

Mobility of negative charges on the melting curve in bcc ^3He

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(Submitted 22 October 1992; resubmitted 6 November 1992)

Pis'ma Zh. Eksp. Teor. Fiz. **56**, No. 11, 595–599 (10 December 1992)

The mobility of negative charges on the melting curve in bcc ^3He crystals has been measured in the pressure interval $P_m = 60\text{--}29.5$ atm for the first time. The mobility is constant above 36 atm and falls off sharply below it. The decrease in the mobility at low pressures may indicate that the concentration of vacancies decreases exponentially with decreasing temperature on the melting curve near the minimum on the P - T diagram.

An electron in liquid or solid helium localizes in a cavity with a radius on the order of 10 \AA . The dimensions, structure, and diffusion mechanism of this charged cavity depend on the pressure, the structure of the crystal lattice, and other factors (see the review by Dahm,¹ the papers cited in that review, and also Ref. 2).

According to measurements¹ of the mobility of charges in samples of constant molar volume, the characteristic activation energies (ϵ_-) for the diffusion of negative charges in bcc ^3He at pressures of 40–60 atm are approximately equal to the activation energies (ϵ_3) for the self-diffusion of ^3He atoms (the latter energies have been measured in NMR experiments^{3,4}); they are also approximately equal to the vacancy formation energies ϵ_v , according to x-ray⁵ and heat-capacity⁶ measurements. It can thus be concluded that the diffusion of negative charges, like the diffusion of the host atoms, in bcc ^3He crystals at pressures below 60 atm is controlled by vacancy mechanisms. In other words, we can work from the behavior of the mobility of the charges to draw conclusions about the properties of the charged defects themselves and also the vacancies which are interacting with them in quantum crystals.

The results of various studies^{3–5,7} have shown that the concentration of thermal vacancies on the melting curve, $x_v(T_m)$, remains essentially constant, at about 0.5%, at pressures above 45 atm ($T_m \geq 1.2$ K) in bcc ^3He . This situation is also characteristic of ordinary classical crystals. At low pressures, however, estimates of x_v based on the data of different experiments contradict each other. For example, according to x-ray measurements,⁵ the vacancy concentration on the melting curve is constant at $x_v(T_m) = 0.5\%$ up to the highest molar volumes. Precise measurements of the vacancy component of the temperature dependence of the pressure⁷ indicate that x_v decreases from 0.04% at $T_m = 0.7$ K to 0.004% at $T_m = 0.37$ K. According to estimates based on NMR experiments,⁸ the vacancy concentration is 3.5% at $T_m = 1$ K and increases to 19% at $T_m = 0.3$ K. These estimates of x_v near P_{min} differ by more than three orders of magnitude.

It thus seemed worthwhile to carry on an independent experiment to learn about the properties of the charges and vacancies at low pressures. The literature reveals

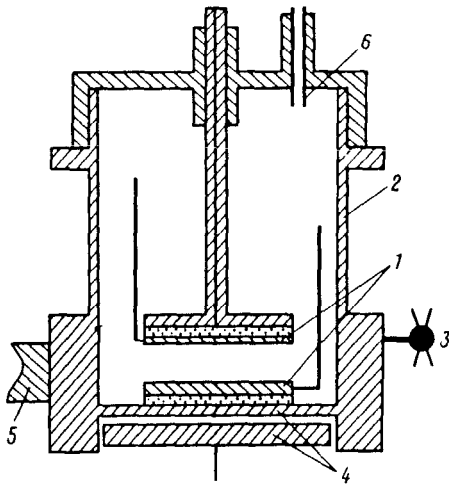


FIG. 1. The test cell. 1—Diode consisting of a radioactive source of charges (the upper electrode) and a metal collector (the lower electrode), both insulated from ground; 2—ampoule; 3—carbon thermometer; 4—capacitive pressure gauge; 5—copper cold line; 6—capillary for filling.

nothing at all about the behavior of the mobility of charges in ${}^3\text{He}$ at pressures below 40 atm.

Experimental procedure. The mobility of the charges was measured by a time-of-flight method. We used the transient characteristics of a diode with a radioactive source of charges, frozen in solid helium. The time of arrival of the charged-particle front at the collector during the stepped application of a voltage corresponds to a peak in the current on the transient characteristic of the diode. The source-collector gap was 2 mm and the range of working fields was 2×10^2 – 10^4 V/cm.

Planar diode 1 is inside a metal ampoule 2 which is 14 mm in diameter and 16 mm long (Fig. 1). The sample temperature is determined from the readings of carbon resistance thermometers 3, which are attached to the outer surface of the ampoule. The pressure in the interior of the ampoule is monitored by a capacitive pressure gauge 4 of the type described in Ref. 9. The sensitive element of the pressure gauge is rigidly attached to the collector, so that changes in the pressure directly in the volume between electrodes can be monitored. The ampoule is cooled by means of a copper cold line 5 running from a container holding liquid ${}^3\text{He}$. The filling capillary 6 connects the ampoule with an external high-pressure system. A thermal bridge (not shown in this figure) makes it possible to monitor the capillary temperature and, where necessary, to block the ampoule with a compressed liquid.

The procedure used to prepare the samples is slightly different from the usual procedures. Since the early studies by Shal'nikov,¹⁰ most experiments on charges in liquid helium have used samples grown from a compressed liquid at a constant pressure. That method is convenient for studying the temperature dependence of the mobility in samples of constant molar volume, $V_{\text{mol}} = \text{const}$. We were interested in the temperature dependence of the mobility on the melting curve, i.e., in measurements at a variable pressure P_m and a variable molar volume V_{mol} . Attempts to use the familiar blocked-capillary method failed. A pronounced trapping of charges by defects was observed in crystals grown at a variable pressure, starting with the first measurements.

An intermediate method proved most successful. The crystal was initially grown at a constant pressure, and the volume between the electrodes was gradually filled. The process was monitored by measuring the collector current at a fixed applied voltage. After the working gap was filled, the temperature of the capillary connecting the ampoule to the external high-pressure system was rapidly lowered and the ampoule was blocked. When the ampoule temperature changed, the volume ratio of the liquid and solid phases then changed, and the pressure at the solid-liquid interface changed correspondingly. Our method differs from the blocked-capillary method in that measurements can be carried out on a sample with a variable molar volume, beginning with fairly high-quality initial samples.

Ideally, the entire working pressure range would have been spanned with two or three samples. However, it turned out that the quality of a crystal suffered significant degradation as the pressure in the ampoule was varied. Because of a trapping of charges by defects (presumably dislocation walls), the transient characteristics of the diode became greatly distorted in comparison with the original characteristics. The appearance of defects was a consequence of a relaxation of a nonuniform stress in the sample. Even when the ampoule was cooled at a rate of 50 mK/h, the pressure interval which could be spanned with one sample was less than 6 atm.

The working temperature interval in this experiment was $T_m = 1.6\text{--}0.4$ K ($P_m = 60\text{--}29.5$ atm). Measurements were carried out on 20 samples with various initial pressures in order to minimize the effects of nonuniform stress. One indication of the reliability of the results is that the P - T diagrams constructed from the measured pressure and temperature during the course of the experiment differed from the equilibrium P - T diagram¹¹ by less than 20 mK. Another indication of the reliability is the circumstance that the mobilities measured on crystals with different initial solidification pressures conformed to a single curve in the region of overlap; i.e., they corresponded to samples of the same molar volume.

Results and discussion. Figure 2 shows the mobility of the negative charges as a function of the temperature on the melting curve in bcc ³He. Over the temperature interval 0.8–1.6 K (or over the pressure interval 35–60 atm) the mobility of the negative charges, μ_- , remains essentially constant at about 10^{-4} cm²/(V·s). Below 0.8 K it falls off sharply. The decrease amounts to two orders of magnitude over the interval $T_m = 0.8\text{--}0.4$ K. Assuming that this decrease is approximately exponential, we find an activation energy of 6 ± 1 K for the process from the slope of the $\mu_-(1/T_m)$ line.

How do we explain this temperature dependence? We first consider the plateau, i.e., the part of the $\mu(T_m)$ curve above 0.9 K. Under the assumption of a vacancy mechanism for the diffusion, the diffusion coefficient for particles of atomic sizes is written as $D_i = A_i x_v D_v$, where x_v and D_v are the concentration and diffusion coefficient of the vacancies, and the factor A_i depends on the particular mechanism by which the vacancy interacts with the particle. We mentioned above that the vacancy concentration $x_v(T_m)$ is essentially constant in the plateau according to the data of various experiments, while the activation energies for the diffusion of matrix atoms (ϵ_3) and of charges (ϵ_-) in samples of constant molar volume agree within ± 1 K with the vacancy formation energy ϵ_v . The product $A_i D_v$ thus depends only weakly on the

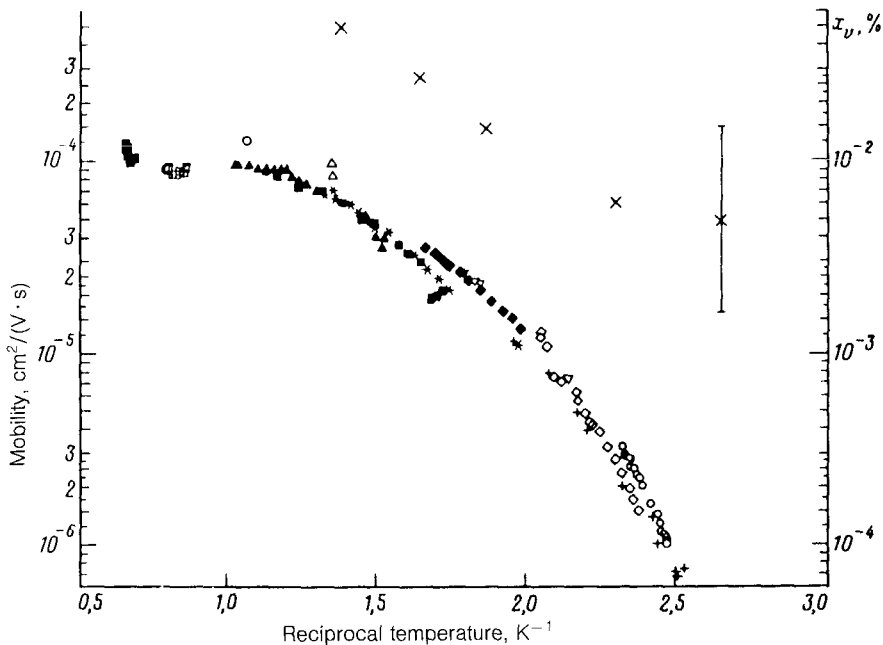


FIG. 2. Mobility of negative charges in bcc ${}^3\text{He}$ on the melting curve versus the reciprocal temperature. The symbols of different shapes correspond to samples grown at different initial pressures. The crosses show the vacancy concentration on the melting curve, in percent, according to estimates of Ref. 7.

temperature at a constant molar volume. We know from NMR measurements³ that the self-diffusion coefficient of ${}^3\text{He}$ atoms, $D_3 = A_3 x_v D_v$, also depends weakly on T_m on the melting line at pressures $P_m > 52$ atm, having a value $D_3 = 10^{-7}$ cm²/s. There is thus reason to believe that, again in the case of a varying molar volume, all three factors— A_i , x_v , and D_v —depend only weakly on the temperature on the melting line. Comparing D_3 from Ref. 3 with D_- , we find the plausible estimate $A_- \approx 0.1A_3$. The diffusion coefficient for the negative charges can be estimated from our measurements with the help of the Einstein equation $D_- = \mu kT/e$.

As we mentioned earlier, the conclusions drawn regarding the properties of vacancies in bcc ${}^3\text{He}$ on the basis of the results of different experiments are contradictory at low pressures. If we assume that at low pressures the motion of a negatively charged cavity is still controlled by the interaction with vacancies, and if we assume that the properties of the vacancies do not change substantially, i.e., that the factors A_i and D_v still depend only weakly on T_m , then we conclude that a decrease in the mobility μ_- indicates a decrease in the vacancy concentration on the melting curve near the minimum on the P - T diagram. This $x_v(T_m)$ behavior agrees qualitatively with the $x_v(T_m)$ behavior found from isochoric measurements⁷ (the crosses in Fig. 2).

The approximately exponential decrease in the number of vacancies on the melting line in ${}^3\text{He}$ at low pressures may be a consequence of particular features of the P - T

phase diagram of a quantum crystal. At relatively high pressures ($P > 40$ atm), where P_m and the molar volume of the crystal, V_{mol} , vary rapidly with the temperature, the vacancy concentration $x_v(T_m)$ remains essentially constant, as it does in ordinary crystals. In contrast with ordinary materials, however, ^3He solidifies at a pressure $P_m \approx 29$ atm at absolute zero. At low temperatures there is shallow minimum on the P - T diagram. In this region, the molar volume of the crystal changes only slightly as we move along the melting curve: For example, as T_m is halved from 1.6 to 0.8 K, the molar volume V_{mol} increases from 22.0 to 24.1 cm^3/mole , while over the interval 0.6–0.4 K it increases from 24.6 to 24.9 cm^3/mole . We know¹² that the formation energy of thermal vacancies, ϵ_v , is determined primarily by the molar volume of the sample, V_{mol} , and the vacancy concentration in a sample of constant volume ($V_{\text{mol}} = \text{const}$) falls off exponentially with decreasing temperature, $x_v \sim \exp(-\epsilon_v/T_m)$. At temperatures near the minimum, where motion along the melting line is essentially motion along an isochore, we would thus expect the concentration of thermal vacancies to fall off exponentially as T_m is lowered along the melting line.

It would be interesting to test the validity of these arguments on the melting line of hcp ^4He , in which case the molar volume again remains essentially constant below 1 K.

We wish to thank E. N. Solov'ev and A. V. Lokhov for assistance in the preparation for the experiments and in the experiments themselves.

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Translated by D. Parsons