

ment⁸ may be due to fringing fields of the magnets used, which can induce several sorts of instability. A z -pinch confinement technique¹⁰ seems to avoid these difficulties altogether by substituting a guide field B_θ for a uniform B_z . Any instabilities present are retarded by the inertia of the strong B_θ field.

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Ionic Mobilities in Solid Helium*

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Measurements of charge-carrier mobilities in solid helium have been made which are the first to be sufficiently extensive to allow conclusions to be drawn concerning the fundamental nature of the transport processes involved. Positive and negative charge carriers are quite different physical entities but in each case our data are able to provide quantitative support for a particular model to describe the carrier and its motion.

The first ionic current measurements in solid helium were reported by Shal'nikov and co-workers,¹⁻³ who measured the current-voltage (I - V) characteristics of a diode filled with solid helium. Ions of both signs were produced near a cathode coated with a low-energy β source, and the results obtained are indicated schematically in Fig. 1(a). For the plane-parallel geometry employed, the current I in the quadratic space-charge-limited region is given by⁴

$$I = 9\mu V^2 A / 32\pi L^3, \quad (1)$$

where μ is the mobility, V is the applied voltage, A is the collector area, and L is the electrode spacing. Although Shal'nikov and co-workers⁵ have also reported mobilities obtained using a time-of-flight technique, no measurements to date have been sufficiently extensive to elucidate the nature of the carriers or the mechanisms which control their motion.

To provide such data we have measured the mobilities of positive and negative carriers in the hcp phase of solid ⁴He over an extensive temperature range, viz., $1.1 < T < 3.6$ K. We measured the space-charge-limited current in a diode sim-

ilar to the one described by Shal'nikov¹ and obtained the mobility with the use of Eq. (1). The I - V characteristics are measured at each temperature, and a typical plot of $I^{1/2}$ versus V is shown in Fig. 1(b). This technique enables one to make measurements on carriers with transit times up to 10 h. Diode spacings of 0.121 and 0.458 cm are used, and crystals are grown from a copper cold finger located directly below the diode. Our criteria for a good crystal are that the steady-state current be established in a time τ_s not more than 50% greater than the transit time and that the plots of $I^{1/2}$ versus V are linear. In crystals strained by rapid cooling the mobility becomes field dependent, and τ_s is increased. Many precautions are taken, particularly in temperature regulation, to insure accurate results, and the experimental details will be presented elsewhere.

The temperature dependencies of the mobilities to within 10 mK of the melting curve are presented in Fig. 2, together with the few data points available from Ref. 4. Relative errors for a given crystal are less than the width of the symbols on the graph except where error bars are shown.

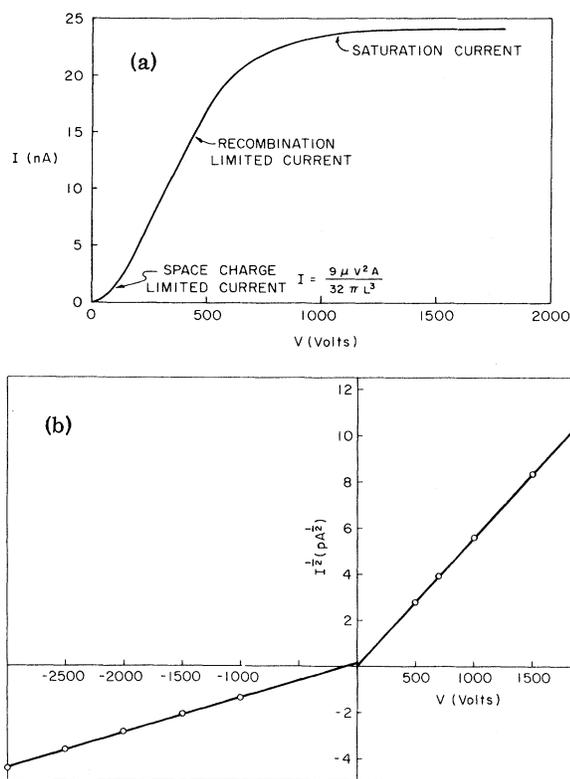


FIG. 1. (a) I - V characteristic of a diode in helium gas at 4.2 K and 1 atm pressure. (b) $I^{1/2}$ versus V in the space-charge-limited region at 3.0 K and a molar volume of 17.9 cm^3/mole .

On four good crystals grown at 20.6 cm^3/mole the variation in mobility from crystal to crystal at a given temperature was $\pm 25\%$, and our absolute errors are estimated as $\pm 50\%$. The data presented are from individual runs with a 0.121-cm diode spacing. Space-charge currents varied as L^{-3} to within our ability to reproduce values with a given diode spacing. The drastic temperature dependences displayed in Fig. 2 suggest thermally activated mechanisms for the motion of the carriers near the melting curve. In analyzing

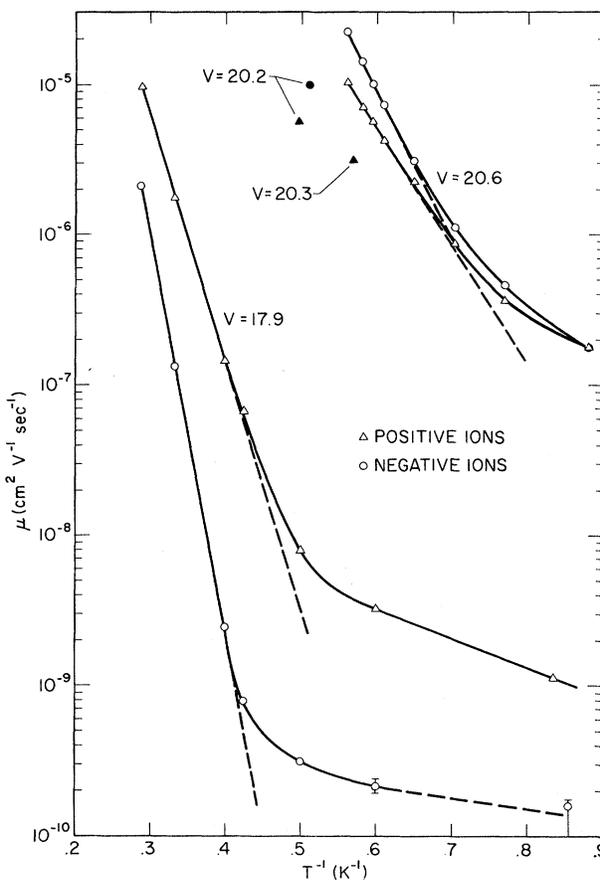


FIG. 2. Ionic mobility μ versus inverse temperature. Molar volumes in cm^3/mole . Closed symbols, data from Ref. 4.

our data we relate the mobility to the diffusion coefficient of an ion with the Einstein relation $\mu = (e/kT)D$, and assume D to obey an Arrhenius equation of the form

$$D_{\pm} = D_{0\pm} \exp(-\Delta_{\pm}/kT). \tag{2}$$

Values of Δ_{\pm} and $D_{0\pm}$ extracted from plots of $\ln(\mu T)$ versus T^{-1} are set out in Table I. We caution the reader that a temperature-dependent D_0 will yield

TABLE I. Activation energies Δ and diffusion-coefficient prefactors D_0 for ions, ^3He atoms, and adatoms in solid ^4He for two molar volumes V .

Entity	V (cm^3/mole)	Δ/k (K)	D_0 ($10^{-4} \text{ cm}^2/\text{sec}$)	D_{0s} ($\text{cm}^2/\text{sec}^{-1}$)
Negative ion	17.9	62 ± 1	340 ± 200	2.0 ± 1.1
	20.6	25 ± 2	20-140	0.15-1
Positive ion	17.9	40 ± 1	3 ± 2	...
	20.6	20 ± 2	0.4-3.8	...
^3He atom	20.6	16 ± 2	0.2-1.5	...

slightly different values, and we proceed to a discussion of the specific mechanisms involved.

We shall assume the positive carrier, in analogy with Atkins's successful model⁶ in liquid helium, to consist of an atomic or molecular ion surrounded by an enhanced density due to electrostriction. The ion is then bound to the density hump it creates with a binding energy equal to the difference between the electrostatic energy with and without the presence of the density hump. In this model the density and pressure vary approximately as the inverse fourth power of the distance from the center of the charge. If one assumes a continuum model up to atomic dimensions, then the pressure p_0 at an atomic spacing from the charge is increased by approximately 400 atm. An obvious mechanism of positive-charge transfer in the solid consists of an electron hopping from a neighboring lattice site to neutralize the charged complex at the center with a subsequent rearrangement of the lattice. However, the density dependence of the activation energy near the melting curve is inconsistent with this model.

Another promising mechanism for the motion of the positive ion is vacancy diffusion. The measured activation energy is then associated with the enthalpy $H_v(p)$ of formation of a vacancy. Guyer⁷ has suggested that the effective radius for the positive ion might be defined as the distance to which vacancies can penetrate the density hump. Alternatively, vacancies could penetrate to the center of the ion if they had an enthalpy $H_v(p_0)$. The parameters associated with the positive-ion motion may be compared with the corresponding values for a ³He atom in a solution of 1.94% ³He concentration in hcp ⁴He measured with nuclear-resonance techniques.⁸ The motion of the ³He atoms was attributed to vacancy diffusion and led to the values of Δ and D_0 given in Table I. Neither a large effective radius (small D_{0+}) nor a large value of $H_v(p_0)$ is consistent with our data. The actual process may be a combination of these two factors with vacancies penetrating to within a couple of atomic spacings from the core of the ion. However, this rather close agreement between these parameters does give strong, quantitative support for a simple vacancy-diffusion model for the motion of the positive carrier in solid helium. The qualitative variation of the positive-ion mobility with temperature at various molar volumes and the associated activation energies can be obtained from the space-charge-limited currents measured by Shal'nikov at a con-

stant voltage (see Figs. 4 and 6 of Ref. 2). The variation of Δ_+ with pressure is consistent with a vacancy-diffusion mechanism, and positive-ion mobility measurements may prove to be the most accurate method of measuring the activation energy associated with vacancy diffusion in ⁴He.

The activation energies associated with the motion of the two carriers near the melting curve differ, indicating that the negative ion differs in character from the positive ion. Furthermore, the data exclude a free-electron model. We assume as our model for the negative ion an electron trapped in a void,⁹ identical to an excess electron in liquid helium. The most common mechanisms of void motion in solids applicable to the present case, where atoms are excluded from the interior of the void by the electron's zero-point pressure, are surface diffusion and volume vacancy diffusion. Expressions for the mobility of voids due to these processes have been calculated by many authors,¹⁰ and an analysis of these expressions show that the large experimental value of the frequency factor D_{0-} is inconsistent with a volume-vacancy-diffusion mechanism. However, this large value is consistent with the surface-diffusion model.

We use the following expression for surface diffusion:

$$\mu_s = (3ea^4/2\pi kTR^4)D_s, \quad (3)$$

where a is the interatomic spacing, R is the radius of the void, and D_s is the surface-diffusion coefficient which may be written¹¹ as

$$D_s = \alpha d^2 \nu \exp(-\delta G/kT). \quad (4)$$

Here α is a constant of order unity, d is the average jump distance along the surface, ν is taken as the Debye frequency, and δG is the change in the Gibbs free energy required to create and move a diffusible atom. We rewrite this expression as $D_s = D_{0s} \exp(-\delta H/kT)$, with

$$D_{0s} = \alpha d^2 \nu \exp(\delta S/k). \quad (5)$$

Here δH and δS are the respective changes in the enthalpy and entropy. Our experimental values of D_{0s} , obtained with the use of Eq. (3) and a pressure-dependent void radius⁹ at 20.6 cm³/mole of $\sim 9 \text{ \AA}$, are listed in Table I. These values may be compared with a value of $\alpha d^2 \nu \approx 10^{-3} \text{ cm}^2 \text{ sec}^{-1}$. These comparatively large values of D_{0s} are consistent with experimental values for metals,¹² and explanations have been proposed by a number of authors^{11,13,14} in terms of the diffusion of adsorbed atoms. Basically, adatoms and

adatom complexes are assumed to translate freely on a surface of area d^2 with $d \gg a$. This leads to a large jump distance and a large entropy change associated with the vibrational and translational degrees of freedom.

Quantitative agreement with the experimental values of D_{0s} can be obtained in the present case with the use of a model in which the entropy change and jump distance are enhanced by assuming that the void surface is partially smoothed by the zero-point pressure of the electron and by treating the adatom as a particle constrained to move on the surface of a sphere. A better understanding of the surface-diffusion process, however, awaits more exact experimental values and a theoretical treatment of the void surface.

The activation energy associated with the negative-ion motion is larger than the enthalpy required to create a vacancy and within the framework of our model includes the work performed against pressure exerted by the electron in the adatom creation process. Our data provide strong, qualitative support for the void model of the negative ion and for its motion by adatom diffusion. We believe that the negative ion provides a small, spherical, uniformly sized void free of surface impurities for the controlled study of voids in solids, and also provides experimental data on the *only free surface of solid helium*.

The ionic mobilities at lower temperatures are not understood. Gjostein¹² explains surface diffusion in metals at lower temperatures by the diffusion of surface vacancies. A charge-hopping model, possibly via atomic exchange, is a possible mechanism for the motion of the positive ion.

In conclusion, the diffusion coefficients of both carriers obey an Arrhenius equation of the form $D = D_0 \exp(-\Delta/kT)$ to within 10 mK of the melting curve. The parameters describing the positive-ion mobility suggest a vacancy-diffusion mechanism. Experimental evidence supporting the void model of the negative ion is presented, and the void motion is consistent with a surface-diffusion

mechanism.

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