Among the first is the fact that the number of research workers in the field was small and that some of them almost simultaneously left it. Schubnikov disappeared, Mendelssohn concentrated a large part of his attention on the superfluid properties of helium II while—as already mentioned—I returned to magnetism. As to the properties of alloys, I feel that the lack of metallurgical facilities and experience also weighed heavily. The rapid advance of the years 1932–1936 was consolidated by the appearance of Shoenberg's excellent monograph. But, though much further valuable work, not discussed in the present rapid and personal review, was carried out, this consolidation did not lead to a concentrated attack on the remaining problems before the outbreak of war. The rapid development during the war of high-frequency electronics led to developments in new directions, mainly achieved in Cambridge and the U.S.A. But it is not my task to describe those or to discuss the important advances made by Landau, Ginzburg, and by a number of British and U.S.A. theorists after the war.

Prewar Work on Superconductivity as Seen from Oxford

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My own recollections, like those of Professor Gorter, are of necessity subjective and personal. They cannot, and should not, claim to provide a complete and balanced account.

In 1933, just thirty years ago, I went from Breslau to Oxford to install the first helium liquefier in England. The liquefier, incidentally, was one of a race of small scale devices which were being developed by Simon and his school in Berlin and, later, in Breslau. Nowadays, it is not generally appreciated that one of the main reasons for making these miniature liquefiers was the scarcity of helium gas, which had to be extracted laboriously from Monazite sand. The arrival of the first American helium from gas wells was an important event, and I went down to the Berlin Customs House to open the crate. But then the official wanted to open the cylinder too, to see whether it contained liquor. He only relented when I assured him that I should be most disappointed if it did.

At the same time that we set up small scale liquefiers at Berlin University in the late twenties, Meissner at the Physikalisch Technische Reichstanstalt installed a conventional type of helium plant, and it was agreed that the field of research should be divided up. Meissner would concentrate on superconductivity, while we were to continue the thermodynamic work of the Nernst Laboratory. Thus, my only excursion into superconductivity before 1933 was a thermodynamic one, and it did not turn out too well. The idea was to look for a discontinuity in the specific heat at the transition point. Unfortunately we chose lead, because it already becomes superconductive at 7.2°K. Not unnaturally (we were then looking for something of the order of RT), we found that it was not there.[1] The small discontinuity which must be there was completely swamped by the enormous lattice specific heat, and has not been observed by direct experiment to this day.[2]

When, a little later, Keesom and van den Ende[3] discovered such a discontinuity in tin, I therefore read their paper with special interest. My attention was drawn to two low specific heat values just below T_c which they ascribed to the accidental presence of a magnetic field. This immediately suggested to me that there must exist a magnetocaloric effect which might be useful for making temperatures below 1°K. That was, of course, before the first paramagnetic cooling experiments.

As I was leaving the University library after reading Keesom's paper, I had to duck a few bullets which were flying through the streets of Breslau, heralding the approach of Nazi rule, and it became clear that the cooling experiment would have to be deferred a little. In fact, it was eventually carried out at Oxford in 1934.

However, my first experiment in Oxford dealt with

1 K. Mendelssohn and F. Simon, Z. Physik. Chem. B16, 72 (1932).

2 After my talk I was informed by Dr. C. Shiffman that, together with J. F. Cochran and M. Garber, he had just measured this anomaly directly.

3 W. H. Keesom and J. N. van den Ende, Comm. Phys. Lab. Univ. Leiden, No. 219b (1932).

another aspect of superconductivity. The first paramagnetic cooling had just been carried out in Leiden. There was the prospect of a new temperature range below 1°K, but Oxford had no suitable magnet. I resolved to remedy this deficiency by making a superconductive one-stroke flux pump,[4] consisting of one large loop and a solenoid in series. This appeared quite a feasible proposition, since a few years earlier de Haas and Voogd[5] had reported threshold values of more than 20 kilogauss in lead–bismuth alloys. A prototype device was constructed, but its performance was far inferior to the field which I had expected according to Silsbee's rule. I was puzzled by this failure, but must admit that I lost interest in the scheme because, in the meantime, Kurti and Simon had arrived in Oxford. They planned to work on paramagnetic cooling and a magnet was bought for them. In retrospect, and having regard to recent work on this alloy, one wonders why the performance was so very poor. The only suggestion which I can offer is that the solenoid was not made of drawn wire, but cut from a cast cylinder.

Coming back to the prediction of magnetocaloric cooling and to the thermodynamic considerations mentioned by Professor Gorter, you may wonder how we all could fail to predict the Meissner effect. I think the reason is that none of us checked the details of the experiment[6] with a lead sphere—which unfortunately was hollow—and that we all took this result for granted: that the path of a persistent current is frozen into the superconductor.

The publication of Meissner's discovery practically coincided with the arrival of another refugee from Germany in Oxford. This was Fritz London. He immediately began to take an interest in superconductivity and, when early in 1934 he was joined by his younger brother, their work on the electromagnetic theory progressed rapidly. I knew Heinz London from Breslau, where in 1931 he had started work on a thesis concerning alternating currents in superconductors. The original suggestion for it had come from Schottky, at Siemens, who for technical reasons wanted to know whether there would be a production of heat. Delayed by poor cryogenic facilities and—let us admit it now, after more than thirty years—by a certain lack of manual dexterity, Heinz London became impatient and attacked the problem theoretically. Soon he realized that the current must penetrate a little into the superconductive metal, and postulated the correct formula for the depth. He was quite unaware of Mrs. de Haas–Lorentz' paper of 1925, and so clearly were Becker, Heller, and Sauter[7] who, in 1933, published the same result. Heinz London, however, went further. For him, finding the penetration depth was only a step towards the solution of his problem. Assuming a two-fluid model with superconductive and normal electrons, he predicted that at high frequencies the former would be unable to shield the latter completely, and that Joule heat would be developed. The experiment is not a simple one, requiring a short wave technique which was very undeveloped before the war, and its success eluded Heinz London until 1940.[8]

The immediate reaction of almost everyone to the Meissner effect was to try to fit it into Maxwell electrodynamics but, with the permeability changing to zero, the equations became indeterminate. It was Fritz London who, approaching the problem from his brother's acceleration equation, came to the conclusion that superconductivity demanded an entirely new relation in which the current is connected not with the electric but with the magnetic field. The form which contains the Meissner effect is the famous equation $-\Lambda c \, \text{curl} \, \mathbf{J} = \mathbf{H}$, where the constant Λ accounts for the penetration depth. The Londons' work was completed, essentially in its final form, in the autumn of 1934, when their famous paper[9] was submitted to the Royal Society, to be published in March 1935. In a number of subsequent papers they elaborated their theory, and Fritz, in particular, tried to discover its significance for the still outstanding microscopic interpretation of superconductivity.

At the same time, in 1933 and 1934, I was busy with a host of experiments. There was first of all the predicted magnetocaloric effect which Miss Judith Moore (later Mrs. Hull) and I could verify on tin in the winter of 1933/34.[10] By that time, the practical use of this cooling method had become less important than it had been two years earlier because of paramagnetic cooling. While these experiments got under way, a Canadian research student, J. D. Babbitt, and I set out immediately after Meissner's publication to repeat his observations, using solid and hollow tin spheres as samples, and bismuth probes as indicators.[11] The effect was there, and it was more pronounced in the solid than in the hollow sphere. Yet

[4] K. Mendelssohn, Nature **132**, 602 (1933).
[5] W. J. de Haas and J. Voogd, Comm. Phys. Lab. Univ. Leiden, No. 208b (1930).
[6] H. Kamerlingh Onnes, Comm. Phys. Lab. Univ. Leiden, Suppl. No. 50a (1924).
[7] R. Becker, G. Heller, and F. Sauter, Z. Physik **85**, 772 (1933).
[8] H. London, Proc. Roy. Soc. (London) **A176**, 522 (1940).
[9] F. and H. London, Proc. Roy. Soc. (London) **A149**, 71 (1935).
[10] K. Mendelssohn and J. R. Moore, Nature **133**, 413 (1934).
[11] K. Mendelssohn and J. D. Babbitt, Nature **133**, 459 (1934); Proc. Roy. Soc. (London) **A151**, 316 (1935).

there were two disturbing features in which our results differed from those of Meissner. Even in the solid sphere the expulsion of flux was incomplete, and it extended over an interval of field, although we measured the flux change equatorially. As we were to learn soon, the reason for this was that we had used polycrystalline tin of commercial purity.

Our results permitted the conclusion that the incomplete Meissner effect was not anything fundamental, but due to the retention of normal regions within the sample. The next step, therefore, was to employ still simpler geometry, i.e., needle-shaped specimens oriented in the field direction, and to extend the work to other materials. By hitting upon a simple technique of measurement, we were able to make rapid progress, and we soon had results showing a whole spectrum of behavior, from a complete Meissner effect in pure mercury to a complete freezing in of flux in alloys. In between, there were all the intermediate stages, showing clearly that the presence of even a small proportion of a second constituent caused radical departure from the ideal behavior. Moreover, we found that, in those cases which differed from ideal behavior, there were two critical fields instead of one. There was the field at which the electrical resistance became normal, and for which we retained the term "threshold field" (now H_{c2}), and a much lower value at which magnetic flux first began to penetrate the sample. This we called the "penetration field" (now H_{c1}).

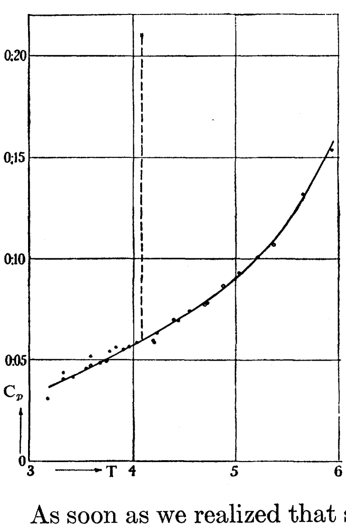

FIG. 1. The specific heat of the alloy $PbTl_2$. The dotted line indicates the discontinuity which had to be expected according to Rutgers' formula.

As soon as we realized that superconductive alloys behave differently from pure metals, our minds went back to the ill-fated solenoid, and we began to wonder whether the high threshold values reported in Leiden might have a quite different significance from the low values, such as in pure mercury or tin. There was an interesting way of testing it, which did not require the use of a strong magnet, and that was by measuring the specific heat. According to Rutger's formula, alloys should have an enormous discontinuity at the transition point. We chose $PbTl_2$ and no such discontinuity was found.

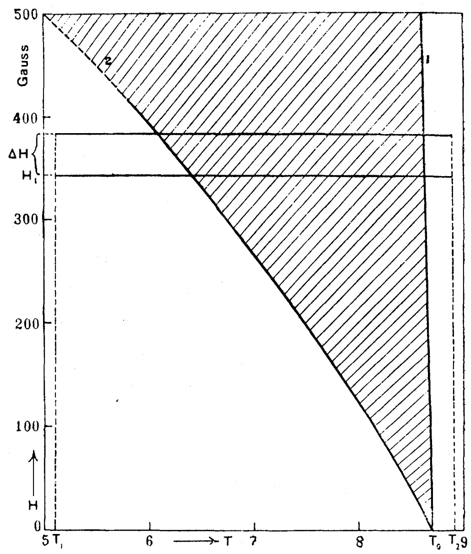

FIG. 2. Experiment to test the sponge model. A Pb–Bi alloy was warmed in a constant field H_1. Flux changes were recorded when a small additional field ΔH was switched on or off. Curve 1 denotes H_{c2} and curve 2 H_{c1}.

There were only three pairs of hands to do the work, but we were lucky, as well as enthusiastic, and within a few months, by the autumn of 1934,[12] we were able to publish the magnetic as well as the calorimetric data. Others were clearly just as keen, and were thinking on similar lines, and early in the next year similar results were reported from Leiden, Toronto, and Kharkov. Shubnikov, in particular, had also hit on checking Rutgers' formula, obtaining the same result but on a different alloy, which was most encouraging.

In the meantime we were trying to take the next step, which was to fit our results together somehow, and to find an explanation for all the diverse observations. The failure of Rutgers' formula showed that Gorter's thermodynamic treatment was not directly applicable to alloys. Therefore, the high threshold value was not a property of the whole alloy but confined to regions in it, and the retention of flux showed that these regions were multiply connected. Thus, the sponge model came into being, and the

[12] T. C. Keeley, K. Mendelssohn, and J. R. Moore, Nature **134**, 773 (1934).

next step was to test it. This again was very simple. We warmed an alloy sample slowly in a constant magnetic field, switching on and off a small additional field and recording the flux changes each time. When passing the penetration curve, we saw flux entering on switching on, but not leaving when switching off; we were filling up the meshes of the sponge with flux. This went on until we reached the threshold curve. However, on cooling in a field, flux was neither entering nor leaving; the sponge was soaked magnetically to capacity. So, by early April 1935,[13] we had enough confidence in the sponge model to publish it. We were able to report further confirmation of it, based on calorimetric data, on May 30th at a discussion on superconductivity held at the Royal Society under the chairmanship of McLennan.[14] Meissner, Keesom, Brillouin, and the Londons contributed to the discussion at which still another laboratory entered the ring. The first experiments on superconductivity from Cambridge were reported by Shoenberg.

We were very curious about the nature of the filaments of high threshold value in our alloys, and here the frequent discussions with the Londons proved a great benefit. Fritz London was usually reluctant to leave his study at the top of Headington Hill, and had the curious habit of conducting lengthy scientific conversations over the telephone, but they lost nothing by this of the clear and precise formulation of which he was master. The Londons pointed out that, according to their theory, superconductors of the size of the penetration depth must have threshold fields higher than the bulk material, and I concluded that regions of strain such as produced by impurities or other lattice imperfections were forming thin filaments which gave rise to the sponge. This meant that the sponge can take up an appreciable fraction of the material, a point which has often been missed in recent discussions on the subject. The results on alloys led Gorter[15] and, independently, Heinz London[16] to theoretical discussions of filamentary models which are tantamount to type II superconductors, and the latter correctly postulated a negative surface tension for the realization of this condition. Sponge structure was also found in fairly pure tantalum[13,17] and from then on we went in search of better and better specimens of hard metals, to see whether, in the end, they would exhibit the same pattern as the soft ones.

One of the highlights of the prewar years was the Sixth International Low Temperature Conference in the Hague in 1936. It provided a forum for discussion at which people from nearly all the world's laboratories met each other, many for the first time. I say "nearly all" laboratories, because the Russian colleagues who had signified their intention to attend failed to turn up. We were given the glib explanation that they were too busy to attend meetings. I was particularly disappointed because Shubnikov's group in Kharkov was carrying out work[18] very similar to ours, but communication had been limited to exchanges of reprints with greetings scribbled on them. As it happened, Shubnikov was arrested shortly afterwards on charges from which he was to be posthumously exonerated twenty years later. His series of superb experiments thus came to an abrupt end (probably in 1937) without allowing him time to give an interpretation of his observations. They formed the starting point of Abrikosov's theory in 1957.

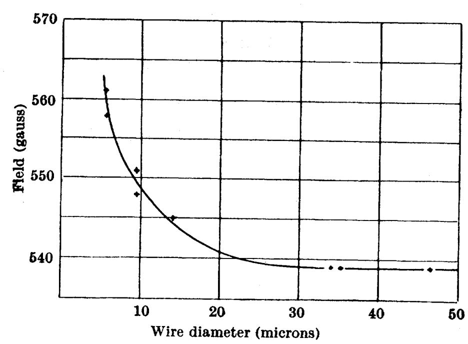

Fig. 3. Enhancement of the critical field of pure lead with decreasing wire diameter.

By 1936 systematic work on metals with controlled and graded amounts of impurity had provided us with more knowledge on sponge structure, including a phenomenon which has recently become known as "flux jumps." However, the most important piece of information, the enhancement of threshold field with diminishing size, remained a theoretical prediction. The trouble was that thin superconductive deposits on metal substrates were useless, being suspect of alloying; and that vacuum evaporation technique was in its infancy. Finally, with great skill and patience, Rex Pontius, a research student from Idaho, succeeded in 1937 in making strain-free wires of pure lead down to 5-micron diameter, and these showed the enhancement pre-

[13] K. Mendelssohn and J. R. Moore, Nature **135**, 826 (1935).
[14] K. Mendelssohn, Proc. Roy. Soc. (London) **A152**, 34 (1935).
[15] C. J. Gorter, Physica **2**, 449 (1935).
[16] H. London, Proc. Roy. Soc. (London) **A152**, 650 (1935).
[17] K. Mendelssohn and J. R. Moore, Phil. Mag. **21**, 532 (1936).
[18] L. V. Shubnikov, V. I. Khotkerich, J. D. Shepelev, and J. N. Rjabinin, Physik. Z. Sowjet Union, Suppl., p. 39 (1936).

dicted by the London theory.[19] A few years later, Shalnikov,[20] in Moscow, and H. London and others[21] in Bristol, made very much thinner films with threshold fields which approached those of alloys. Pontius[22] provided yet another test of the sponge model by finding the hysteresis in heat conductivity measurements which we had expected.

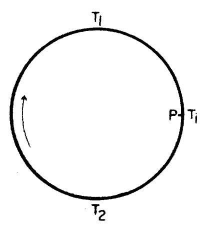

FIG. 4. The arrangement for measuring the specific heat of the persistent current consisted of a superconductive ring, held at different temperatures, T_1 and T_2, at opposite ends of a diameter. The temperature was recorded at T_i when the direction of the current was reversed. From the fact that no change in temperature took place, it was concluded that the entropy of the electrons engaged in a supercurrent is zero.

There remained Heinz London's first problem of high frequency heating in superconductors. With another research student, John Daunt, we attacked this in 1936 from the short wave end by using infrared absorption, and we found indeed that as much energy is absorbed in the superconductive as in the normal state.[23] The onset of absorption at waves of 20-cm length was ultimately found by Heinz London himself, in 1940. A few years earlier, in 1938, Daunt and I had tried the meter region for which we had equipment, but obtained a negative result which we never published. However, a chance observation at these experiments led us to the work on helium films, which was soon to occupy most of our time. But before this, one more step forward in superconductivity was to come.

Two years earlier, Daunt and I had become interested in deriving thermodynamic data from our measured critical field curves,[24] and noted that at low temperatures the electronic entropy difference between the two states approached γT, and we began to suspect that, even at finite temperatures, the entropy of the system of superconductive electrons might be zero. To test this, we measured the heat carried by a persistent current, and found that it is indeed zero. Switching our thoughts to the corresponding problem in superfluid helium, we found, in 1939, the mechanocaloric effect.

[19] R. B. Pontius, Nature **139**, 1065 (1937); Phil. Mag. **24**, 787 (1937).

[20] A. Shalnikov, Nature **142**, 74 (1938).

[21] E. T. S. Appleyard, J. A. Bristow, H. London, and A. D. Misener, Proc. Roy. Soc. (London) **A172**, 540 (1939).

[22] K. Mendelssohn and R. B. Pontius, Phil. Mag. **24**, 777 (1937).

[23] J. G. Daunt, T. C. Keeley, and K. Mendelssohn, Phil. Mag. **23**, 264 (1937).

[24] J. G. Daunt and K. Mendelssohn, Proc. Roy. Soc. (London) **A160**, 127 (1937).

With the conviction that war was approaching fast, and with the most interesting problems of superfluidity and superconductivity on our hands, we found ourselves working against time. We could not stop to publish more than a short note[25] on the zero specific heat of the persistent current, leaving the full paper until after the war. Thus, the suggestion that superconductivity is distinguished by an energy gap was published in 1946[26] instead of 1938.

There was, as Professor Gorter has mentioned, a great deal of discussion with our friends in Leiden, but the closest contact naturally formed with Shoenberg and Allen in Cambridge. Even if the rail connection between the two ancient centers of learning was practically nonexistent, they are within easy motoring distance of each other. When David Shoenberg bought an old car, his first—and, I regret to recall, unsuccessful—run was to Oxford. Much of his early work was concerned with accurate and careful measurements of the magnetization curves of spheres and rings.[27] The problem of small dimensions he approached in a very original manner by using colloids and emulsions.[28] From his stay in Russia in the late thirties he brought back accounts of Landau's theory of the intermediate state, and of other Soviet work which he published in his book. In those days also, Henry Boorse came to Cambridge to look for a resistance anomaly in aluminium.

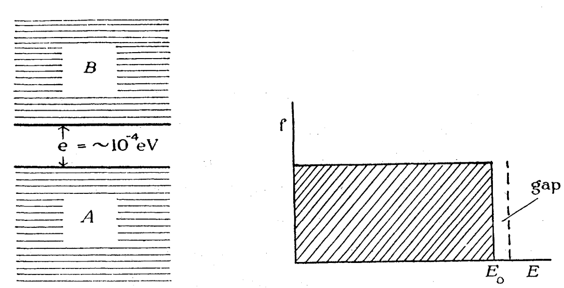

FIG. 5. The empirical model suggested for a superconductor on the basis of the result that the entropy of the supercurrent is zero.

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

[25] J. G. Daunt and K. Mendelssohn, Nature **141**, 116 (1938).

[26] J. G. Daunt and K. Mendelssohn, Proc. Roy. Soc. (London) **A185**, 225 (1946).

[27] D. Shoenberg, Proc. Roy. Soc. (London) **A155**, 712 (1936).

[28] D. Shoenberg, Proc. Roy. Soc. (London) **A175**, 49 (1940).

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. It needed radar, rockets, and atomic bombs to loosen up the stringency of prewar finance, which now must appear as an equal folly to the young scientific generation of the sixties.

HIGH FIELD SUPERCONDUCTIVITY

CHAIRMEN: *J. E. Kunzler, C. J. Gorter, C. P. Bean, J. K. Hulm*

Type II or London Superconductors

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I. INTRODUCTION

When a superconductor is placed in a magnetic field two competing processes take place. On the one hand, according to the London equation, which we may write

$$\nabla^2 H = H/\lambda_L^2 , \qquad (1)$$

the superconducting state attempts to exclude the magnetic field H from all but a thin penetration layer of thickness of the order of λ_L, the London penetration depth. On the other hand, in accordance with the concept of spatial coherence introduced by Pippard,[1] any local perturbation of the superconducting order parameter caused by the magnetic field spreads out to a distance of the order of ξ, the range of coherence, from the center of the disturbance.

It was recognized for some time[2] that Eq. (1) gave no indication of the existence of the positive interphase surface energy which is required in order to explain the Meissner effect, and it was left to Pippard[3] to point out that a plane interphase boundary would indeed have a positive surface energy if ξ were greater than λ_L. Thus, the sign of $\xi - \lambda_L$ was destined to play a crucial role in determining the type of magnetic behavior shown by a superconductor. Following, with slight modification, an initiative due in particular to a number of Russian authors, it seems fitting to call type I superconductors, for which $\xi > \lambda_L$, Pippard superconductors, and type II superconductors, for which $\lambda_L > \xi$, London superconductors.

The reversible magnetic behavior of Pippard superconductors has been understood for some time and measurements on the structure of the intermediate state of such superconductors enabled their positive surface energy to be determined even before its origin was fully understood. For a Pippard superconductor lying in a field H_e the magnetization curve has the familiar triangular shape, all trace of superconductivity disappearing at the critical field H_c, defined by

$$F_n - F_s = -\int_0^\infty M dH_e = \frac{H_c^2}{8\pi} , \qquad (2)$$

where M is the magnetization, and F_n and F_s are the free energies per unit volume of the normal and superconducting states, respectively. There are many indications, as yet mostly of a qualitative character, which suggest that flux trapping in Pippard superconductors is due to the pinning down of interphase boundaries by extended defects in the specimen.

As early as 1935, when it was known that many alloys did not conform to the Pippard or "ideal" type

[1] A. B. Pippard, Proc. Roy. Soc. (London) **A203**, 210 (1950).
[2] F. London, *Superfluids* (John Wiley & Sons, Inc., New York, 1950), Vol. I, pp. 125–130.
[3] A. B. Pippard, Proc. Cambridge Phil. Soc. **47**, 617 (1951).