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- <sup>1</sup>R. H. Kingston, Proc. IRE 50, 472 (1962).
- $2N. M.$  Kroll, Phys. Rev.  $127, 1207$  (1962).
- ${}^{3}S.$  A. Akhmanov and R. V. Khokhlov, Zh. Eksperim. i Teor. Fiz. 43, 351 (1962) [translation: Soviet Phys. — JETP 16, 252 (1963).
- <sup>4</sup>J. A. Armstrong, N. Bloemberger, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).
- 5W. H. Louisell, Coupled Node and Parametric Electronics (John Wiley & Sons, Inc., New York, 1960).
- ${}^3C.$  C. Wang and C. W. Racette, Appl. Phys. Letters 8, 169 (1965).
- ${}^{7}$ A. Ashkin and G. D. Boyd, private communicatio  ${}^{8}$ K. Nassau and H. J. Levinstein, to be published. <sup>9</sup>G. D. Boyd, R. C. Miller, K. Nassau, W. L. Boyd,
- and A. Savage, Appl. Phys. Letters 5, 234 (1965).  $^{10}$ R. C. Miller, G. D. Boyd, and A. Savage, Appl. Phys. Letters 6, 77 (1965).
- <sup>11</sup>A. A. Ballman, J. Am. Ceram. Soc. 48, 112 (1965).
- ${}^{12}K$ . Nassau, to be published. <sup>13</sup>R. C. Miller, Phys. Rev. 134, A1313 (1964).
- i4L. F. Johnson, Proceedings of the Third Internation-
- al Conference on Quantum Electronics, Paris, 1963,
- edited by P. Grivet and N. Bloembergen (Columbia Uni-
- versity Press, N $\epsilon$ w York, 1964), p. 1021.
- $^{15}$ M. Born and E. Wolf, Principles of Optics (The Macmillan Company, New York, 1964).
- <sup>16</sup>D. A. Kleinman, Phys. Rev. 126, 1977 (1962).
- $17$ J. A. Giordmaine, Phys. Rev. Letters 8, 19 (1962); P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Letters 8, 21 (1962).  $^{18}A$ . Ashkin, private communication.

QUANTIZED VORTICES AND THE SUPERFLUID HELIUM ANALOG OF THE ac JOSEPHSON EFFECT

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In a recent paper Richards and Anderson have reported the observation of an effect in superfluid helium which they have interpreted in close analogy to the ac Josephson effect in superconductors.<sup>1</sup> Their interpretation is based on an equation which has been derived using the concept of a complex order parameter for the liquid. It is the purpose of this note to point out that this equation can also be derived from the conventional equations of two-fluid hydrodynamics combined with the assumption of quantization of vorticity in the superfluid. Although the hydrodynamic approach makes no reference to an order parameter, it is believed that this approach is closely related to the order-parameter approach and that the two points of view complement one another.

Let us consider the situation shown in Fig. 1, in which two reservoirs of superfluid helium are connected by a small circular orifice of radius b. Richards and Anderson point out that if there exists a difference  $\Delta \mu$  between the chemical potentials of the two baths, the relative phase of the order parameters of the baths will vary with time at a mean rate  $\omega = \Delta \mu / \hbar$ . Thus, if the two baths are at the same temperature and if there is a difference of head  $\Delta z$ between them, the relation  $\omega = mg\Delta z/\hbar$  will hold. Here  $m$  is the mass of the helium atom and  $g$  the acceleration of gravity. Richards



FEG. 1. Sketch illustrating the possible steady-state production of vortex rings as superfluid helium flows from the inner to the outer reservoir.

and Anderson suggest that the time variation of relative phase can be accounted for by the motion of quantized superfluid vortices across or away from the orifice at a rate  $\nu = \omega/2\pi$  $=mg\Delta z/h$ , if the vortices are taken to be singly quantized. ' We present below two related hydrodynamic derivations of this last equation, one in terms of a simple physical model of possible vortex motion and the other by means of a more rigorous, general argument.

In order to fix our ideas, let us begin with the simple model. Suppose that as downward flow through the orifice takes place, in the process of equalizing the level difference between the two baths, a steady state has been reached, and that in this steady state the average superfluid flow velocity in the orifice  $\bar{v}_s$  is constant. Following one of the suggestions of Richards and Anderson, let us assume that vortex rings of radius  $b$  are being formed at the orifice at a steady rate  $\nu$  and propagated downward as shown in Fig. 1. Let us throughout the argument assume that  $b$  is so small in relation to the size of the reservoirs that the slow change in  $\Delta z$  taking place during the flow can be neglected.

Now let us determine the rate of vortex ring formation in the following way. Assume that the pressure drop  $\Delta P = \rho g \Delta z$ , where  $\rho$  is the total fluid density, occurs right at the orifice. Then the net force exerted on the superfluid in the orifice by this pressure drop is simply  $\rho_{\rm s}g\Delta z\pi b^2$ , where  $\rho_{\rm s}$  is the superfluid density. If we equate this force to the product of the local impulse  $p$  needed to form each vortex ring and the rate of production  $\nu$ , we get the relation

$$
\rho_s g \Delta z \pi b^2 = p \nu.
$$

It may be noted that the remainder  $\rho_{n}g\Delta z\pi b^{2}$ of the total force on the liquid in the orifice due to the pressure drop acts on the normal fluid and merely gives rise to viscous flow of that component through the orifice.

For a superfluid vortex ring of radius  $b, p$ is given by the equation

$$
p = \rho_{\rm c} \kappa \pi b^2,
$$

where  $\kappa$  is the circulation about the core of the ring.<sup>3</sup> Taking  $\kappa$  to be  $h/m$ , one quantum unit, we have the result

$$
v = mg\Delta z/h
$$

The last equation is seen to be the same equation that Richards and Anderson have obtained for singly quantized vortices.

Now let us consider a more general derivation of the same result. Following Landau and Lifshitz, we write the equation of motion for the superfluid as

$$
\frac{\partial \vec{v}_S}{\partial t} + \text{grad}\left(\frac{\mu}{m} + \frac{v_S^2}{2}\right) = 0,
$$

where  $\bar{v}_s$  is the superfluid velocity.<sup>4</sup> If  $\bar{v}_s$  is irrotational except for the presence of vortices we may write

$$
\overline{\mathbf{v}}_e = \mathbf{grad}\varphi,
$$

where  $\varphi$  is a multivalued velocity potential. Then the relationship

$$
\text{grad}\left(\frac{\partial \varphi}{\partial t} + \frac{\mu}{m} + \frac{v_s^2}{2}\right) = 0
$$

is valid everywhere except at the cores of the vortices.

Let us take the line integral of both sides of this equation along some arbitrary but stationary path through the fluid connecting the two reservoirs. Such a path is shown in Fig. <sup>1</sup> running from point 1 to point 2. Since the end points lie in regions where the fluid is essentially at rest we can neglect  $v_s^2/2$ , and we then have the equation

$$
\frac{d(\varphi_2-\varphi_1)}{dt}=-\frac{(\mu_2-\mu_1)}{m},
$$

which holds at all times except at the moments when vortices cross the path of integration.

Now without regard to the equation above, we can argue that since  $\varphi_2-\varphi_1$ , equals the line integral of  $\bar{v}_s$  along the path, the time-average value of  $\varphi_2-\varphi_1$  must, in steady state, remain constant. Thus, the changes in  $\varphi_2-\varphi_1$  which take place in the intervals of time between successive vortex crossings, and which are given by the equation above, must just be cancelled on the average by the changes in  $\varphi_2-\varphi_1$  which occur during the vortex crossings. Each time the contour is crossed by a vortex of circulation  $\kappa$ ,  $\varphi_2-\varphi_1$  must suddenly increase by an amount  $\kappa$ , the sign of the change given here being chosen to agree with the changes which would occur in the situation shown in Fig. 1. Therefore, during the interval between successive crossings,  $\varphi_2-\varphi_1$  must change on the average by an amount  $-\kappa$ , and as a result, vortices must cross the path of integration at an average rate  $\nu$  given by

$$
-\nu\kappa=-\Delta\mu/m,
$$

where  $\Delta \mu = \mu_2 - \mu_1$ . From this equation we are led once again to the result  $\nu = mg\Delta z/h$  that Richards and Anderson have obtained by other means.

The most interesting feature of this second argument is that, like the Richards and Anderson argument involving the order parameter, it is not based on a detailed model for vortex motion. The generality of these arguments indicates, as Richards and Anderson suggest, that there may be a number of repetitive vortex motions possible in steady state in addition to the simple ring motion proposed earlier. The similarity of the results of the general hydrodynamic argument and the order-parameter argument suggests that there exists a close and fundamental relation between these two points of view. This suggestion is supported by the fact that, at least in the low-density limit, the phase of the complex order parameter of a gas of weakly interacting Bose particles can be identified with  $m\varphi/\hbar$ .<sup>5</sup>

Let us conclude by returning to the vortexring model in order to point out that it leads to a prediction for the average steady-state superfluid flow velocity in the orifice,  $\bar{v}_s$ . If we equate the free energy available per unit time, due to flow from a bath of higher chemical potential to one of lower, mith the energy required per unit to create the vortex rings, we obtain the relationship

$$
\overline{v}_s \pi b^2 \rho_s g \Delta z = E \nu,
$$

where  $E$  is the energy needed to form a single superfluid vortex ring.  $E$  is given by

$$
E = \frac{\rho_s \kappa^2 b}{2} \left[ \ln \frac{8b}{a} - \frac{7}{4} \right],
$$

where  $a$  is the radius of the vortex core.<sup>3</sup> Substituting for  $\nu$  the expression derived earlier and solving for  $\bar{v}_s$  we obtain

$$
\overline{v}_s = \frac{\hbar}{mb} \left[ \ln \frac{8b}{a} - \frac{7}{4} \right].
$$

Using the Richards and Anderson value of b  $= 7.5 \times 10^{-4}$  cm and the Rayfield and Reif value

of  $a = 1.3 \times 10^{-8}$  cm, we calculate  $\overline{v}_s = 2.4$  cm  $\sec^{-1}$ .

Richards and Anderson observe a net steadystate velocity of flow through the orifice,  $\bar{v}$ , of  $\approx 27$  cm sec<sup>-1</sup>. A rough calculation indicates that at the temperature of  $1.15^{\circ}$ K, where this observation was made, viscous flow of the normal fluid makes only a small contribution to the total flow observed. Thus this value of  $\bar{v}$ provides an approximate experimental value for  $\bar{v}_s$ . The fact that this experimental  $\bar{v}_s$  is an order of magnitude greater than our theoretical estimate may cast some doubt on the vortex-ring model for the details of the vortex motion. However, it is possible that the  $\bar{v}_s$ calculated here should only be regarded as a lower limit for the ring formation to proceed, much in the same way that for a classical fluid flowing in a circular pipe the critical Reynolds number serves only to give a lower bound for the velocity at which turbulence will appear.<sup>6</sup>

Richards and Anderson observe that  $\bar{v}_s$  may be several orders of magnitude lower after vigorous agitation of the bath by means of the quartz oscillator. This observation seems consistent with the energy considerations presented here. If a large amount of vorticity due to another source is already present in the liquid, the vortex motion needed to maintain the flow in steady state may require the creation of very few new vortices. Since in that case the rate at which energy mould have to be supplied to the superfluid would be much reduced,  $\bar{v}_s$  might be correspondingly reduced.

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<sup>&</sup>lt;sup>1</sup>P. L. Richards and P. W. Anderson, Phys. Rev. Letters 14, 540 (1965).

<sup>&</sup>lt;sup>2</sup>Vortices in superfluid helium have generally been thought to exist preferentially in singly rather than multiply quantized form. For this reason we have restricted our considerations to singly quantized vortices. Such vortices appear to suffice for the explanation of the experimental results of Richards and Anderson, although this point is perhaps not really clear.

 ${}^{3}$ G. W. Rayfield and F. Reif, Phys. Rev. 136, A1194  $(1964).$ 

<sup>4</sup>L. D. Landau and E. M. Lifshitz, Fluid Mechanics (Addison-Wesley Publishing Company, Inc., Reading,

Massachusetts, 1959), Scc. 130.  ${}^{5}$ See, e.g., E. P. Gross, J. Math. Phys. 4, 195 (1963); P. S. Hohenberg and P. C. Martin, Phys. Rev. Letters

12, 69 (1964); A. L. Fetter, Phys. Rev. 138, A429 (1965). Reference 4, Sec. 29.

## CHANGING PROPERTIES OF METALS BY FERROELECTRIC POLARIZATION CHARGING

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This Letter describes a new method of studying the properties of metals and other conductors by inducing large electric charges on them. The method involves depositing a thin film of the substance to be studied onto a ferroelectric crystal and measuring the film properties while the polarization of the substrate is directed toward and away from the film. The difference in film properties between the two substrate polarization states is the quantity of interest. This technique has produced changes of  $2\%$  in the resistance of 100Å-thick gold films on BaTiO<sub>3</sub>, of 0.0013°K in the superconducting transition temperature of 160A-thick tin films on triglycine sulfate (TGS), and of as much as 0.7% in the reflectivity of 200Å-thick silver and 150A-thick gold films.

The ferroelectric polarization effect, like the similar field effect,<sup>1-4</sup> is described by the relation

$$
\sigma = \Delta D_n = \Delta (E + 4\pi P)_n \tag{1}
$$

for the density  $\sigma$  of charge on a capacitor plate as a function of the discontinuity  $\Delta D_n$  in the normal component of the electric displacement.  $E$  is the electric field and  $P$  is the polarization. Previous charge changing has been effected by varying the term  $E$  in (1) (field effect); the present method varies the term  $P$  (at negligible values of  $E$ ) by reversing a ferroelectric capacitor. The first advantage of the ferroelectric method is that the charge densities are much higher since  $4\pi P$  for common ferroelectrics (e.g., BaTiO<sub>3</sub>) is  $3 \times 10^8$  V/cm compared to the breakdown strength of usual dielectrics  $(\sim 10^6 \text{ V/cm})$ . The reversal of polarization of BaTiO, changes the charge induced on its electrodes by  $3\times10^{14}$  electrons/cm<sup>2</sup>, or by  $3\times10^{20}$ electrons/cc if the electrode is 100  $\AA$  thick. The second advantage of the ferroelectric method is that the effects are measured, after the polarization is reversed, with no electric field

applied to the dielectric. Since the quantities measured are ordinarily very small electric voltages or currents, this can be a great advantage.

In order to test the hypothesis that the changes of film properties are due to charging, several experiments using the electrical resistivity as the property of interest were performed. Films were evaporated onto both sides of etched ferroelectric single crystals in a conventional high-vacuum plater at about  $10^{-6}$  Torr. The conductivity of the film was measured by a fourelectrode method at constant current with the polarization of the ferroelectric directed toward or away from the film, using a potentiometer principle to detect the small voltage changes precisely. The amount of charge needed to reverse the polarization was always measured during reversal and was always found to equal the product of the electrode area on the crystal times twice the published polarization of the crystal (26  $\mu$ C/cm<sup>2</sup> for BaTiO<sub>3</sub> and 2.8  $\mu$ C/cm<sup>2</sup> for TGS). The resistance changes were believed to occur in the films rather than in the substrates because the ferroelectric crystals used have resistivities of  $\geq 10^{12}$  Q cm and because even if extra impurities are diffused into  $Bario<sub>3</sub>$ , the carrier mobilities in doped BaTiO<sub>3</sub> are so low  $(\sim 10^{-3}$  cm<sup>2</sup>/V sec)<sup>5</sup> that such an impurity layer would not show observable resistance changes on polarization reversal of the substrate.

Variations were made of film material, substrate material, temperature of measurement, and film thickness. The films used were gold and gadolinium (both  $n$ -type conductors), germanium and tellurium (both  $p$ -type). The charging hypothesis predicts that electrons will be added to a film when the ferroelectric polarization is directed toward it and that these extra electrons will lower the resistance of an  $n$ type film and raise the resistance of a  $p$ -type film. This predicted sign of resistance change