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Propagation of Density Fluctuations in Liquid Rubidium: A Molecular-Dynamics Study*

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Liquid rubidium has been simulated on a computer. Density fluctuations in the computer liquid are essentially identical with those in the real liquid as measured by neutron inelastic-scattering experiments. Hence the interaction potential used for the simulation should be considered reliable for the study of liquid rubidium. Density waves are found to propagate even at wavelengths as small as the nearest neighbor distance in the liquid, a property not found in liquid argon.

In a liquid-argon-like system, molecular-dynamics (MD) calculations indicate that Brillouin side peaks in the spectrum of density fluctuations disappear already at a wavelength λ of about 8 times the first-neighbor distance in the liquid.[1] For shorter λ the fluctuations are over-damped oscillations with a single, λ-dependent decay time.[2]

MD calculations in a liquid-rubidium-like system are reported here. They show clear Brillouin peaks for λ almost as short as the first-neighbor distance; the λ dependence of the width and intensity of these peaks shows features of interest to the theory of density fluctuations in monatomic liquids.

The interparticle potential for liquid rubidium, obtained by Price and co-workers,[3] has an oscillatory decay to zero and, in this respect, is very different from a Lennard-Jones (6-12) potential which has been found suitable for the study of liquid-argon-like systems. Since the spatial and temporal behavior of density fluctuations, apart from its dependence on temperature and density, is determined by the nature of the interparticle interaction, a MD calculation with a potential qualitatively different from the Lennard-Jones

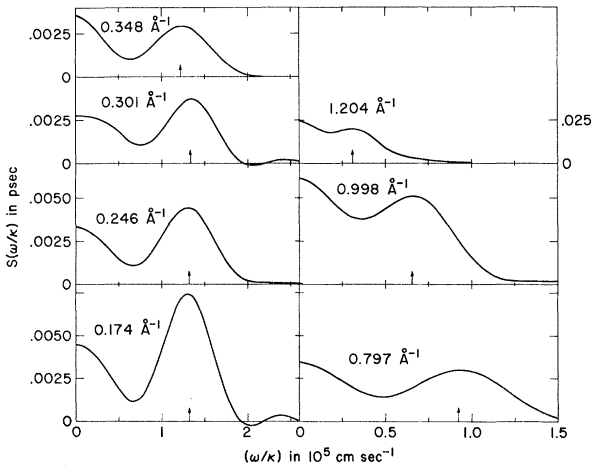

FIG. 1. $S(\kappa,\omega)$ for a few selected values of κ. Note that the abscissa is a velocity. The velocity of sound is seen to be 1.32×10^5 cm sec^{-1}. Note also that the vertical scale for $\kappa = 1.204$ Å^{-1} is different.

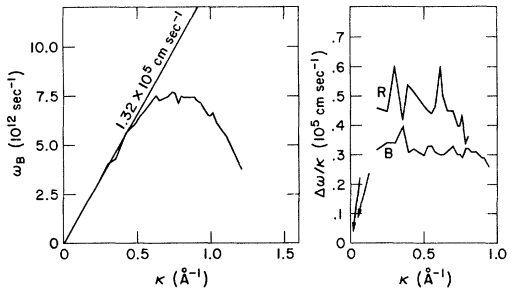

FIG. 2. Frequency ω_B of the Brillouin peak as a function of κ. No peak is present beyond 1.2 Å^{-1}. Half-widths $\Delta\omega_R$ and $\Delta\omega_B$ of the Rayleigh and Brillouin peaks are also shown. The arrows at the origin indicate symbolically the long-wavelength limits of $\Delta\omega_R/\kappa$ and $\Delta\omega_B/\kappa$.

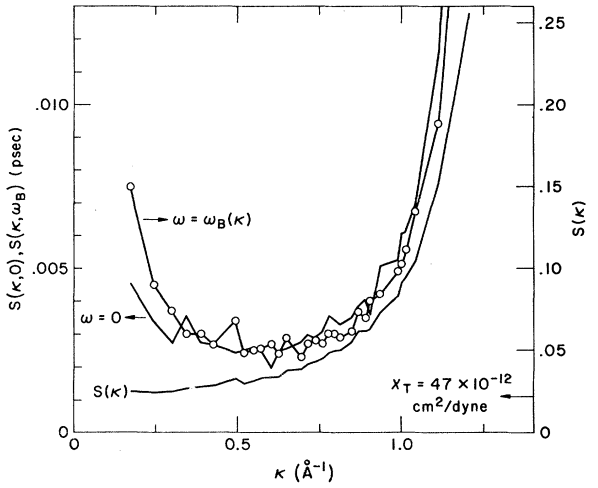

FIG. 3. $S(\kappa,\omega=0)$ and $S(\kappa,\omega=\omega_B)$, left-hand scale. Structure factor $S(\kappa)$, right-hand scale. $S(0)$ gives the value of χ_T shown (Ref. 4).

potential gives us a clear idea of the extent to which density fluctuations are sensitive to the interparticle potential. On the other hand, comparison with neutron inelastic-scattering data enables us to judge the suitability of the potential for the study of real liquid rubidium.

Using the potential obtained by Price and coworkers,[3] an MD calculation on 500 particles has been performed for liquid Rb at 1.502 g cm^{-3} and 319°K. Density fluctuations of the wave vector $\kappa = 2\pi/\lambda$ were computed from

$$Q(\vec{\kappa},t) = N^{-1/2}\sum_j \exp(i\vec{\kappa}\cdot\vec{r}_j),$$

by calculating its autocorrelation

$$F(\kappa,t) = \langle Q(\vec{\kappa},0)Q^*(\vec{\kappa},t)\rangle,$$

and the Fourier transform

$$S(\kappa,\omega) = \pi^{-1}\int_0^\infty \cos(\omega t)F(\kappa,t)\,dt.$$

In Fig. 1 $S(\kappa,\omega)$ is shown for a few values of κ. Only at $\kappa > 1.2$ Å^{-1} do the Brillouin peaks merge completely into the central Rayleigh intensity region. From Fig. 1 it is seen that the velocity of sound is 1.32×10^5 cm sec^{-1}.[4]

The dispersion of the velocity of propagating density fluctuations is shown in Fig. 2. It is remarkably similar to the average dispersion curve in a polycrystal[5] but the differences are to be kept in mind: Firstly, here the dispersion curve disappears *before* κ reaches the value (1.54 Å^{-1}) where $S(\kappa)$ [$\equiv F(\kappa,0)$], the structure factor, is a maximum, and secondly, the central intensity region in $S(\kappa,\omega)$ is totally absent in a polycrystal. The half-width of the Brillouin line, $\Delta\omega_B$, is taken from the right part of each spectrum (i.e., one assumes that the central region has disappeared before $\omega = \omega_B$). This is clearly unambiguous up to about $\kappa = 0.8$ Å^{-1}. $\Delta\omega_B/\kappa$ is shown in Fig. 2. It is clear that $\Delta\omega_B$ is proportional to κ and not κ^2 as in the strictly hydrodynamic region. If the width is written as $\kappa^2\eta(\kappa)$, it implies a "viscosity" $\eta(\kappa)$ decreasing as κ^{-1} in the region beyond $\kappa = 0.1$ Å^{-1}. The half-width of the central line, $\Delta\omega_R$, is also shown in Fig. 2. $\Delta\omega_R/\Delta\omega_B$ is roughly 1.5 up to $\kappa = 0.7$ Å^{-1}, after which $\Delta\omega_R$ tends to increase less rapidly.

$S(\kappa,\omega=0)$ and $S(\kappa,\omega=\omega_B)$ are shown in Fig. 3. These are consistently the same to within statistical fluctuations, except for $\kappa < 0.25$ Å^{-1} where the latter is much larger. For $\kappa > 0.8$ Å^{-1} the opposite occurs. Figure 3 also shows the structure factor $S(\kappa)$. Its maximum occurs at $\kappa = 1.54$ Å^{-1}. The isothermal compressibility χ_T is also indicated in Fig. 3.[4]

From the point of view of the theory of liquids, these results show that we need to understand the connection between the input—namely, the temperature, the density, and the pair potential —and the resulting persistence (as in Rb) or other behavior (as in a Lennard-Jones liquid) of short-wavelength density fluctuations.

There is close agreement between the MD results presented above and the experimental results of Copley and Rowe,[6] except in the quasielastic region. (The relevant experimental difficulties which may be a cause of this discrepancy are discussed by Copley and Rowe.[6]) The dispersion curves are essentially identical (see Fig. 4), and, as will be shown in another paper,[7] for $\kappa > 1$ Å^{-1} the MD results are in close agreement with experimental results[8] (in the quasielastic

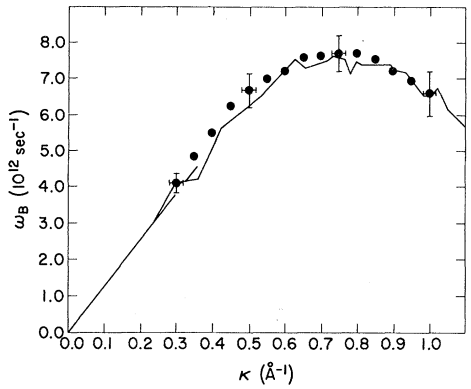

FIG. 4. Comparison of MD results with experiment (Ref. 6); ω_B is the frequency of the Brillouin peak; filled circles, taken from Ref. 6.

region as well). We conclude that the potential[3] used here for liquid Rb gives a very faithful description of density fluctuations in that liquid.

Thus, for further development in the theory of liquids and for an understanding of liquid alkali metals, rubidium appears to be a good material for intensive study.

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Calorimetric Investigation of the Continuous Demagnetization of Ce Impurities in Superconducting La, Th Alloys

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A calorimetric investigation of the magnetic-nonmagnetic transition of an impurity (Ce) in a superconducting binary-alloy matrix (La, Th) is reported for the first time. The continuous demagnetization of the Ce ions with increasing Th composition is documented in terms of the specific heat jump at the superconducting transition temperature T_c as a function of T_c. A recent calculation by Müller-Hartmann and Zittartz for superconducting Kondo systems describes these data remarkably well out to ~ 70 at.% Th, but fails at higher Th compositions.

A recent report of data on the superconducting transition temperature T_c for La, Th alloys containing dilute quantities of Ce has documented the transition of the solute Ce from a magnetic state in La to a nonmagnetic state in Th.[1] This documentation, however, was solely in terms of the initial depression of T_c of the La, Th matrix with increasing Ce impurity concentration n, $\Delta T_c/n$. Starting from the matrix-impurity system *La*Ce, the depression steepens sharply as the Th content increases, then beyond a maximum at about 45 at.% Th it sweeps back to a shallow slope for *Th*Ce. Such behavior had been observed previously, not as a function of alloying but of pres-