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Weak Electrolytes, Brownian Motion, Vortices in Superfluid Films, and Odins Aker

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A brief sketch of the author's days at Yale as Lars Onsager's last physics student is followed by the contributions of the Onsager school to our current understanding of persistent current decays and vortex pinning in very thin superfluid films. The resulting theory is an interplay of three subjects that were dear to Onsager's heart: electrolytes, vortices in superfluids, and Brownian motion. The discussion also surveys a topic of current interest, the role played by defects and boundaries in producing the "stiffness" that characterizes superfluids. The article ends with a few words about the author's connection to Norway.

KEY WORDS: Quantized vortex; superfluid; electrolyte; Brownian motion; dissociation/recombination; vortex pinning.

1. ONSAGER'S LAST PHYSICS STUDENT

The first time that I met Lars Onsager was in April 1965, at the University of Kentucky, where I was a senior physics major heading for graduation and then to Yale. The First (and last) Kentucky Conference on Phase Transitions was organized by an extremely inspiring lecturer, a theoretical astrophysicist named Wendell C. DeMarcus.[1] DeMarcus, a former east Tennesseean, was the reason that I (an east Kentuckian) was headed for Yale. He later told me that he had probably spent more research time (and a lot of enjoyable private time as well) with Lars than anyone else had. Nothing of theirs was ever published, as the work was done at places like Oak Ridge and Livermore and was classified. DeMarcus was very conservative, I had strong left-wing sympathies and later became politically active, but in spite of our mutual antipathy for each other's politics we had

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a very strong and very good relationship until he died prematurely young of a heart attack in 1983.

The next time I met Onsager was two years later, after I had passed the Ph.D. Qualifying Exam at Yale. It was time to choose a thesis advisor, and DeMarcus was there on sabbatical visiting his colleague and former thesis advisor Rupert Wildt. He encouraged me strongly to go to Lars and ask him if he had an interesting problem for me to work on.

As I quickly learned, it was dangerous to visit Lars: he was always available and held marathon sessions in which he talked to students about whatever problem he happened to the thinking about, as if we were at his intellectual level. After about an hour of his description of ice (John Nagle's name kept popping up, as it did in his University of Kentucky lecture), he finally stopped describing the physics and asked me: "Do you know why the dielectric constant of water is so high?!" I mentally surveyed the escape route to the door, but remained glued to my seat and managed a very weak, "No." I was terrified that he would ask me another question that would require a real answer, but he only continued to describe the ice problem. Finally he paused long enough to give me an opening (he was always relighting his pipe because, once he won the struggle to light it, he perennially forgot to draw enough to keep it lit). I quickly asked him what I would need to know in order to work on such a problem. "Only a little bit of partial differential equations!" he replied with his famous mischievous grin.

Finally, I asked for and received copies of two articles[2,3] that illustrated the method I would need to learn in order to work on the ice problem. Those two articles were *uralt* and looked extremely boring: no creation and annihilation operators, no group theory, not even any quantum mechanics. Only a description of old-fashioned Brownian motion theory applied to weak electrolytes! There was only one basic equation, the Smoluchowski equation[3]

$$\frac{\partial}{\partial t} = D\nabla \cdot (\nabla f + f\nabla u/kT) \tag{1a}$$

[3] For two charges of opposite sign in a combined Coulomb and external potential, he solved this equation exactly in three dimensions to calculate the electric field dependence of the dissociation and recombination rates. Prior to that time, some chemists had spculated that the enhanced conductance of weak electrolytes in an electric field could not be accounted for by the Coulomb attraction alone! Furthermore, when the Chemistry Department at Yale asked him for piece of work to call his "dissertation" so that he would not be an un-degreed professor, it was the mathematics of this piece of work that he handed to them. They didn't understand it and gave it to the Mathemtics Department for an evaluation. When the Math Department offered to award him the degree, Chemistry did not hesitate further and awarded it first. The dissertation lies in Yale's Rare Book Collection, the Bieneke Library.

I was depressed and went back to DeMarcus, whose lectures on classical theoretical physics I was attending in the Astronomy Department while learning about continuous groups and Lie algebras in a physics course. I told him the problem Lars had described (I did not dare call him Lars—he was always "Professor Onsager" to me) looked too boring to me. He recommended that I go back to "Lars" (Wendell was on an easy first-name basis with him) and ask him if he had a problem in superfluids. So I did.

Onsager did not hold it against me when I told him that the problem he had described was not for me, and even reassured me, looking quite serious, that one should *only* work on problems that look interesting to a person. When I asked if he had a problem in superfluid physics, he said, "Well, yes, there is this little problem with vortex lines in helium." As he described the problem, it sounded interesting enough, but when I asked for articles that would explain the method of approach, he gave me exactly the same two articles as before! That was the beginning of my long and initially unwanted association with weak electrolytes and Brownian motion theory.

The next fall, Onsager got the Nobel Prize in chemistry. Many of my fellow students congratulated me as if I had gotten the prize, or at least had something to do with it. I thought that was a bit strange, but that luck stayed with me for a long time, as colleagues always enjoyed introducing me as "Onsager's student." Apparently, one became elevated in other peoples' minds just by the mere fact of association with Lars. When finally, after many years, the association ceased to be announced, I knew that I had developed a strong enough reputation of my own (in Norway, it is still mentioned from time to time that I am Onsager's last physics student, but not in the countryside, where no one has heard of Lars, and where you are judged strictly according to your own faults, a democratic aspect of Norwegian life that Lars himself was much in tune with).

Conveniently enough, I graduated just as Lars retired. He recommended that I spend a few years in Europe, but I did not understand how that would work. He said, "Oh, we'll just call up and get you a Fullbright!" This was not a joke, for he also told me that he regarded my dissertation as a lot better than most of the stuff that came out "next door," meaning the Physics Department, of which I was a member. His recommendation was to go to Cambridge, Leiden, Göttingen, or Rome. Apparently, he thought that I should spend some time talking to experimenters. Unfortunately, as he later found out, Fullbrights must be applied for in the preceding fall, not in the later spring, so I took a postdoc at Rutgers with Mike Stephen and Elihu Abrahams instead, enduring the pain of the New Brunswick area in favor of excellent theoretical physics, and also deferring my European experience until much later in life (my first trip outside the

United States was to Norway). Lars had also invited me to accompany him to Coral Gables as his postdoc, and it was a little sad when, one day as he sat reading a journal in the Chemistry Library, I finally got up the courage to tell him I had decided to go to Rutgers instead. His response, smiling in a slightly sad but still congratulatory way, was: "Oh,... so you're leaving the program." I was, in that minute, extremely proud to have been a part of "the program."

2. WEAK ELECTROLYTES IN TWO DIMENSIONS

Two years later, just before I left Rutgers for Houston, I made copies of two articles by Kosterlitz and Thouless[4] on the two-dimensional Coulomb gas and a related problem of vortices in the plane. It was equilibrium statistical mechanics, and they had used the renormalization group (which had learned about at Rutgers) to predict that there is a critical temperature T_c such that for $T < T_c$ all charges of opposite sign are bound pairwise so that the gas becomes completely neutral. They also argued unconvincingly that the response to a weak external electric field should be dielectric. Because by necessity I had had to understand much more than I wanted to about weak electrolytes, I knew very well that two thermally associated charges could dissociate in the presence of a weak electric field (the so-called Wien effect[2]). During my first summer in Houston, I used Eq. (1a) and Lars' method to calculate the capture and escape rates approximately, to lowest order in a weak external electric field, in order to back up my speculation quantitatively. In Onsager's words, I had used a "hammer and tongs method," matching the leading terms in asymptotic expansions for the short- and long-distance solutions of a boundary-value problem at the saddle point of the potential of the combined electric and Coulomb fields.

I sent the article to *Physical Review Letters* and finally got back the following responses: one referee wrote that "This isn't physics, he's only solving boundary value problems." Another said that the article was too late, that interest in the Kosterlitz–Thouless theory had come and gone so that the article was no longer "topical" enough to warrant publication in such a prestigious journal. I finally gave up and sent the article to *Journal of Physics C*, where the reviews were written in a much more polite style, really in an English style, and also pointed out that the article was both unusual and interesting. So, my results were published there[5] and, of course, went largely unread. The main result was later reproduced independently by other authors[6] who derived the correct pair kinetics for point vortices, and who eventually acknowledged the relationship to my earlier work[7,8] once I pointed it out to them.

In order to exhibit the electrolytic response of the Kosterlitz–Thouless model, and to prepare the reader for the rest of this discussion, I now sketch the Onsager method[2,9,10] of formulating dissociation–recombination problems, which is much more general than Kramers' one-dimensional treatment and even includes Kramers' method as a special case whenever "ionic dissociation" is determined by a saddle point and the current density of the escaping particle is approximately one-dimensional. Whenever the current density is not strongly one dimensional, then the Bjerrum radius[2] or some other symmetry condition must be used to define "ionic association" or "thermal association."

The pair correlation function $f(r, \theta)$ for a pair of opposite charges in two dimensions separated by a distance r satisfies the Smoluchowski equation

$$\frac{\partial f}{\partial t} = D \nabla \cdot e^{-U} \nabla f e^{U} \tag{1b}$$

where D is the diffusion coefficient and $U = u/kT$ is the dimensionless interaction energy, the potential energy of a pair of charges divided by kT. In the present case, $U = \lambda \ln(r/a) - x/x_c$ if $r \geqslant a$, where $\lambda = q^2/kT$ is the dimensionless coupling constant ($\lambda \geqslant 4$ is the condition for the spontaneous thermal collapse of free charges into bound pairs in the Kosterlitz–Thouless theory). With an external electric field E along the x axis, $x_c = q/E$ is the location of the saddle point of the potential U.

Bound pairs are accounted for by the density

$$\nu = \int f \, d^2 r \tag{2}$$

where the integration is effectively over the region $r \leqslant x_c$, and where $f \approx A r^{-\lambda}$ is a Boltzmann distribution. In order to solve the dissociation–recombination problem, one must compute the rate $P\nu$ at which the thermally-bound pairs of opposite charge escape from each other and become free, and also the rate An^2 at which free pairs of opposite charge and density n associate and so become bound at short distance $r \leqslant x_c$. The field-dependent dissociation and recombination coefficients in the kinetic equation

$$\frac{dn}{dt} = -\frac{d\nu}{dt} = P\nu - An^2 \tag{3}$$

are computed in the Onsager method by solving two separate boundary-value problems (the famous boundary-value problems mentioned above):

from the conditions $f=0$ at infinity and a finite current $J_r<0$ at the origin, one computes the escape rate

$$J_r = -Pv = -\int_C \mathbf{j}\cdot\hat{r}\,dl \tag{4}$$

where C is any closed loop that encloses one charge at $r=0$ and

$$\mathbf{j} = De^{-U}\nabla f e^{U} \tag{1c}$$

is the current density. In order to compute the rate at which free charges associate to form bound pairs, one takes as boundary conditions $f=n^2$ at infinite separation, but with a finite current

$$J_r = An^2 = \int_C \mathbf{j}\cdot\hat{r}\,dl \tag{5}$$

For the two-dimensional Coulomb problem, the approximate solution to lowest order in the external field E predicts that the free charge density n varies as $E^{\lambda/2}$ for weak fields E, which shows that the system's response is *not* dielectric but is *weak electrolytic* instead. In the language of low-temperature physics, the corresponding superfluids are not absolutely stable (an ideal fluid is absolutely stable) but are only metastable, which means vortices can be created and annihilated and that any persistent current will dissipate if you can afford to wait long enough to see it happen.

3. SUPERFLUIDS VS. IDEAL FLUIDS

A vortex in an ideal fluid experiences no force and simply moves with the local velocity of the flow. In two dimensions, there is a Hamiltonian theory of point vortices because a Hamiltonian is just the generalization to phase space of the idea of a stream function.[11] The Hamiltonian theory of vortex motions in two dimensions had been formulated in very general and very useful terms by 1943,[12] and the canonically conjugate coodinates and momenta are simply the Cartesian position coordinates of a vortex. That fact was used by Onsager to explain how large vortices in a normal fluid like the atmosphere may be formed by the coalescence of smaller ones, at high energies.[13]

The required Hamiltonian is just the potential energy of interaction of N point charges in two dimensions (which is a simple logarithmic interaction in the absence of boundaries), the circulation κ_i being essentially the "charge" of the vector field $\mathbf{v}$ which is the local velocity of the fluid. In the language of particle mechanics, the Hamiltonian is therefore purely "potential": there is no kinetic energy term for the charges because vortices are massless (the interaction energy of the charges *is* the kinetic energy of the flow field). In the language of Newton's laws, there is no acceleration term, so that the dynamics follow from force balance alone. Such a fluid experiences no dissipation at all: vortices can neither be created nor annihilated because the distance between them is conserved. This is not a model of a superfluid, because a superfluid is unstable against vortex creation and annihilation through the phase slip mechanism.[14] Another way to say it is that the distance between vortices is not conserved in a superfluid, but that alone, of course, does not guarantee superfluidity.

In order to describe the motion of a vortex in a superfluid, you must introduce a drag force on the vortex, a force opposite to its velocity, which velocity is produced by a field of the other $N-1$ vortices along with any external flow that is imposed experimentally (linear hydrodynamics). But whenever you try to impede a vortex along the x direction it experiences a lift force in the y direction (to the zeroth approximation, an airfoil is a vortex line[15]). Consequently, a pair of vortices with opposite circulation that experience drag will tend either to separate or else come together, depending upon the interplay of the vortex-pair Hamiltonian and the external flow rate. The resulting *pair* kinetics (although not the kinetics of a *single* vortex) becomes *identical* with the kinetics of a pair of Coulomb charges in an external electric field along the y axis whenever the external flow V_s is in the x direction. In other words, the effective potential energy that one needs in order to apply Eq. (1b) to vortex pair dissociation and recombination is exactly that of the corresponding Coulomb problem,

$$\begin{aligned} U &= \lambda \ln(r/a) - y/y_0 \qquad \text{if} \quad r \geqslant 1 \\ U &= 0 \qquad\qquad\qquad\qquad \text{if} \quad t \leqslant a \end{aligned} \tag{6}$$

where $\lambda = \sigma\kappa^2/2\pi kT$ is the dimensionless vortex pair coupling constant, σ is the areal superfluid density (roughly speaking, $\sigma \approx \rho_s d$, where ρ_s is the superfluid density in three dimensions and d is the film thickness), $\kappa = h/m \approx 10^{-3}\ \text{cm}^2/\text{sec}$ is the quantum of circulation, and a is the vortex core radius and is likely due to zero-point fluctuations.[16] With the externally imposed superflow along the x axis, the "effective electric field" is along the y axis and so $y_0 = \kappa/2\pi V_s$ is the location of the saddle point of the potential U, which delineates free from thermally associated vortex pairs because there is no meaningful Bjerrum radius for the two-dimensional Coulomb potential. We can take $U \approx 0$ for $r \leqslant a$ because in that case the vortices effectively annihilate each other (there is no kinetic energy of the flow field whenever two vortices with opposite circulation sit on top of each other). On the other hand, there is a thermal distribution of tightly bound pairs: at a finite temperature T, the energy required to create a vortex pair at separations $r \approx a$ is vanishingly small. Furthermore, the pair separation r is not conserved, but circulation is conserved because only pairs with opposite circulation are created via thermal fluctuations in the heat bath.

If there is no drag, the case of an ideal fluid, then two vortices of opposite circulation simply translate at constant velocity with conserved separation. In a superfluid, the drag may be provided by the substrate, by thermal excitations in the helium film, or by both. In both cases, the "charges" are massless particles.

One main lesson that I learned from Lars is that one must be very careful when comparing theoretical predictions with experimental measurements. This is the central thread that runs throughout the discussion that follows.

4. PERSISTENT CURRENT DECAYS IN THIN SUPERFLUID FILMS

Whereas Reppy's group had concentrated upon experiments with oscillating substrates, requiring a time-dependent treatment of the Brownian motion problem,[6] Eckholm and Hallock provided the only hard and reliable test of the so-called "DC solutions," namely, measurements of the decay rates for persistent currents.[17] In that case, the experimental setup is roughly as follows: first, you drive the flow across the film long enough to prepare it in a steady state where it has a spatially uniform and time-independent velocity V_{s0}. Then, you measure the velocity V_s as a function of time as it is allowed to decay. The results are shown as collections of data points in Figs. 1a and 1b for films of thickness d ranging from about six to about ten monolayers. More accurate experiments have not yet been performed, although, as we shall point out below, there is a need for them.

In order to understand the experimental results, consider first the theoretical prediction where the film is assumed to be infinite in extent, where boundaries are presumed to play no role whatsoever. In that case, Onsager's method yields

$$\frac{d\tilde{n}}{d\tilde{t}} = \tilde{V}^{\lambda} - \tilde{n}^2 \tag{7}$$

where we use dimensionless variables

$$\tilde{n} = n/n_{f0}, \qquad \tilde{V} = V_s/V_{s0}, \qquad \tilde{t} = t/\tau \tag{8}$$

$n = n(V_s)$ is the free vortex density for a spatially uniform average superfluid velocity $V_s = V_s(t)$, $n_{f0} = n(V_{s0})$ is the free vortex density whenever there is initially a steady state with velocity $V_s = V_{s0}$, $\tau = (2\pi D\lambda n_{f0})^{-1}$ is the characteristic time scale for both dissipation and free vortex density relaxation whenever a flow that is prepared in the steady state is permitted to decay, and $D \approx \kappa \approx 10^{-3}\ \text{cm}^2/\text{sec}$ is the diffusion constant for a vortex.

Integration of Eq. (7) yields the prediction for the rate at which the free vortex density decays with time. The first term on the left-hand side is the dissociation rate for bound vortex pairs and the second one is the recombination rate fo free vortices with opposition circulation. In order to go further, one must use the phase slip mechanism[14] in the form

$$\frac{dV_s}{dt} = -|\kappa|\, j_y \tag{9a}$$

to compute how the separation and escape to infinity of pairs along the y axis causes the superfluid velocity, which is taken here in the x direction, to decrease as time goes on. In (9a), j_y is the current of free vortices transverse to a superflow V_s along the x axis, and the mechanism states that one gets a phase slip of 2π in the wave function every time that a bound pair of opposite circulation dissociates and goes off to infinity along the y axis. In dimensionless variables, the phase slip equation becomes

$$\frac{d\tilde{V}}{d\tilde{t}} = -\tilde{n}\tilde{V} \tag{9b}$$

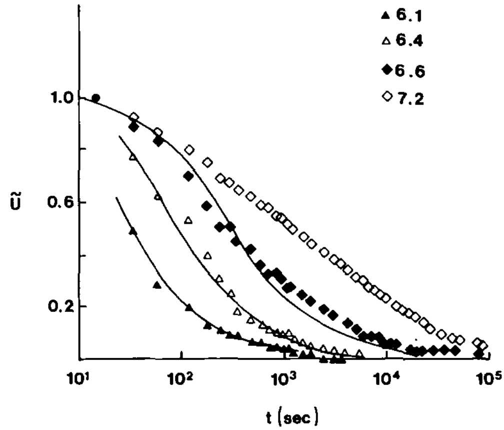
and Eqs. (7) and (9b) must be solved simultaneously. A steady-state approximation whereby one uses $dn/dt \approx 0$ is impossible because both n and V_s have the same relaxation time τ, a fact that was ignored in some early papers on the subject. In fact, the exact solution of (7) and (9b) is given by[19]

$$\tilde{n} = \tilde{V}(\tilde{n}^2(0) + 2(1 - \tilde{V}^{\lambda-2})(\lambda-2)^{-1})^{1/2} \tag{10a}$$

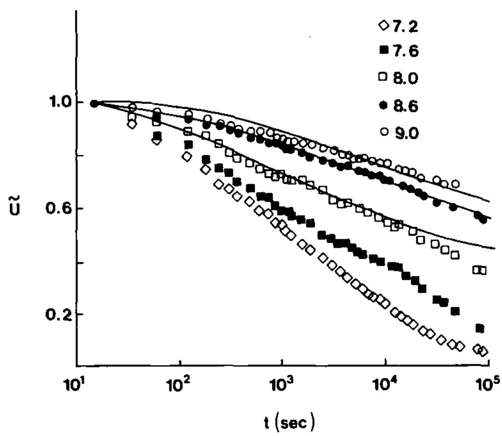
The coupling constant λ is known for various temperatures and film thicknesses from second sound measurements,[17] and $14 < \lambda < 25$ for the films under consideration. Under these circumstances, where also $\tilde{n}(0) \approx 1$ (the flow is prepared in a steady state initially), we have $\tilde{n} = \tilde{V}$, which yields

$$\tilde{V}(t) \approx (1 + \tilde{t})^{-1} \tag{10b}$$

The theoretical results are shown as the solid curves in Fig. 1a and indicate that the theory of vortex pairs in the unbounded plane accounts for the decays of the three thinnest measured films.[19]

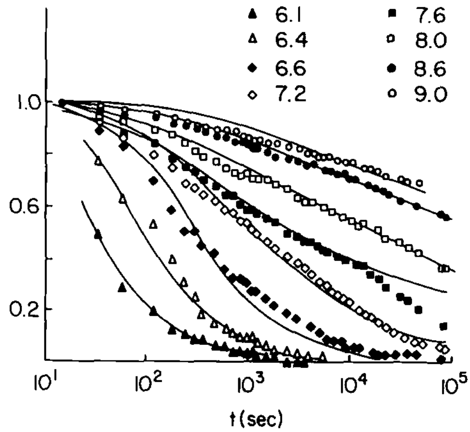


(a)



(b)

Fig. 1. (a) Experimental data (from ref. 17) on persistent current decays for films with thickness 6.5, 6.8, 7.0, and 7.7 monolayers. Only the three thinnest films decay in agreement with the long-time behavior $\tilde{V} = \tilde{t}^{-1}$ predicted by the theory of vortices in the unbounded plane (solid curves). (b) Experimental data for films with thicknesses 7.7, 8.1, 8.6, 9.2, and 9.6. The solid curves represented the effect of a single film edge and are approximately logarithmic whenever $\tilde{t} \ll 1$. (c) Experimental persistent current decays for films with thickness ranging from 6.5 to 9.6 monolayers. The solid lines represent the predictions of the theory based upon pinning sites with a single characteristic size b. The three thickest films are consistent with infinite b, which would represent the effect of a film edge, and the crossover decay $d \approx 7.7$ is now accounted for.



(c)

Fig. 1. *(Continued)*

The reason that the three thickest films cannot be fit by this results[19] is that they are in agreement with a very slow logarithmic decay (see Fig. 1b) that would follow *if* there were two different relaxation times, with the vortex density relaxing fast relative to the superfluid velocity, so that the steady-state relation $\tilde{n}^2 \approx \tilde{V}^\lambda$ would hold, yielding

$$\tilde{V} = (1 + t/\tau')^{-2/\lambda} \approx 1 - (2/\lambda)\ln(t/\tau') + \cdots \qquad (11a)$$

This result is, of course, completely impossible to obtain from the pairing theory because there is only *one* relaxation time in that theory. More important is that the crossover decays ($d = 7.2$) are not accounted for at all by either (10b) or (11a).

In order to get a second relaxation time into the theory, consider a film of finite width W with infinitely long parallel boundaries at $y = 0$ and W. Equation (7) is then replaced by

$$\frac{d\tilde{n}}{dt} = \tilde{U}^\lambda - \tilde{n}^2 + G\tilde{U}(\tilde{U}^{\lambda/2} - \tilde{n}) \qquad (12)$$

where G is dimensionless and is given by $G = V_{s0}/\kappa W n_{f0}$. For $G \ll 1$, the predictions of the unbounded film theory are recovered, as is required for the case of the thinnest films. In addition, if $G \gg 1$, then we can make the required steady-state approximation $\tilde{n}^2 \approx \tilde{V}^\lambda$ to obtain the logarithmic decays characteristic of the two thickest films, as is indicated in Fig. 1b.[20] In the limit where $G \gg 1$ the theory is equivalent to a semiinfinite film with a single infinite boundary, a single film edge at $y = 0$.[21]

Note that for the film with a thickness of 8 monolayers, a misfit with the theory begins to show up at long times, and the theory cannot *at all* be made to account for the crossover behavior exhibited by the data for 7.2 monolayers. In other words, something fairly interesting happens as the film thickness is varied, while the original idea of vortex pairs where boundaries play no important role holds *only* for the thinnest measured films, where one effectively loses the superfluidity because the velocity decays much too fast. One can well wonder for a long time why this should or even *can* be the case. The reason, which the author only realized spontaneously after describing his state of confusion to Y. Shapira one evening during dinner at the 1983 Geilo meeting, is stated near the end of this section.

The next inventive step was made by Brown and Doniach,[22] who proposed vortex pinning as the missing mechanism. Their results were also based upon Onsager's method, and they were able to fit the data with several free parameters (too many for my taste), but their method involved certain assumptions along with the neglect of a phenomenon that left their treatment of vortex pinning open to criticism.[20]

Once you decide to consider pinning, then there are two kinds of pinning sites, those with and those without a net quantized circulation. The Brown–Doniach approach was later shown to be equivalent to treating the former in a certain approximation that is not altogether consistent aside from the fact that it ignores the charged sites altogether. What is beyond criticism is their idea that pinning was the right thing to look at.

A pinning site is essentially any region in the film where the superfluid density, due to substrate defects, is significantly reduced locally from its average value σ. As an idealization that makes the theory tractable, Brown and Doniach restricted their theoretical considerations to sites where the superfluid density completely vanishes over a closed region of characteristic diameter b. To handle these internal boundaries better than they did, one must use Lin's Hamiltonian theory.[12] Using Lin's method, one derives the interaction energy of a vortex with the (images in the) internal boundary that one needs in order to write down the Smoluchowski equation:

$$U_{\text{eff}} = \sigma\kappa[\psi_0(\mathbf{r}) + \kappa g(\mathbf{r}, \mathbf{r})/2] \tag{13a}$$

where

$$g(\mathbf{r}, \mathbf{r}) = \lim_{\mathbf{r}' \to \mathbf{r}} \left[G(\mathbf{r}, \mathbf{r}') - \frac{1}{2\pi} \ln |\mathbf{r} - \mathbf{r}'| \right] \tag{13b}$$

G is the stream function for a single vortex,

$$\Delta G(\mathbf{r}, \mathbf{r}') = -\frac{1}{2\pi}\,\delta(\mathbf{r}-\mathbf{r}') \tag{14}$$

and must satisfy the condition that it is constant on all internal and external boundaries of the fluid, a pinning site being simply an internal boundary. ψ_0 is the stream function for a uniform superflow past the pinning site in the absence of vortices, and therefore satisfies Laplace's equation with the boundary condition of constancy at fluid boundaries. Far from any pinning sites,

$$\psi_0(\mathbf{r}) \approx -\hat{\mathbf{z}} \times \mathbf{V}_s \cdot \mathbf{r} \tag{15}$$

Consider next only circular sites with radius b, meaning that we assume that the pinning centers can be characterized by a single parameter. If a site is "uncharged" meaning that it has not trapped a free vortex and therefore has no net circulation, then any free vortex is attracted to that site via a weak dipolar interaction, which we can write in polar coodinates as

$$U_{\text{dip}} = \frac{1}{2}\ln\left(1-\frac{b^2}{r^2}\right) - \frac{\sin\theta}{y_0}\left(1-\frac{b^2}{r^2}\right) \tag{16}$$

If, on the other hand, a site has already trapped a vortex, then that site has the circulation of the trapped vortex and has a dimensionless interaction energy

$$U_{\pm} = \frac{1}{2}\ln\left(r-\frac{b^2}{r^2}\right) - \frac{\sin\theta}{y_0}\left(r-\frac{b^2}{r^2}\right) \tag{17}$$

with any other free vortex. Changed sites becomes dipolar (neutral) via dissociation, and vice versa. Now, we must integrate the system of coupled kinetic/phase slip equations

$$\begin{aligned} d\tilde{V}/dt &= -\tilde{n}\tilde{V} \\ d\tilde{n}/dt &= [n_0^2(2\pi a V_{s0}/\kappa)^{\lambda}\,\tilde{V}^{\lambda}/\tilde{n}_{f0}^2 - \tilde{n}^2] + G[\tilde{n}_f^2 - \tilde{n}^2] \end{aligned} \tag{18}$$

where

$$G = A_{\pm} A_{\text{dip}} N_b [\pi D \lambda (R_{\pm} + R_{\text{dip}} + A_{\pm} n + A_{\text{dip}} n)]^{-1} \tag{19}$$

is a time-dependent dimensionless factor that measures the influence of pinning relative to vortex pair processes. Here, A_{dip} and R_{dip} are the recombination and dissociation coefficients for dipolar sites with density N_{dip} (sites with no net circulation), while $A_{\pm}$ and $R_{\pm}$ are the corresponding coefficients for capture and escape of vortices from pinning the sites with density $N_\pm$ and a net circulation $\pm\kappa$. $N_b = N_{\text{dip}} + N_\pm$ is the total pinning site density and is conserved. Also,

$$n_F(V_s) = (R_\pm R_{\text{DIP}}/A_\pm A_{\text{DIP}})^{1/2} \tag{20}$$

is the *steady-state* prediction for the free vortex density in the presence of a steady flow V_s, and $\tilde{n}_f = n_f(V_s)/n_f(V_{s0}) = n_f(V_s)/n_{f0}$ is the dimensionless free vortex density in that steady state.[23] The kinetic equation for the rate at which pinning site occupancy changes with time has been eliminated in favor of a steady-state relation because a comparison of relaxation times shows that that density relaxes rather rapidly relative to the free vortex density and superfluid velocity $\tilde{n}$ and $\tilde{V}$. Furthermore, since $N_\pm/N_{\text{dip}} \approx 10^{-13}$ under the conditions of the experiment, we have also set $N_\pm \approx 0$.[23]

If $G \ll 1$, then pinning can be ignored. In this case, the superfluid velocity and free vortex density are characterized by bulk pair processes with the same relaxation time, yielding [see (10b)]

$$\tilde{V} \approx (1 + t/\tau)^{-1} \tag{10b}$$

which describes the three thinnest films. The comparison with experiment is shown as the three solid curves for $d = 6.1$, 6.4, and 6.6 in Fig. 1c.[23] As we noted above, the relaxation to normal behavior is so fast in the absence of pinning that these films do not show a superfluid response. One cannot use an equilibrium theory like the Kosterlitz–Thouless theory to decide whether a given system is or is not a superfluid; in modern language, one must have a "stiffness," which is a nonequilibrium idea. In other words, the effects of substrate defects or boundaries are absolutely essential in order to get the slow logarithmic decay[18] characteristic of the "stiffness" that we call "superfluidity."

If $G \gg 1$, then the pinning terms drive the free vortex density into a quasi-steady state where [see (11a)]

$$\tilde{V} \approx (1 + t/\tau')^{-2/\lambda} \approx 1 - (2/\lambda)\ln(t/\tau') + \cdots$$

holds and yields the fits shown for $d = 8.6$ and 9 in Fig. 1c. For $d = 8$, the observed decay is logarithmic, but the theory deviates from logarithmic behavior at large times: with the relaxation time required to make the solution logarithmic at short times and thereby fit most of the slope, the logarithmic approximation in (11a) fails at large times where

$$\tilde{V} \approx (1 - t/\tau')^{-2/\lambda} \approx (t/\tau')^{-2/\lambda} \tag{11b}$$

must be used instead. Whether the same misfit of theory and experiment might well arise for the two thicker films if the measurements were extended to much larger times is unknown, as the required more accurate extention of Eckholm and Hallock's experiments has not been carried out.

In order to describe the smooth crossover from bulk pair processes and rapid decay to the pinning-dominated superfluid regime, it is necessary to assume that the pinning site radius increase from $b \approx 10$ Å for the films with thicknesses $d \approx 6.1$–7.6 monolayers, is 25 Å for 8 monolayers, and can be set equal to any number greater than or equal to 500 Å for the two thickest films (including infinity, suggesting that film edges and not internal pinning may well begin to take over and dominate in vortex creation and annihilation).[23] The result is shown for $d = 7.2$ in Fig. 1c. For $d = 7.6$, the long-time behavior predicted by the theory is roughly logarihmic and (still) does not agree with the experimental data.

Now, we arrive at the answer to a very interesting question: what is the reason for the rapid change by several orders of magnitude in the dimensionless control parameter G as the film thickness d is varied from 6 to 10 monolayers at fixed temperature T or, dimensionlessly stated, while the dimensionless vortex pair coupling constant λ is varied from 14 to 25? Aside from factors that are only details,

$$G \approx \frac{N_b V_{s0}}{\kappa} e^{C\lambda} \tag{21}$$

where $C\lambda \approx E_c/kT$ is essentially the dimensionless core energy of a vortex and is roughly proportional to d, the length of vortex line (bent lines with significant curvature are energetically so expensive to create as to be negligible). Therefore, the reason why pinning or, more likely, edge effects dominate the thick films is simply that it is a lot less expensive to create one length of vortex line near a boundary compared with two in the bulk. Fluctuating bound vortex–image pairs are created thermally near a boundary, and there is a Boltzmann distribution of them (circulation is conserved on the average), just as fluctuating tightly bound pairs are created as thermal equilibrium fluctuation in the bulk. Conversely, it becomes easy enough to make pairs with shorter lengths of vortex line in the thin films.

So far, λ has been varied at constant temperature T only by varying the film thickness d. But as the result (21) is based upon dimensionless variables, we see that one should be able to create the crossover from pair-dominated to pinning-dominated vortex kinetics by varying the temperature T while holding the film thickness d fixed. So far as we know, this prediction remains untested by an experiment. Another interesting possibility is to make better superfluids from thin films by making the substrate microscopically dirty enough, but one would need sites with a characteristic radius about 500 Å in order to get the logarithmic decays that are the signature of superfluidity.

Finally, although the crossover phenomenon discussed above is not sharp and is certainly not a phase transition, it is energetically analogous to the crossover from a grain-boundary-dominated first-order transition (low dislocation core energy) to a hexatic phase (large dislocation core energy) in the theory of two-dimensional melting.

5. NORWAY

In 1983, I traveled to Norway for the first time. The change fell to me because Arne Skjeltorp's name, which I remembered from Yale, appeared on a poster advertising the famous Geilo meeting which takes place in odd years in April. I was interested in visiting Norway, not because of Onsager, but because of a deep interest in the culture that gave birth to my surname. It is the anglicized form of "MacAmhlaib" (and worse spellings), which is only the old Celtic translation of Olafson, meaning ancestors' relic (Lars' slightly younger brother Per told me this when I visited him in Oslo in 1985, but I already knew it by that time). As another physicist once put it, the name comes from the times when Norwegians were persistent visitors to the British Isles.

"Onsager" is the (Danish-influenced) Bokmål spelling of Onsaker, which was taken from the name of the old family farm (there is such a place-name near Hønefoss, but I do not know if it is the right one). The name indicates that the farm was, as Per Onsager put it, "a gods place" in Heidensk times, for Onsaker is a foreshortening of "Odins Aker," which mans "Odin's place." According to Snorre,[24] the idea of Odin traveled north through Germany and Denmark and entered Sweden and then Norway during the Folkevandring time, where he used his wisdom to triumph over all of the old local gods to become the main god, akin perhaps to the way that Lars triumphed over physics. Unfortunately, in disagreement with Snorre's wonderful account of Nordic mythology, the scant evidence available points to Thor, not Odin, as having held the high seat at Gammel Uppsala in Sweden and at several other god-sites[25] in Norway in the Viking era. Surely, Lars would have liked to set us straight with his own opinion on this matter!

At any rate, my first visit to Norway did nothing to extinguish my strong affinity for Norwegian ways and even had the opposite effect. I therefore began to study and learn "Norwegian"[4] by taking classes in

[4] Bokmål, also the Oslo dialect, passes formally as "Norwegian": Norway has two main languages, 28 strong dialects, and a host of subdialects, making conversation extremely difficult but always interesting. In Viking times, it was said that Sogn dialect would take you all the way to Mikligård (Constantinople). It certainly would have taken you to Normandy as well.

Houston in the fall of the 1984, and then took a sabbatical at the Institutt for energiteknikk, lecturing at the University of Oslo in the fall of 1985, with much help from Tormod Riste and Gerd Jarrett. Tormod got me a nice NORDITA stipend and I also got a small Haakon Styri Fellowship from the American–Scandinavian Foundation. Both were based upon my proposal to lecture and continue research on nonlinear dynamics and deterministic chaos, which I has just begun "to do my homework on" (as Lars might have put it) in the time when interest in two-dimensional superfluids began once more to vanish. I have visited IFE every summer since 1985, save 1986, with earlier financial support from NORDITA and Norges Allmenvitenskapelige Forskningsråd (NAVF) for my work on nonlinear dynamics, then later with support (through IFE) from Petrofina/FINA NORWAY for using ideas from dynamics and fractals for the modeling of flow through porous media.

I have benefited enormously from my time spent in Norway, at IFE (the writing of my first book[26] began during that 1985 sabbatical and continued during later visits), from our visits (my wife also now speaks some Norwegian) to a certain old farm that lies perched on a small, steep green meadow high above Eidfjord, and from our time in the mountains.

The first and last sections of this paper about my Onsager days and times in Norway were taken from a larger article written in Norwegian for the sole surviving Norwegian physics journal.[27]

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