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ИНСТИТУТА  
ЯДЕРНЫХ  
ИССЛЕДОВАНИЙ  
ДУБНА

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THE TEMPERATURE DEPENDENCE  
OF THE CORRELATED AND INDIVIDUAL  
MOTIONS IN LIQUID SODIUM

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## INTRODUCTION

The inelastic neutron scattering is proved to be one of the most powerful techniques to study the microscopic behavior of the density fluctuations in liquids particularly liquid metals.

From the incoherent, neutron scattering one can get information about the microdynamics of the individual atoms while the correlated motions are studied on the coherent scattering of neutrons.

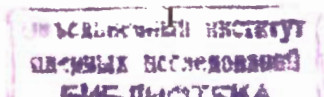
Sodium is monoatomic with the isotopic abundance 100%  $^{23}\text{Na}$  so that only the scattering from one single isotope is involved therefore the incoherent and coherent scattering functions  $S_i(Q, \omega)$  and  $S_c(Q, \omega)$  providing informations about individual and collective motions in liquid Na cannot be experimentally separated.

Several papers on liquid sodium have been published in the literature but no one followed such a separation [1-8]. In this paper we present the full pattern of  $S_i(Q, \omega, T)$  and  $S_c(Q, \omega, T)$  obtained from the mean field theory [9], viscoelastic [10] and memory function approaches [11].  $S_i(Q, \omega, T)$  was obtained on the basis of the three-pole approximation [12]. The structure data ( $S(Q)$ ,  $g(r)$ ,  $\Phi(r)$ ) asked in these theories have been discussed in the paper [13]. In an earlier paper [8] the total scattering function  $S(Q, \omega)$  at  $T=413\text{K}$  experimentally measured is well correlated with the calculations from the quoted models.

## COHERENT AND INCOHERENT SCATTERING FUNCTIONS. RESULTS

Since the sodium is both a coherent and an incoherent scatterer the scattering function  $S(Q, \omega)$  has to be understood as a sum of two components.

$$\sigma_t S(Q, \omega) = \sigma_c S_c(Q, \omega) + \sigma_i S_i(Q, \omega) \quad (1)$$



The  $\sigma_c$ ,  $\sigma_i$  and  $\sigma_t$  are the coherent, incoherent and total nuclear scattering cross sections.

The main quantities necessary in the theoretical models calculations are the fourth order moments. For classical monoatomic liquids there can be written as follows:

$$\langle \epsilon^4 \rangle = \frac{\hbar^4 Q^4 k_B T}{M^2} [3k_B T + n P(Q)] \quad (2)$$

$$P(\vec{Q}) = \int d\vec{r} g(\vec{r}) [(1 - \cos(\vec{Q}\vec{r}/Qr)] (\vec{Q} \cdot \nabla)^2 \Phi(r)$$

with  $g(r)$  the radial distribution function,  $\Phi(r)$  the pair interaction potential. The other constants have their usual meanings ( $\epsilon = \hbar\omega$ ).

$$\langle \epsilon^4 \rangle_1 = \langle \epsilon^2 \rangle_1 [3 \langle \epsilon^2 \rangle_1 + \langle \hbar^2 \Omega^2 \rangle]$$

$$\Omega^2 = \frac{4\pi n}{3M} \int dr g(r) \left[ \frac{\partial^2 \Phi(r)}{\partial r^2} + \frac{2}{r} \frac{\partial \Phi(r)}{\partial r} \right] \quad (3)$$

$$\langle \epsilon^2 \rangle_1 = \hbar^2 Q^2 k_B T / M$$

As we already stated from inelastic scattering (INS) on liquid Na it can be determined the total neutron scattering function  $S(Q, \omega)$  only.

To get further beyond the observations obtained from INS two ways namely MD simulation and calculation in terms of models could be used as supplement for neutron data.

We have reliable experimental data for  $S(Q, \omega)$  at 413K and they represent a good test to check the model calculations. A good agreement have been obtained, and we concluded that  $S_c(Q, \omega)$  and  $S_i(Q, \omega)$  can be calculated in the same way at other temperatures if the structure data are known. The

temperature dependence of the static structure factor  $S(Q)$  and the radial distribution function  $g(r, T)$  was reported in [13]. A Lennard-Jones effective pair potential  $\Phi(r)$  is taken into calculation. With the known form of  $S(Q, T)$ ,  $g(r, T)$  and  $\Phi(r, T)$  extensive calculations in terms of the models [9-12] were made. The full  $S_c(Q, \omega)$  surface in the range in which it was calculated for  $T=100-600^\circ\text{C}$  is shown in Fig.1.

The main maximum located at about  $Q \approx 2 \text{ \AA}^{-1}$  decreases constantly with the increasing of the temperature. A strong correlation between the dynamic and the static structure is observed. The collective excitations showing up very clearly in  $S_c(Q, \epsilon)$  as peaks give a well defined dispersion relation  $\epsilon = \epsilon(Q)$ . The three models used to calculate  $S_c(Q, \epsilon)$  gives closed result and in Fig.1 are shown only the data obtained from the model of the paper [10].

From the  $S_c(Q, \epsilon)$  it was determined the temperature dependence of the spectral function of the current correlation functions:

$$J_c(Q, \epsilon) = \frac{\epsilon^2}{Q^2} S_c(Q, \epsilon) \quad (4)$$

The dispersion of the collective modes is determined from the  $Q$  dependence of the  $\epsilon_m = \hbar\omega_m$  corresponding to the main maximum in  $J_c(Q, \epsilon)$ . This dispersion relations are plotted in Fig.2. The slope of these curves extrapolated to the limit  $Q \rightarrow 0$  gives the velocity of the sound  $v_s$ .

The values of  $v_s$  obtained in this way are within the range  $v_s(100^\circ\text{C}) = 2580 \text{ m/s}$  and  $v_s(600^\circ\text{C}) = 2120 \text{ m/s}$ . These data are compared to the result of the paper [14]. Here  $v_s$  is obtained for  $T$  near the melting point from the generalized hydrodynamics by means of the formula:

$$v_s = \left[ \frac{C_P k_B T}{C_V M S(0)} \right]^{1/2} \approx 2270 \text{ m/s}$$

In the range of the first Brillouin zone our data for  $T = 100^\circ\text{C}$  are in good agreement with the dispersion relation for solid sodium presented in the paper [15]. The authors used the static screening function of the electron gas in conjunction with the theory of Vasco Taylor and Keech [16].

From [16] we estimated  $\omega_m = 2.32 \cdot 10^{13} \text{ s}^{-1}$  and our data for  $T = 100^\circ\text{C}$  give  $\omega_m = 2.56 \cdot 10^{13} \text{ s}^{-1}$ .

The full  $S_1(Q, \epsilon)$  surface in the ranges in which it was calculated is shown in Fig.3 for  $Q=(0.3-15)\text{\AA}^{-1}$  and  $\epsilon=(0-18.45) \text{ meV}$  at  $T = 100-600^\circ\text{C}$ . For the sake to not complicate the figures  $S_1(Q, 0)$  is not presented.

At the transition from small  $Q$  values to higher ones  $S_1(Q, \epsilon)$  has to show a change from a Lorentzian to a Gaussian behavior (at  $Q \rightarrow \infty$ ). It is observed that this transition is faster than it is expected. This observation is in agreement with the paper [17] where the total scattering function was measured at three temperatures. The function  $S_1(Q, \omega)$  was obtained after what  $S_c(Q, \omega)$  was subtracted by means of the model.

#### CONCLUSIONS

A full surface in the interest ranges ( $Q, \epsilon$ ) for coherent and incoherent scattering functions  $S_c(Q, \epsilon)$  and  $S_1(Q, \epsilon)$  on liquid Na is presented. The data calculation performed are in terms of several dynamic theoretical models which do not involve any adjustable parameters. The only data needed in models, the static structure factor  $S(Q)$  and the radial distribution function  $g(r)$  have been obtained from neutron diffraction experiments.

The whole dynamics of the liquid sodium in the range of temperature  $T = 100-600^\circ\text{C}$  is shown in Fig.1-3. The static structure of liquid sodium can be well described with a hard sphere potential while the dynamics with a Lennard - Jones one.

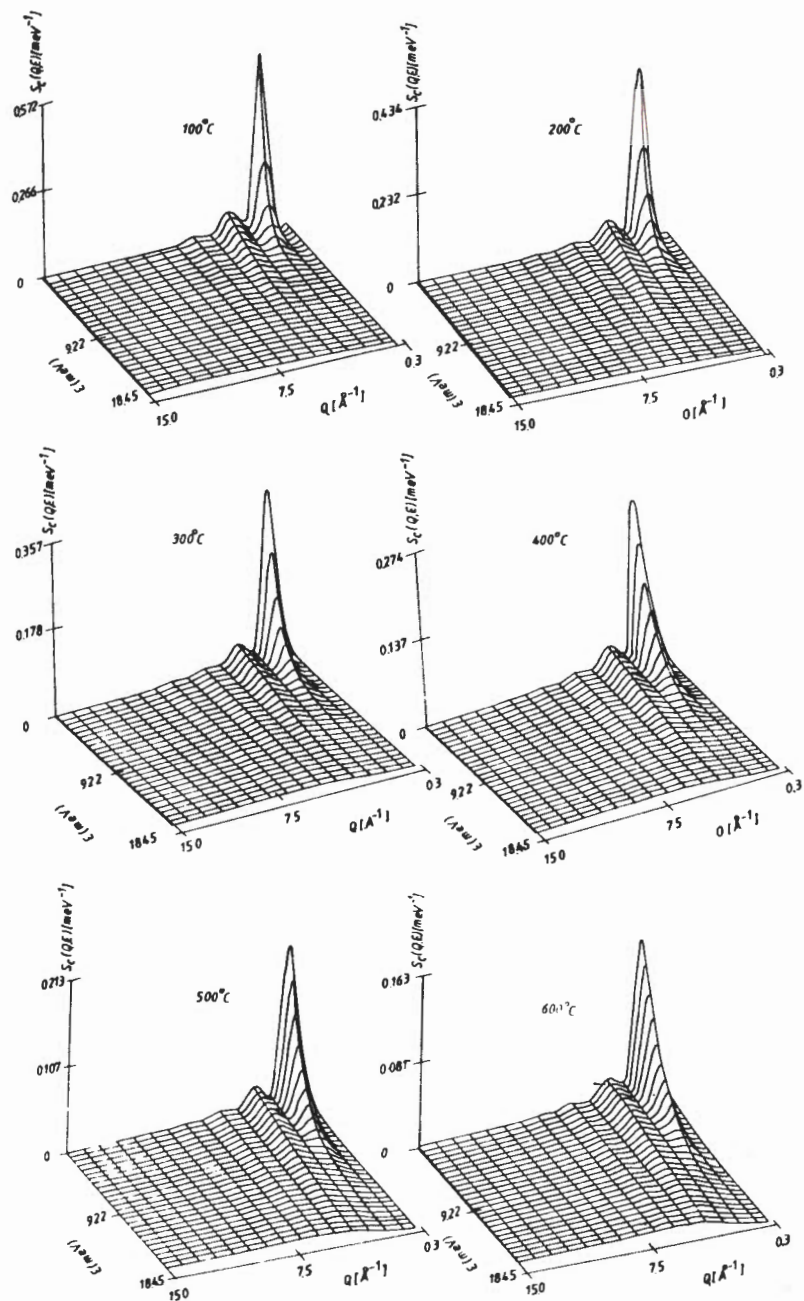


Fig. 1. Three-dimensional plot of the coherent scattering function for liquid sodium at  $T = 100-600^\circ\text{C}$

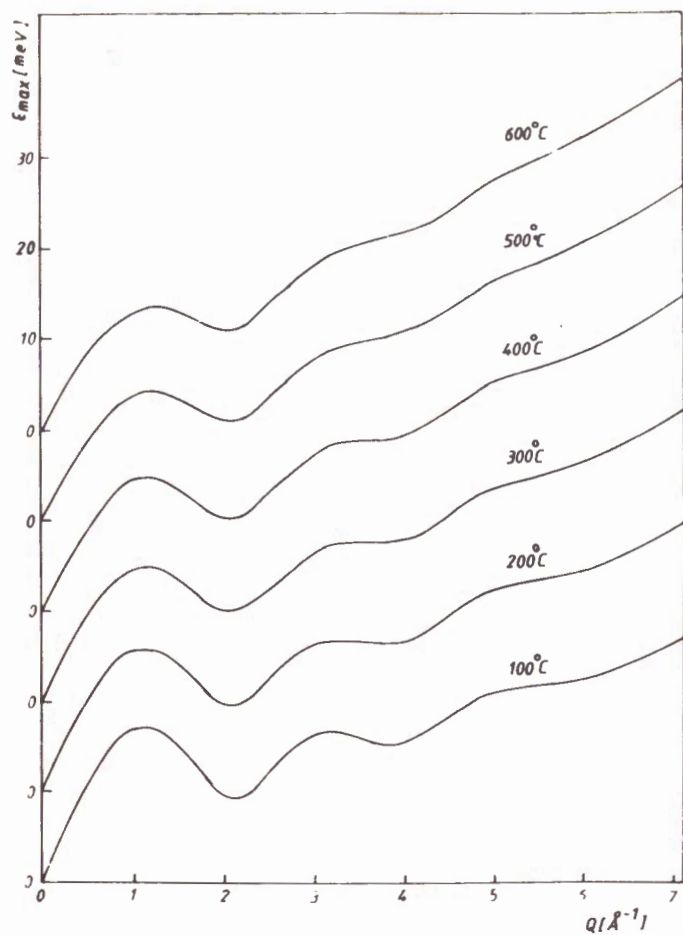


Fig. 2. Dispersion relations  $\epsilon_m(Q)$  from the maximum of the spectral function  $J_c(Q, \epsilon)$  for  $T = 100-600^\circ\text{C}$

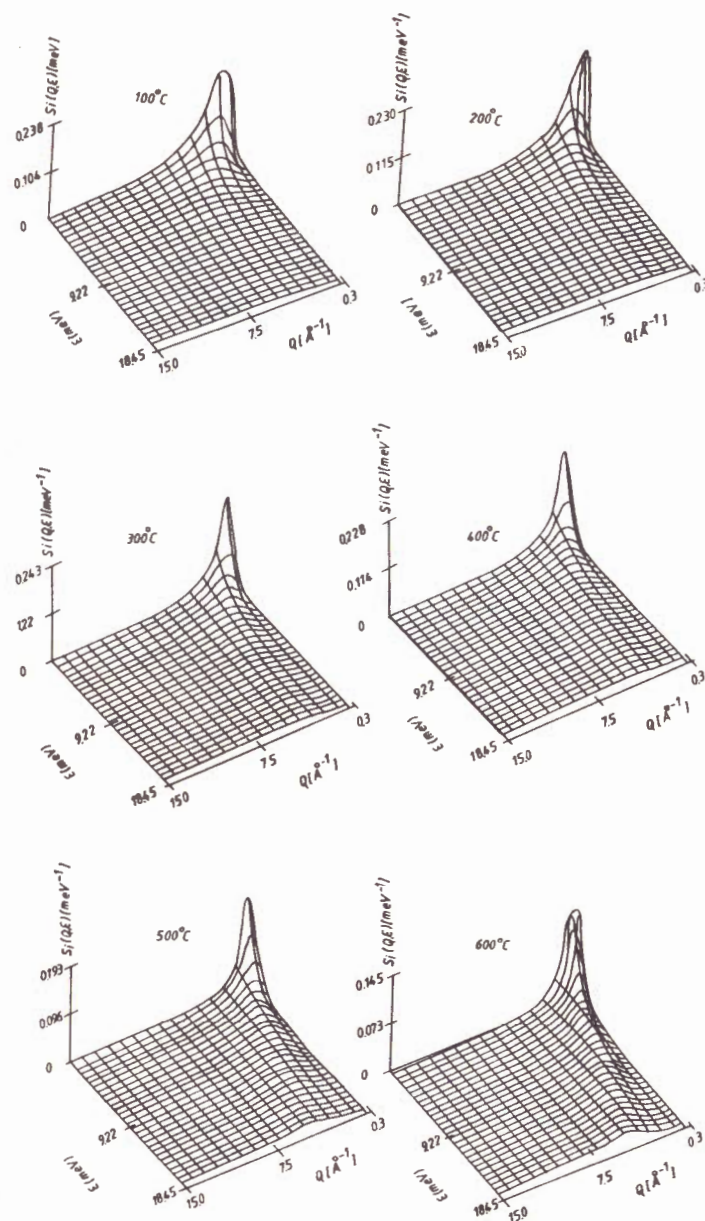


Fig. 3. Three-dimensional plot of the incoherent scattering function for liquid sodium at  $T = 100-600^\circ\text{C}$

These data allow for further calculation of the space-time correlation functions  $G_1(r,t)$  and  $G(r,t)$ .

The structural transformation observed in [18] at various temperatures cannot be explained with hard sphere or a continuous pair potential.

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Падуреану И. и др.

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Температурные зависимости коллективных  
и индивидуальных движений в жидком натрии

Получены функции  $S_c(Q, \epsilon)$  и  $S_i(Q, \epsilon)$  жидкого натрия для когерентного и некогерентного рассеяния нейтронов в области температур 100–600°C. Дисперсионные соотношения коллективных мод определены из зависимости  $\epsilon_m(Q)$ , соответствующей главному максимуму корреляционной функции продольных токов  $J_c(Q, \epsilon)$ .

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The Temperature Dependence of the Correlated  
and Individual Motions in Liquid Sodium

A full surface in the  $(Q, \epsilon)$  space for coherent and incoherent scattering functions  $S_c(Q, \epsilon)$  and  $S_i(Q, \epsilon)$  on liquid Na in the temperature range  $T = 100-600^\circ\text{C}$  is presented. The dispersion of the collective modes at various  $T$  are determined from the  $Q$  dependence of  $\epsilon_m(Q)$  corresponding to the main maximum of the longitudinal current correlation function  $J_c(Q, \epsilon)$ .

The investigation has been performed at the Laboratory of Neutron Physics, JINR.

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