Journal of Low Temperature Physics, Vol. 110, Nos. 1/2, 1998

Self-Diffusion in ^{3}He crystals

N.V. Zuev, V.V. Boiko, N.E. Dyumin, and V.N. Grigor'ev

B. Verkin Institute for Low Temperature Physics and Engineering, National Academy of Sciences, 310164, Kharkov, Ukraine

Measurements of self-diffusion in bcc ^{3}He 24,20-24,80 cm^3/mole were carried out by a new method in the temperature range 0.4 - 0.8 K. The vacancy diffusion coefficient was obtained by comparison of the self-diffusion data and the vacancy specific heat. It is found out that the vacancy diffusion is independent of temperature, because of spin disorder in this region. The data obtained shows that vacancies in bcc ^{3}He are wide-gap quasiparticles.

PACS numbers: 67.60 Fp, 64.70 Ja.

1. INTRODUCTION

Diffusion phenomena in solid helium belong to the field of physical studies whose development was determined by the ideas formulated by I. M. Lifshitz and A. F. Andreev. The publication in 1969 of the article "Quantum Theory of Defects in Crystals" by Andreev and Liftshitz[1] changed radically the existing concepts concerning the diffusion process and determined the direction of research work in the field of quantum crystal physics for many years. Solid helium was always regarded as a typical representative of this class of crystals, but prior to the publication by Andreev and Lifshitz[1] no manifestations of quantum effects in its macroscopic properties had been predicted. In Ref. 1, several such effects (quantum diffusion, zero-point vacancies, and the possibility of superfluidity of crystals) were predicted simultaneously, which stimulated experimental and theoretical investigations in solid helium.

The main peculiarities of quantum diffusion have been observed experimentally after a comparatively short time. In accordance with the predictions of Andreev and Lifshitz, it was found that ^{3}He impurities in hcp crystals of ^{4}He are narrow-band quasiparticles whose motion is determined

0022-2291/98/0100-0103$15.00/0

by gas laws. It has been calculated theoretically that vacancies in solid helium must be wide-band quasiparticles, and it is reasonable to expect strong quantum phenomena in the diffusion of vacancies.

Experimental confirmation of this conjecture is long overdue, owing to the lack of direct methods for investigating of the motion of vacancies. We have recently implemented a method for the investigation of mass transfer in solid helium by measuring the motion of a porous membrane under the influence of small forces not exceeding the yield point of helium crystals. Such measurements yield direct information about vacancy mobility. This procedure has been used in previous work to gather data on the diffusion of vacancies in solid ^{4}He in the temperature interval 1.3 - 1.7 K in hcp and bcc phases near the melting curve.[2,3] The results, combined with data on other properties attributable to vacancies, have been interpreted self-consistently within the framework of the wide-gap vacancy model.

Diffusion processes in solid ^{3}He have several other unique characteristics associated with the presence of the nuclear spin. The disorder of the ^{3}He spins at $T > 1$ mK is known to narrow the vacancy band gap[4] and to result in the formation of distinctive vacancion polarons at low temperatures.[5] On the other hand, the presence of the spin can be exploited to perform NMR spin diffusion measurements. The first evidence of the role of vacancies in solid ^{3}He was obtained by Reich back in 1963.[6] However, the strong exchange interaction, is the main factor contributing to spin diffusion precisely in the case of interesting large molar volumes, and it throws any attempt to ascertain the role of vacancies in this case. Our newly developed method is free of this shortcoming, and the opportunity to compare the results of diffusion measurements with NMR data in ^{3}He is particularly stimulating.

2. EXPERIMENTAL TECHNIQUE

The cell used by us for studying the velocity of helium flow is described in detail in Ref. 2. The basic element of the cell is a paralell-plate measuring capacitor consisting of a flexible membrane and a massive stationary electrode. The membrane is a stretched porous film made of aluminized terylene (the film thickness is 10 μm, the pore diameter - 2 μm, and the membrane porosity - 18%), which can move in solid helium under the action of the force emerging between capacitor plates connected to a dc source. The gap between the capacitor plates under zero voltage is - 16 μm. The cell is also equipped with a capacitive pickup which makes it possible to measure the pressure in the sample with a resolution of 1.6 atm/pF.

The cell is fixed to the low-temperature part of an ^{3}He refrigerator, which uses adsorption pump. The temperature in the cell was determined from the vapor pressure over liquid ^{3}He in the evaropation chamber by taking into account the small difference in temperatures between the cell and the chamber, which are measured in a separate experiment with a partial filling of the cell with solid ^{3}He. The crystallization pressure in the cell and the vapor pressure in the evaropation chamber were measured simultaneously. The investigated ^{3}He crystals, with molar volumes of 24.2 - 24.8 cm^3/mole were grown at practically constant volume. The experiments were made in the temperature interval 0.7 - 0.45 K, bounded by the melting point of the sàmple from above and by the resolution in the measurement of the flow velocity (which was equal to $\pm 2 \cdot 10^{-10}$ cm/s) from below.

3. RESULTS AND ANALYSIS

The data on flow velocity were obtained from capacitive measurements of the displacement of the membrane in solid helium under the action of small forces during about 60 min at each temperature point. It was found that the velocity depends linearly stress up to values of P $\simeq 8 \cdot 10^3$ dyne/cm^2, which is typical of the vacancy mechanism for mass transfer. At higher stress, the velocity increases more rapidly, indicating a contribution from dislocations in the mass transfer. In addition, the experiments reveal a stress threshold $P_0 \simeq 1.8 \cdot 10^3$ dyne/cm^2, below which no displacement was observed. This threshold can be associated with the possible emergence of crystal boundary in the membrane pores, since the crystallization in pores during crystal growth must occur after the crystal has filled the entire cell volume. This is due to nonwetting of pore walls by solid helium, the crystallization in a pore of radius r requires an excess pressure $\Delta P \simeq 2\sigma/r$, where σ is the surface tension. It should be noted that the obtained value of the stress threshold coincides with the excess pressure if the value of ΔP is calculated by using the experimental value of $\sigma = 0.060 \pm 0.011$ erg/cm^2 for ^{3}He.[3] A similar situation also takes place for ^{4}He crystals.[4]

The temperature dependence of the flow velocity was measured mainly in the region of linear dependence of the velocity on stress at $7 \cdot 10^3$ dyne/cm^2. The velocity decreases sharply with temperature, indicating that the mass is transferred though the membrane diffusely by thermally activated vacancies. Proceeding from this assumption, we determined the atomic self-diffusion coefficient D_s which is connected with the velocity v_m of the membrane through the following relation:[3]

$$D_s = v_m LkT/[(P - P_0)\Omega H], \qquad (1)$$

where L is the thickness of the membrane, H its porosity, Ω the atomic volume, T is the temperature, and k is Boltzmann's constant. The values of

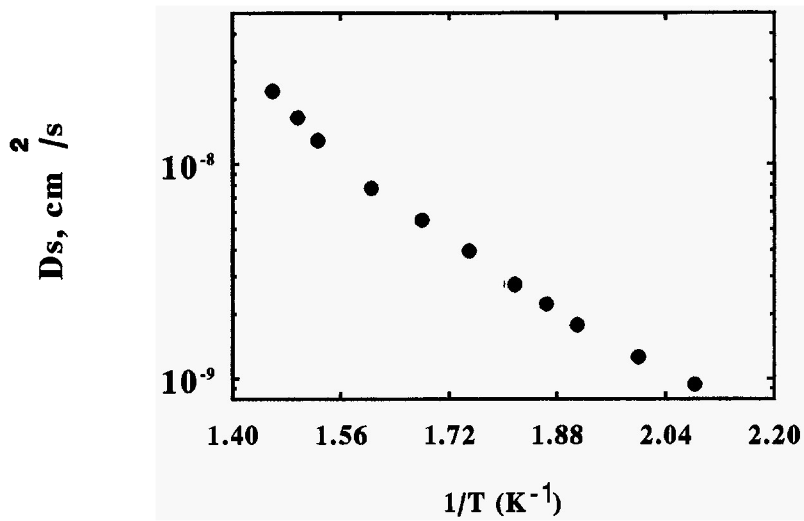

Fig. 1. Self-diffusion coefficient of helium atoms as a function of the inverse temperature for a molar volume of 24.40 cm^3/mole.

D_s determined by formula (1) are presented in Fig.1 as a function of inverse temperature for a molar volume of 24.40 cm^3/mole.

It can be seen that in these coordinates the Arhenius law is not observed: there is not only one mechanism of mass transfer indicating a nonclassical mechanism of vacancy diffusion in the ^3He crystals under investigation. In order to determine the diffusion mechanism, we must know the vacancy diffusion coefficient D_{vac} and its temperature dependence because this dependence is completely determined by the type of vacancy motion. For example, in the case of the above-the-barrier motion of vacancies, the diffusion coefficient must increase exponentially with temperature, while in the case of tunneling the value of D_{vac} must be independent of temperature in ^3He crystals.[2]

Using the obtained values of D_s we can determine the vacancy diffusion coefficient from the following relation:

$$D_s = D_{vac} x_{vac}, \tag{2}$$

where x_{vac} is the concentration of vacancies.

In this case, it was found that D_v does not depend on temperature, which can be regarded as an evidence of tunnel motion of vacancies. This conclusion is also confirmed by the results of similar processing of the data on spin diffusion obtained by Reich.[6] A comparison of these data with the results of calculations made by Landesman[7] made it possible to obtain much more reliable values of the vacancion band width which also turned out to be larger than the temperature. The obtained results are presented in Table 1.

Table I

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

An interesting problem concerning ^{3}He is vacancion polarons which were first considered by Andreev.[5] In this case, the band motion of vacancies is impossible in view of the spin disorder, and the formation of a region with ferromagnetic spin ordering around a vacancy turns out to be energetically favorable in the case of large-band width and low temperature. The presence of such vacancion polarons affects the static and kinetic properties of solid helium. For example, their effect on spin diffusion was considered by Biushvili and Tugushi[8] and on the mobility of vacancies by Iordanskii.[9] In several publications, attempts were made to observe vacancion polarons (see also Ref. 10), but no reliable evidence of such an observation has been obtained.

In conclusion, we can state that the available data on vacancy diffusion can be described self-consistently by the model of wide-band vacancions whose motion is restricted by disordered spins in ^{3}He. However, the latter statement requires further experimental and theoretical verification. The most important experiments will be those in ^{3}He at $T < 0.3K$, in which we can expect a manifestation of the effect of vacancion polarons predicted by Andreev.

ACKNOWLEDGMENTS

This research is supported, in part, by CRDF grant N UP1-297 Prop.1531

REFERENCES

1. A.F. Andreev, I.M. Lifshitz, *Zh. Eksp. Teor. Fiz.* **56**, 2056 (1969).
2. N.E. Dyumin, N. V. Zuev and V.N. Grigor'ev, *Sov. J. Low Temp. Phys.* **19**, 23 (1993).
3. E.Rolley at al. *Europhysics Letters.* **8**, 523 (1989).
4. W.F. Brinkman and T.M.Rice, *Phys. Rev.* **B 2**, 1324 (1970).
5. A.F. Andreev, *JETP Lett.* **24**, 564 (1976).
6. H.A. Reich, *Phys. Rew.* **129**, 630 (1963).
7. A. Landesman, *JLTP* **17**, 365 (1974).
8. L.L. Buishvili, and A.I. Tugushi, *JETP lett.* **24**, 564 (1976)
9. S.V. Iordanskii, *JETP Lett.* **26**, 171 (1977)
10. M.E.R. Bernier, and E.Suaudeau, *Phys. Rev.* **B38**, 784 (1988)