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STUDY OF STRUCTURAL AND DYNAMICAL PROPERTIES OF LIQUID METALS

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ABSTRACT

In this review recent experimental and theoretical research of static and dynamical phenomena in liquid metals is discussed. Using experimental data for the structure factor $S(\kappa)$ we describe correlation functions, and using statistical theories we discusse different approaches to the pair interaction potential $\varphi(r)$. The dynamical structure factor $S(\kappa,\omega)$ in liquid metals is also described by neutron scattering techniques and various theoretical models.

INTRODUCTION

The interest for liquid metals dating from late sixtieth has been increased in the last decade because of the improved neutron scattering techniques and wider posibilities in the interpretation of experimental data. All information collected from the experiments on liquid metals is related to the research of the structure and dynamics and may contribute to a better understanding of the nature of liquids state in general. Besides, investigation of liquid metals is important for different applications, especially in the nuclear technology.

Of the three principal phases of matter the liquid state is the least understood, because of its complicated nature. The gaseous phase is a disordered state, whose interatomic forces are very week, because of the large relative distances between particles. Besides this fact gaseous state can be described in a predominant number of cases, by the classical statistical physics. The solid state, in spite of very strong interatomic interactions, because of its higher internal symetry in comparison with other two phases can be studied by powerful theoretical approaches(1). On the other hand in the liquid state there are very strong interatomic forces and in addition is a typical many particle disordered system. Furthermore it is impossible to define a complete Hamiltonian of the system. To overcome these difficulties statistical theories have been applied for solving the equation of state, as it will be explain latter on. Without going into ditailes of these theories it is usefull in this paper to underline several different branches of research in physics of simple liquids, of which it is important to mention the following:

1. Theoretical research of static and dynamic properties of monoatomic liquid metals.

2. X-ray and neutron scattering studies of liquid metals.

3. Computer experiments.

Firstly we shall briefly discuss the computer simulation studies of many - particle systems. There are two basic methods for such a kind of research: the Monte Carlo (MC) method and the molecular dynamics method (MD). Both methods are used for studing monoatomic liquid metals, the former for equilibrium characteristics and the latter for time dependent properties, as it have been described by several authors(2,3).

In order to achieve a better understanding of the complicated nature of the liquid state, in the next sections we are going to discuss the both meantioned approaches.

STATIC PHENOMENA

Many - particle systems, like simple monoatomic liquids (aluminium, lead, tin) can be described by the equation of state(4):

$$\frac{dU(r)}{dr} = \frac{d\varphi(r)}{dr_1} + \frac{1}{\rho^2}\int \frac{n_3(r_1, r_2, r_3)}{g(r_{12})} \cdot \frac{\partial\varphi(r_{13})}{\partial r_1} d^3r_3 \qquad (1)$$

Hence, to get the pair interaction potential $\varphi(r_{12})$ it is necessary to solve the equation of the state. However, it is a very complex task, which requires knowledge of the three - atom correlation function $n_3(r_1, r_2, r_3)$ that is not accessible from the experiment. Nevertheless, there are some possibilities of solving the equation (1). One way is to replace the three - body correlation function with the product of pair correlation functions$^{(5)}$, which are obtainable from experimental data:

$$n_3(r_1, r_2, r_3) = \rho\, g(r_{12})g(r_{23})g(r_{13}) \qquad (2)$$

Using equation (2) it is possible to get the so-called Born-Green solution of the equation (1).

There is also a possibility to replace the many - particle correlation functions with the Ornstein-Zernike$^{(5)}$ approximation for the total correlation function h(r):

$$h(r_{12}) = c(r_{12}) + \rho \int_0^{\infty} c(r_{13})h(r_{23})d^3r_3 \qquad (3)$$

According to the Ornstein-Zernike equation (3) the total correlation function $h(r_{12})$ is decoupled into its direct and indirect components, as it is shown in Fig. 1. When the density ρ tends to zero total correlation function reduces to the direct correlation function c(r). The equation (3) should be taken as the definition of direct correlation function c(r). It describes the correlation between a two atoms in the presence of the remaining (N-2) atoms.

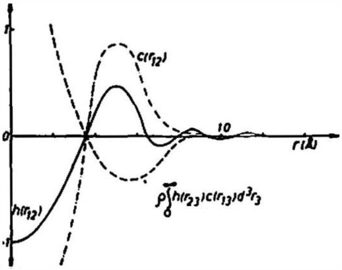

Fig. 1. Direct and indirect components of the total correlation function.

So far we have introduced, the principal correlation functions, but their numerical evaluation is possible only from a knowledge of the static structure factor S(κ). The structure factor can be deduced from X-ray and neutron diffraction measurements, or it can be calculated if the interaction potential φ (r) is of a hard core type. Both ways are fairsible, but in this work we shall discuss with the experimental one.

For the experimental conditions for the structure factor measurements we refer to the existing papers$^{(6,7)}$, since we would rather discuss the interpretation of experimental data. Analysis of the experimental data the following corrections are the most important: for background, for self--shilding , for absorption , for inelastic scattering and for multiple scattering. It should be emphasized that the correction for the multiple scattering is particularly important for the small κ-values, where the cross-section is very low. All these corrections are necessary to performe if one wants to get a precise S(κ) for a wide κ-region, Cf fig. 2. Applying the

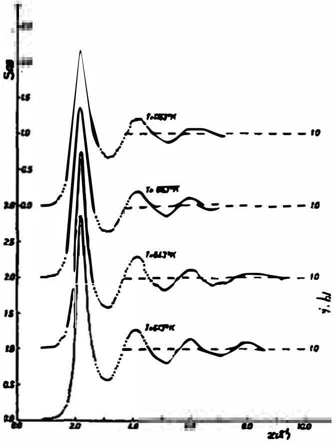

Fig. 2. The static structure factor of liquid lead at four temperatures.

Fourie transform for the corrected static structure factor S(κ), all correlation functions can be calculated$^{(8)}$. It is important to point out that this numerical integration introduce spurious ripples in a correlation function, which is caused by an uncertainty in measurements of S(κ), for very small and very large κ-values.

Knowing correlation functions g(r), h(r) and c(r)it is easy to calculate the pair interaction potential φ(r), for the metals of low densities, using certain statistical theories. Because of their simplicity the most frequently applied are Percus-Yevick and the Hypper-netted-chain (HNC) theories. There is a paper $^{(9)}$ at this conference in which will contain more details about the calculation of the interatomic potential.

DYNAMICAL PHENOMENA

Inelastic neutron scattering technique is the most powerful tool, available at present, for investigation of dynamical properties of liquids state. The observation time of neutrons is about 10^{-12} sec. and the relaxation time τ of the processes in liquid metals is of the same order of magnitude. The dynamical behaviour of liquid metals is related to the amplitudes of disturbances and to the relaxation time, and can be distinguished within three regions: $\omega\tau<1$, $\omega\tau\approx1$ and $\omega\tau>1$. In these regions the NMR, Rahman and Brillouin methods can be successfully applied as well.

In the process of the inelastic neutron scattering, of the momentum transfer $\hbar\kappa$ and the energy transfer $\hbar\omega$, take place in such a way that κ can be defined by the relation:

$$\kappa = \left[2k_0^2 - \frac{2m}{\hbar}\omega - 2k_0\sqrt{k_0^2 - \frac{2m}{\hbar}\omega}\cdot\cos\Theta_{sc}\right]^{1/2} \qquad (4)$$

where Θ_{sc} is the angle of scattering. The equation (4) defines the so-called kinematic allowed regions in the (κ,ω) space, which are shown in Fig. 3.

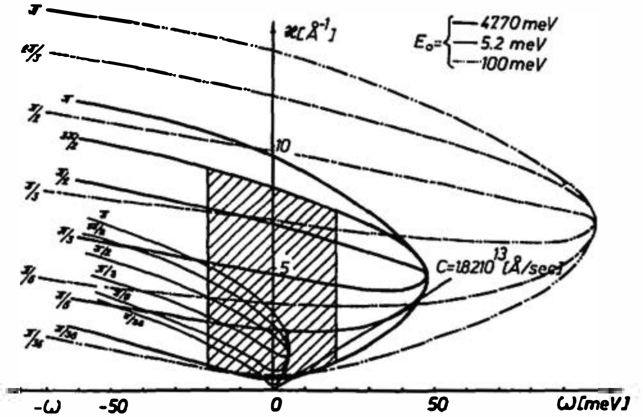

Fig. 3. The kinematic allowed region, for three typical values of E_0.

The Fig. 3. illustrates an efficiency of the neutron scattering technique for a study of the evidence of collective modes of vibration in a liquid, the optical density of thermal excitations and the structure of monoatomic liquid metals.

By the inelastic neutron scattering it is possible to extract the dynamic structure factor $S_{coh}(\kappa,\omega)$ and a self-scattering function $S_s(\kappa,\omega)$. The measurements of the scattering laws are described by many authors(10,11) and we shall not go into this matter.

The scattering laws can be obtained from a measurements(12) according to the formula:

$$J(\Theta,\hbar\omega) \rightarrow \frac{d^2\sigma}{d\Omega dE} = \frac{\sigma_{coh}}{4\pi}\frac{k_o}{k}S_{coh}(\kappa,\omega) + \frac{\sigma_s}{4\pi}\frac{k_o}{k}S_s(\kappa,\omega) \qquad (5)$$

The measurements of $\frac{d^2\sigma}{d\Omega dE}$ can be performed by a hybride time-of-flight spectrometer with a high resolution(12,13), or by the classical cold neutron technique, which is illustrated in Fig. 3. These measurements require similar corrections like it those mentioned for the static structure factor measurements.The recently developed Monte Carlo method for multiple scattering (MSCAT)(14) is frequently used for deducing the scattering laws. It should be pointed out that there are a rather big uncertainties for the mesurement points in (κ,ω) space for a large energy transfer $\hbar\omega$ and for small and large values of κ(10,11).

In the last decade a pronounced theoretical interest in research of dynamics of the liquid metals has been evident, and numerous models appeared for description of the scattering law $S(\kappa,\omega)$. The most general approach to this problem is the kinetic theory of liquids(15,16). All others models (the mean field theory, the viscoelastic theory generalized Langevin method) are a special cases of a kinetic theory(17).

The scattering laws obtained either theoretically or experimentally, depends only on the dynamics of the scattering, and can be related to the moment theorems(12):

$$<\omega^n(\kappa)> = \omega^n S(\kappa,\omega)d\omega \qquad (6)$$

For a high temperature, at which liquid state of metal exist, the static quantities could be defined from the scattering law using the moment theorems introduced by De Gennes(18):

$$\langle\omega^0_{coh}\rangle = S(\kappa)$$

$$\langle\omega^2_{coh}\rangle = \frac{\kappa^2}{M\beta S(\kappa)}, \qquad \beta = (k_B T)^{-1}$$

$$\langle\omega^4_{coh}\rangle = \frac{\kappa^2}{M\beta}\left[\frac{3\kappa^2}{M\beta} + \frac{\rho}{M}\int d^3r g(r)\ (1-\cos\kappa r)\ (\kappa\nabla)^2\varphi(r)\right] \qquad (7)$$

In Fig. 4. the experimental and theoretical second and forth moments of the coherent scattering law, for liquid lead, as function of κ are compared. It is evident, from Fig. 4, that De Gennes narrowing, near the first peak of the static structure factor S(κ), exceedingly pronounced.

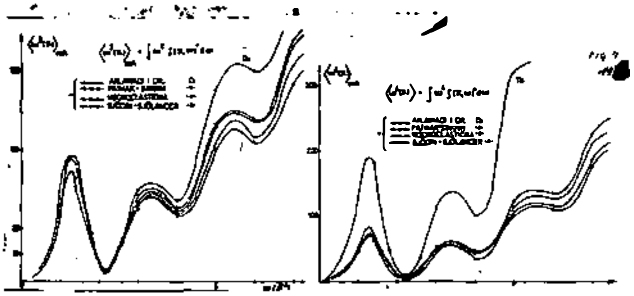

Fig. 4. The second and forth moments for liquid lead calculated from different theoretical models(12).

At the end of this lecture if should be concluded that from the static and dynamic structure factor we can learn more about interatomic forces, atomic motions and the relation between micro structure and the transport coefficients.

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