*Permanent address.

[1]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

[2]D. J. Scalapino, J. R. Schrieffer, and J. W. Wilkins, Phys. Rev. 148, 263 (1966).

[3]W. L. McMillan, Phys. Rev. 167, 331 (1968).

[4]J. W. Garland and K. H. Benneman (unpublished).

[5]N. F. Mott, Advan. Phys. 13, 325 (1964).

[6]A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice Hall, Englewood Cliffs, N. J., 1963).

[7]J. J. Hopfield, Phys. Rev. 186, 443 (1969).

[8]The temperature Green's function for an electron is given by

$$G_{s's}(x', x) = -\langle T_\tau(\varphi_{s'}(x')\varphi_s^+(x))\rangle,$$

where T_τ is the Wick ordering operator and

$$\varphi_s^+(x) = e^{(H-\mu N)\tau}\varphi_s(\vec{r})e^{-(H-\mu N)\tau}$$

is the Heisenberg operator for the creation of a particle; H is the Hamiltonian, N is the number operator, and μ denotes the chemical potential. The angular bracket represents a thermal average over a grand canonical ensemble.

[9]The Fourier transforms are found in Ref. 6, p. 120 ff. They can be written as follows:

$$G_{s's}(x', x) = \frac{1}{\beta}\sum_{j',=-\infty}^{+\infty} G_{s's}(\vec{r}', \omega_{j'};\ \vec{r}, \omega_j)\, e^{-i(\omega_j\tau-\omega_{j'}\tau')},$$

$$I_{s_1' s_2' s_1 s_2}(x_1', x_2', x_1, x_2) = \frac{1}{\beta^3}\sum_{j_1', j_1, j_2', j_2=-\infty}^{+\infty} I_{s_1' s_2' s_1 s_2}$$

$$\times(\vec{r}_1', \omega_{j_1'}; \vec{r}_2', \omega_{j_2'}; \vec{r}_1, \omega_{j_1}, \vec{r}_2, \omega_{j_2})$$

$$\times\exp[-i(\omega_{j_1}\tau_1+\omega_{j_2}\tau_2-\omega_{j_1'}\tau_1'-\omega_{j_2'}\tau_2')].$$

[10]W. Kohn and J. M. Luttinger, Phys. Rev. Letters 15, 524 (1965); J. M. Luttinger, Phys. Rev. 150, 202 (1966).

[11]A discussion of these corrections is found in Ref. 2. They are negligible as long as typical phonon energy ω_0 is much less than characteristic electron energies.

[12]J. Appel and H. Heyszenau, Phys. Rev. 188, 755 (1969).

[13]We may note that U_c, here the pertinent Coulomb parameter for superconductivity, can be very different from the effective Coulomb parameter which enters the condition for ferromagnetism. The latter is given by

$$U_{\rm eff} = U/1 + \tfrac{1}{2}U\int_{\epsilon>0}[N(\epsilon)/\epsilon]\, d\epsilon;$$

cf. C. Herring, *Magnetism IV*, edited by G. T. Rada and H. Suhl (Academic, New York, 1966), p. 222.

[14]G. Gladstone, M. A. Jensen, and J. R. Schrieffer, in *Superconductivity*, edited by R. D. Parks (Dekker, New York, 1969), Vol. 2, p.721.

[15]T. des Cloizeaux Phys. Rev. 129, 554 (1963); T. Callaway (private communication).

[16]B. T. Matthias, Phys. Rev. 97, 74 (1955).

[17]T. H. Geballe, B. T. Matthias, G. W. Hull, Jr., and E. Corenzwit, Phys. Rev. Letters 6, 275 (1961); Phys. Rev. 129, 1025 (1963).

[18]N. F. Berk and J. R. Schrieffer, in *Proceedings of the Tenth International Conference on Low Temperature Physics, Moscow*, 1966, Vol. IIA, edited by M. P. Malkov (Proizvodstrenno-Izdatel'skii Kombmat, VINITI, Moscow, 1967), p. 150.

[19]S. Doniach, Phys. Rev. Letters 18, 554 (1967).

[20]N. F. Berk and J. R. Schrieffer, Phys. Rev. Letters 17, 433 (1967).

PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Surface Density of Normal Metal in the Intermediate State of Superconducting Aluminum†

G. A. Wilkinson

University of Pittsburgh, Pittsburgh, Pennsylvania 15213

(Received 31 March 1971)

Surface-impedance measurements have been made at 0.5 MHz on the intermediate state of aluminum and indicate an increased fraction of normal metal near the surface as compared with the bulk. This result confirms Landau's prediction of the surface structure in the intermediate state and suggests a similar interpretation for Pippard's early observation of the surface resistance of tin at 10 GHz. Surface-impedance techniques are thus seen to provide accurate information on domain broadening near the surface, and to corroborate the effective-field reduction there suggested qualitatively by bismuth-probe and magneto-optic methods.

The nonbranching Landau theory of the intermediate state[1] of an infinite parallel-sided slab of a type-I superconductor in a magnetic field H applied normal to the slab surface postulates a structure consisting of alternating laminar layers of normal and superconducting metal parallel to the field direction. The azimuthal orientation of the parallel-layer structure is arbitrary, but can be imagined to be set up, in practice, by a slight tilt of the applied field from the direction normal to the slab. The thicknesses a_N and a_S of the normal and superconducting domains give rise to a total repeat distance a whose scale depends on the surface energy $+\frac{1}{2}\mu_0 H_c^2\Delta$ between the normal and superconducting phases as follows:

$$a = a_N + a_S = [\Delta d/\phi(h)]^{1/2} , \qquad (1)$$

where d is the slab thickness, $h = H/H_c$, and $\phi(h)$ is a function calculated by Lifshitz and Sharvin[2] having a maximum value of about 0.022 near $h = 0.45$. For an aluminum slab 1 cm thick, Faber's[3] value of $\Delta = 1.8 \times 10^{-4}\ (1 - t^4)^{-1/2}$ cm yields a minimum repeat distance of about $0.9(1 - t^4)^{-1/4}$ mm. This result (1) is based on Landau's calculation of the shape of the normal-superconducting interface as that surface on which the tangential magnetic field is just H_c. The interface is rounded towards the superconducting phase within a depth $\sim a \ll d$ from the slab surface on account of the demagnetizing effect of the sharp edge which would otherwise exist at the junction of the interface and the slab surface. The detailed expression for the profile $y(x)$ of the interface is given implicitly by equations in a parameter ζ:

$$x = \pi^{-1} ah \{\cosh^{-1}(\zeta/\zeta_0)^{1/2} - (\zeta_0 + 1)^{1/2} \cosh^{-1}[\zeta(\zeta_0 + 1)/\zeta_0(\zeta + 1)]^{1/2}\} , \qquad (2)$$

$$y = \tfrac{1}{2} a_S - \pi^{-1} ah\, \zeta_0^{1/2} \tan^{-1} \zeta^{-1/2} ,$$

where x is the distance measured normally into the slab, y is the position of the interface measured normal to the laminae relative to the central plane of the adjacent superconducting lamina, and $\zeta_0 = \frac{1}{4}(h^{-1} - h)^2$. At the junction of the interface and the slab surface, $x = 0$ and the parameter $\zeta = \zeta_0$, and the broadening of the normal domains near the surface leads to a fraction f_N' of normal metal at the surface which exceeds that in the bulk ($f_N = a_N/a = h$) by

$$f_N' - f_N = [a_S - 2y(0)]/a = \pi^{-1}(1 - f_N^2) \tan^{-1}[2f_N/(1 - f_N^2)]. \qquad (3)$$

Near the normal state $f_N \to 1$ this yields the result

$$f_N' = 1 - (\tfrac{1}{2} + 2/\pi)(1 - f_N)^2 + O\left((1 - f_N)^4\right) ,$$

so that the approach to the normal state occurs with zero gradient in f_N. We shall see later that this characteristic gives rise to a horizontality in the plot of surface impedance versus field at H_c. The result (3) is actually insensitive to the *absolute* scale of the thicknesses of the laminae and thus also to the surface energy, depending only on the *relative* scale f_N, since this is also the case for the shape of the interface. The theory strictly applies only to the case of a slab, but we shall have occasion to compare it with the case of a cylinder in transverse field, for which

$$f_N = 2h - 1 \quad (\tfrac{1}{2} < h < 1) .$$

We have made measurements of the surface reactance of an electropolished aluminum cylindrical single crystal of residual resistivity ratio equal to 10^4, of diameter 9 mm, length 15 cm, and orientation [100] situated in a magnetic field transverse to the axis of the crystal, and parallel to a [100] direction. The ends of the crystal were rounded to help reduce magnetic hysteresis and flux trapping. The specimen was installed in a conventional He^3 cryostat, and the impedance measurement made in terms of the resonant frequency of a transistorized Clapp oscillator whose tank circuit consisted of a copper coil wound on a former around the specimen and shunted by a 2000-pF capacitor so as to resonate at about 0.5 MHz. The coil was wound of 325 turns of 36 AWG copper wire with a gap separation from the specimen surface of about 0.5 mm and had a high-frequency inductance $\sim 50\ \mu$H. The resonant frequency could be measured to ~ 0.1 Hz using a Hewlett-Packard Model 5235B crystal-controlled counter, and is such that a change in frequency is proportional to the change in surface reactance of the specimen:

$$\delta\nu = -\,\delta X/\mu_0\gamma , \qquad (4)$$

where γ has the dimensions of a length, and depends on the coil geometry. In passing, we should note that there was in practice an extra small, but not negligible, contribution to Eq. (4) arising from the changing surface resistance of the intermediate state which was sufficient to change the loading of the oscillator. This is a disadvantage of this "active" type of measurement which does not afflict a "passive" measurement of the coil reactance. However, we shall see that this effect should not significantly affect the present measurements where the change in impedance of the intermediate state is due solely to the change in the fraction of normal metal of fixed surface impedance. The change in resistance is always proportional to the change in reactance, and thus produces to first order a change in frequency also proportional to the change in reactance, the only result of which is to modify the effective coil constant γ slightly without altering the relative scaling of the frequency shift throughout the intermediate state.

In Fig. 1 we show the results of measurements of the frequency shift as a function of the transverse field applied by means of a large pair of Helmholtz coils external to the cryostat, at a specimen temperature of $t = T/T_c = 0.298$. The vertical arrows show the value of H_c (and hence $\frac{1}{2}H_c$) measured as that field applied parallel to the specimen axis just sufficient to take the oscillator frequency to its normal-state value in increasing field, and the straight line represents the reactance expected on the basis of the fraction f_N of normal metal in the bulk. It will be seen that in increasing fields there is some superheating just above $\frac{1}{2}H_c$ and in decreasing fields some supercooling just below H_c similar

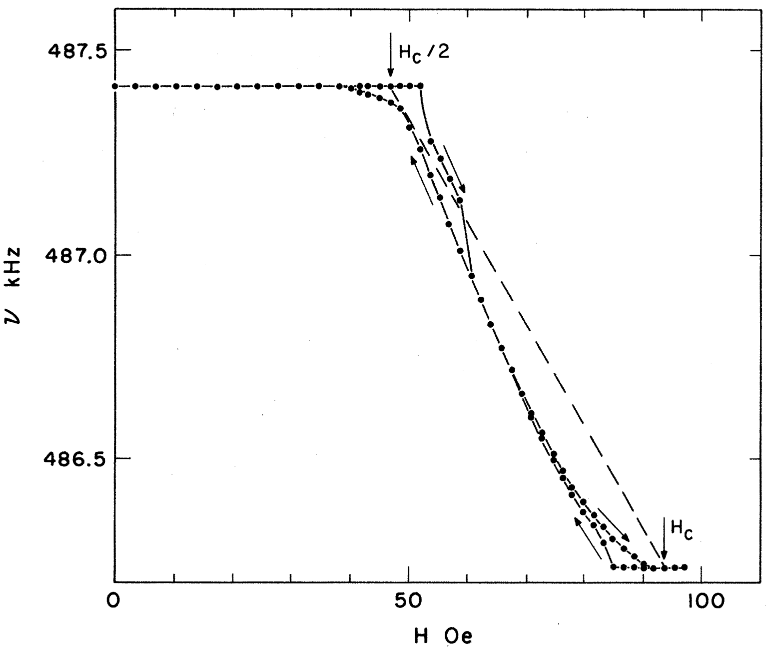

FIG. 1. Resonant frequency in increasing and decreasing transverse magnetic fields at $T/T_c = 0.298$.

to the behavior of the magnetization of the intermediate state noted by Garfunkel.[4] It is reasonable to consider the equilibrium curve to be approximated by the field-decreasing curve below about $\frac{3}{4}H_c$ and by the field-increasing curve above $\frac{3}{4}H_c$. It will be noted from Fig. 1 that the former section of curve shows some flux trapping below $\frac{1}{2}H_c$, while the latter section approaches the normal state almost horizontally, making the determination of H_c from this curve difficult. Regarding the flux trapping below $\frac{1}{2}H_c$, it should be said that in this region flux expulsion was still occurring, albeit slowly, with a time constant of the order of 10 min, and that if one had waited long enough, the curve would probably have approached the ideal; typically, data were taken at 4–5-min intervals in this region. This feature became much diminished at higher temperatures, and in any case the portion of the curve just before flux trapping starts appears to extrapolate correctly to $\frac{1}{2}H_c$. The supposed equilibrium curve at this and other temperatures seemed quite reproducible from run to run, and it would appear reasonable to regard it as truly representing the equilibrium behavior.

The most noticeable feature of the curve is its deviation from the straight-line behavior towards the normal-state reactance. In Fig. 2 we replot these results together with those for two higher temperatures in terms of a modified fraction f'_N of normal metal. There is no significant temperature dependence observable. Measurements made on a chemically polished aluminum sample of similar dimensions and purity to the [100] crystal, but containing several large crystallites of random orientations, showed almost identical behavior to that reported in the present results, suggesting that they are insensitive to differences in crystal orientation and surface-polishing procedure. There was also no change in behavior on doubling the rf measuring field [$\sim 5\times10^{-2}$ Oe $\sim 5\times10^{-4}H_c(0)$] set up by the oscillator parallel to the specimen axis; this would

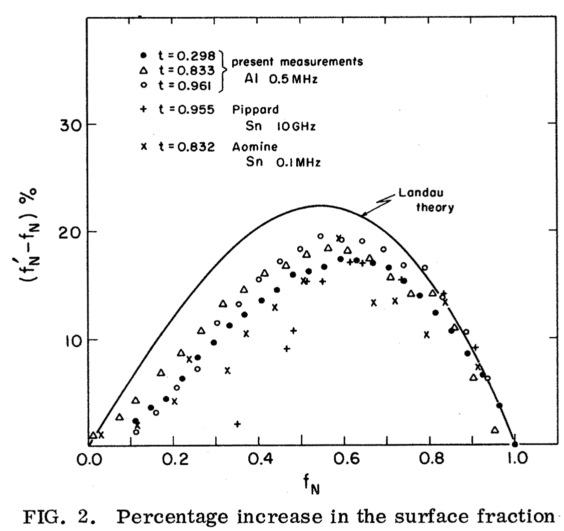

FIG. 2. Percentage increase in the surface fraction of normal metal in the intermediate state as a function of the bulk value. The points show the present results on Al at 0.5 MHz for $T/T_c = 0.298$, 0.833, and 0.961; also shown are Pippard's surface resistance results on Sn at 10 GHz for $T/T_c = 0.955$, and Aomine's surface reactance results on Sn at 0.1 MHz for $T/T_c = 0.832$. The curve is the Landau theory for an infinite slab.

argue against any second-harmonic effects of the type observed in indium by McLachlan[5] introduced by oscillation of the laminae at this frequency.[6] We should like to suggest that the explanation of these results may well lie simply in the geometrical increase in the fraction of normal metal present at the specimen surface owing to the demagnetization of the superconducting domains, since the rf currents flow within a skin depth (~ 2.7 μm for aluminum of RRR = 10^4 and at 0.5 MHz) of the surface in the normal domains and within a penetration depth (~ 50 nm) in the superconducting domains. Both these depths are considerably less than the region over which the interface is rounded ($\sim a \sim 0.9$ mm). For comparison,[7] in Fig. 2 is also shown the result (3) of the Landau theory for an infinite slab, together with some measurements by Pippard[8] of the surface resistance at 10 GHz of a pure-tin cylinder 93 μm in diameter and 1.4 cm long, at $T/T_c = 0.955$. At higher fields his results agree well with ours, but at lower fields they fall below ours, since in decreasing fields his transition to the superconducting state behaved nonideally, occurring at about $0.55H_c$. In addition, it should also be remarked that the diameter of Pippard's specimen was sufficiently small that the thickness of the laminae estimated from Eq. (1) is about 35% of the specimen diameter at $h = 0.5$. Thus at lower fields his specimen may be approaching the region where the superconducting domains become thicker than the specimen and the phase interfaces become nearly flat. In this limit $f'_N \to f_N$, but in the absence of a theory of the interface shape as a function of slab thickness it is hard to know how important this effect is for Pippard's specimen. Pippard's conclusion that in increasing fields the transition to the normal state occurred at $0.94H_c$ may be accounted for by the horizontality of his curve near H_c also noted in our measurements and predicted by the Landau theory.

Neither our results nor Pippard's show complete agreement with the Landau theory especially at lower fields; our maximum deviation from linearity is about + 18% at $f_N \sim 0.6$ compared with about + 22% at $f_N \sim 0.55$ for the Landau theory. However, perfect agreement is scarcely to be expected on account of the differing geometries in the two cases. Qualitatively, one might expect the rounding of the laminae to be diminished at those parts of the cylinder surface parallel to the applied field, occasioning a decrease in the excess of normal metal at the surface. The deviation at low fields conceivably might also arise from the onset of branching in the narrow normal domains, though this possibility is usually discounted in superconductors of ordinary size and of as large a surface energy as aluminum. It is also rather questionable how accurately the laminar model applies to the experimental structure, since we have made no attempt to set up a laminar structure by slow rotation of the transverse field in the manner suggested by Walton.[9] Nevertheless, it is possible that this kind of measurement takes a rather more suitable average over the domains than do measurements of thermal and electrical resistivities. We believe that the absence of significant temperature dependence of the excess reactance probably argues in favor of a geometrical cause rather than the possibility of boundary scattering which is known to be important for thermal[9] and electrical[10] resistivities. In this respect there is an important difference between our measurements and Pippard's, since in ours the rf currents flow parallel to the normal-superconducting interfaces, while in Pippard's the current flow was normal to the interfaces. In the latter case boundary scattering could be operative, although the relative agreement with our results argues against this.

Finally, comment must be made on whether other means of investigating the relative scale of the surface structure in the intermediate state, such as the bismuth-probe and magneto-optic techniques, can reveal the surface domain broadening. It has been suggested[11] that the latter is indeed responsible for the fact that apparent surface fields sometimes as low as $0.1H_c$ in the normal domains have been observed using these two techniques. No quantitative analysis of these effects appears yet to have been attempted, however, since the resolution of most experiments to date has been limited by probe size or magneto-optic film thickness which are often not much smaller than the domain thickness. The effect could arise from an unresolved branching in the apparently normal regions. We should like to recommend surface-impedance measurements of the type discussed here both as being highly sensitive and as taking a very suitable average over the surface domain structure.

We wish to thank F. H. Rogan for preparing the aluminum sample, and Professor M. P. Garfunkel for a stimulating discussion of the results and their interpretation.

†Work supported by the National Science Foundation.

[1]L. D. Landau, Phys. Z. Sowjetunion 11, 129 (1937); L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, New York, 1960), pp. 182–185.

[2]E. M. Lifshitz and Yu. V. Sharvin, Dokl. Akad. Nauk SSSR 79, 783 (1951).

[3]T. E. Faber, Proc. Roy. Soc. (London) A248, 460 (1958).

[4]M. P. Garfunkel, Phys. Rev. 87, 108 (1952).

[5]D. S. McLachlan, Physik Kondensierten Materie 7, 226 (1968).

[6]It should be noted that significant oscillation of the laminae is not to be expected under the conditions of the present experiment, as is also the case for the results of other workers to be quoted later, since in all these instances the frequency used is sufficiently high that the normal-state skin depth $\delta \ll a$. In this limit, the length of the curved phase interface within a skin depth of the surface may be calculated from Eq. (2) to be $\sim (a\delta 2)^{1/3}$. The oscillation amplitude of this section of interface is limited by eddy-current damping in the adjacent normal domain [A. B. Pippard, Phil. Mag. 41, 243 (1950)], and is expected to be $\sim \delta H_{\mathrm{rf}}/H_c$. This gives a fractional contribution to the surface impedance $\sim (\delta/a)^{2/3} \sim 2\%$ for the present measurements.

[7]It has recently come to the author's attention that the measurements by Aomine [T. Aomine, J. Phys. Soc. Japan 25, 1585 (1968)] of the surface reactance at 0.1 MHz of a pure tin cylinder of 3-mm diameter using a method similar to that described here appear to show the same effect. These results are also included in Fig. 2 and, although somewhat scattered, are in good agreement with the data on aluminum.

[8]A. B. Pippard, Proc. Roy. Soc. (London) A203, 210 (1950).

[9]A. J. Walton, Proc. Roy. Soc. (London) A289, 377 (1965).

[10]I. L. Landau, Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu 11, 437 (1970) [Sov. Phys. JETP Letters 11, 295 (1970)].

[11]J. D. Livingston and W. De Sorbo, *The Intermediate State in Type I Superconductors*, Vol. 2, of *Superconductivity*, edited by R. D. Parks (M. Dekker, New York, 1969), Chap. 21, pp. 1251 and 1252.

PHYSICAL REVIEW B VOLUME 4, NUMBER 7 1 OCTOBER 1971

Flux-Flow and Fluctuation Effects in Granular Superconducting Films*

P. M. Horn and R. D. Parks

Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

(Received 23 April 1971)

Results are reported on the flux-flow state in granular aluminum films. Anomalously small values of depinning current are reported and attributed to the fact that the temperature-dependent coherence length is appreciably larger than the grain size in these films. Near T_c, the energy dissipated in vortex flow D ($D \propto d\rho_f/dH$, where ρ_f is the flow resistivity) was found to vary exponentially with temperature. This anomalous temperature dependence suggests that the major source of energy dissipation in this regime is the interaction of the vortex current fields with thermodynamic fluctuations. For sufficiently large values of $R_{\square}^{N}/\epsilon$ [$R_{\square}^{N}$ = normal-state resistance per square and $\epsilon = (T_c - T)/T_c$], the ρ_f-vs-H curves were found to be nonlinear in a manner not reported before in conventional flux-flow experiments. This curvature is attributed to the above-mentioned interaction between the vortices and fluctuations.

I. INTRODUCTION

Resistivity and dissipation in the flux-flow state of superconductors have been the subject of much theoretical and experimental work.[1] In this paper we discuss the low-field flux-flow characteristics of granular Al films and contrast our results with the work by other authors on both bulk samples[1] and thin films.[2-5]

Section I of this paper will serve as an introduction and a brief review of the pertinent work of the past. In Sec. II we discuss current-voltage characteristics including (i) pinning at low current densities and (ii) nonlinear effects at high currents. Section III contains work on the flux-flow resistivity, and Sec. IV discusses the interplay of flux-flow and fluctuation effects close to the superconducting transition temperature T_c.

In the presence of an applied magnetic field H a bulk type-II superconductor or a thin superconductor in a perpendicular magnetic field allows flux to penetrate in the form of quantized Abrikosov vortex lines. Then if a transport current of sufficient magnitude is applied, steady-state vortex flow of velocity v is obtained where v is determined by equating the net force on a vortex $f = f_L - f_P$ (f_L being the Lorentz force $J\phi_0/c$ and f_P the pinning force) to a viscous drag force ηv:

$$\eta v = J\phi_0/c - f_P , \tag{1}$$

where $\phi_0 = hc/2e$ is the flux quantum. The electric field E_0 arising from motion of the flux lines is given by

$$E_0 = n(v/c)\phi_0 = (v/c)H , \tag{2}$$

where n is the vortex density such that $n\phi_0 = B \cong H$ (in a situation such as the present one where the macroscopic diamagnetism of the sample can be ignored). Defining the pinning independent-flow resistivity ρ_f as $\partial E_0/\partial J$, we obtain

$$\rho_f = \phi_0 H/\eta c^2 = DH/J^2\phi_0 , \tag{3}$$

where D is the total power dissipated in vortex flow. Most of the interesting physics lies in un-