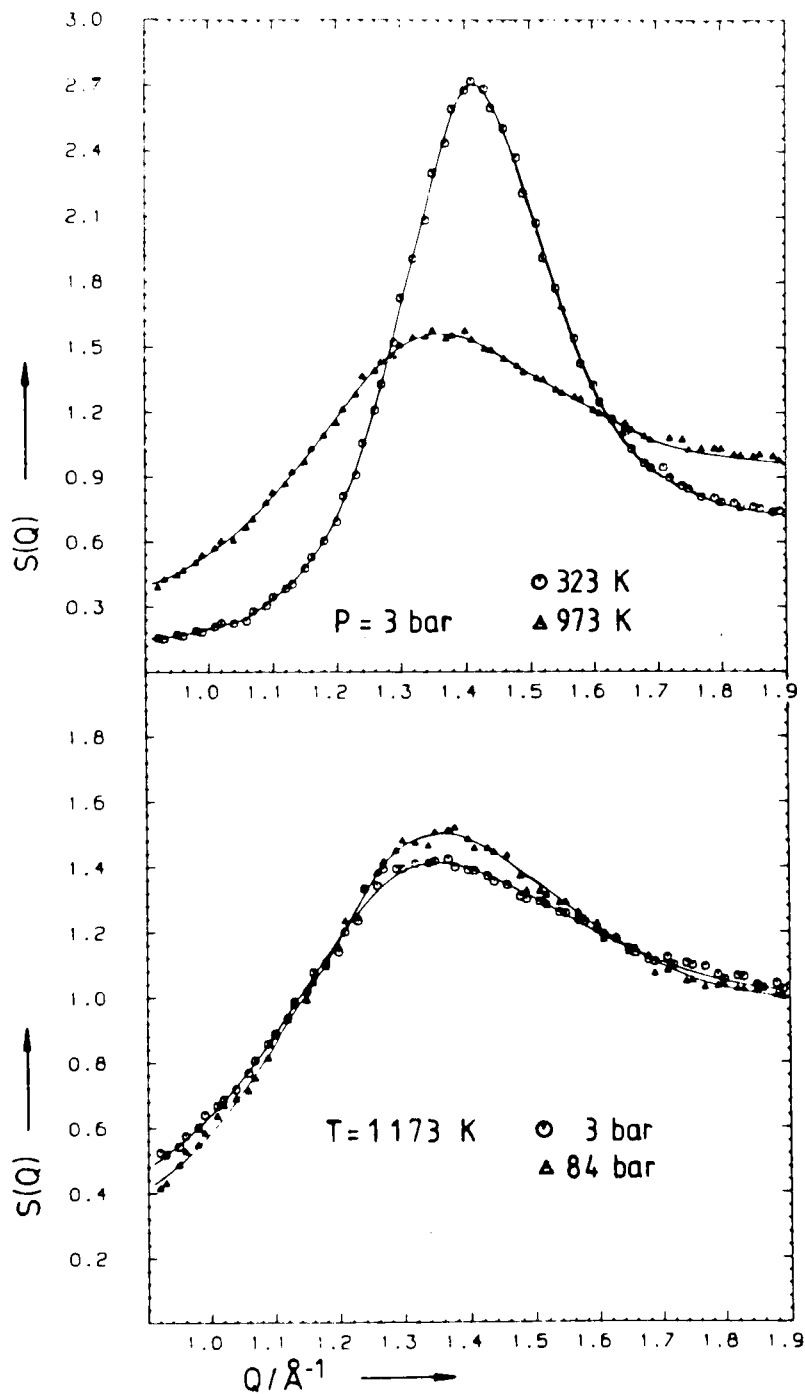


Figure 5 Temperature and pressure effect on the region of the main peak of the structure factor $S(Q)$ of liquid cesium.

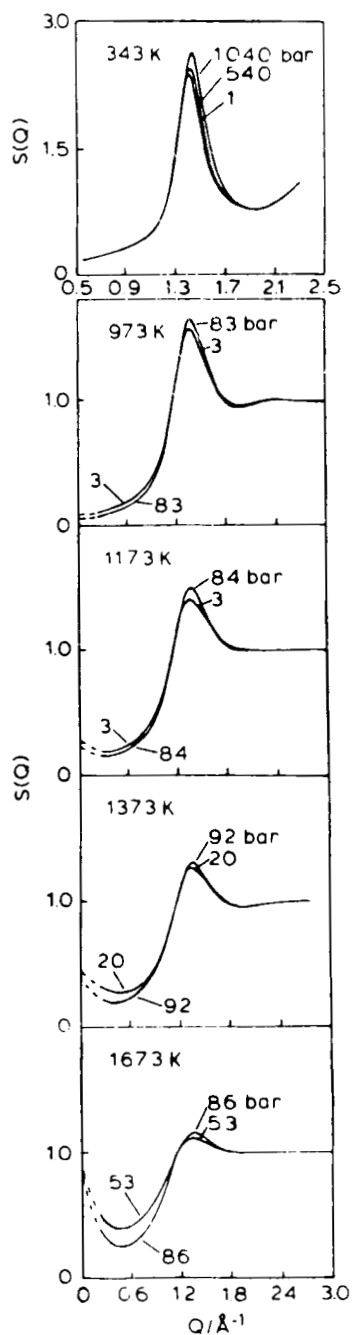


Figure 6 Structure factors $S(Q)$ of expanded liquid cesium as a function of temperature and pressure. The dashed lines at low Q are extrapolations to the compressibility limit.

simply to scale all distances with the density dependence of k_F . This behaviour leads to the following expression for the density derivative of $S(Q)^{35}$:

$$\left(\frac{\partial S(Q)}{\partial d}\right)_T = -\frac{Q}{3d} \cdot \left(\frac{\partial S(Q)}{\partial Q}\right)_{d,T} \quad (2)$$

Egelstaff *et al.* verified experimentally that liquid rubidium near the melting point satisfies Eq. 2. The prediction of the UFM (Eq. 2) is compared with the experimental data of cesium in Figure 7. As can be seen, the UFM fits the experimental data for cesium at $T = 343$ K (mean density $\bar{d} = 1.87 \text{ gcm}^{-3}$), i.e., close to the melting point, also reasonably well. However, it is obvious from Figure 7 that its validity breaks down with increasing temperature. It has been shown that the UFM does not agree with data for the density dependence of $S(Q)$ of liquid argon, neon or a Lennard-Jones model fluid^{36–38}, because the number of nearest neighbours changes with density in these cases, whereas the distance of the nearest neighbours remains virtually constant. The data for liquid cesium at $T = 973$ K ($\bar{d} = 1.46 \text{ gcm}^{-3}$) and $T = 1173$ K ($\bar{d} = 1.34 \text{ gcm}^{-3}$) shown in Figure 7 do indeed show features which are similar to those observed for the Lennard-Jones fluids.

The contrast between the behaviour of the liquid metals and the Lennard-Jones fluids has been analyzed in a number of theoretical papers^{36–40}, which use a selection of model potentials (hard-sphere, Yukawa, Lennard-Jones and the Price potential), each of which is solvable within the mean-spherical approximation or the Percus-Yevick approximation. The results of these papers can be interpreted that two characteristic features of the effective interaction potential of metals are mainly responsible for an experimentally observed UFM-behaviour: the softness of the repulsive core and the presence of Friedel oscillations in the tail of the potential.

The failure of the UFM for liquid cesium at conditions $T \geq 973$ K can thus be explained by the disappearance of these features which are typical for a nearly-free electron metal²¹. At slightly higher temperatures the breakdown of the nearly-free electron (NFE) approach of the Faber-Ziman model for the electrical conductivity is observed (see Figure 4).

As already noted, the electron mean free path approaches a value which becomes comparable with that for the mean interatomic distance at these temperatures and densities. A small electron mean free path, however, corresponds to a blurring of the Fermi surface, which has not been included in the calculation of the NFE-model^{41,42}. It has been shown for a point model⁴³ that such a blurring damps out the oscillatory behaviour of the ion-ion interaction potential and makes at the same time the repulsive part harsher. Both effects can thus lead to a breakdown of the UFM.

A further interesting feature of Figures 6 and 7 is the shape of $S(Q)$ and its density derivative in the low Q region for the temperatures $T = 1373$ K and $T = 1673$ K, which correspond to the reduced temperatures $\Delta T/T_c$ of 0.29 and 0.13, respectively. At such large distances from the critical point, insulating fluids like argon do not show such a marked enhancement in $S(Q)$ for small Q . One explanation of this effect, which has been discussed by Kahl and Hafner⁴⁴, is that it reflects the strong density dependence of the attractive interaction when the screening is reduced as the metal-nonmetal transition is approached.

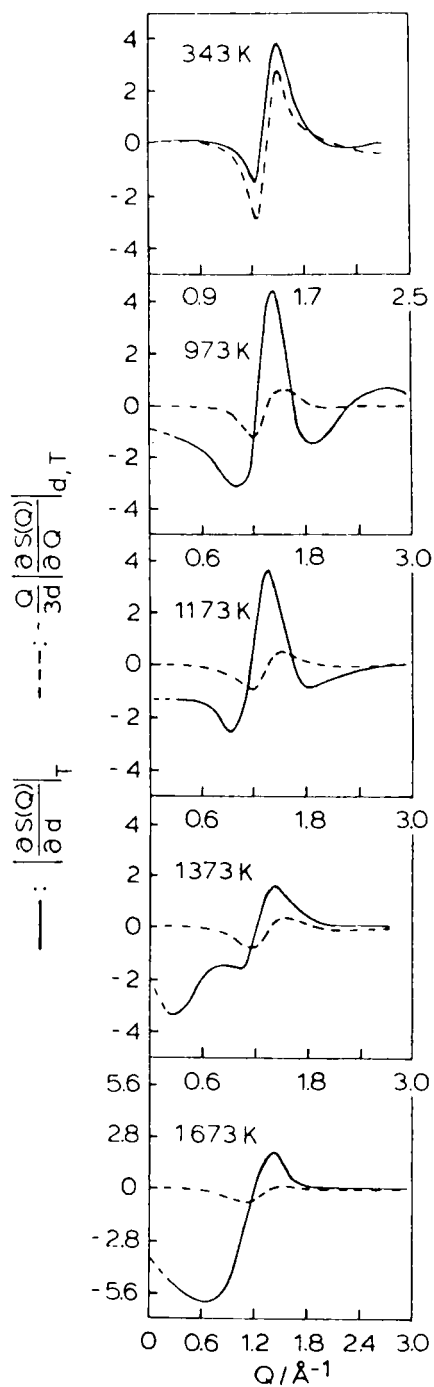


Figure 7 The density (full line) and wavenumber (dashed line) derivative of the structure factor $S(Q)$ of liquid cesium (see Eq. 2) at different temperatures along the liquid-vapour coexistence curve.

3 THEORETICAL CALCULATIONS OF THE STRUCTURE OF EXPANDED LIQUID METALS

Whilst a considerable amount of theoretical work has been devoted to the structural properties of liquid alkali metals at high densities, i.e. close to their melting point^{21,45-47}, the structure of expanded liquid metals is less well explored. In particular, all theoretical attempts employed up to now to explain the structure of expanded metals have used nearly-free electron theory in describing the interatomic forces in connection with computer simulation techniques or liquid state theory.

Mountain⁴⁸ used Price's density dependent potential^{49,50} and the Monte-Carlo method for his calculation of the structure factor of expanded liquid rubidium. Because of the poor agreement found between the calculation and experimental structural data available at that time²⁹, Mountain claimed that the Price potential is too strongly repulsive and does not reliably incorporate the density dependence into the potential. On the other hand, the agreement with the more recent and more accurate data on expanded rubidium¹⁴ is better, except for the low Q -region at the highest temperatures studied.

A similar approach was used by Tanaka⁵¹ employing a molecular dynamics calculation based on a slightly different effective pair potential $\varnothing(R)$, the optimized Heine-Abarenkov potential as proposed by Shaw⁵², using Singwi's⁵³ formula for the dielectric screening function of the valence electrons. The calculations showed that the minimum of $\varnothing(R)$ becomes deeper with decreasing density, whereas the position of the minimum does not shift very much. Good agreement is found with the experimental results for liquid rubidium¹⁴ at moderate temperatures and momentum transfers, but the low Q -region of $S(Q)$ is also not adequately reproduced.

Bratkovsky *et al.*⁵⁴ calculated the structure factor of expanded liquid rubidium in the Percus-Yevick approximation, using a potential as proposed by Animalu and Heine^{55,56}. They also found that under expansion of the liquid metal the repulsive core size decreases and the minimum depth of the effective interaction potential increases. They observe good agreement with the data of Franz *et al.* for liquid rubidium¹⁴ (which cover the range from 350 K ($d = 1.46 \text{ gcm}^{-3}$) up to 2000 K ($d = 0.54 \text{ gcm}^{-3}$) at conditions near the liquid-vapour coexistence curve) up to $T \leq 0.7 T_c$, i.e., up to a temperature of about 1400 K, corresponding to a density of about 0.97 gcm^{-3} . On further approaching the critical point, discrepancies at small Q -values become significant. One reason probably is that the Percus-Yevick approximation fails to describe the behaviour of fluids in the critical region correctly, but the discrepancies might also arise from the use of the Animalu-Heine-potential, which might become inappropriate at the low densities.

McLaughlin and Young⁵⁷ have calculated the interatomic interaction potential of expanded rubidium employing the pseudopotential technique, based on a softened hard sphere reference system following the WCA method⁵⁸, the attractive tail has been incorporated in the mean density approximation⁵⁹. They observed, as the density is reduced, that the attractive tail deepens, whereas the core stays approximately constant. At the higher temperatures, the potentials become harder in the neighbourhood of the effective collision diameter. They also compared their results

with the most recent neutron scattering data of Franz *et al.*¹⁴ The experimentally observed $S(Q)$'s can be understood quite well within their model up to the highest temperatures, where the attractive tail of the potential becomes essential in describing the low Q behaviour of $S(Q)$.

Kahl and Hafner⁴⁴ also performed a theoretical calculation of the structure factor of expanded liquid rubidium at conditions near the liquid-vapour coexistence. Their calculation is based on the optimized random-phase approximation and on an effective interatomic pair potential derived from pseudopotential perturbation theory⁴⁷. The pair potential is split into a repulsive short-range interaction and an attractive long-range part following the WCA convention. The repulsive part consists of an effective hard-sphere interaction with corrections for the softness being performed by the blib function method⁴⁷. The long-range attractive interaction has been solved in the optimized random-phase approximation. The authors find the following. The oscillatory part of the interaction potential plays an important role in determining the medium Q -range structure of liquid rubidium. The importance of these Friedel-oscillations in determining the liquid structure has also been pointed out by Cummings and Egelstaff^{36,39}. In course of the expansion of liquid rubidium, the nearest neighbour distance R_1 is essentially constant, in agreement with the experiment¹⁴. The strength of the interatomic potential increases strongly with decreasing density. Only for temperatures $T \leq 1400$ K ($d = 0.97 \text{ gcm}^{-3} = 3.3d_c$) good agreement between theory and experiment is found. For higher temperatures, the density dependence of the interaction potential is evidently underestimated within the applied model. In this region, the electronic mean-free path is of the order of magnitude of the interatomic spacings, so that the applied linear screening and weak-scattering formalism break down. This breakdown has been observed for expanded liquid cesium also at a density of about 3 to 4 times d_c ⁵. The screening functions would have to be corrected for short range electronic mean-free paths in the calculation. The true interatomic potentials are probably more attractive at the higher temperatures than the ones given by the linear-response calculation used by the model.

Khanna and Bretonnet⁶⁰ calculated $S(Q)$ of liquid expanded rubidium by using the analytical form for the direct correlation function of the one-component plasma reference system and the random-phase approximation to consider the electronic screening⁶⁰⁻⁶² and derived the expression of the long-wavelength limit $S(0)$ of the structure factors. They were able to describe the experimental $S(0)$ up to about 1800 K with their model by using the dielectric screening functions of Geldart and Vosko or Ichimaru and Utsumi^{63,64}.

Recently, Naito and Yokoyama⁶⁵ applied a method that combines features of the mean-spherical approximation and the phonon model⁶⁶ for calculating the effective interaction potential from the observed low-angle structural data of liquid rubidium. Their effective pair potential can adequately explain the observed structure factors, which have been calculated in the Percus-Yevick approximation, for all momentum transfers up to 1900 K. However, at their highest temperature $T = 1900$ K, the interatomic potential becomes markedly different from the other calculations, particularly the core size shrinks drastically. The authors explain this effect in conjuncture with a metal-nonmetal transition.

In conclusion, it seems that the structure of expanded liquid rubidium can be explained reasonably well within the framework of a nearly-free electron potential at least up to densities of about 3 to 4 times the critical density. However, further theoretical work seems necessary which comments on the problems encountered with the deviation from the linear-screening theory and which takes into account the effects of short mean free paths. Further theoretical developments can now also be tested with the new structure data of expanded liquid cesium⁵. Especially the measured temperature- and pressure-derivatives of the structure factors, which exist now for a wide density region, will allow a crucial proof of the applied model. The density derivative of $S(Q)$ will probably allow to extract more quantitative information on the triplet-correlation function.

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