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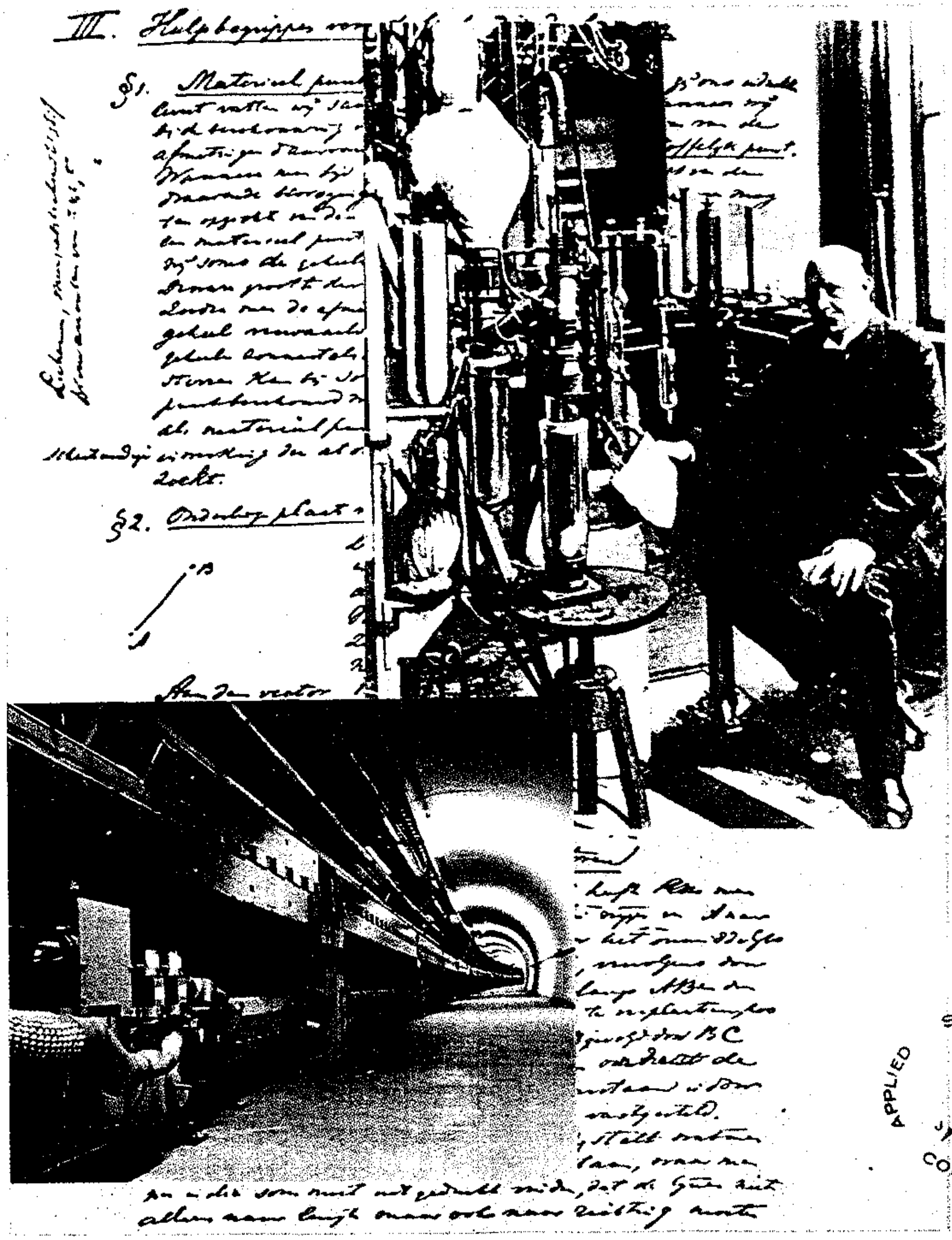

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TYPE II SUPERCONDUCTIVITY: QUEST FOR UNDERSTANDING

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Abstract

By 1941, many of the essential experimental features of type II superconductivity had already been observed (de Haas and Voogd, Shubnikov et al., Keesom and Desirant). Moreover, truly remarkable progress had been made toward theoretical understanding based on negative interphase surface energy considerations (Gorter, H. London). However, a competing explanation, the filamentary sponge model, was proposed (Mendelssohn) in an attempt to explain magnetic hysteresis effects which tended to obscure the intrinsic thermodynamic character of type II superconductivity. This filamentary sponge model is now known to be of only very restricted applicability, but for more than two decades it enjoyed wide acceptance, so much so, that when the ultimate theoretical basis for type II superconductivity was formulated in the 1950's (Ginzburg and Landau, Abrikosov, Gorkov (GLAG)), it was largely ignored. With the discovery of the practical supermagnet potential of type II superconductors (Yntema, Kunzler et al.), interest in achieving deeper understanding of high-magnetic-field superconductivity was reawakened. Only then was the power of the GLAG formalism very belatedly recognized, both with respect to near-ideal type II superconductors (Goodman) and with respect to non-ideal materials of technical interest (Berlincourt and Hake). Rapid experimental and theoretical progress followed on a number of significant aspects, including flux trapping, flux creep, and flux flow (Yntema, Anderson, Kim, Hempstead, Strnad), and surface superconductivity (Saint-James and de Gennes). Indirect "observation" of Abrikosov's vortex lattice was soon accomplished by neutron scattering techniques (Cribier et al.) and by nuclear magnetic resonance techniques (Pincus et al.). Finally, a more direct magnetic decoration technique (Essmann and Träuble) yielded remarkably graphic and incontrovertible pictoral confirmation of the Abrikosov vortex lattice.

Introduction

The discoveries of Yntema[1] and of Kunzler, Buehler, Hsu, and Wernick[2] demolished a myth. Despite earlier contrary indications, it was suddenly found that large critical current densities could, after all, be supported in superconductors at high magnetic fields. Well, almost suddenly. Yntema's work went virtually unnoticed at first. But imagine the excitement when it was ultimately realized that the possibility of generating high magnetic fields at unprecedented economy might be within our grasp!

Whenever such a profound change in perspective occurs, the human species feels a compulsion to account for it, and in the attempt often rushes headlong in the wrong direction. So it was in this instance, as we attempted to adapt a part of an old myth, viz., the sponge model, to explain a new reality. Little matter that even before the work of Kunzler et al. there already existed a rigorous theoretical structure, which was capable of accounting for much of the remarkable behavior of high-magnetic-field superconductors. I refer, of course, to the Ginzburg-Landau (GL) theory,[3] to Abrikosov's theory[4] of type II superconductivity, to Gorkov's reconciliation[5] of the GL theory (and hence of Abrikosov's theory) with the Bardeen-Cooper-Schrieffer (BCS) microscopic theory, and to Gorkov's derivation of the critical parameters of the GL theory in terms of readily measurable normal-state quantities.

This paper focuses mainly on the experimental and conceptual factors which culminated in the development and confirmation of the Ginzburg-Landau-Abrikosov-Gorkov (GLAG) theory. But I also mention the sponge model, for, prior to its being supplanted by the GLAG theory, its seductive, but misleading, counterpoint profoundly influenced investigations of alloy superconductivity over more than a quarter of a century.

Foundations

As early as 1935 many of the experimental features typical of type II superconductivity had already been observed. An alloy superconductor, subjected to a magnetic field, had been shown initially, i.e., at low fields, to resist magnetic field penetration in a manner generally indistinguishable from that for a pure superconductor. However, in the alloy magnetic flux penetration was observed to commence at a lower field than was typical for a pure metal. Moreover, the magnetic flux penetration, instead of being abrupt and complete, was gradual, and it extended to exceptionally high magnetic fields, where the normal state was finally restored. In decreasing fields hysteresis was evident, and at zero applied field the alloy was left with more trapped flux than was typically the case for a pure metal. These features are all depicted in Figure 1.

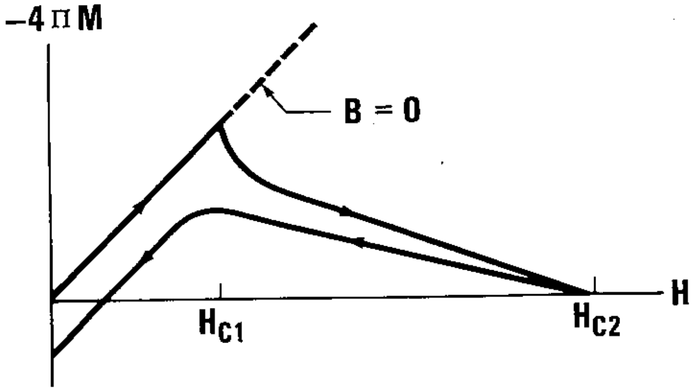

Figure 1. Magnetization (M) versus magnetic field (H) for typical non-ideal type II superconductor.

The absence of electrical resistance, characteristic of superconductivity, had been shown for modest measuring currents to persist all the way to the magnetic field H_{c2}, where flux penetration was

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complete. As early as 1930, in studies of Pb-Bi alloys, de Haas and Voogd[6] had established that this transition could occur at magnetic fields as high as 2.1T (or 2.3T when extrapolated to absolute zero). However, attempts at Leiden[7] and Kharkov[8] in 1935 to exploit alloy superconductors for production of high magnetic fields were thwarted when the combined magnetic field-critical current density performance of those early specimens was shown to be disappointingly poor.

Nevertheless, in the year 1935 there was keen scientific interest in alloy superconductivity, and prophetic concepts emerged from attempts to account for the high transition fields. Of central importance was H. London's prediction[9] that a superconducting film of thickness much less than the penetration depth would exhibit a longitudinal transition field much higher than that of its bulk counterpart. This followed from the fact that in this geometry the film only slightly distorts the applied field, and hence more slowly expends its superconducting condensation energy establishing shielding currents as the applied magnetic field is increased.

Aspects of this circumstance were seized upon in 1935 by Gorter,[10] by H. London,[9] and by Mendelssohn[11] to account for the survival of superconductivity in alloys at high magnetic fields. It is evident that they were very much aware of each other's work. Both Gorter[10] and H. London[9] reasoned that a bulk, homogeneous alloy superconductor might achieve a state of lower magnetic field distortion, and hence lower energy, simply by subdividing into thin superconducting lamina or into needles parallel to the applied field and separated by normal layers of extremely small thickness and that this would allow the survival of superconductivity at higher fields.

In 1950 H. London's brother, F. London,[12] described the probable structure as "small superconducting domains somewhat like a mosaic crystal," and elsewhere in the same monograph, he first described fluxoid quantization.[13] With some refinement and fusion of these highly intuitive ideas the Abrikosov vortex structure might have been anticipated. Instead, the vortices were destined to appear only several years later in a purely mathematical solution of the GL equations.

Additional concepts indicative of remarkable foresight were evident in Gorter's 1935 paper.[10] For his laminar model he predicted that the transition field would be given by $\frac{\ell}{k}H_o$, where ℓ is the penetration depth, H_o is the thermodynamic critical field, and k is a "minimum size for the superconductor." If Gorter's k is identified with the quantity we now call the coherence length, ξ, and if modern notation is utilized, his transition field becomes $\frac{\lambda}{\xi}H_c$. This is within a factor of $\sqrt{2}$ of the Abrikosov value for the upper critical field H_{c2}. Not bad for 1935! Equivalently, Gorter argued that the thin high-field laminar structure would form for $k < \ell$ (i.e., $\xi < \lambda$), a condition he identified with alloys, that for "$k > \ell$ (i.e., $\xi > \lambda$) there will be no tendency to form very small supraconductive regions" in agreement with the behavior of (type I) pure metal single crystals. H. London's more mathematical analysis[9] invoked interphase surface energy considerations to account for the same distinction between alloys and (type I) pure metals, and he pointed out the equivalence of his surface energy classification scheme and Gorter's $k <$ or $> \ell$ scheme. Gorter noted, however, that "The behavior of supraconductive alloys is certainly explained far from completely by these remarks, which do not even offer a suggestion why ℓ (i.e., λ) should be especially large or k (i.e., ξ) especially small for an alloy...." That would have to await subsequent advances by Pippard.[14,15]

Gorter and H. London both recognized that their approach shed no light on the cause of the observed magnetic hysteresis and trapped flux in alloys, but they did anticipate that inhomogeneities would probably have to be invoked in some manner to account for these features.

It is important to emphasize at this point that the thin super-normal structure hypothesized by Gorter and by H. London to account for alloy superconductivity would appear for a perfectly homogeneous alloy, i.e., no inhomogeneities whatsoever were required for it to occur. In stark contrast Mendelssohn[11] proposed to account for high-magnetic-field superconductivity purely in terms of the inhomogeneities. In his words, "We think that all experimental results so far obtained on impure metals and on alloys can be explained by their inhomogeneity which causes the formation of a 'sponge' of higher threshold value."

There was some justification for this perspective, inasmuch as the highest-field material known at that time, the Pb-Bi alloy studied by de Haas and Voogd,[6] was a eutectic, i.e., it was composed of a finely divided mixture of two separate and different-composition phases. Mendelssohn seemed to imply that the high threshold field could either be intrinsic for the material of the sponge or could follow if the thicknesses of the superconducting sponge regions were of the order of, or less than, the penetration depth. Finally, the multiple connectivity of the sponge provided a ready explanation for the observed magnetic hysteresis and trapped flux.

The crucial experiment to distinguish between the Gorter-H. London model and the sponge model would be one on highly homogeneous defect-free alloys. For such materials negligible flux trapping would be expected. According to Gorter and H. London survival of superconductivity to high magnetic fields would still be expected, whereas, according to the sponge model behavior typical of pure metal (type I) single crystals would be expected.

Just such an experiment was reported in 1937 by Shubnikov, Khotkevich, Shepelev, and Rjabinin.[16] They studied the magnetization M(H) curves of carefully prepared single-phase, single-crystal alloys in the systems Pb-Tl and Pb-In. They speculated on the existence of a critical alloy composition (between 0.8 and 2.5 atomic % Tl in Pb) which would mark the boundary between what we now know as type I and type II behavior. For the more concentrated alloys their data showed near-ideal type II behavior. In fact, they remarked, "Such unusual magnetic properties of superconductors cannot be explained by hysteresis phenomena, inasmuch as it is just at high increasing and diminishing fields that the phenomenon is quite readily reversible and the hysteresis is quite low." With insight prophetic of developments to follow some 25 years later, Shubnikov et al. noted (1) that $\int MdH$ should represent the superconducting state condensation energy, (2) that even though the alloy upper critical field H_{c2} greatly exceeded the field H_c characteristic of a typical pure metal, the condensation energies for the two cases were comparable and depended upon temperature in the same way, and (3) that, therefore, the zero-field specific heat jump in an alloy superconductor should be comparable to that of a pure superconductor.

With this last point Shubnikov et al. were the first to appreciate the thermodynamic character of alloy superconductivity and to understand the folly of earlier frustrated specific heat investigations[17,18] which sought the enormous specific heat jump which would be expected were complete flux exclusion to persist all the way to the highest alloy transition field.

The specific heat jumps characteristic of a type II superconductor and consistent with the perspective of Shubnikov et al. were first observed in 1941 in studies by Keesom and Desirant[19] on impure Ta, which we would now characterize as having a kappa value of $K \sim 2.5$. (See Figure 2.) While Keesom and Desirant noted the inapplicability of thermodynamics based on complete flux exclusion, they made no mention of any possible relation of their results to the predictions of Shubnikov et al.

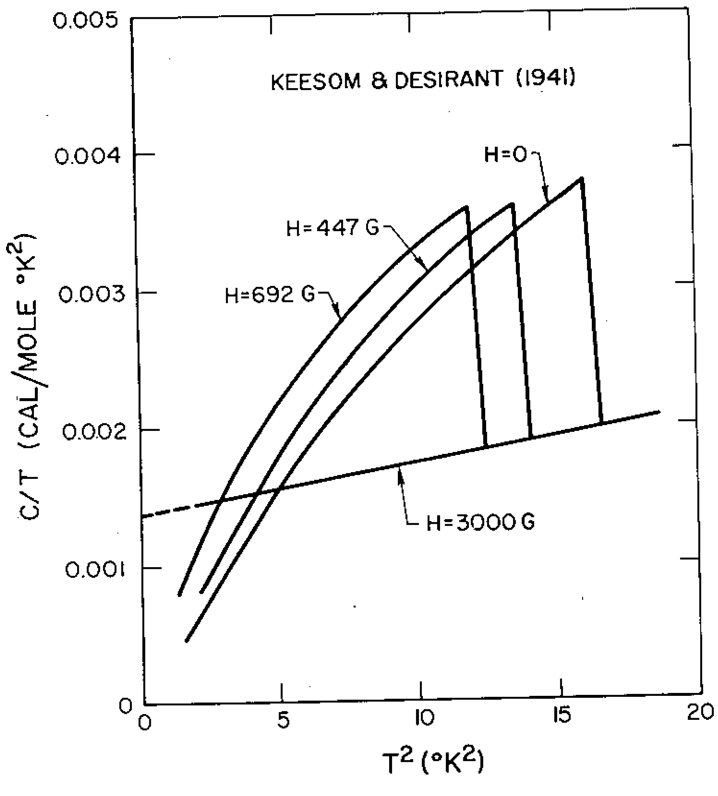

Figure 2. Dependence of specific heat (C) on temperature (T) and magnetic field (H) for impure Ta for which $K \sim 2.5$.

Thus, by 1941 the magnetic and calorimetric character of type II superconductivity had been observed, and the thermodynamic consequences of reversibility had been recognized by Shubnikov et al. In fact, in 1937 the latter authors had accurately characterized ideal type-II superconductivity and they had attributed it to the bulk homogeneous alloy, not to the defect structure. Thus, the missing link had been supplied, and it unequivocally favored the Gorter-H. London theory over the Mendelssohn sponge model.

But then history took a strange turn, for although Shubnikov et al. had done the crucial experiment and had interpreted it correctly, their paper made no mention of the Gorter-H. London theory (although there was reference to other work by Gorter[20]) nor of the Mendelssohn sponge (even though Shubnikov et al. ruled out inhomogeneity as the basis for the observed high-field superconductivity and even though they referenced two works by Mendelssohn and Moore[18,21] which mentioned the sponge model). Even more curiously, the work of Shubnikov et al. was almost universally ignored, even in the Soviet Union. Only some twenty years later did it finally receive due attention when Abrikosov compared his theory with the data of Shubnikov et al. in his epic paper on type II superconductivity.

Could early communications really have been that poor? Although the paper of Shubnikov et al. appeared only in the Russian language, portions of it had been reported earlier in English.[8] Moreover, Shubnikov had spent some years engaged in low temperature physics research at Leiden, where he and W. J. de Haas had discovered the magnetoresistance oscillations now known as the Shubnikov-de Haas effect. And so Dutch low temperature physics researchers doubtless maintained communications with Shubnikov. Why didn't Gorter and H. London seize the Soviet results as confirmation of their theory? And why did Shubnikov et al. fail to exploit this new-found understanding? While the answer to the first of these questions remains a puzzle, an answer to the second question is found, at least in part, in a letter I received from Mendelssohn in 1963. A portion follows:

> "It was extremely nice of you to send me a copy of your own paper, as well as a translation of Shubnikov's paper published in 1937. This is indeed of considerable help in assessing the earlier developments. At that time the Stalin Purge was only beginning, and I was very puzzled at the blanks I drew in trying to get in touch with Shubnikov. In 1957 Landau introduced me in Moscow to Shubnikov's widow, Olga Trapeznikova, who also is a physicist. She told me that her husband had just been exonerated posthumously from all charges. This made it possible for Abrikosov to refer to Shubnikov's papers, since up to then Soviet etiquette required that anyone who had disappeared in the purges had never lived."

A eulogy by Balabekyan[22] in 1966 reveals that Shubnikov was unjustly arrested in 1937, that he was sentenced to 10 years imprisonment, and that he died in 1945.

Other factors surely left their impact on the superconductivity research of that era. There were the tumultuous events of World War II. And, of course, the rather meager data on ideal type II superconductors could easily have been obscured by the plethora of data on non-ideal type II superconductors and on two-phase alloys. Perhaps

Mendelssohn[23] sensed the nature of those times better than is possible for those of us who were not a part of the very small community of low temperature physicists of that era. In 1964 he philosophized:

> "Today it is often said that if research on superconductive alloys had been pushed only a little farther in the thirties, the exciting developments of superconductive magnets would have taken place twenty-five years sooner. I don't believe this is true. Those of us who can recapture the mentality of a quarter of a century ago know that even the discovery of high current densities would have remained just another curiosity. The contemplation of the required technical effort in cryogenics and of all the ancillary development would have appeared to us as outrageous folly."

Ginzburg-Landau Theory

It was not until 1950, i.e., thirteen years after the work of Shubnikov et al., that another theoretical clue to the true nature of type II superconductivity appeared, again in the Soviet Union. In their very remarkable macroscopic theory of superconductivity Ginzburg and Landau[3] introduced the parameter kappa, or K, which provided a measure of the surface energy at the interface between normal and superconducting phases. The case $K < 1/\sqrt{2}$ corresponded to a positive interphase surface energy, and as we now know the corresponding solutions to the GL equations account very nicely for a number of features of ideal type I superconductors, including the magnetic-field-induced first order phase transition, with its prominent "superheating" and "supercooling" phenomena. Ginzburg and Landau noted that for $K > 1/\sqrt{2}$ the interphase surface energy would be negative and hence that for this case superconductivity would occur at fields above the thermodynamic critical field. Curiously they concluded that "It has not been necessary to investigate the nature of the state which occurs when $K > 1/\sqrt{2}$, since from the experimental data .... it follows that $K << 1$." This despite the fact that the experimental data of Shubnikov et al. demanded explanation in just such terms! Meanwhile, in Great Britain, Pippard[14,15] was explaining on very intuitive grounds that the short electron mean free paths in alloys and films would lead to negative interphase surface energy. Why were these various perspectives not swiftly reconciled in a unified theory of type-II superconductivity? An amusing account from Pippard's perspective appears in his paper on "The Historical Context of Josephson's Discovery."[24] It says in part:

> "So in the early 1950's there was a certain amount of conflict which wasn't helped, incidentally, by the fact that Ginzburg kept on writing small papers in which he said it would be much better if we interpreted the electronic charge as not being exactly e, but e times a small numerical factor which might be as large as 2! He didn't say it was exactly 2; instead he wanted to introduce a fudge factor of (say) 1.6, and Landau kept on telling him he couldn't just put in arbitrary numbers, and muttered darkly about gauge invariance going wrong if you did."

Later, with the advent of the BCS theory, that double electronic charge would, of course, be placed on a firm theoretical foundation.

But, what of the disconnect between GL and the experimental work of Shubnikov et al.? Was it merely a case of theoreticians being unaware of experimental results, or, as Mendelssohn implied, did Shubnikov's imprisonment play a role? One can only speculate.

Abrikosov's Theory

The next advance toward understanding was soon triggered by experimental investigations by Zavaritski.[25] In 1952 he reported studies on the critical fields of superconducting films. For pure, well-annealed crystalline films of Sn and Tl his measurements exhibited good accord with the GL theory predictions for the case $K < 1/\sqrt{2}$, i.e., for thick crystalline films the magnetic-field-induced transitions were of the first order, and only below a certain thickness did the transitions become second order. In contrast, for amorphous films of the same pure metals, deposited and measured immediately at low temperatures, second order transitions were observed for all thicknesses. These results were a topic of discussion between Zavaritski and his colleague Abrikosov, and they recognized the similarity of the amorphous pure-metal results to the behavior of alloys. In the words of Abrikosov:[26]

> "Discussing with Zavaritski the possible origin of this discrepancy, we came to the idea that the approximation $K << 1$ based on the surface tension data (where K is the Ginzburg-Landau parameter) could be incorrect for objects such as low-temperature films. Particularly one could suppose that $K > 1/\sqrt{2}$. According to Ginzburg and Landau, the surface energy should be negative under these conditions. Intuitively it was felt that in this case the phase transition in a magnetic field would always be of second order, and this was in fact what Zavaritski observed."
>
> "When I calculated the dependence of the critical field on the effective thickness with $K > 1/\sqrt{2}$, it appeared that the theory corresponded to the experimental data. This gave me the courage to state in my article of 1952 containing this calculation that apart from ordinary superconductors whose properties were familiar, there exist in nature superconducting substances of another type, which I proposed to call superconductors of the second group (now called type II superconductors). The division between the first and the second group was defined by the relation between the quantity K and its critical value $1/\sqrt{2}$."

In the 1952 publication[27] to which Abrikosov alluded above he predicted that the upper transition field in a bulk type II superconductor would be given by $H_{c2} = \sqrt{2}\, K\, H_c$. Next, Abrikosov decided to explore the nature of the phase which existed just below H_{c2} in a type II superconductor, and he soon concluded that there would be a periodic distribution of current, magnetic field, and superconducting order parameter, which he named the "mixed state." Landau, who was Abrikosov's mentor, took interest in the work and encouraged Abrikosov to publish his results for the immediate vicinity of H_{c2}. However, Abrikosov wished to explore the mixed state over the total range of its existence in magnetic field strength. In the words of Abrikosov:[26]

> "At this time I became ill and had to stay in bed for almost three months. One day Landau visited me. The conversation, as in most cases, concerned everything but physics, and Landau sipped with great pleasure from a glass of glühwein, which was not at all like him. And then suddenly I destroyed all this paradise by telling him what I had invented for the mixed state, namely, the elementary vortices. As Landau's eyes fell on the London equation with a δ function on the right-hand side, he became furious. But then, remembering that an ill person should not be bothered, he took possession of himself and said, 'When you recover we shall discuss it more thoroughly.' Then he hastily bade farewell and disappeared."
>
> "He did not come to me any more. When I felt better and appeared at the Institute and tried to tell him again about the vortices, he swore rather ingeniously. At that time I was still very young and did not know the temper of my teacher well enough. He had seen in his life many kinds of pseudoscience, and this made him suspicious toward unusual statements. However, by making some effort and disregarding the noise which he made, one could always 'drag' him through any reasonable idea. But at that time I sadly put my calculations in my table drawer 'until better times.'"

Thus, the ultimate reckoning with type II superconductivity was again postponed as Abrikosov busied himself with other problems. It was, in fact, some three years before Abrikosov's "better times" arrived. Late in this period Landau and Lifschitz sought unsuccessfully to describe the state of superfluid helium in a rotating vessel. This problem was solved, however, by Feynman,[28] who hypothesized that the single-quantum superfluid vortices, first conceived by Onsager,[29] would appear in the rotating vessel. This solution was accepted readily by Landau. And Abrikosov of course immediately recognized the analogy with his type II superconductivity theory. According to Abrikosov:[26]

> "When Landau began to praise Feynman's work I asked him, 'Dau, why are you ready to accept the vortices from Feynman while you flatly rejected the same idea from me?' Landau answered, 'You had something different.' 'Well then, look, please,' I said, and produced my calculations from the drawer. This time no objections followed. We discussed the subject very thoroughly and Landau's remarks were very useful."

Abrikosov's publication of this work[4] included the now well-known vortex lattice (Figure 3), a thorough and compelling comparison of his theory with the experimental data of Shubnikov et al.,[16] and recognition that macroscopic inhomogeneities, which were ignored in his theory, most probably accounted for remnant magnetic moments. Thus Abrikosov had captured the essence of type II superconductivity in purely mathematical solutions to the GL equations for the case $K > 1/\sqrt{2}$.

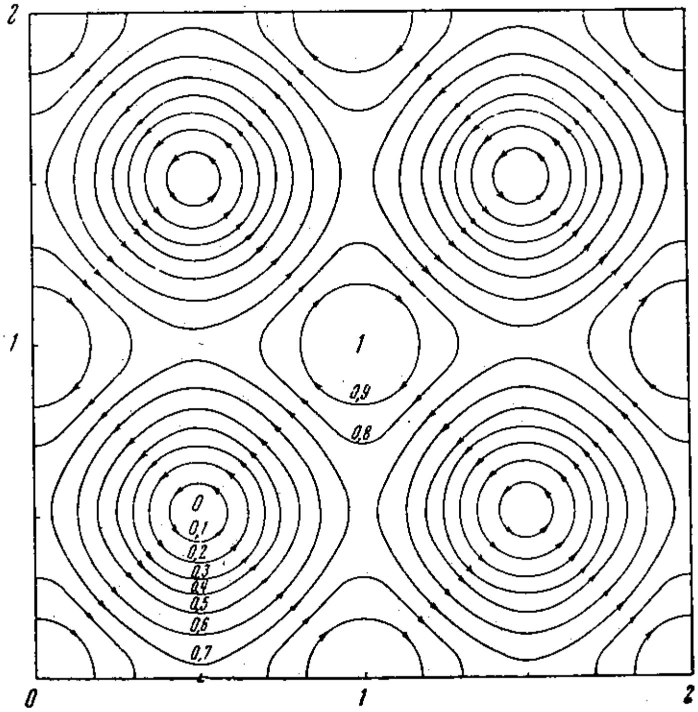

Figure 3. The Abrikosov vortex lattice.

This remarkable advance was compounded when, in 1959, Gorkov[5] showed that in the local limit the GL equations could be derived from the Bardeen-Cooper-Schrieffer microscopic theory of superconductivity. His work made it possible to deduce theoretically, values for K and for the upper critical field, H_{c2}, using experimentally determined values of normal-state parameters. For the case of short electron mean free path it was necessary only to know the normal-state electrical resistivity, the normal-state electronic specific heat coefficient, and the superconducting transition temperature. But, ironically, the world took little, if any, notice of the very powerful but highly abstract GLAG formalism.

Experimental Confirmation of GLAG

Although most experimentalists appeared to be oblivious to the GLAG theory, they were nevertheless making progress in the laboratory. In 1955 there was Yntema's demonstration[1] of high critical supercurrent densities at modest magnetic fields, and, after a 27-year hiatus the high-magnetic-field limit for existence of superconductivity was again being advanced. In 1957, in studies[30,31] of U-Mo and U-Nb alloys at Atomics International, I observed transition fields which, when extrapolated to absolute zero, approached 3.6T, or nearly 60% higher than the corresponding value reported for Pb-Bi alloys in 1930 by de Haas and Voogd. Still higher transition fields were apparent in measurements[32] which Hake, Leslie, and I carried out on Ti-Mo alloys in 1959, but limits on the available measuring field precluded our determination of actual values.[33]

Our high-field studies at Atomics International in 1959 were carried out at very modest current densities (tens of A/cm^2) and with the belief that the high transition fields were attributable to the filaments of the Mendelssohn sponge, which we believed to be incapable of supporting high current densities. Then in 1960 Bozorth, Williams, and Davis[34] published a highly hysteretic magnetization curve for Nb$_3$Sn, which today we would immediately recognize as that of a nonideal type II superconductor, and they concluded that the critical field was 7T. With the advantage of today's hindsight one may deduce from the observed hysteresis that the specimen was supporting a critical current density of $\sim$ 6,000 A/cm^2 at 7T, not impressive by today's standards, but it would have been regarded as remarkable had it been recognized at that time.

Other rapid progress in the realization of high current densities was evident. Beginning in 1959, Autler[35] made impressive progress on Nb supermagnets, apparently unaware of Yntema's earlier work, and a year later Kunzler, Buehler, Hsu, Matthias, and Wahl[36] developed a Mo$_3$Re alloy supermagnet which generated fields up to 1.5T. All of these developments were overshadowed, however, by the startling announcement by Kunzler, Buehler, Hsu, and Wernick[2] that dissipationless current densities of 10^5 A/cm^2 could be sustained in Nb$_3$Sn at fields as high as 8.8T!

During the immediate flurry of activity following this advance the experimental community, still completely oblivious to the GLAG theory, invariably invoked the Mendelssohn filamentary sponge as the basis for explanation of all manner of phenomena. The appeal of the sponge model doubtless stemmed from its simplicity and the plethora of adjustable parameters which could be invented at will, so long as no one actually "saw" and quantitatively described the phantom filaments.

Ironically, when a challenge to the sponge model did appear, some four months after the discovery of Kunzler et al. it was not based on the GLAG theory. Rather it appeared in a paper in which Goodman[37] invoked the old Gorter-H. London laminar model, together with the negative surface energy rationale of Pippard, to account with modest success for the near-ideal type II magnetization curves observed by Calverly and Rose-Innes[38] for single crystals of Nb-Ta alloys. Although Goodman was evidently unaware of the GLAG theory, his approach was conceptually closely attuned to it. However, by the time Goodman's report of this work appeared in print he had "discovered" GLAG. At the IBM Conference on Fundamental Research on Superconductivity held at Yorktown Heights in June 1961 he showed[39] that the GLAG theory was superior to his own in its ability to account quantitatively for the behavior of Pb-Tl alloys. Whereas Abrikosov had simply deduced values for K directly from the magnetization curves measured by Shubnikov et al., Goodman was able to invoke the Gorkov microscopic extension[5] of Abrikosov's theory. By using measured values for the electrical resistivities of Pb-Tl alloys together with extrapolated values for other pertinent electronic parameters Goodman succeeded in calculating values for the upper critical fields with noteworthy success. He was also able to test the GLAG theory for a U-Mo alloy. Using my data for transition temperature and normal state electrical resistivity together with normal-state electronic specific heat data he and his colleagues[40] had obtained prior to his "discovery" of the GLAG theory, he deduced a theoretical upper critical field of 2.7T at 1.2K. This fell at the center of the magnetic field range over which I had observed the magnetic-field-induced restoration of resistivity in this alloy at the same temperature and for a measuring current of 4 A/cm^2.

The corresponding K value was 65, or a factor of ten greater than for the Pb-Tl alloys! In hindsight, we know that Goodman's paper marked a highly significant turning point in the linkage of the GLAG theory to high-magnetic-field superconductivity. But, at that time it was pretty much ignored, almost as if it were not a serious enough threat to the filamentary sponge model to merit its being refuted! But I did take some notice of it, if for no other reason than Goodman's use of my U-Mo data. In discussing Goodman's results with him immediately after his talk I cautioned him that, while his observation might indeed be very significant, it could also have been simply a fortuitous coincidence, for the magnetic-field-induced resistive transitions I had observed were sensitive functions of measuring current density. Further, I suspected that for different magnetic field and current orientations, and for different mechanical and metallurgical treatments I might have observed significantly different transition fields. After all, cold working was well known to affect very markedly the resistive transition field in Nb.[41] Goodman accepted my comments with equanimity, acknowledging that the issue was not yet settled.

Throughout the latter part of 1961 Goodman's conjecture was largely ignored. Like most other superconductivity researchers in those very exciting times my colleague Richard Hake and I were swept up in the stampede to build practical supermagnets and to find still better supermagnet materials. We enjoyed an acrimonious patent interference with Kunzler and Matthias over Nb-Zr alloys.[42,43] This was to end nearly five years later with award of a patent to Bell Telephone Laboratories and a royalty-free license to Atomics International. (During this "dispute" Kunzler and his colleagues graciously provided my research group with very-high-purity, high-perfection metal single crystals for use in our de Haas-van Alphen effect studies!)

In late 1961 Hake and I upgraded our experimental capabilities at Atomics International, and with the help of a 16T pulsed magnet, we investigated a great multitude of ductile transition metal alloy superconductors. To our surprise we found Nb-Ti alloys to be superior to Nb-Zr alloys for supermagnet applications,[44-46] thus rendering the Nb-Zr patent interference quite pointless. Nb-Ti alloys had of course been studied superficially earlier both at Atomics International[47,48] and at Bell Telephone Laboratories,[49-52] but those investigations had failed to reveal the superior high-magnetic-field, high-current-density potential of these alloys, and so they had been by-passed in the excitement over Nb$_3$Sn and Nb-Zr.

But more important from the scientific

perspective, Hake and I sought deeper understanding of the basic mechanisms of high-magnetic-field superconductivity. In the course of this work we found that so long as a very low measuring current density ($\gtrsim 10$ A/cm^2) was used, the resistive transition field, H_r, for a given concentrated alloy composition proved to be nearly independent of the degree of mechanical working and of the relative orientations of field and current (Figure 4). These results suggested that the low-current-density resistive critical field was intrinsic, i.e., that it was related to fundamental bulk electronic properties rather than to the more capricious features of the inhomogeneities of a supposed filamentary sponge structure. It also suggested that Goodman's interpretation of the U-Mo alloy data in terms of the GLAG theory might, in hindsight, be fully justified!

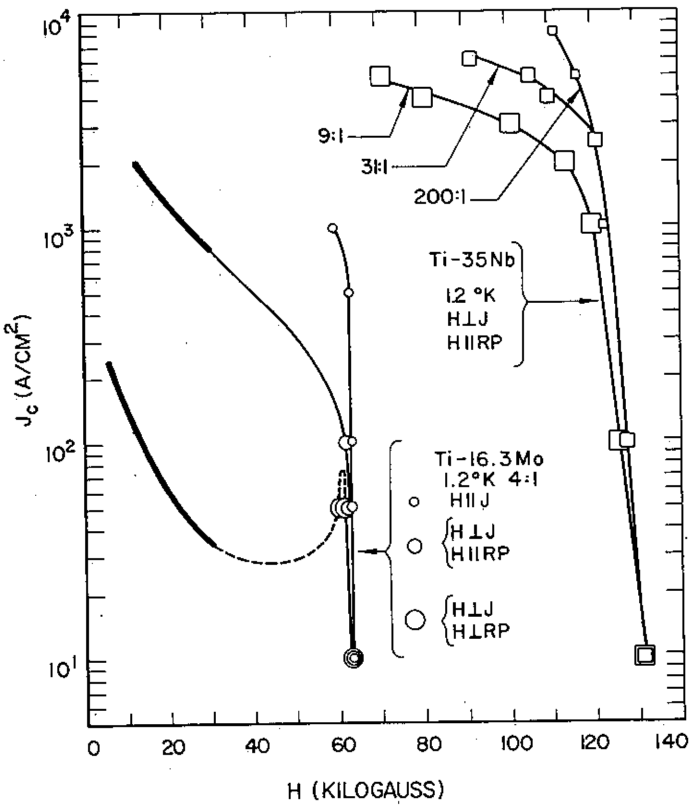

Figure 4. Illustrations of independence of low-current-density resistive critical fields upon cold working and relative orientations of magnetic field (H), current (J), and rolling plane (RP) defect structure. Ratios indicate cold-rolling thickness reductions. (After Berlincourt and Hake, 1962).

Encouraged, Hake and I undertook a serious comparison of our low-current-density resistive critical field data on U-Mo, Ti-Mo, and Ti-V alloys with upper-critical-field predictions of the GLAG theory. This was possible, because the necessary data on superconducting transition temperature, normal-state resistivity, and electronic specific heat were by then available for all three of these alloys. For some ranges of alloy composition we found remarkably good quantitative agreement with the GLAG theory predictions with no arbitrary adjustable parameters. For those compositions the evidence in support of the GLAG theory was compelling.

In a post deadline paper[44] presented at the April 1962 Washington, DC meeting of the American Physical Society we described the new-found virtues of Nb-Ti as supermagnet material and the remarkable success of the GLAG theory in accounting for high-magnetic-field superconductivity. To my surprise both results were virtually ignored. The only person to express any interest at all was Goerge Yntema. He had come to the meeting to describe his ideas[53] on the possible existence of supercurrent vortices in alloy superconductors. He had arrived independently at this conjecture by analogy with studies he had conducted on rotating superfluid helium some years earlier. Now returning to low temperature physics after having spent some years in operations research, he had been unaware of Abrikosov's supercurrent vortices.

Wishing to convince a larger audience, Hake and I submitted a regular contributed paper[45] on our work to the Evanston meeting of the American Physical Society scheduled for June 1962, and we bombarded other superconductivity researchers with preprints of it. But, in spite of our enthusiasm for the GLAG theory we acknowledged some shortcomings. Although the GLAG theory predictions of upper critical fields showed excellent accord with experiment for some ranges of alloy composition there were discrepancies approaching a factor of two for other compositions. Ironically, the resolution of this discrepancy rested upon concepts about which Pippard and Heine[54] had speculated four years earlier, before anyone imagined that critical fields as great as 10T might be possible. They had pointed out that in a superconductor the energy gain resulting from electron spin alignment along an applied magnetic field would, at 10T, become comparable to the opposite-spin-paired superconducting state gap energy. That this kind of magnetic-field-induced depairing might impose a limitation on filamentary sponge superconductors was proposed independently by Chandrasekhar[55] and by Clogston.[56] Upon receiving preprints describing their work, Hake and I immediately recognized that this paramagnetic consideration had also been ignored in the formulation of the GLAG theory and that it would likewise impose limits in the case of interest to us. The discrepancies we had encountered were now explicable. As shown in Figure 5, our experimental upper critical field data for Ti-V (rectangles) are closely approximated by the GLAG theory predictions for H_{c2} for high concentrations of V. For other compositions the paramagnetic limiting field H_p more closely approximates the experimental data.

With this problem resolved in July 1962, and confident that we now understood the basis for high magnetic field superconductivity, Hake and I rushed a manuscript to Physical Review Letters. To our dismay it encountered two referees deeply committed to the filamentary sponge model. One responded as follows:

> "Although it is alleged that the independence of H_r on H, J and rolling plane at low current supports the GLAG theory, this fact can just as well be explained by the filamentary theory. At the very low current densities, where all filaments can be active and where the important fact is the best existing filaments and not their number, there will always be some filaments properly oriented (parallel to the applied field) that will yield the same critical field H_r regardless of orientation. As a matter of fact, this

also explains why the number 10 amp./cm^2 cannot be taken for all alloys as this number will depend on the degree of anisotropy, number of filaments, etc. Figure 2 can be explained by the filamentary theory as well $H_f = k\ H_c$ where H_f is the filamentary critical field. H_c the bulk critical field and k a constant depending on the size of the filaments, the coherence length and the penetration depth. As H_c peaks between 4 and 5 e/a so will H_f. Finally, phenomena such as flux trapping, anisotropy, peak effect can be explained by the filamentary theory and not by the GLAG theory. Actually, ... the GLAG theories and filamentary models may both be correct but the GLAG model fits the more homogeneous and soft hard superconductors. There is no sharp line and negative surface energy may be needed to realize the filamentary structure."

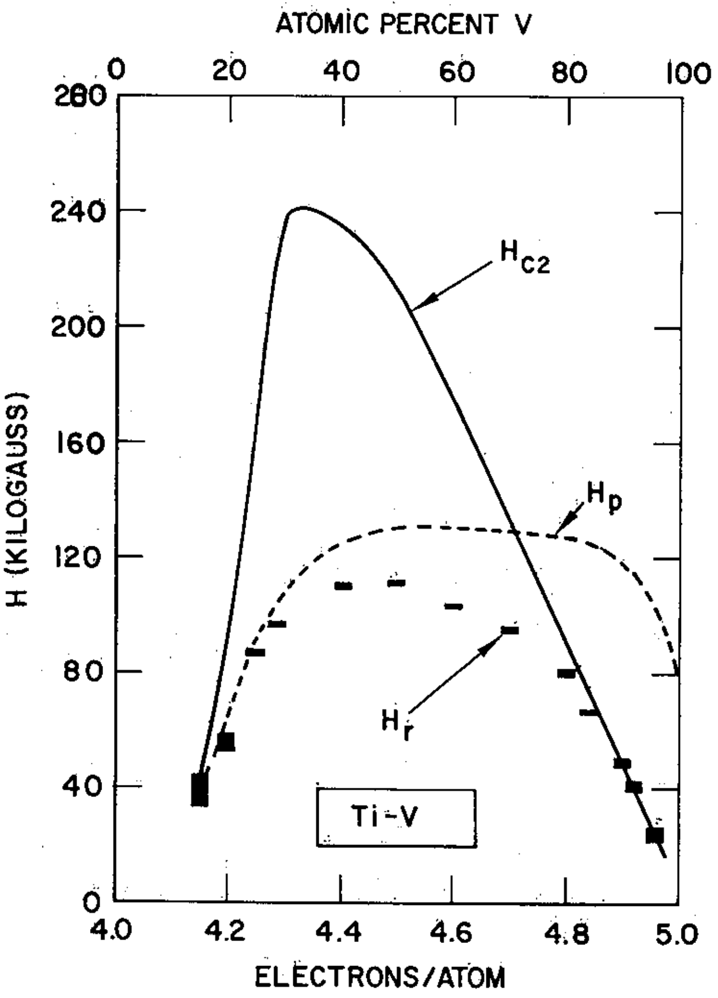

Figure 5. Experimentally determined transition field, H_r versus composition for Ti-V alloys, compared with theoretical values for H_{c2} (from GLAG theory) and for H_p (the paramagnetic limiting field). (After Berlincourt and Hake, 1962).

That rebuke typifies both the confusion of that era and the tenacious grip of the sponge model on the minds of many researchers at that time. Still another illustration is revealing. In April 1962 Morin et al.[58] obtained high-magnetic-field specific heat data on V_3Ga which were the higher-field equivalent of the 1941 Keesom and Desirant data[19] on impure Ta. The new V_3Ga data were interpreted by the authors as follows:

> "All of the results reported here can be interpreted by assuming that the sample contains a large number of filaments (probably dislocations) whose effective diameters are sufficiently large (but less than the penetration depth) that most of the sample appears superconducting. Because of the structure and low compressibility of V_3Ga, this assumption has been shown to be reasonable by Hauser and Helfand. However, it is expected that a perfect single crystal of V_3Ga (free of dislocations) would behave more like a 'soft' or nearly ideal superconductor and have a critical field of the order of 6 kgauss at 0°K."

In contrast, Hake and I[46,57] interpreted these experimental results as striking confirmation of the GLAG theory as did Goodman.[59]

In any event, anticipating that we might encounter resistance from referees we had in July 1962 mailed preprints of our Physical Review Letters submission[57] to a wide audience, and so it mattered little that publication of our work was delayed until October. The tide was turning in favor of the GLAG theory, and by September, when I reported[46] on our results at the Eighth International Conference on Low Temperature Physics in London, it was evident that the abandonment of the filamentary sponge in favor of the GLAG theory was assured.

Of course a number of scientific issues remained to be resolved, particularly with regard to mechanisms for current stabilization, but at least the effort was now focussed on the proper direction. In rapid succession flux pinning,[60] flux creep,[61,62] and flux flow[63] phenomena were investigated both experimentally and theoretically with great success. Each such advance could be neatly rationalized and shown to be compatible with the GLAG theory. There were a few seeming incompatibilities, but these soon found explanation.

In one such instance two of my colleagues, Joseph and Tomasch, were puzzled by the torques they observed acting on thick (thickness much greater than coherence length) Pb-Tl films placed at various orientations relative to an applied magnetic field.[64] For the applied field perpendicular to the plane of the specimen the observed transition field (H_{c2}) was in good accord with earlier bulk determinations for the same alloy composition. However, much to their surprise they found that when the applied field direction approached the plane of the specimen total destruction of superconductivity required a much higher applied field strength (Figure 6). As they described their unexpected results to me, I asked if their observed value for the parallel case might perhaps be a factor of exactly 1.69 too high. Surprised, they answered in the affirmative and asked how I could possibly have guessed. The explanation was really quite simple, for a day earlier I had received a preprint from Saint-James and de Gennes[65] who had solved the GL equations for the case of a superconductor-vacuum interface parallel to an applied magnetic field (recall that Abrikosov had in effect considered only the case of the superconductor filling all space). Thus, Saint-James and de Gennes "discovered" the superconducting sheath theoretically, and now its experimental confirmation was almost immediate, and to an accuracy of better than 1%!

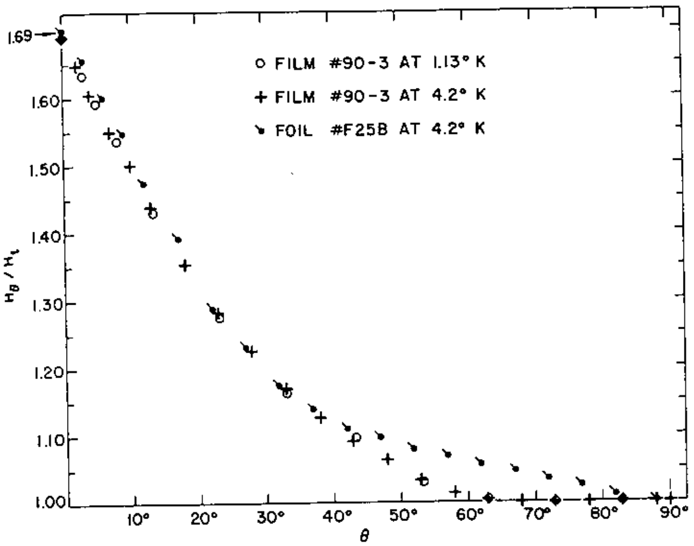

Figure 6. Ratio of transition field (H_θ) to transition field (H_t) for $\theta = \frac{\pi}{2}$ versus θ (angle between magnetic field and plane of film) for thick Pb-Tl films. The ratio 1.69 for θ = 0 matches exactly the theoretical prediction for the superconducting sheath. (After Tomasch and Joseph, 1964.)

Such striking confirmation of the GL equations added luster as well to Abrikosov's vortex lattice. After all, both the vortex lattice and the sheath were simply physical manifestations of different and purely mathematical solutions to the GL equations which arise for different boundary conditions. It thus appeared almost inevitable that the vortex lattice would ultimately be "observed" experimentally.

The first measurements were indirect, in that the vortex lattice structure was inferred from neutron scattering studies[66] and also from nuclear magnetic resonance measurements.[67] Then in 1967 Essmann and Träuble[68] developed a magnetic decoration technique, which, with the help of an electron microscope, yielded direct images of the vortex lattice structure (Figure 7). This very elegant demonstration was, however, almost anticlimactic, for by then we *knew* that the vortex lattice just *had* to be there!

Without question, the success of the quest for understanding of type-II superconductivity represents a truly remarkable achievement of the human intellect. But, to quote Pippard[24] from a different context:

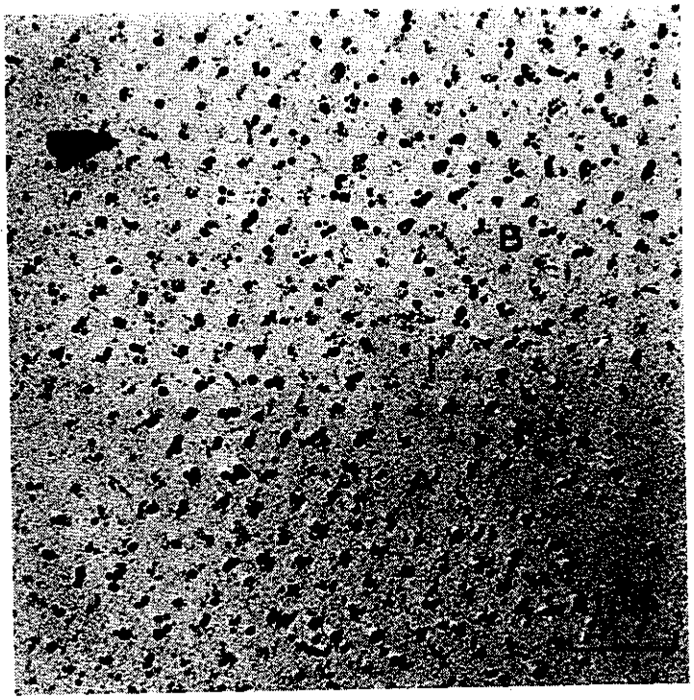

Figure 7. Triangular vortex lattice for type II superconductor as revealed by magnetic decoration technique. (After Essmann and Träuble, 1967.)

> "If we wish to boast of our achievements, let us not point to the unerring pursuit of truth by a logically faultless thinking-machine, but to the more astonishing way in which truth can be caused to emerge from the toils of error and stupidity."

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