Sample and Swenson,<sup>15</sup> and by Jarvis, Ramm, and Meyer<sup>19</sup> for the Grüneisen gamma defined by

 $\gamma = (V/C_{\nu}) (\partial P/\partial T)_{\nu}$ 

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## THEORY OF THE ONSET OF SUPERFLOW\*

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Spontaneous production of quantized vortices in He II by thermal fluctuations is considered. Implications of this theory for superflow in finite channels are discussed.

In a recent communication<sup>1</sup> we examined the problem of the nucleation of quantized vortex rings by ions. There we presented arguments to suggest that the smallest vortex rings are rotons and that larger rings form higher-energy states which are filled by collisions. In order to fix our ideas and notation about superflow, consider first spontaneous vortex production in an unbounded fluid, a concept introduced by Iordanskii,<sup>2</sup> and applied to the interpretation of the decay of persistent currents by Langer and Fisher,<sup>3</sup> Fisher,<sup>4</sup> and Langer and Reppy.<sup>5</sup> Rotons in a counterflow will be polarized against the oncoming superfluid and will become rings if they can fluctuate over a saddle point *C* in momentum space constituting a free-energy barrier  $\Delta F$ . We shall show elsewhere that the probability per unit time of such a diffusion taking place may be estimated by Brownian-motion theory of vortex rings to be

$$P = \frac{\Lambda_c kT}{4\pi \mu^{1/2} p_0^2} \left[ \frac{v_s p_0 / kT}{\sinh(v_s p_0 / kT)} \right] \left( \frac{\omega_c}{s_c^2} \right) e^{-\Delta F / kT}, \tag{1}$$

where  $\Lambda_c$  is the diffusivity constant for vortex rings (and includes contributions from rotons, phonons, and solvated He<sup>3</sup> atoms as explained in Ref. 1),  $v_s$  is the relative velocity of the counterflow,  $\mu$  and  $p_0$  are the effective mass and momenta of rotons,  $\omega_c$  and  $S_c$  are the principal curvatures of momentum space at the saddle point. The derivation of (1) follows from considerations similar to those leading to Eq. (133) of Donnelly and Roberts.<sup>6</sup> In a counterflow the density of rotons is given by

$$N_r = 2(2\pi kT)^{3/2} \mu^{1/2} p_0 h^{-3} v_s^{-1} \sinh(p_0 v_s / kT) \exp(-\Delta/kT).$$
<sup>(2)</sup>

The rate of production of vortex rings per unit volume is therefore given by

$$\nu \equiv PN_r = (2\pi)^{1/2} (kT)^{3/2} \Lambda_c h^{-3} \omega_c S_c^{-2} \exp(-\Delta F / kT),$$
(3)

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where  $\Delta F$  is now corrected for  $\Delta$ . Equation (3) coincides with Iordanskii's expression below  $(3.14)^2$  (except for a factor of  $\sqrt{2}\pi^2$  whose origin is not clear to us).

Experiments on the decay of persistent currents use small  $v_s$  and large kT, hence we may regard the quantity in square brackets in (1) as unity and write

$$p = f \exp(-\Delta F / kT), \tag{4}$$

where to a reasonable approximation

$$f = \frac{\beta \rho_s \kappa^3 kT}{64\pi^2 \rho_0^2 [2\mu v_s^5]^{1/2}} \left[ \ln\left(\frac{c\kappa}{4\pi v_s a}\right) \right]^2,$$
(5)

and

$$\frac{\Delta F}{kT} = \frac{\rho_s \kappa^3}{16\pi kT v_s} \left[ \ln\left(\frac{c\kappa}{4\pi v_s a}\right) \right]^2 - \frac{\Delta}{kT} \,. \tag{6}$$

In these equations  $\Lambda_c = \beta \rho_c^{1/2}$  where  $p_c = p_s \kappa \pi R_c^2$ ,  $R_c$  is the radius of the ring at C,  $c = 8/\sqrt{e}$ , and a(=1.28 Å) is the radius of the vortex core. Equation (6) contains the velocity

$$v_{\rm th} = \rho_s \kappa^3 / 16\pi kT \tag{7}$$

which might be called the "thermal characteristic velocity." Typical values for  $v_{\rm th}$  are 188 m/ sec at  $1.1^{\circ}$ K, 46.5 m/sec at  $2.0^{\circ}$ K, and 2.2 m/ sec at 2.17°K. These high velocities contain the essence of the difficulty of vortex nucleation. For, unless  $v_s \approx v_{th}$ , appreciable rates of vortex production will not occur. For example at 2.1°K we find [on a somewhat more precise extimate than is given by (6)]  $\nu = 2.90 \times 10^{-73}$  for  $v_s = 200$ cm/sec,  $\nu = 1.03$  for 388 cm/sec, and  $\nu = 1.07$  $\times 10^{20}$  for  $v_s = 600$  cm/sec. If we adopt  $\nu = 1$  as a rate characteristic of values of  $v_s$  observed in the laboratory, we find the critical velocity is almost an order of magnitude too large to agree with the experiments of Kukich, Henkel, and Reppy<sup>7</sup> and Notarys.<sup>8</sup> It may be noted, however, that the values of  $fN_r$  given by (5) (e.g.,  $fN_r$  $=6.04 \times 10^{35} \text{ sec}^{-1} \text{ cm}^{-3} \text{ at } v_s = 388 \text{ cm/sec}$  are of the order of those estimated by Langer and Fisher<sup>3</sup> and stated by them to be a characteristic atomic frequency. We see that since  $\Lambda$  and  $N_r$  vary rapidly with temperature,  $fN_r$  should be extremely temperature dependent and (5) provides a means of estimating this dependence as well as those changes created by pressure variations, He<sup>3</sup> concentrations, etc., as outlined in Ref. 1.

Consider now the effect of boundaries on the spontaneous nucleation process discussed above. For simplicity we suppose the fluid is confined

to a tube of radius  $R_0$  and is at rest. We imagine a ring to nucleate from a roton at the center of the tube polarized along its axis, to a circular ring whose plane is perpendicular to the axis of the tube on which it is centered. This model is common to studies of the Feynman critical-velocity mechanism by Fineman and Chase<sup>9</sup> and Gopal.<sup>10</sup> We will follow the analysis of the latter author assuming, as he did, that the rings are classical "solid core" rings when their radius R is large compared with their cross section a. We imagine that for  $R \leq a$  the spectrum has a roton well. Also, by analogy, for  $R \leq R_0 - a$  the quantum pressure effects associated with healing at the wall and the image of the ring produce an analogous minimum which we call the "image well." Following Gopal, we assume this occurs at  $R_I \cong R_0 - a$ , and evaluate the energy  $E_I$  and momentum  $p_I$  of that state accordingly. We assume for simplicity that the curvature  $\partial^2 E / \partial p^2$ of the image well is  $1/\mu^{1/2}$ . These considerations lead to the curve sketched in Fig. 1. The theory here applies strictly only to those rotons on the axis of the tube but we apply it (perhaps less accurately) to all rotons in the tube. The energy of a ring in an unbounded fluid is a monotonically increasing function of R; the situation in a tube is crucially different. The confinement of the vortex flow to the tube depresses E at large R, and creates a single maximum C between the roton and image wells, and nucleation can occur in either direction over this barrier. By "nucleation outwards" we will mean the nu-



FIG. 1. Schematic diagram of the dependence of the energy of a vortex ring, centered on the axis of a tube, as a function of the radius R of the ring. The curve is also, parametrically, a dispersion curve since  $p \propto R^2$ . The region in which classical theory is applied is indicated, and the roton and image wells greatly exaggerated for clarity.

cleation of vortex rings in the image well by rotons in the interior. By "nucleation inwards" we mean the reverse process of the collapse of rings from the well into rotons in the interior. In either process, the probability of nucleation is independent of the sense of circulation. Once a nucleation has taken place, however, a flow  $v_s$  is created in the tube, and this enhances the probability that a nucleation of the opposite sense will take place next (see below). The mechanism, then, is self-correcting and on average maintains the original state of superfluid at rest. The time scale of this process is, for nucleation outwards, of order  $1/(\nu V)$  where V is the volume of helium in the apparatus and  $\nu$  is the rate parameter introduced above. For nucleation inwards, it is of order  $1/(\nu_s S)$  where S is the surface area of the tube,  $v_s = P_V S_V$  where  $P_V$  is the probability for inward nucleation and  $S_V$  is the density of vortex line on the walls. We show below that in the presence of a superflow the rate of production of vortices by either of these processes adjusts itself in such a way as to reduce the superflow. We may infer that if  $\tau$  is "sufficiently small," say 1 second, the phenomenon of superfluidity will be greatly inhibited. This is the well known "suppression of the  $\lambda$  point," or more accurately, the onset temperature  $T_0$  for superflow.

The free energy F is simply the kinetic energy of superflow and can, in the classical range, be obtained from Gopal's analysis.<sup>10</sup> For  $E_I$ , we have taken Gopal's estimate

$$E_I = \frac{1}{2} \rho_s \kappa^2 R_I (\ln 2 + 7/4). \tag{8}$$

It is clear that since  $E_I \gg \Delta$ ,  $\Delta E_I \ll \Delta E_R$  (cf. Fig. 1) and the probability of nucleation inwards greatly exceeds that of nucleation outwards. Balancing the inward rate  $(2\pi)^{-1}S_V\Lambda\omega_A\omega_c \exp(-\Delta E_I/kT)$  with the outward rate  $(2\pi)^{-1}N_r\pi R_0^2\Lambda\omega_A\omega_c$  $\times \exp(-\Delta E_R/kT)$  we find the equilibrium density of vortex rings on the wall to be (in units cm<sup>-1</sup>)

$$S_V = (\mu kT / 2\pi)^{1/2} \hbar^{-3} (\rho_0 R_0)^2 \exp(-E_I / kT).$$
(9)

Whether such a population as (9) exists may well depend on how the sample is prepared.

For P we find a one-dimensional approximation suffices:

$$P = (2\pi)^{-1} \Lambda_c \omega_A \omega_c \exp(-\Delta E_R / kT), \qquad (10)$$

and we use (10) to evaluate  $T_0$  as that temperature at which, on the average, one vortex per unit volume per unit time is fluctuating over the barrier at C in Fig. 1. We have ignored inward nucleations in this definition, but it is easily shown that were they added, the change in the estimate of  $T_0$  would be negligible.

Nothing in the discussion above implies that the superfluid is destroyed above  $T_0$ . Rather any attempted superflow in this range would rapidly decay owing to fluctuations. An alternating flow such as is induced by third or fourth sound of frequency f will become strongly damped at a temperature such that  $f \approx \nu$ . But  $\nu$  increases so rapidly with T that third and fourth sound should extinguish at nearly the same temperature as direct flow. Evidence for this is seen in Fig. 6 of the article by Guyon and Rudnick.<sup>11</sup>

 $T_0$  is plotted in Fig. 2 as a function of temperature and tube diameter,  $d = 2R_0$ . With the reservation that the core radius *a* must be an increasing function of *T* (we adopt *a* = 1.28 Å throughout), the onset curve is in gratifying agreement with the experiments of Guyon and Rudnick<sup>11</sup> and Fokkens, Taconis, and de Bruyn Ouboter.<sup>14</sup> The latent heat of the vortex cores will contribute a specific heat anomaly with a maximum in the interval  $T_0 < T < T_{\lambda}$ .

Now consider the situation if the normal fluid is at rest, but the superfluid is moving everywhere with the uniform velocity  $v_s$  (>0) down the tube (plug flow). The difference in free energy between this state and one including a vortex of radius *R* coaxial with the tube is related to E(R), the expression for  $v_s = 0$ , by

$$F(R) = E(R) + \vec{\mathbf{v}}_{s} \cdot \vec{\mathbf{p}}(R), \qquad (11)$$



FIG. 2. Calculation of  $T_0$  as a function of  $d=2R_0$ from the condition  $\nu=1$  with a=1.28 Å. The experimental onset temperatures indicated by rectangles and one circle were obtained by Guyon and Rudnick, Ref. 11, and the squares indicate unsaturated film measurements by Fokkens, Taconis, and de Bruyn Ouboter, Ref. 14.

where  $\tilde{p}(R) = \rho_s \tilde{\kappa} \pi R^2$ . For  $\kappa < 0$ , the last term of (11) is negative so that  $F(R) \le E(R)$  and the probability of outward nucleation is increased relative to its value for  $v_s = 0$ . Since, however, |p(R)|/E(R) increases monotonically with R, the probability of inward nucleation is decreased. Conversely, for  $\kappa > 0$  these conclusions are reversed. Outward nucleation becomes less probable. There are, then, four fluctuating processes, inward and outward nucleation of rings of positive and negative circulation. The dominant processes, the outward nucleation of rings of negative  $\kappa$  and the inward nucleation of rings of positive  $\kappa$ , act in a sense to destroy  $v_s$ , which will ultimately cease unless an agency acts to maintain it (i.e., a source of thermodynamic potential). Superfluidity is observed in the laboratory when the time scale for this decay greatly exceeds the duration of the experiment.

If we compare the present nucleation process with that for an unbounded fluid we see that the energy barrier at *C* is always lower for outward nucleation of rings of negative  $\kappa$  than it is at the same  $v_s$  in the Iordanskiĭ process. For large flows (compared with the Feynman velocity  $\kappa/$  $4\pi R_0$ ) *C* moves away from the wall (Fig. 1), image effects are less important, and the results are asymptotically the same as Iordanskiĭ's theory would predict. For small flow rates  $R_c(v_s)$ lies close to  $R_c(0)$ , the maximum of E(R) shown in Fig. 1. This prevents the critical ring from ever exceeding the size of the tube, a problem raised by Notarys.<sup>8</sup>

We may further, in extension of our ideas of the onset of superflow, define  $T_0(R_0, v_s)$  as that temperature at which  $\nu = 1$ . Figure 3 illustrates this generalization. For  $v_s = 0$  the curve coincides with that of Fig. 2. The curves for  $v_s$ = 350 cm/sec and 700 cm/sec agree asymptotically with Iordanskii's theory, and in this region of relatively large channels it is correct to speak of an "intrinsic" critical velocity, i.e., one independent of channel size. This is no longer appropriate for  $d \leq 800$  Å.

Let us now consider the process of "cooling through the onset temperature  $T_0$ ." While  $T > T_0$ the processes of creation and destruction of vortex rings occur so rapidly that the populations in the fluid and on the walls can adjust continuously to the changing temperature. Below  $T_0$ , however, this process will be so slow that further cooling is adiabatic (no matter how slowly it is carried out on the laboratory time scale) and populations on the walls are now "frozen" to their val-



FIG. 3. Generalized onset temperature for  $v_s = 0$ , 350, and 700 cm/sec as a function of channel size. The whole family of curves  $T_0(R_0, v_s)$  may be thought of as a map of critical velocities.

ues at  $T_0$  as roughly estimated by (9). Normally, as we have seen, the numbers of vortices of positive and negative circulation should be equal:  $S_V^+ = S_V^-$ . If on cooling a superfluid gyroscope through  $T_0$  fluctuations should leave one sign of vortex predominant, a test would reveal a spontaneous persistent current of magnitude  $\kappa(S_V^+$  $-S_V^-)$  to have been generated, as indeed Mehl and Zimmerman have reported.<sup>13</sup>

Now suppose, for example, we were to rotate a gyroscope with 200 Å channels at a speed corresponding to  $v_s$  = 700 cm/sec, cool down to a temperature below  $(T_{\lambda} - T_0) = 200$  mdeg K, and stop. A persistent current of magnitude 700 cm/ sec will be observed corresponding to a population difference  $(S_V^+ - S_V^-)$  induced by the rotation. If, now, the temperature is raised to  $(T_{\lambda})$ -T) = 60 mdeg K, fluctuations will occur, and the persistent current will decay, at first rapidly, then more slowly until the critical value of  $v_s$ (350 cm/sec) is reached and passed. This decay has been observed experimentally by Kukich, Henkel, and Reppy under the general conditions described.<sup>7,5</sup> As one would expect, the rate of decay reported by them is greater, the higher the temperature.

While the qualitative effects described here are in striking accord with experiments, the calculated velocities are still higher than those observed experimentally. We believe core-size corrections and density-of-states arguments will improve the agreement but we must await better experimental knowledge of these quantities and their variation with temperature. \*Research supported by the Air Force Office of Scientific Research under grant No. AF-AFOSR 67-1239, and by the National Science Foundation under grant No. NSF-GP-6473.

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<sup>12</sup>It may be argued that for a ring near the wall  $p = \rho_s \kappa \pi (R_0^2 - R^2)$ . Since we are interested only in freeenergy <u>differences</u>, the alternate definition is immaterial.

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## DIVERGENT FLUCTUATIONS IN SUPERCONDUCTING FILMS\*

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We have observed anomalously broad resistive transitions in superconducting films and have shown that the widths gradually reduce to the usual Aslamazov-Larkin value when a magnetic field is applied parallel to the film. These results are explained by adding to the Aslamazov-Larkin theory another fluctuation contribution to the conductivity of the type first proposed by Maki, which for thin films is logarithmically divergent with a cutoff proportional to the pair-breaking interactions.

Theoretically, fluctuations in thin superconducting films tend to diverge. In fact if one assumes that long-range superconducting order exists, then the fluctuations would actually diverge, thereby destroying such order.<sup>1,2</sup> Naturally one would also expect to see some evidence of this divergence in the fluctuations above the mean-field critical temperature  $T_c$ , although perhaps not in the leading order. However, the first successful theory of the rounding of the resistive transition, that of Aslamazov and Larkin (AL),<sup>3</sup> contained no divergence above  $T_c$  and was in excellent agreement with the experimental results of Glover<sup>4</sup> on very dirty films of strong-coupling superconductors. In contrast, the early experimental work of Strongin et al.<sup>5</sup> followed by the definitive experiments of Masker and Parks<sup>6</sup> showed that the resistive transitions in aluminum could be anomalously large, even more than an order of magnitude larger than the

AL value for low-resistance films. A second theory of the resistive transition by Maki,<sup>7</sup> which appeared shortly after AL, was divergent when applied to films. In the form first proposed by one of us<sup>8</sup> the divergence is cut off by a pairbreaking interaction such as electron-phonon scattering or magnetic fields. The extra term of Maki was predicted to decrease relative to the AL term as stronger magnetic fields are applied parallel to the film. We now wish to report experimental confirmation of this theory, the first experimental evidence identifying the tendency of fluctuations to diverge in superconducting films away from  $T_c$ . Furthermore we have observed the same tendency in lead films although the enhancement of the transition width is smaller. The AL result was recovered near the transitions with no significant reduction due to strong coupling.

The AL theory for weak-coupling superconduct-