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Diffusion flow of ^{3}He bcc crystals

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As known, the vacancy mechanism of mass transfer in solids is one of the most fundamental ones. For classical crystals, vacancies play an important role at high enough temperatures only, when diffusion is rather intensive. In quantum crystals, according to Andreev-Lifshitz theory [1], vacancies are tunnelling which results in a high vacancy mobility and, hence, in a high pliability of the crystal. Unfortunately, vacancies are spinless and there are no reliable methods to study the vacancy diffusion (such as NMR).

Recently, we have developed a new method suitable for the investigation of vacancy diffusion by measuring the displacement of a thin membrane under constant force. The main advantage of the method in comparison with other ones used to measure displacements of macroscopic bodies in solids [2] is the reduced characteristic dimensions of micron scale (the membrane thickness is $\sim 10\,\mu$m, the pore diameter is near $2\,\mu$m). This allowed us to measure membrane displacements under very low loads.

The main element of the experimental cell described in Ref. 3, is a planar capacitor filled with solid helium. The capacitor consists of an aluminized porous lavsan membrane and a massive metallic electrode. The force acting on the membrane is supplied by dc voltage. The membrane displacement is registered by measuring the capacity of the device.

In this work we studied solid ^{3}He at molar volumes 24.2–24.8 cm^3/mole. The results obtained are published elsewhere [4]. The self-diffusion coefficient for ^{4}He atom was previously [5] measured at different molar volumes of solid ^{4}He. Based on these data we carried out an analysis of the applicability of different theoretical models for vacancy motion, taking also in account heat capacity, lattice parameter, and dP/dT data. As was demonstrated, all experimental data (except the results of x-ray experiments) can be described self-consistently in the frame of the model of wide-band vacancions with the mobility limited by one-phonon processes.

For a more detailed study of the role of vacancies in mass transfer, experiments with crystalline ^{3}He seem to be very important because diffusion processes in ^{3}He have an additional facet due to the nuclear spins available.

The disorder of ^{3}He spins at $T > 1$ mK is known to narrow the vacancion band gap [6] and to result in the formation of distinctive vacancion polarons at low temperatures [7]. On the other hand, the presence of the spin can be employed to perform NMR spin diffusion measurements. The first evidence of the role of vacancies in solid ^{3}He was obtained by Reich as far back as in 1963 [8]. However, the strong exchange interaction, which is the main factor contributing to spin diffusion, just in the case of large molar volumes, which are most promising for quantum effect observation, thwarts any attempt to ascertain the role of vacancies in this case. Our newly developed method is free of this shortcoming, and the possibility of comparing the results of diffusion measurements with NMR data of Thompson and collaborators [9] and Grigor'ev and collaborators [10] offers an extra stimulus to do investigations in ^{3}He.

The experiments were carried out in the temperature range 0.48 to 0.70 K. The upper limit of this range is due to melting, while the lower one is connected with the resolution of velocity measurements, which was $\pm 2\cdot 10^{-10}$ cm/s.

A characteristic indicator of diffusion decay is a linear dependence of the strain rate on the stress. Our primary concern, therefore, was to investigate the steady-state velocity of the membrane as a function of the applied pressure. The dependence was found to be linear up to pressures of $(6–7)\cdot 10^3$ dyn/cm^2. In contrast, the corresponding critical stress for fcc ^{4}He is ~2.5 times as high. The deviations from linearity at large stresses are associated with the appreciable contribution of dislocation creep. As in the series of experiments with ^{4}He, we detected the existence of a threshold stress $P_0 = 1.8\cdot 10^3$ dyn/cm^2, below which mass transfer is not observed. Note the agreement of the P_0 value with the pressure required for the crystal to grow inside the pores of the membrane. All the

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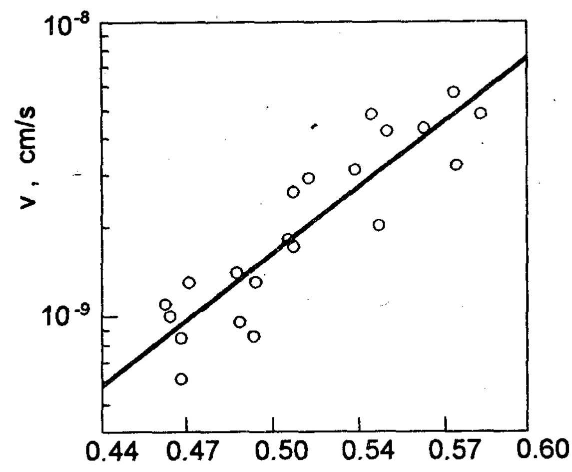

Fig. 1. Velocity of the membrane vs temperature for a molar volume of 24.60 cm^3/mole.

subsequent measurements were performed at pressures below $7 \cdot 10^3$ dyn/cm^2.

The experimentally determined membrane velocities as a function of the temperature for samples having molar volumes 24.60 cm^3/mole are shown in Fig. 1, which exhibits an abrupt increase in the membrane velocity as the temperature rises. Figure 2 shows the self-diffusion coefficient as a function of the inverse temperature, as evaluated from [11]

$$D_s = \frac{vLkT}{(P - P_0)\Omega H} \ . \qquad (1)$$

Here v is the velocity of the membrane; L is the thickness of the membrane; H is the porosity; Ω is the atomic volume; k is the Boltzmann constant; and P is the stress applied to the crystal. The self-diffusion coefficient for atoms is related to the vacancy diffusion coefficient D_v by the obvious relation:

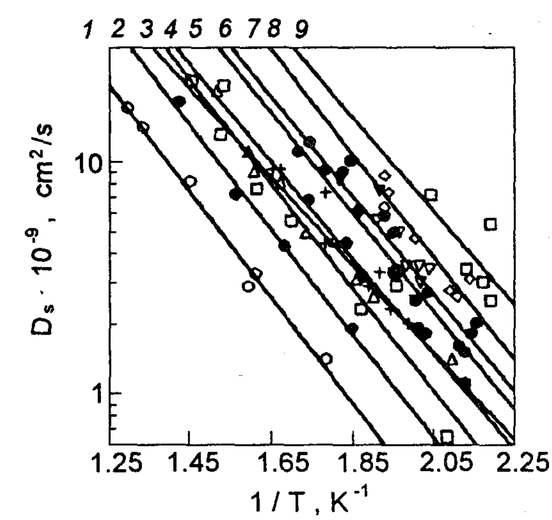

Fig. 2. Self-diffusion coefficient of ^{3}He atoms vs reciprocal temperature for molar volumes, cm^3/mole: 24.20 (*1*), 24.32 (*2*), 24.40 (*3*), 24.47 (*4*), 24.53 (*5*), 24.60 (*6*), 24.64 (*7*), 24.72 (*8*), 24.80 (*9*).

$$D_s = X_v D_v \ , \qquad (2)$$

where X_v is the number density of vacancies, which must be known before the behavior of D_v can be determined.

As it was noted previously, the x-ray measurements fall out of the self-consistent picture of known experimental data. For this reason, in the vacancy diffusion calculations we used our data on the self-diffusion coefficient for ^{3}He atoms and the data on the vacancy concentration obtained from heat capacity measurements. If we use the standard parabolic dispersion law for wide-gap vacancions, the vacancion-related heat capacity is described by

$$C_v = R\left[\left(\frac{\Phi}{T}\right)^2 + \frac{3\Phi}{T} + \frac{15}{4}\right]X_v \ , \qquad (3)$$

where R is the universal gas constant; the vacancy density,

$$X_v = 4\left(\frac{T}{\pi\Delta_v}\right)^{3/2} \exp\left(-\frac{\Phi}{T}\right) \ , \qquad (4)$$

contains two fitting parameters. Equation (3) has been used to interpolate Greywall's vacancy-related heat capacity data for our molar volumes according to the formulas of Ref. 12. The parameters Δ_v and Φ obtained in such a way are given in Table 1.

As is usual in the processing of exponential functions, the error in the determination of the coefficients is much higher than that for the exponent. As a result, Δ_v is determined only to within a factor of ~1.5–2. The quantity Δ_v can be determined more

Table 1

Parameters Φ and Δ_v for various molar volumes of solid ^{3}He

V, cm^3/mole	Φ, K	Δ_v, K
24,20	4.26	3.23
24.32	4.07	3.36
24.40	3.96	3.45
24.47	3.85	3.59
24.53	3.78	3.61
24.60	3.69	3.68
24.64	3.65	3,69
24,72	3.58	3.72
24.80	3.47	3.89

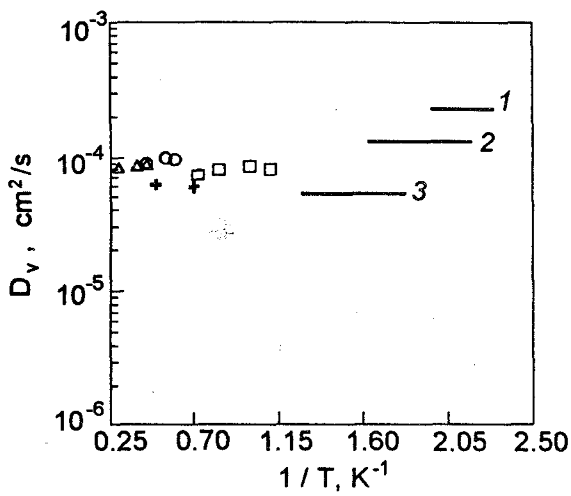

Fig. 3. Vacancy diffusion coefficient vs reciprocal temperature for molar volumes, cm^3/mole: 24.80 (*1*), 24.60 (*2*), 24.20 (*3*). Data of Ref. 8 for molar volumes, cm^3/mole: 19.32 (△); 20.12 (○); 20.10 (+); 22, 48 (□).

reliably from the vacancion diffusion coefficient, which can be expressed on the basis of the accepted model in terms of experimental data on D_s and C_v and the reliably determined parameter Φ.

The following equation is obtained directly from Eqs. (2) and (3):

$$D_v = \frac{D_s R}{C_v}\left[\left(\frac{\Phi}{T}\right)^2 + \frac{3\Phi}{T} + \frac{15}{4}\right] . \qquad (5)$$

Equation (5) and the experimental values of D_s and C_v have been used to find the vacancion diffusion coefficients for the nine investigated molar volumes.

The resulting temperature dependence of D_v for our three molar volumes 24.20; 24.60 and 24.80 cm^3/mole is shown in Fig. 3. One can see that D_v is virtually temperature independent for all the samples and that this can be regarded as evidence of the tunneling motion of vacancies in solid ^{3}He.

In conclusion, our analysis of known data on vacancy diffusion gives us ground to conclude that vacancies in solid helium are wide-gap quasi-particles. The investigation of diffusion at lower temperatures might provide clearer manifestations of the quantum nature of vacancy motion.

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