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Self-diffusion in liquid potassium

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Abstract. The natural line of the incoherent quasi-elastic neutron scattering law of liquid potassium has been obtained from experimental double-differential scattering cross-sections at the temperatures of 340, 440, and 550 K for wavevector transfers in the range of $0.27 \leq Q \leq 1.13 \text{ \AA}^{-1}$. The full width at half maximum of this line as a function of Q^2 has been analysed on the basis of several diffusion models. It has been found that the self-diffusion process in liquid potassium has an individual and activationless character and can be approximately described by means of the mode-coupling theory at temperatures noticeably more than the melting point.

1. Introduction

There are numerous experiments on the inelastic neutron scattering by condensed matter which have given extensive and detailed information about structural and dynamic properties of different substances, among them liquid alkali metals: rubidium (Copley and Rowe 1974a, b, Pilgrim *et al* 1991), sodium (Rapeanu *et al* 1981, Gläser and Morkel 1984, Söderström and Dahlborg 1984, Morkel and Gläser 1986, Morkel *et al* 1987), and caesium (Bodensteiner *et al* 1990, 1992). In the absence of data for liquid potassium, we have performed an inelastic neutron scattering experiment for potassium melt at the temperatures of 340, 440, and 550 K (the melting temperature of potassium is 336.7 K). The analysis of the incoherent inelastic part of the double-differential scattering cross-section (DDSCS) of slow neutrons, the frequency spectrum of the velocity autocorrelation function, and the temperature dependence of a number of microscopic dynamic characteristics of liquid potassium were presented in our earlier work (Zaezjev *et al* 1994a).

The objectives of the present work are the analysis of the incoherent quasi-elastic component of the DDSCS and obtaining from these data information on the self-diffusion process in liquid potassium.

2. Experiment

The experiment was carried out at the DIN-2PI double time-of-flight neutron spectrometer of direct geometry (Taran 1992) set up at the IBR-2 pulsed reactor of the Joint Institute for Nuclear Research in Dubna. A more detailed description of the spectrometer, the melt sample and experimental conditions are presented by Zaezjev *et al* (1994b). Briefly, the spectrometer is placed in the horizontal plane. Neutrons from the water moderator pass by the first vacuum flight path of 18.8 m in the length to the chopper which monochromatizes them. Then, neutrons are scattered with the sample investigated and pass through the second vacuum flight path of 7 m in length towards the detector system with ^3He counters.

Two background choppers and a special system for simultaneous measurements of an effect and background ensure favourable background conditions.

The scattering cross-section of potassium includes coherent and incoherent components; the latter is much below the former: $\sigma_c = 1.73$ b, $\sigma_i = 0.25$ b (Sears 1984). Therefore, in order to observe and analyse the incoherent scattering, we should operate in the range of the neutron wavevector transfer, Q , located on the left of the static structure factor, $S(Q)$, main peak (see figure 1). For this purpose, the incident neutron energy of $E_0 = 4.0$ meV (elastic peak resolution, $\Delta E_0 = 0.16$ meV) and eight scattering angles in the interval of $11^\circ \leq \Theta \leq 48^\circ$ have been chosen, resulting in the range of $0.27 < Q_0 < 1.13 \text{ \AA}^{-1}$ for the neutron elastic scattering and $S(Q_0) \leq 0.15$. The energy transfer was -1.5 – 100 meV at every scattering angle.

The melt sample is constructed as a loop since it has been designed to be used for study of the potassium–oxygen system at a variable oxygen concentration. In the present experiment, it is filled with pure (99.9%) potassium. The part of the sample placed directly into the neutron beam is a thin-wall (0.2 mm) cylinder made from iron of 16 cm in height and 12 cm in diameter. The cylinder has been tightly nested in an aluminium jacket of 1 mm in thickness. Nevertheless, this container gives a noticeable contribution to the elastic scattering (up to 30% in the most unfavourable conditions: $T = 550$ K, $\Theta = 48^\circ$) because of very low potassium scattering cross-section. It was estimated that about 25% of incoming neutrons have been scattered by the sample.

Two heaters are located at the top and the bottom of the sample such that they are outside the neutron beam. The temperature of the sample is fixed with an accuracy of ± 1 K. In order to minimize the background of the heating system two cadmium screens towards the detectors are used.

3. Data processing

The experimental spectra have been initially corrected for the background, the beam attenuation in the sample and the container, the container scattering, and the detector efficiency. Then, they have been converted to the DDSCS form and absolutely normalized to the vanadium standard.

Further, the treatment of the experimental data has involved the extraction of the natural line of the incoherent quasi-elastic scattering law (IQESL) from the DDSCSs obtained above. To do this, the following is required.

(i) To separate the incoherent quasi-elastic part from the total DDSCS, taking into account that the both quasi-elastic and inelastic neutron scattering in potassium includes coherent and incoherent components. In so doing, it should be remembered that the separation of the DDSCS into inelastic and quasi-elastic components can be done only under the assumption that the vibrational and diffusive motions of particles in a substance are uncorrelated (Springer 1972).

(ii) To turn the incoherent quasi-elastic component of the DDSCS measured at fixed scattering angle into the IQESL at constant value of $Q = |k - k_0|$ where k and k_0 are the neutron wavevector before and after scattering respectively. Such a procedure is required for the subsequent model analysis of results.

(iii) To take into account the spectrometer resolution and to turn the ‘apparatus’ curves to the natural lines of the IQESL.

The separation of the incoherent quasi-elastic scattering component from the experimental DDSCS has been performed by the factor method which goes back to Slaggie’s

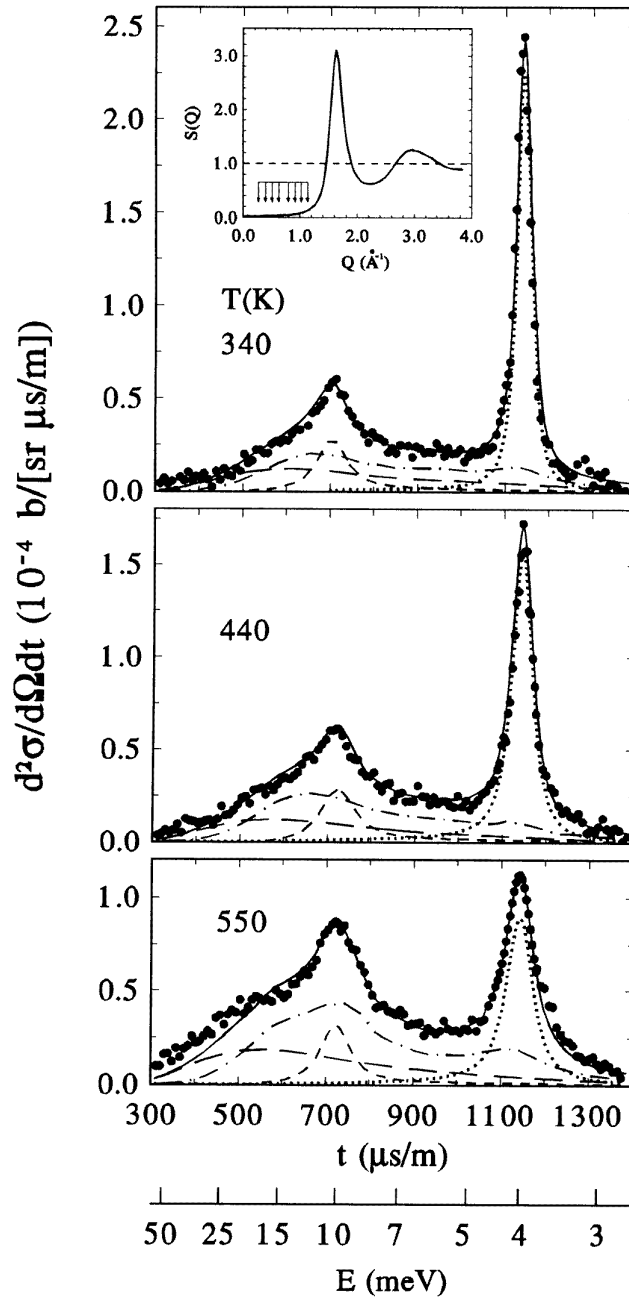


Figure 1. The DDSCS of liquid potassium $\Theta = 26^\circ$. The experimental points are described by the full curve which represents the total calculated DDSCS. The dotted curve shows the quasi-elastic scattering component. The dot-dashed, short-dashed, and long-dashed curves represent the inelastic incoherent, coherent, and multiple-scattering components respectively. The static structure factor of liquid potassium (van der Lugt and Alblas 1985) is shown in the inset. Arrows indicate Q_0 values corresponding to the elastic scattering for eight scattering angles: 11, 16, 21, 26, 33, 38, 43, and 48° .

work (1967). In the framework of this method, we suppose that the relation between the quasi-elastic scattering component and the total DDSCS must be equal in calculation and experiment. Hence, the quasi-elastic scattering component of the total experimental DDSCS is expressed as

$$(d^2\sigma/d\Omega dt)_{q.el}^{exp} = (d^2\sigma/d\Omega dt)_{tot}^{exp} \times F \quad (1)$$

where the factor

$$F = (d^2\sigma/d\Omega dt)_{q.el}^{calc} / (d^2\sigma/d\Omega dt)_{tot}^{calc} \quad (2)$$

is the ratio of the model calculated incoherent quasi-elastic DDSCS and the total DDSCS, Ω is the solid angle and t is the neutron time of flight.

The calculations have been carried out by means of a computer code (Novikov *et al* 1986). It calculates the DDSCS and its partial contributions ('one phonon', 'multiphonon', quasi-elastic) in the solid-like approximation on the basis of a certain dynamic model of a scatterer, taking into account effects of the sample dimensions (self-shielding, multiple neutron scattering) and the spectrometer resolution. The computation includes an iteration procedure which provides a desirable degree of agreement between experiment and calculation by sequential correction of the initial model. This approach is based on the work of Macdougall (1962).

In so doing, a frequency spectrum of the velocity autocorrelation function for liquid potassium obtained by molecular dynamics simulation (Gonzalez Miranda 1986) has been used to model the incoherent inelastic scattering and to evaluate a Debye–Waller factor. The component of incoherent quasi-elastic scattering has been modelled under the assumption that the natural line of the IQESL for a simple liquid has Lorentzian shape (Gurevich and Tarasov 1968). Both inelastic and quasi-elastic coherent effects have been accounted for on the basis of the viscoelastic model (Lovesey 1971) which was used for description of the coherent neutron scattering in liquid sodium (Morkel and Gläser 1986, Morkel *et al* 1987) and caesium (Bodensteiner *et al* 1992). Two parameters of this model, the radius of the pair correlation maximum, r_0 , and the Einstein frequency, ω_E , are given in table 1. The value of r_0 is estimated from the maximum of $S(Q)$. The meaning of ω_E at 340 K is evaluated from the article by Rani *et al* (1989).

Table 1. The parameters of the viscoelastic model for potassium.

T (K)	r_0 (Å)	ω_E (10^{12} s $^{-1}$)
340	3.9	8.5
440	3.9	9.4
550	3.9	9.6

Some examples of the experimental and calculated DDSCSs as well as their components calculated in the manner mentioned are shown in figure 1. One can see that the coincidence between the experiment and calculation takes place. It is also seen that the contribution of inelastic and coherent components to the common intensity of the quasi-elastic peak is rather modest. Thus, some uncertainties inevitably introduced to the modelling process of these components are unable to change remarkably the characteristics of the IQESL (amplitude, half-width and shape).

The experimental incoherent quasi-elastic DDSCS obtained by (1) and (2) is transformed to the energy scale and then into the IQESL:

$$S_{q.el}^{exp}(Q, \omega) = (d^2\sigma/d\Omega d\omega)_{q.el}^{exp} (4\pi/\sigma_i) k_0/k \quad (3)$$

where $\hbar\omega = E - E_0$ is the neutron energy transfer. Then, the values of the IQESL at different scattering angles have been converted by the interpolation procedure to the form at constant Q .

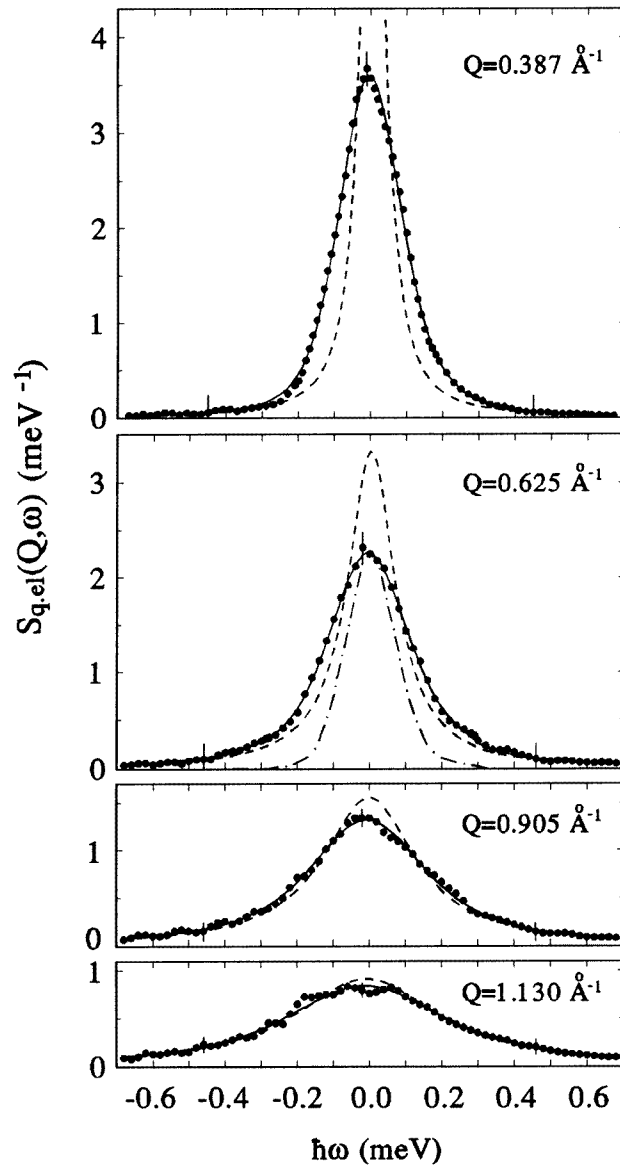


Figure 2. The IQESL for constant values of Q at $T = 340$ K. The full curves represent the optimal fitting of the experimental points by equation (4). The broken curves are the IQESL natural lines. The dot-dashed curve represents the spectrometer resolution function.

Examples of the experimental IQESL for several values of Q at 340 K are shown in figure 2. These curves are free from multiple-scattering effects but contain the resolution

ones.

To eliminate the resolution effects on the experimental IQESL, the iterative procedure based upon the least-squares method algorithm has been used. It provides the optimal description of the experimental IQESL for convolution of the model curve (Lorentzian) and the spectrometer resolution function, $R(E_0, \omega)$, measured with an appropriate vanadium sample (see figure 2):

$$S_{q.el}^{exp}(Q, \omega) = S_{q.el}^{nat}(Q, \omega) \otimes R(E_0, \omega) \quad (4)$$

where

$$S_{q.el}^{nat}(Q, \omega) = \exp(-2W)[C(Q)\Delta E(Q)/2]/([\Delta E(Q)/2]^2 + (\hbar\omega)^2). \quad (5)$$

Here, $\exp(-2W)$ is the Debye–Waller factor, $C(Q)$ is the normalizing constant, and $\Delta E(Q)$ is the full width at half maximum (FWHM) of the IQESL natural line. The experimental values of $\Delta E(Q)$ have been taken as the initial ones.

It is seen that the coincidence between the experimental and approximating curves is satisfactory. The good description of the natural line of the IQESL with a single Lorentzian seems to be evidence that coherent effects do not reveal any remarkable impact on the quasi-elastic scattering region. To confirm this assumption, the superposition of two Lorentzians corresponding to the incoherent and coherent scattering contributions has been fitted to the IQESL natural line:

$$S_{q.el}^{nat}(Q, \omega) = \exp(-2W) \times \left(\frac{\sigma_i}{\sigma_i + \sigma_c} \frac{C_i(Q)\Delta E(Q)/2}{[\Delta E(Q)/2]^2 + (\hbar\omega)^2} + \frac{\sigma_c}{\sigma_i + \sigma_c} \frac{C_c(Q)\Delta E(Q')S(Q)/2}{[\Delta E(Q')/2]^2 + (\hbar\omega)^2} \right) \quad (6)$$

with

$$Q' = Q/[S(Q)]^{1/2}. \quad (7)$$

The ‘coherent’ Lorentzian is represented here in the form proposed by Sköld (1967). The analysis of such a procedure shows that the addition of the second Lorentzian gives no improvement to the description of the experiment. The FWHM of the ‘coherent’ Lorentzian is 10–20 times greater than that of the ‘incoherent’ one, and its amplitude is less than 1–2% of the amplitude for the ‘incoherent’ Lorentzian.

4. Results and discussion

The integral intensity of the incoherent quasi-elastic scattering can be written as (Gurevich and Tarasov 1968)

$$(d\sigma/d\Omega)_{q.el} = (\sigma_i/4\pi) \exp(-2W) \quad (8)$$

with

$$2W = \overline{u^2} Q^2 \quad (9)$$

where $(\overline{u^2})^{1/2}$ is the mean-square amplitude of atomic vibrations. We have calculated the values of (8) by the numerical integration of an area under the experimental quasi-elastic DDSCS within the limits, assuming a loss of the area less than 2%. Represented in a semilogarithmic scale (see figure 3), the experimental results demonstrate approximately linear dependence on Q^2 , and enable us to estimate values of $(\overline{u^2})^{1/2}$ and σ_i . The former is shown in table 2. These amplitudes appear to be in good agreement with values calculated earlier (Zaezjev *et al* 1994a), using the frequency spectrum of the velocity autocorrelation function for the melt.

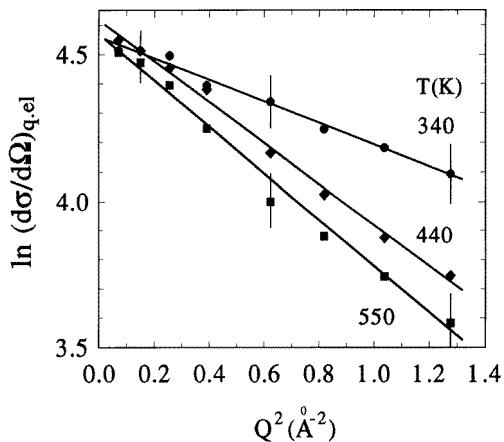


Figure 3. The integral intensity of the experimental quasi-elastic DDSCS in a semilogarithmic scale at different Q^2 values for three temperatures.

Table 2. The mean square amplitudes of potassium atomic vibrations.

T (K)	$(\overline{u^2})^{1/2}$ (Å)
340	0.60 ± 0.07
440	0.84 ± 0.05
550	0.89 ± 0.05

The estimated value of σ_i is equal to 0.26 ± 0.02 b and agrees well with that of 0.25 b from Sears (1984).

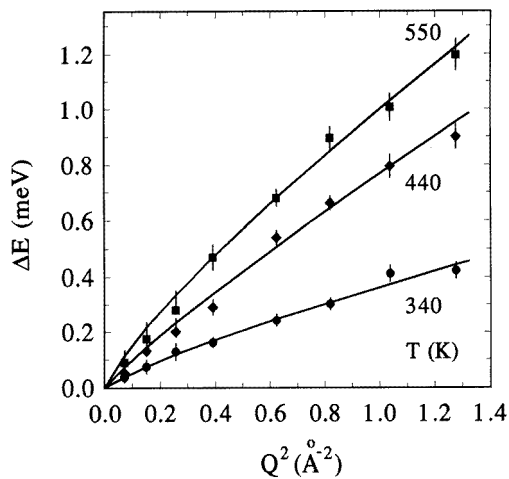


Figure 4. The FWHM of the IQESL natural line at different values of Q^2 . The full curves describe the experimental points in the context of the mixed diffusion model (Oskotskii 1963).

The FWHM of the IQESL natural line as a function of Q^2 is represented in figure 4 for three temperatures. Unfortunately, the FWHM can be determined only in the range of wavevector transfer restricted by $Q \leq 1.13 \text{ \AA}^{-1}$. The reason is a high value of the Debye–Waller factor, considerable broadening of the quasi-elastic peaks and coherent effects increasing with angle and temperature increase, so that the separation of the quasi-elastic component from the total DDSCS at $Q > 1 \text{ \AA}^{-1}$ becomes practically impossible.

Experimental data of $\Delta E(Q^2, T)$ have been analysed by two ways. The first one describes these data in the context of the simplest self-diffusion models: the jump diffusion model (Singwi and Sjolander 1960), the effective mass model (Egelstaff and Schofield 1962), and the mixed diffusion model (Oskotskii 1963). The latter presumes the self-diffusion process is the superposition of jump and continuous mechanisms. In statistical analysis, it has been deduced that preference should be given to the mixed diffusion model.

For this model at $\tau_0 \gg \tau$ the IQESL natural line has the Lorentzian form with the FWHM

$$\Delta E(Q^2) = (2\hbar/\tau_0)[1 + D_0 Q^2 \tau_0 - \exp(-2W)/[1 + (D - D_0)Q^2 \tau_0]]. \quad (10)$$

Here, τ_0 is the residence time of the atom, τ is the time of jumping, D_0 is the coefficient of continuous (collective) diffusion, and D is the total coefficient of self-diffusion. Considering D as known (Gerl and Bruson 1985), we will obtain $\tau_0(T)$ and $D_0(T)$ for three temperatures. The values of these parameters are represented in figure 5.

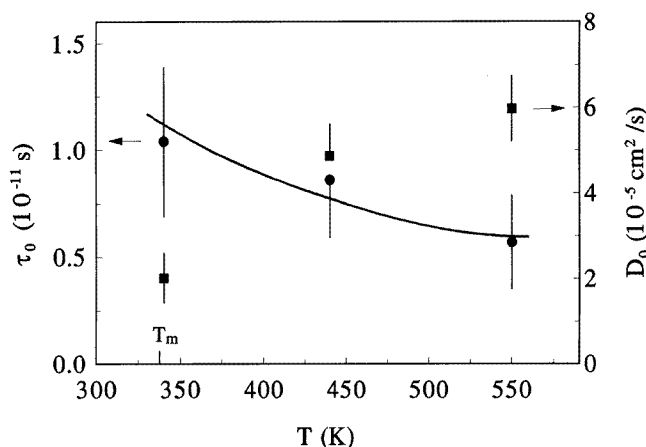


Figure 5. The temperature dependence of the residence time of the atom, τ_0 , and the coefficient of continuous diffusion, D_0 . The experimental points of τ_0 are fitted by equation (11) (Frenkel 1955).

Although the description of the experimental FWHM by equation (10) is satisfactory, there are some reasons to apply this model for liquid potassium with precaution.

First, the residence time at $T = 340$ K seems to be too high: $\tau_0 = 10^{-11}$ s, whereas the estimation (Cocking 1968, Randolph 1968) of that for liquid sodium gives $\tau_0 \approx (2-5) \times 10^{-12}$ s at the melting point and just above. If we suppose the jump diffusion is an activation process (Frenkel 1955)

$$\tau_0 = \tau'_0 \exp(E_{jp}(T)/k_B T) \quad (11)$$

where τ'_0 is the period of particle vibration near the temporary equilibrium centre, k_B is the Boltzmann constant, we estimate the activation energy of jumps, E_{jp} , as 49.2 meV from

the data of figure 5. At the temperature of 550 K this value is near the mean energy of atomic thermal motion of $k_B T = 47.4$ meV, so that the notion of activation process loses sense.

Second, it is known that the experimental coefficients of self-diffusion in liquid alkali metals are in agreement with the ones calculated on the basis of the hard-sphere model (Protopapas *et al* 1973, Ogloblya *et al* 1987). This points to the absence of the remarkable influence of collective effects on the self-diffusion process in these substances.

So, the individual activationless mechanism of so-called 'passive diffusion' (Frenkel 1955) plays an important role in the atomic diffusion of liquid potassium at least for temperatures noticeably higher than the melting point. Such diffusion takes place due to the vacancy character of the thermal expansion of liquid alkali metals, which proceeds with decreasing coordination number, but no considerable change of the interparticle distance (Matsuda *et al* 1991).

Therefore, we choose another way to analyse the experimental data of $\Delta E(Q, T)$, namely, its comparison with the predictions of the mode-coupling theory (MCT) (see Verkerk 1990 and references therein), which is based on the hydrodynamic approach. The fruitfulness of the MCT for the description of self-diffusion in liquid alkali metals has been convincingly demonstrated on the example of sodium by Montfrooy *et al* (1986) and by Morkel *et al* (1987). The MCT predicts deviation of $\Delta E(Q^2)$ from a straight line corresponding to simple diffusion: $\Delta E(Q^2) = 2\hbar D Q^2$, due to effects of the mode coupling. In reduced units we have (Morkel *et al* 1989)

$$\Delta E/2\hbar Q^2 = D[1 - aQ + O(Q^{3/2})]. \quad (12)$$

The maximum of the IQESL is

$$\pi\hbar Q^2 S_{q,el}(Q, 0) = (1/D)[1 + bQ + O(Q^{3/2})] \quad (13)$$

where

$$a = (D/2\omega_0)^{1/2} \quad b = (2D/\omega_0)^{1/2} \quad \omega_0 = 72(\pi\rho D/k_B T)^2(D + \nu)^3 \quad (14)$$

ρ is the density, and ν is the kinematic viscosity. The mode-coupling effects slow the self-diffusion and the simple diffusion case is the limit of that at $Q \rightarrow 0$, according to equations (12) and (13).

It has been shown (Alder and Wainwright 1970, Morkel *et al* 1989) that equations (12) and (13) are valid only for the limited wavevector transfer range of $0 \leq Q \leq Q_L$ where

$$Q_L = (10D\tau_c)^{-1/2} \quad (15)$$

and τ_c is the atomic collision time in the liquid. The evaluation of Q_L for potassium at $T = 340$ K gives $Q_L \approx 0.6 \text{ \AA}^{-1}$.

Our experimental data of $S_{q,el}(Q, 0)$ in reduced form (the left-hand part of equation (13)) for three temperatures are presented in figure 6. The straight lines of the right part of equation (13) are obtained with tabular values of ρ , ν (Shpil'rain *et al* 1985a, b), and D (Gerl and Bruson 1985). The shaded areas correspond to the self-diffusion coefficient uncertainty. As seen for 440 and 550 K, our data within the experimental errors are close to the MCT predictions in the Q range of our experiment ($0.27 \leq Q \leq 1.13 \text{ \AA}^{-1}$). They are also in agreement with the data of slow-neutron scattering on liquid sodium by Montfrooy *et al* (1986) and by Morkel *et al* (1987) who concluded that the MCT properly describes the IQESL for $0 < Q < 1.5 \text{ \AA}^{-1}$. The comparison of the MCT effects in liquid alkali metals is shown in figure 7. One can see that the deviation from Fick's law in potassium is somewhat more pronounced than that in sodium for equal reduced temperatures. It should be expected that the strongest and weakest mode coupling are in caesium and lithium respectively.

This is the result of particular combinations of thermo-physical properties of these liquids reflected by the values of a and b in equations (12) and (13).

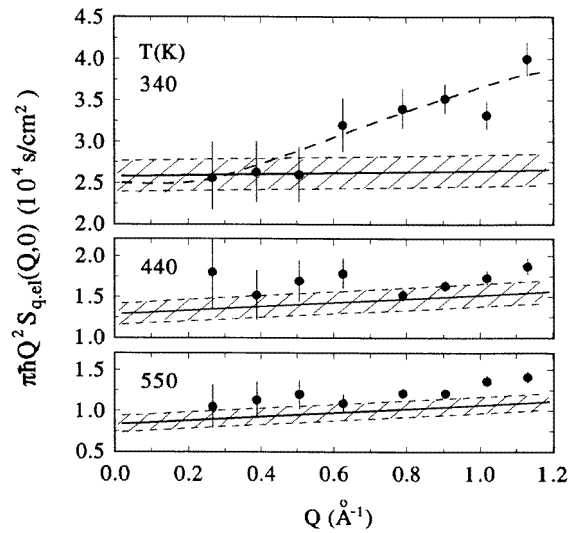


Figure 6. The quantity $\pi\hbar Q^2 S_{q,el}(Q, 0)$ at different Q values. Points with error bars represent the experiment. The straight lines take the form of the right-hand part of equation (13) with tabular values of ρ , ν (Shpil'rain *et al* 1985a, b), and D (Gerl and Bruson 1985). The shaded areas correspond to the uncertainties given for the self-diffusion coefficient. The broken curve is obtained from the mixed diffusion model (Oskotskii 1963).

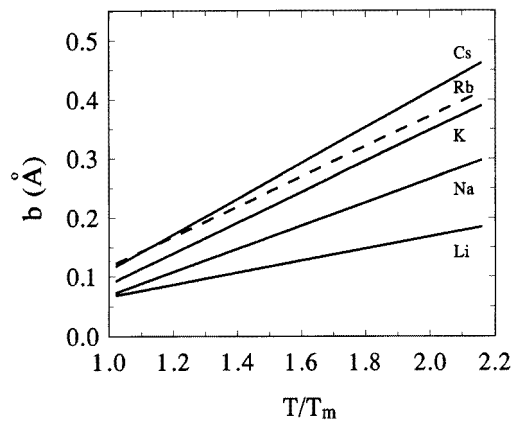


Figure 7. Estimations of the coefficient b in equation (13) for liquid alkali metals as a function of reduced temperature. The parameters of ρ , ν , and D are taken from Shpil'rain *et al* (1985a, b) and Gerl and Bruson (1985) respectively.

At 340 K, the coincidence of the experiment and the MCT calculation is achieved only at small Q . This should be expected because the mode-coupling effects are strongly diminished due to small values of a and b (equations (12)–(14)) (Bosse *et al* 1979, Morkel

et al 1987) for the temperature in the vicinity of the melting point (the lowest temperature of the given experiment corresponds to $T/T_m = 1.01$). At the same time, the mixed diffusion model (see the broken curve in figure 6) fits the experimental data rather satisfactory at this temperature. It can be supposed that the solid-like features in melt self-diffusion near the melting point are responsible for the deviation of the $\Delta E(Q^2)$ function from the straight line corresponding to the simple diffusion.

5. Conclusion

One can conclude that the self-diffusion in liquid potassium at temperatures of 340–550 K is of a complex character. It has an individual and activationless mechanism and can be approximately described by the mode-coupling theory for temperatures relatively far from the melting point. Being applied to reliable experimental data, such a model analysis enables us to determine simultaneously a self-diffusion coefficient and a kinematic viscosity, as was done for liquid sodium by Morkel and Gronemeyer (1988). The solid-like features of the self-diffusion in liquid potassium appear near the melting point, that explains the character of the $\Delta E(Q^2)$ function for 340 K.

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