Motion of charged low-angle grain boundaries in solid helium

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Studies of quasiperiodic ionic-current bursts superimposed on a steady-state space-charge-limited current in strained crystals of solid helium located in a diode are presented. A model involving the motion of a charged small-angle grain boundary which relaxes to its equilibrium configuration after releasing the charge at the anode explains all features of the data. An estimate of the binding energy of positive ions to dislocations is obtained, and conclusions are drawn regarding differences in the mechanisms by which negative ions move in hcp ^{4}He and bcc ^{3}He at large molar volumes.

I. INTRODUCTION

At a constant voltage, unstrained crystals of helium located in a diode with a sufficient supply of charge carriers at one electrode, exhibit steady-state space-charge-limited currents which are constant in time. On the other hand, when ions are injected into sufficiently strained crystals of helium at a constant potential difference, current bursts with a sharp leading edge and a nearly exponential relaxation to steady state are observed.[1] In a limited temperature range these bursts are quasiperiodic with an average period which varies exponentially with the inverse temperature and applied electric field. In this paper, we will characterize the properties of these current bursts and explain them with a model in which charged low-angle grain boundaries move until they reach the collector and are discharged. We will also discuss the implications of our observations on the mechanisms for negative-ion motion since these transients were observed in negative-ion currents in bcc ^{3}He at large molar volumes but not in hcp ^{4}He.

II. EXPERIMENT AND RESULTS

Crystals of solid helium were grown between the plates of a plane parallel diode which was mounted vertically just above a cold finger from which crystal growth was initiated. The cathode consisted of a 0.41-cm^2 titanium tritide beta source backed with a 1-cm^2 copper-clad epoxy board. Ions were created within $\approx$ 10 μm of the source by beta radiation. The separation between the cathode and a 1-cm^2 stainless-steel anode was d = 1.1 mm. Severe strains were introduced into a crystal either by locally melting a region of solid around a heater located behind the diode and near one edge, or by blocking the fill capillary just before completion of crystal growth in the chamber containing the diode. A more detailed description of our cell is given elsewhere.[2,3] All the crystals studied here were hcp ^{4}He at a molar volume of $V_m = 20.5$ cm^3. Current bursts have also been observed[4] in bcc ^{3}He at $V_m = 21.6$ cm^3 for both ion species.

For space-charge-limited currents in a trap-free insulator, the current, I, is related to the ionic mobility, μ, and the voltage V, as

$$I = g\mu V^2 \quad , \tag{1}$$

where g is a constant depending upon the electrode geometry and the dielectric constant of the insulator. If charge traps are present, the current decreases according to the relation[5,6]

$$I = g\mu V^2(1-\chi) \quad , \tag{2}$$

where $\chi = \rho_t/\rho$ is the ratio of the trapped to total charge densities[7]. In carefully grown virgin crystals, the I-V characteristics obeyed Eq. (1). Following a severe strain, the current was considerably reduced, the $I^{1/2}$-V plots were nonlinear and intercepted the abscissa at a voltage $V_i \gtrsim 100$ V, and abrupt, quasiperiodic, current bursts appeared. For the one crystal, crystal 1, on which a complete set of I-V characteristics were taken before and after straining at $V > V_i$, the fractional change in current $\Delta I/I = \chi$ was independent of voltage at the lower values of voltages and temperatures. Since the total charge in the diode varies as V, through the relation $Q = CV$, where C is an effective capacitance, the trapped charge in the crystal also varied as V in this regime. Above a voltage $V_1(T)$ which decreased with increasing temperature, the fraction of trapped charge decreased with voltage.

Figure 1(a) provides examples of the current bursts in their least ambiguous form. They are characterized here with respect to their amplitude A, period τ, and the burst duration δt, defined as the full width at half maximum. The bursts have a sharp leading edge

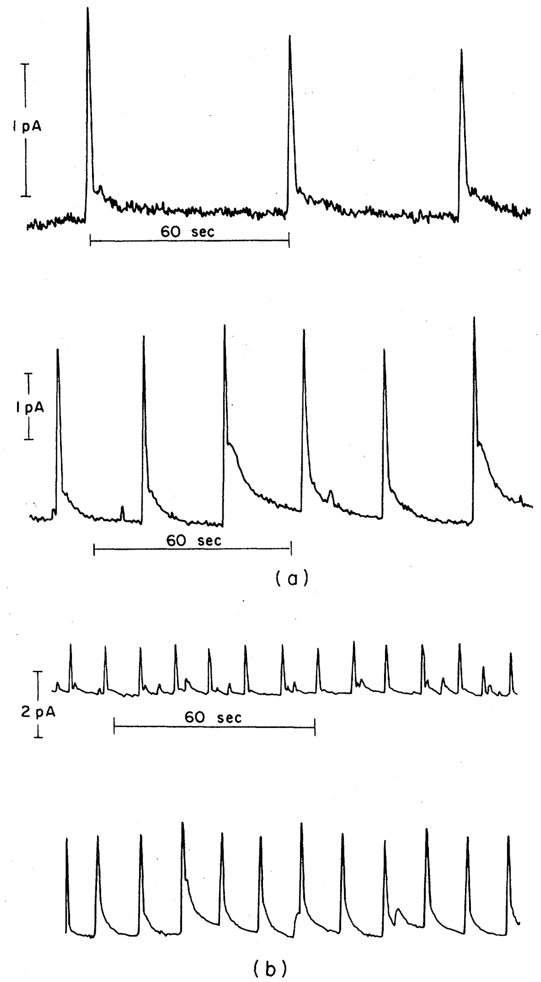

FIG. 1. (a) Examples of well defined current bursts at 1.5 K: upper trace—400 V; lower trace—600 V; Crystal 3. (b) Examples of currents bursts at 600 V and 1.6 K before (upper curve) and after (lower curve) overnight annealing at 1.6 K. The melting temperature is 1.8 K. The scales apply to both traces. Crystal 3.

and a rapid decrease in current followed by exponential decay to the background current. The amplitude at which the exponential decay set in varied with the experimental conditions.

The data indicate that the actual configuration of the crystalline imperfections affected the behavior of these transients. This has the result that two different strained crystals at the same temperature and voltage did not exhibit identical current-burst patterns, complicating a quantitative investigation of the problem. Indeed, as the two curves in Fig. 1(b) indicate, allowing a crystal to anneal overnight at a temperature not far from its melting point produced changes in the shape of the current bursts.

Not all of the data have the well articulated form shown here. At some values of temperature and voltage the current bursts exhibited far less regularity. Also, on occasion, well behaved transients of differing period and amplitude coexisted at certain temperatures. It is a measure of the complexity of the experimental problem that some crystals exhibited well defined current bursts over a narrow range of voltages but over a wide range of temperatures, while for others, the opposite was true. However, a study of selected strained crystals has yielded plots of the experimental parameters which do exhibit a functional consistency over an extended range of temperature and voltage. The behavior of the current bursts which were well defined only over a more limited range of T and V was consistent with these results.

The temperature and voltage dependencies of the period for two crystals are shown in Figs. 2 and 3, respectively. The inverse period varies as

$$\tau^{-1} \propto \exp(-\mathcal{E}/T) , \tag{3}$$

$$\tau^{-1} \propto \exp(V/V_0) , \tag{4}$$

where $\mathcal{E} \simeq 20$ K and is greater than the activation energies Δ associated with the positive-ion mobility characterizing the virgin state of these crystals which were in the range[8] 12–15 K. (All energies are given in degrees kelvin.) The value of V_0 for crystal 2 obtained from Fig. 3 is $\simeq 200$ V which corresponds to a field of $E_0 \simeq 1800$ V/cm. The activation energy $\mathcal{E}$

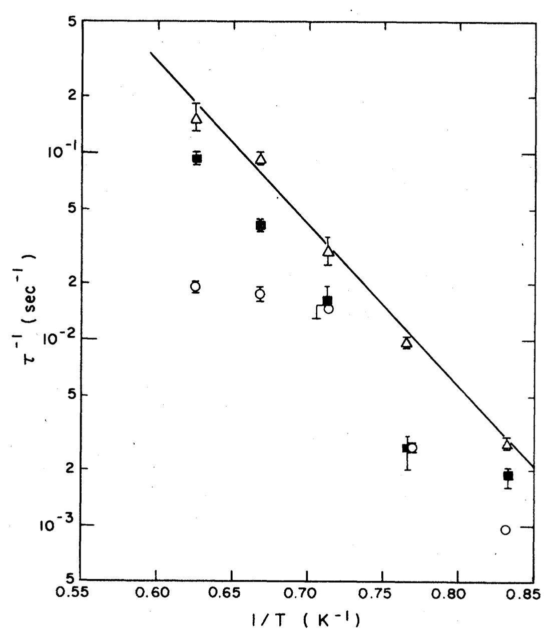

FIG. 2. Plot of τ^{-1} vs T^{-1} for several values of V. Crystal 3. The symbols are ○—400 V; □—600 V; Δ—800 V. The solid line is a least-squares fit of the data to a linear plot at 800 V.

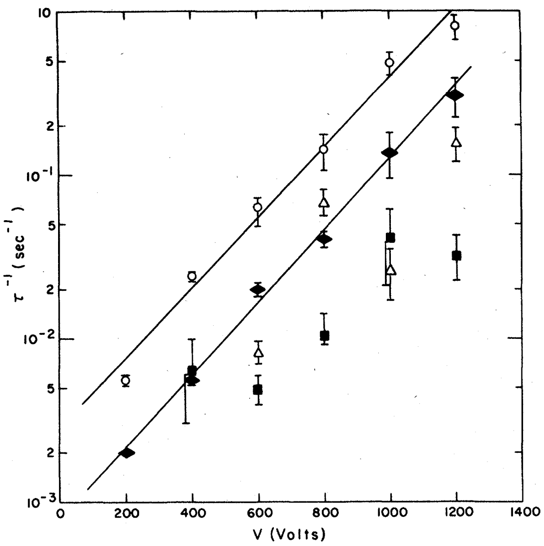

FIG. 3. Plot of τ^{-1} vs V for several values of T. Crystal 2. The symbols are: □—1.4 K; Δ—1.5 K ; □—1.6. K; O—1.7 K. The solid lines are least-squares fits of the data to a linear plot at 1.6 K and 1.7 K.

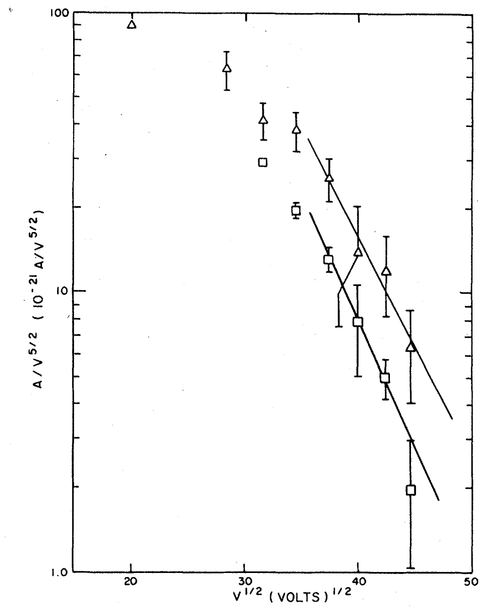

FIG. 4. Plot of $A/V^{5/2}$ vs $V^{1/2}$ for two temperatures. Crystal 1. The symbols are Δ—1.5 K; □—1.6 K. The solid lines are least-squares fits to the data points above 1300 V.

for a number of crystals was in the range of 20 K while the value of V_0 was less reproducible. For example, Lau[2] presents data at 1.3 K from a crystal grown in the same diode which if force fitted to Eq. (4) would yield a value of $V_0 = 600$ V. The value $V_0 = 200$ V was both the minimum and the most frequently observed value of V_0. The ratio of τ to the space-charge-limited ionic transit time, $T_0 = 4d^2/3\mu V$, was typically $\sim 10^2$ for positive ions in hcp ^{4}He and on the single trace which could be analyzed [4] for negative ions in bcc ^{3}He at 300 V and 1.2 K was $\simeq 6$. In the one crystal for which we had sufficient data to compare the burst duration and the space-charge transit time, T_0, the ratio $\delta t/T_0 \approx 2/3$ was temperature and voltage independent.

Because of the variations in the shape of the bursts, we were unable to determine accurately the deposited charge $\delta Q = A\,\delta t$ as a function of voltage. Since in those cases where δt was well defined, δt was proportional to $T_0 \propto (\mu V)^{-1}$, we take δQ to be proportional to $A/\mu V$. The quantity A/V had a maximum at a voltage V_m which increased with decreasing temperature. The value of V_m at a given temperature varied from crystal to crystal and was typically in the range of 500 −1000 V. In order to facilitate a comparison with our model, we have plotted the quantity $A/V^{5/2}$ as a function of $V^{1/2}$ in Fig. 4. The value of $A/V^{5/2}$ decreases approximately as $\exp(-\beta V^{1/2})$ at large V, where β is a constant. The value of A at low voltages and low temperatures was as large as 5% of the current in the unstrained crystal. Values of δQ as large as 4×10^7 charges were observed. At a given temperature and voltage, the standard deviation in δQ was typically 20% of the mean value.

III. INTERPRETATION OF RESULTS

These data clearly indicate a periodic sudden release of trapped charge. The exponential relaxation following the bursts is probably due to a reduction of current, according to Eq. (2), as the background trapped-charge distribution is reestablished.

We interpret these data with a model involving a charged low-angle grain boundary consisting of a planar array of edge dislocation lines. The strain field of a dislocation serves as a trap for both species of ions which are very different in structure. The positive ion has an ionized atom or molecule at its core and is surrounded by a density hump caused by electrostriction.[9] The short-range repulsive interaction between an electron and a helium atom results in a void of $\approx$ 9-Å radius surrounding an electron as the negative-ion structure.[10] The negative ion is attracted to a dislocation by displacing a volume in the strain field of the dislocation, and a comparison of the reduction in currents after straining a crystal indicates that the negative ion is more tightly bound than the positive ion.

The method by which we apply stress to our crystals is to force material into the diode from one end. The electrodes provide a frictional restraining force on the crystal. Thus, we expect the largest motion of helium to be in a plane at the center of the diode. After the heater is turned off, the crystal will relax, and it is reasonable to assume that a low-angle grain boundary will have been formed in a plane parallel to the electrodes. The classic experiment of Washburn and Parker[11,12] showed that a low-angle grain boundary can be transported as a whole by the application of a uniform stress.

We suggest that the portion of a low-angle grain boundary within the ion beam will become charged and move in the presence of an applied field. The grain boundary will probably be pinned at some point outside of the diode, and the dislocation lines comprising the boundary will bow out toward the collector until a portion of each line approaches within a few atomic spacings. The lines need not make contact with the collector since the charges can be ejected from the lines by the large image fields. Those charges which move by a vacany-assisted mechanism then move rapidly along the line to be desposited.[13] Once the charges have been released, the grain boundary will return toward the center of the diode by the tension in the lines. It is not essential that the grain boundary be pinned at some point; it could move as a whole, since it will return to a minimum energy configuration which provides a more homogeneous strain on the crystal.

In explaining these data we examine the motion of individual charged dislocation lines. We first explain the temperature and voltage dependence of τ. Consider charges which move by a vacancy-assisted mechanism. A dislocation line will follow the motion of the charges associated with it. Thus the velocity of the dislocation line is proportional to the average drift velocity in the slip plane, which for the positive ion is proportional to the product of the probability that a vacancy occupy a neighboring site $[\exp(-u_{vn}/T)]$ and a Boltzmann factor. It is given by

$$\vec{u} \propto \sum_n \vec{a}_n \exp[-(u_{vn}+B_n-e\vec{E}_l\cdot\vec{a}_n)/T], \qquad (5)$$

where $\vec{a}_n$ is a vector to a near-neighbor site in the slip plane measured from the ion when it is located on the dislocation, u_{vn} is the energy of a vacancy at $\vec{a}_n$, B_n is the change in binding energy of the ion in moving to this site, and E_l is the field at the dislocation. We have not included in Eq. (5) any activation energy for motion associated with a barrier formed by neighboring atoms since atoms are thought to tunnel into vacancies in solid helium.[14,15] We also note that the line tension plays a negligible role for $V \gtrsim 1$ V. Given the scatter in the data, it is sufficient to consider only motion parallel to the Burgers vector. The period is $\tau = \int ds/v$, where the integration is over the path of the dislocation line, and v is the velocity of the line. It is assumed here that the relaxation to equilibrium is fast on the scale of τ. The inverse period is then

$$\tau^{-1} \propto e^{-(u_{vn}+B_n)/T} \sinh V/V_0 \ , \qquad (6)$$

where we define $V_0(\vec{r}) = Td_e(\vec{r})/ea \cos\theta(\vec{r})$, $\theta(\vec{r}) = \angle(\vec{E}_l, \vec{a}_n)$, and $\operatorname{csch} V/V_0 = \langle \operatorname{csch} V/V_0(\vec{r}) \rangle$. The variable effective length is defined as $d_e(\vec{r}) = V/E_l(\vec{r})$, and the average is taken over all charges on the line and over all positions of the line.

The field under space-charge-limited conditions varies as the square root of the distance from the cathode and is equal to $E_c = 3V/2d$ at the collector. Thus, the value of V_0 can take on a range of values for different crystals which is determined by the location and configuration of the uncharged dislocation line. The calculated value of V_0 for $E_l = E_c$, $\cos\theta = 1, T = 1.6$ K, and the bulk value of a, is 280 V. For the range of voltages over which quasiperiodic current bursts were well characterized, the data can be fitted equally well with the forms $\sinh V/V_0$ and $\exp V/V_0$. The activation energy associated with the burst period is then $u_{vn} + B_n - TV/V_0$. The predicted dependences $V_0 \propto T$ and $\mathcal{E} = \mathcal{E}\ (V)$ are not evident in the data but cannot be ruled out because of the scatter in the period. The value of $(\mathcal{E} - \Delta) \simeq 5-8$ K should be given by $B_n - TV/V_0 - \delta u_v$, where δu_v is the cost in energy in transferring a vacancy from a near-neighbor site of the line to the bulk. We estimate B_n to be $\simeq 12-15$ K.

The center of mass of the negative-ion void moves only a distance $\simeq a(a/R)^2$ in a single-vacancy encounter. Therefore, we expect a small value of B_n and a large value of V_0. Furthermore, the energy of a vacancy at a distance R from the line should differ by only a small amount from its bulk value. For negative ions, we predict the line velocity to be proportional to V $[\sinh(V/V_0) \simeq V/V_0]$ and of the order of the ion velocity in the bulk.

Next, we examine the amplitude of the bursts. At small applied fields, charge will be driven toward an isolated dislocation line from the cathode until the field of the charged line E_l is greater than the applied field normal to the line. The value of $E_l(r)$ is a maximum near $r = l$, where r is the distance from the line, and l is the average interelectron spacing on the line. The maximum value is $\approx e/\epsilon l^2$, where ϵ is the dielectric constant. We equate this to $E_c \sin\phi$, where ϕ is the angle between the applied field and the dislocation line, and obtain the linear density of the charges as $n = l^{-1} \simeq (3V\epsilon/2ed)^{1/2} \simeq 10^4$ $V^{1/2}\,\mathrm{cm}^{-1}$, where V is in volts. Thus, a typical charge density for isolated dislocation lines is $\simeq 10^5$ charges per cm.

At large fields, the electric field reduces the barrier holding charges to the line. We assume the potential of a positive charge in both a strain field and electric field at $r \geq r_0$ to be of the form

$$V(r) = -W\frac{a}{r} - eEr\sin\phi \quad , \qquad (7)$$

where W and r_0 are constants. The maximum of this potential is $B_0 - 2(eEaW\sin\phi)^{1/2}$, where B_0 is the binding energy in zero field. The escape rate of charges from the line will vary as

$$\gamma_e \propto n \exp\{-[B_0 - 2(eEaW\sin\phi)^{1/2}]/T\} \quad , \qquad (8)$$

and the equilibrium line charge density is determined by equating γ_e to the rate γ_t at which charges are trapped. The quantity $\gamma_t = jL2b_c$, where j is the current density ($j \propto \mu V^2$), L is the length of the line, and $b_c(n,V)$ is a critical value of the impact parameter b for charges approaching the line from the cathode. Charges with $b < b_c$ will be attracted to the dislocation. If the electric field of the charged dislocation is neglected, the critical impact parameter will be determined by the ratio of the forces due to the strain field and the applied electric field, $Wa/b_c^2 = ceV/d$. Here c is a number of order unity. With this expression for b_c and $n \propto A/\mu V$, we obtain from the relation $\gamma_e = \gamma_t$ the following expression:

$$A/V^{5/2} \propto \exp(B_0 - 2\Delta - \alpha V^{1/2})/T \qquad (9)$$

with $\alpha = (6eaW\sin\phi/d)^{1/2}$. We have used $E = E_c$ and $\mu \propto \exp(-\Delta/T)$.

The effect of the trapped charges on $b_c \lesssim 10$ Å is negligible. For $r << l$, the field of the trapped charge along a line of approach midway between two trapped charges is $E \simeq er/\epsilon l^3$. The component of electric field perpendicular to the path of an ion with an impact parameter b is $E_\perp = eb/\epsilon l^3 \propto n^3$ which is to be compared with the applied field E_A. For a line with maximum charge $E_\perp/E_A \approx b/l$. In this case, the path of the ion will be diverted by an additional amount b in approaching a dislocation from a distance l. In the regime of interest, the density of charges has been reduced from the maximum by a factor $\gtrsim 2$. In addition, there is a considerable further reduction in the linear charge density for a planar array of dislocations, and Eq. (9) is valid.

We next wish to explain the difference in behavior between positive and negative ions. It is generally assumed that positive ions in solid helium move by a vacancy-diffusion mechanism. A comparison of the activation energy associated with negative-ion motion and the creation energy for vacancies[16] suggests that negative ions move by vacancy diffusion in bcc ^{3}He for $V_m \gtrsim 21.5$ cm^3. Sai-Halasz and Dahm[9] excluded a vacancy mechanism for negative ions in hcp ^{4}He on the basis of such a comparison and suggested that the negatively charged void moves by the diffusion of adatoms around the void surface. The enhanced vacancy concentration near the dislocation provides a mechanism for a rapid discharge of negative ions in bcc ^{3}He at large molar volumes and positive ions in solids of either isotope from the end of the line. On the other hand, in thermally activating an adatom onto the negatively charged void surface, work must be done against the electronic pressure, and this process will not be appreciably affected by the presence of a dislocation. A negatively charged dislocation line in hcp ^{4}He should remain in its elongated configuration with charges being replenished along the line as rapidly as they exit at the collector.

This model of the motion of isolated charged dislocations explains the temperature and voltage dependence of the period of the current blips, the nonexistence of these blips for negative ions in hcp ^{4}He, and the high frequency of blips for negative ions in bcc ^{3}He at large molar volumes. The model also gives a reasonable estimate of the parameter V_0 and accounts for the different values of V_0 and V_m obtained from different crystals since both have a dependence on the direction of the applied field. Likewise, it qualitatively explains the temperature dependence of $V_1 \simeq V_m$. It further explains the variation of the charge in the bursts with applied voltage at large voltage, but gives an incorrect quantitative estimate of the charge on the line by a factor of 10^2.

In order to explain the large magnitude of the current bursts, the coherent motion of many dislocations, i.e., a low-angle grain boundary, is required. Coherence is maintained by the fact that if some region of the grain boundary lags, it is brought back into alignment after expelling the charge in order to minimize the strain energy. For a grain boundary with a spacing w between dislocations and $w << l$, the trapped charge density will be proportional to the applied voltage at small voltages as observed, as opposed to a $V^{1/2}$ dependence for isolated lines. The maximum areal density of trapped charges for $s >> w$ is $n_m = V\epsilon\cos\psi/4\pi es$ where ψ is the angle between the electric field and a normal to the plane, and s is the distance of the grain boundary from the collector. The density enhancement for small s is limited by the rate at which charges approach the line. The field E_l can also be enhanced by the induced charge in the anode which explains a value of V_0 smaller than 280 V.

An estimate of the binding energy B_0 of a positive ion to a dislocation line can be obtained from the data. The value of $W\sin\phi$ for crystal 1, determined from Fig. 4 with the use of Eq. (9), is 4 ± 1 K. The value for crystal 2 was 15 ± 5 K. The relative values of $\sin\phi$ for the two crystals can be estimated from the voltage V_m for which n is a maximum. The linear density is an increasing function of $(V\sin\phi)^{1/2}$ at small V and decreases expontentially with $(V\sin\phi)^{1/2}$ at large V. Thus, the quantity $V_m\sin\phi$ should be approximately the same for all crystals. The values of V_m at 1.6 K were $\simeq$ 400 and 850 V, respectively for crystals 2 and 1. Thus, we conclude that $\sin\phi$ is less by a factor of 2 for crystal 1. We estimate the value of W as 10 ± 3 K and $B_0 = B_n + W$ as 20–25 K. This may be compared with the calculated value[17] of $\lesssim 180$ K for the binding energy of a *negative* ion to a dislocation in solid ^{4}He.

The best fits to the data for both crystals yielded a negative value of $B_0 - 2\Delta$ from Eq. (9), but a positive value was well within the estimated error. The parameter Δ for crystal 1 was 13 K and $B_0 - 2\Delta \lesssim 0$ is consistent with the above estimate.

In conclusion, we have explained quasiperiodic current bursts in strained crystals of solid helium by the motion of charged low-angle grain boundaries. An approximate value of the binding energy of a positive ion to a dislocation line $\approx$ 20–25 K is determined. A better quantitative test of the model and a more accurate value of the binding energy could be obtained if crystals could be strained in such a manner as to exhibit a larger voltage and temperature range of well characterized current bursts with considerably less scatter. A study of current bursts for negative ions in bcc ^{3}He would further test this model and might yield a value of the binding energy of a negative ion to a dislocation line. An important conclusion of this work is that negative ions in hcp ^{4}He do not move by a vacancy-assisted mechanism.

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