

Review

The Path to Type-II Superconductivity

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Abstract: Following the discovery of superconductivity by Heike Kamerlingh Onnes in 1911, research concentrated on the electric conductivity of the materials investigated. Then, it was Max von Laue who in the early 1930s turned his attention to the magnetic properties of superconductors, such as their demagnetizing effects in a weak magnetic field. As a consultant at the Physikalisch-Technische Reichsanstalt in Berlin, von Laue was in close contact with Walther Meissner at the Reichsanstalt. In 1933, Meissner together with Robert Ochsenfeld discovered the perfect diamagnetism of superconductors (Meissner–Ochsenfeld effect). This was a turning point, indicating that superconductivity represents a thermodynamic equilibrium state and leading to the London theory and the Ginzburg–Landau theory. In the early 1950s in Moscow, Nikolay Zavaritskii carried out experiments on superconducting thin films. In the theoretical analysis of his experiments, he collaborated with Alexei A. Abrikosov and for the first time they considered the possibility that the coherence length ξ can be smaller than the magnetic penetration depth λ_m . They called these materials the “second group”. Subsequently, Abrikosov discovered the famous Abrikosov vortex lattice and the superconducting mixed state. The important new field of type-II superconductivity was born.

Keywords: type-II superconductivity; Meissner–Ochsenfeld effect; thermodynamics; London theory; Ginzburg–Landau theory; mixed state

1. The Early Period: Kamerlingh Onnes in Leiden

During the last years of the 19th century, **Heike Kamerlingh Onnes** set up a laboratory in Leiden for experimental low-temperature physics, which soon found worldwide recognition. Motivated by the research and ideas of **Johannes Diderik van der Waals** at the University of Amsterdam, Kamerlingh Onnes was interested in the thermodynamic properties of gases and liquids at low temperatures. In 1860, van der Waals published his law of corresponding states. At the time, in Europe a competition had started between several laboratories regarding the generation of low temperatures and the liquefaction of gases. In 1895, **Carl von Linde** in Germany and **William Hampson** in England announced the application of the Joule–Thomson effect for the large-scale liquefaction of gases. In the same year, for the first time, von Linde achieved the liquefaction of air by combining the Joule–Thomson effect with the counter-flow heat exchanger proposed already in 1857 by **Werner Siemens**. Based on this process, the liquefaction of neon, hydrogen, and finally helium can be achieved, with the goal of reaching lower and lower temperatures. On 9–10 July 1908, for the first time, Kamerlingh Onnes’ team achieved the liquefaction of helium as the last remaining noble gas, thereby reaching the record low temperature value of 4 K (−269 °C). Then, in 1911, during the cooling of a mercury sample down to 4 K, Kamerlingh Onnes discovered the phenomenon of superconductivity, i.e., the disappearance of any measurable electric resistance in a metal. On 28 April 1911, he reported this to the Academy in Amsterdam.

Measurements of the electric resistance of metals at distinctly lower temperatures than could be reached before were interesting, since at the time three predictions existed about resistance behavior at

very low temperatures: (1) with decreasing temperature the resistance decreases to reach the value zero; (2) it remains constant; (3) it increases again. The choice of mercury for the first experiment was suggested by the fact that it could be prepared with high purity due to its low melting point. On 8 April 1911, Kamerlingh Onnes and his team consisting of Cornelis Dorsman, Gerrit Jan Flim, and the student Gilles Holst observed how the electric resistance of a mercury sample filling a small glass capillary decreased with decreasing temperature. When the temperature finally reached 4 K, the curve showed a sharp turn down, and the resistance fell to an unmeasurable small value. Initially, there was some irritation, since it was suspected that the electric circuitry would be defect.

Following this discovery of superconductivity in mercury, it was also observed in other metals and alloys. In addition to mercury, during the early period it was also found in aluminum, lead, indium, zinc, and tin. Soon after his discovery of superconductivity, Kamerlingh Onnes looked into the question of whether superconductors can serve for the transport of electric power without any losses. To his great disappointment, he found that the magnetic field of high electric currents is highly detrimental for superconductivity. It turned out that in addition to the critical temperature T_c , which must not be exceeded, there also exists a critical magnetic field $H_c(T)$, above which superconductivity disappears. In Figure 1, we show the temperature dependence of the critical magnetic field $H_c(T)$. Starting from the value $H_c = 0$ at $T = T_c$, with decreasing temperature the critical magnetic field increases, reaching its maximum value at $T = 0$.

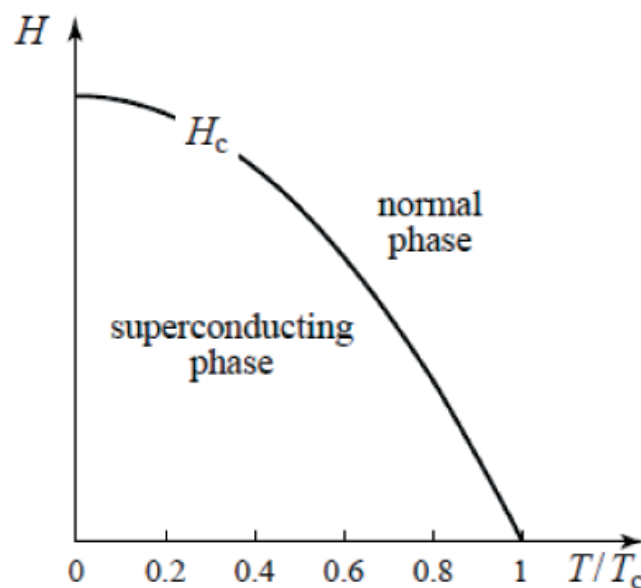


Figure 1. Temperature dependence of the critical magnetic field $H_c(T)$.

2. Meissner–Ochsenfeld Effect: The Physikalisch-Technische Reichsanstalt in Berlin

At the *Physikalisch-Technische Reichsanstalt* (PTR) in Berlin, the developments in the field of low-temperature physics had been followed with great interest. The PTR was founded in 1887 mainly due to the initiative of **Hermann von Helmholtz** and Werner Siemens. It was charged with fundamental research in physics, supplementing the activities of the rapidly growing industry in Germany. In particular, Carl von Linde, as a board member of the PTR, suggested systematic studies at low temperatures and the establishment of a low-temperature laboratory at the Reichsanstalt. So, during his presidency of the PTR, in 1913 **Emil Warburg** charged **Walther Meissner** with the task of installing a liquefier for hydrogen. Walther Meissner had studied mechanical engineering at the *Technische Hochschule* in Berlin-Charlottenburg. Then, he had spent three years studying mathematics and physics at Friedrich-Wilhelms University in Berlin, where in 1907 he obtained his PhD under **Max Planck**. In 1908, he joined the PTR, where he started working in the pyrometry laboratory. After his

new assignment in 1913, Meissner began operating an improved liquefier based on a construction by Walther Nernst. As it did everywhere else, World War One interrupted Meissner's research activities. Then, in 1920 he began thinking about a facility for the liquefaction of helium, and during 1922–1925 he was able to realize this project. Furthermore, Meissner had to spend almost a year for extracting the helium from the air, being strongly supported by the Linde company. On 7 March 1925, helium was liquefied for the first time at the PTR. Now, worldwide the *Reichsanstalt* was the third location where experiments with liquid helium could be carried out, after Leiden in Holland, and since 1923, Toronto in Canada.

Initially, Meissner's experiments concentrated on the electric resistance of materials at low temperatures and the search for new superconductors. He succeeded in discovering superconductivity in tantalum, thorium, titanium, niobium, and likely in vanadium. However, Meissner was always also interested in the fundamental principles of superconductivity. In addition to the electrical conductivity, he turned to the magnetic properties of superconductors. Here, his contacts with **Max von Laue** proved to become highly important. Von Laue had an appointment as a professor at Friedrich-Wilhelms University in Berlin. During his presidency at the PTR 1922–1924, **Walther Nernst** realized that the program at the *Reichsanstalt* would benefit from support in theoretical physics. So, he arranged for von Laue to become a consultant at the PTR, working there one day per week as a theoretician. In the early 1930s, von Laue turned his attention to the demagnetizing effects expected in the case of a perfect electric conductor in a weak magnetic field. Likely because of his contact with Max von Laue, Meissner was planning experiments dealing with the magnetic behavior of superconductors. As chairman of the physics committee of the *Notgemeinschaft* in Germany, von Laue could help in hiring an additional experimental physicist, **Robert Ochsenfeld**, to assist Meissner in these experiments. Ochsenfeld joined the PTR at the end of 1932. He was financed by a program supporting young academics with an opportunity to gain additional research experience during the world economic crisis.

Meissner and Ochsenfeld made an important and unexpected discovery: in the interior of a superconductor, a magnetic field vanishes, being completely expelled by means of superconducting shielding currents flowing along the surface. This became known as the Meissner–Ochsenfeld effect [1]. They performed their experiments with superconducting lead and tin and measured the magnetic field very close to the surface of a superconductor, above and below its critical temperature. In Figure 2, we schematically show the magnetic field expulsion due to the Meissner–Ochsenfeld effect. Max von Laue called the discovery of this effect a turning point in the area of superconductivity. It was the starting signal of important theoretical developments which will be outlined briefly in the next section.

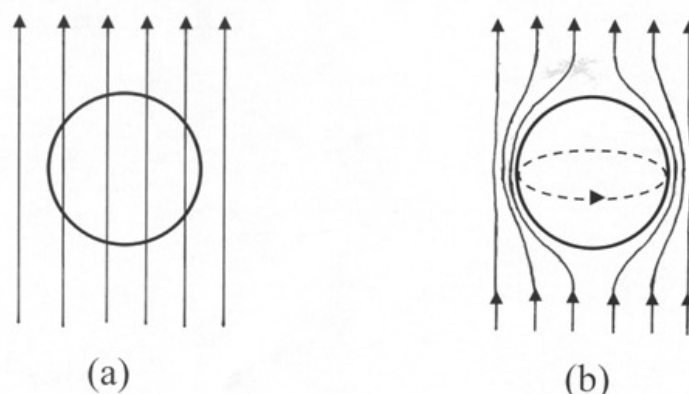


Figure 2. Meissner–Ochsenfeld effect. (a) In the normal state above the critical temperature, the spherical superconductor is completely penetrated by the external magnetic field. (b) Below the critical temperature, the superconductor completely expels the magnetic field from its interior, as long as the critical magnetic field is not exceeded. This field expulsion is effected by superconducting shielding currents flowing along the surface of the superconductor.

3. Theoretical Advances

The existence of the Meissner–Ochsenfeld effect shows that superconductivity represents a thermodynamic equilibrium state, which is reached independently from the path leading to it. In the end, the only requirement is: $T < T_c$ and $H < H_c(T)$. **Cornelis J. Gorter** and **Hendrik B. Casimir** were the first who pointed out this important consequence. In addition, they showed that, based on this effect, the energy difference between the normal and superconducting states can be calculated [2]. It turns out that the difference between the free Gibbs energy density in the normal state, $G_n(T, 0)$, and in the superconducting state, $G_s(T, 0)$, at zero magnetic field is

$$G_n(T, 0) - G_s(T, 0) = \frac{1}{8\pi} H_c^2(T) \quad (1)$$

For a typical value of the critical magnetic flux density $B_c = 10^{-2}$ Tesla, this difference in energy density amounts to $398 \text{ erg/cm}^3 = 2.49 \times 10^{14} \text{ eV/cm}^3$. If we express this energy difference in terms of energy per electron, we obtain the small value of only a few meV per electron. Hence, the theoretical explanation of superconductivity turned out to be quite difficult.

In 1935, the brothers **Fritz London** and **Heinz London** presented a phenomenological theory of superconductivity and the Meissner–Ochsenfeld effect [3]. Their theory also yielded a value of the magnetic penetration depth λ_m within which the superconducting shielding currents are flowing along the surface. In a brief outline of the London theory, we start with the equation for the forces experienced by an electron in the presence of an electric field $\vec{E}$

$$m \frac{\partial \vec{v}_s}{\partial t} = (-e) \vec{E} \quad (2)$$

(m = electron mass; e = elementary charge; $\vec{v}_s$ = velocity of the superconducting electrons). In (2), we have ignored a dissipative part. Introducing the supercurrent density $\vec{j}_s = (-e)n_s\vec{v}_s$, we obtain the relation

$$\vec{E} = [m/(e^2 n_s)] \frac{\partial \vec{j}_s}{\partial t} = \mu_0 \lambda_m^2 \frac{\partial \vec{j}_s}{\partial t} \quad (3)$$

In (3), λ_m denotes the magnetic penetration depth given by

$$\lambda_m^2 = m/(\mu_0 n_s e^2) \quad (4)$$

(n_s = density of the superconducting electrons; μ_0 = permeability of vacuum). Because of Maxwell's equation $\text{curl } \vec{H} = \vec{j}$, one finds for the maximum density j_s of the superconducting shielding current $j_s = H_c/\lambda_m$. On the other hand, Maxwell's equation $\text{curl } \vec{E} = -\frac{\partial \vec{B}}{\partial t}$ together with (3) yields

$$\mu_0 \lambda_m^2 \text{curl} \left(\frac{\partial \vec{j}_s}{\partial t} \right) + \frac{\partial \vec{B}}{\partial t} = 0 \quad (5)$$

(B = magnetic flux density). Ignoring the time-derivative in (5), Fritz and Heinz London postulated a new equation

$$\mu_0 \lambda_m^2 \text{curl } \vec{j}_s + \vec{B} = 0 \quad (6)$$

Maxwell's equation $\text{curl } \vec{H} = \vec{j}$ and the vector relation $\text{curl } \text{curl } \vec{x} = \text{grad } \text{div } \vec{x} - \Delta \vec{x}$ finally yield

$$\Delta \vec{H} = \frac{1}{\lambda_m^2} \vec{H} \quad (7)$$

with the solution

$$\vec{H}(x) = \vec{H}(0) \exp(-x/\lambda_m). \quad (8)$$

Equation (8) indicates that the magnetic field decays exponentially towards the interior of the superconductor, the characteristic decay length being the magnetic penetration depth λ_m . In the limit $T \ll T_c$ typical values of the magnetic penetration depth are in the range $\lambda_m = 40\text{--}80$ nm. In the limit $T \rightarrow T_c$, we have $\lambda_m \rightarrow \infty$.

In 1950, **Brian Pippard** from Cambridge, UK, pointed out that in a superconductor, in addition to the magnetic penetration depth λ_m , there also exists another characteristic length, which limits the spatial sharpness, within which the superconducting properties can change: the superconducting coherence length, ξ [4]. Superconducting properties can change only within a distance larger than the coherence length ξ .

The finite range of coherence in a superconductor also represented an important result of the theory presented in 1950 by the two Russians, **Vitaly L. Ginzburg** and **Lev D. Landau** [5]. The phenomenological Ginzburg–Landau theory describes the superconducting state of the electrons in terms of a macroscopic wave function

$$\psi(\vec{r}, t) = |\psi(\vec{r}, t)| e^{i\phi(\vec{r}, t)} \quad (9)$$

with an amplitude $|\psi(\vec{r}, t)|$ and a phase $\phi(\vec{r}, t)$. The absolute value $|\psi(\vec{r}, t)|$ indicates the local density n_s of the superconducting electrons. The phase $\phi(\vec{r}, t)$ describes the superconducting currents.

In this theory, the density f of the free energy of the electrons is expanded in powers of the “order parameter”, assuming that $\psi(\vec{r}, t)$ remains small, which is valid below and close to T_c . In calculating the minimum value $f(\vec{r}, t)$ of the free energy density in the case of a spatial variation of the order parameter $\psi(\vec{r}, t)$ and of the magnetic field or the vector potential $\vec{A}(\vec{r})$, one obtains the following Ginzburg–Landau equations:

$$\alpha\psi + \beta|\psi|^2\psi + \frac{1}{2m^*} \left(\frac{\hbar}{i} \nabla - \frac{e^*}{c} \vec{A} \right)^2 \psi = 0 \quad (10a)$$

$$\vec{j}_s = \frac{e^* \hbar}{2m^* i} (\psi^* \nabla \psi - \psi \nabla \psi^*) - \frac{e^{*2}}{m^* c} \psi^* \psi \vec{A} \quad (10b)$$

Equation (10a) has the form of a Schrödinger equation with the eigenvalue $-\alpha$ of the energy. The part $\beta|\psi|^2\psi$ represents an effective repulsive potential. Equation (10b) is the quantum mechanical description of a particle current. Both equations apply to particles with mass m^* and charge e^* .

The description of the superconducting state in terms of a macroscopic wave function (9) turned out to be highly fruitful. The explanation of the characteristic lengths $\xi(T)$ and $\lambda_m(T)$, the critical current density j_c , and the quantization of the magnetic flux represent an important result. The minimum value of the magnetic flux in a superconductor, the magnetic flux quantum $\Phi_0 = h/2e = 2.068 \times 10^{-15}$ Tm², simply results from the fact that the macroscopic wave function $\Psi(\vec{r}, t)$ must reproduce itself exactly after the spatial coordinate point has circled once around the magnetic flux region and returned to the starting point.

At the end of this section, we briefly discuss the first valid microscopic theory of superconductivity: the theory proposed by **J. Bardeen**, **L. N. Cooper**, and **J. R. Schrieffer** (BCS theory) [6], even though it did not directly motivate the concept of type-II superconductivity. An important new idea was proposed in 1950 by **H. Fröhlich** [7] and independently by **J. Bardeen** [8]. They recognized that an electron in a crystal lattice is surrounded by a cloud of virtual phonons which are emitted and absorbed continuously. As a result, the crystal lattice is slightly distorted in the neighborhood of an electron. The fact that the interaction of the electrons with the crystal lattice (phonons) plays an important role for superconductivity was been clearly demonstrated by the isotope effect observed in the early 1950s. In this case, it was shown that the critical temperature T_c of superconductivity depends on the nuclear mass M of the crystal atoms. Specially prepared isotopically pure samples of lead, mercury, and tin showed the proportionality

$$T_c \propto \frac{1}{M^\alpha} \quad (11)$$

with $\alpha = 0.5$.

In 1956, **L. N. Cooper** [9] proposed the formation of bound electron pairs in a degenerate Fermi gas, since then referred to as Cooper pairs. The attractive interaction and binding energy of Cooper pairs result from the weak polarization of the crystal lattice, which overcompensates the Coulomb repulsion between the two electrons. Cooper pairs always consist of two electrons having oppositely oriented spin such that the total spin of each pair vanishes. In this case, the Pauli principle does not apply, and all Cooper pairs can occupy the same quantum state described by a macroscopic wave function. BCS theory predicts an energy gap in the energy spectrum of the electrons, which was confirmed in many experiments.

An interesting discussion of “Failed Theories of Superconductivity” has been given recently by J. Schmalian [10]. In his list of world-famous physicists who have failed appear A. Einstein, N. Bohr, R. Kronig, L. D. Landau, F. Bloch, L. Brillouin, W. Heisenberg, F. London, M. Born, H. Fröhlich, and R. Feynman.

4. The Abrikosov Vortex Lattice and the Mixed State

In the 1930s, experiments already indicated that some ideas about superconductivity needed to be modified. Almost simultaneously in Oxford [11,12], Leiden [13], and Kharkov, [14] anomalies were observed in the specific heats and magnetic properties of superconducting alloys. As a result, in 1935 **K. Mendelssohn** proposed his famous sponge model claiming that superconductivity is restricted to a small volume fraction within the material [15].

The relative length scale of the coherence length ξ and the magnetic penetration depth λ_m turned out to become an important issue. It was generally felt that the length difference $\xi - \lambda_m$ must always be positive ($\xi > \lambda_m$), since it determines the energy of the wall separating the superconducting phase from the normal phase of the domain pattern of the intermediate state. The young theoretical physicist **Alexei A. Abrikosov** working at Moscow University achieved an important breakthrough. He was a roommate of **Nikolay Zavaritzkii**, who worked at the Kapitza Institute for Physical Problems and performed experiments with superconducting thin films. Abrikosov examined the predictions of the Ginzburg–Landau theory regarding the dependence of the critical magnetic field on the film thickness and the temperature. Motivated by these experiments, in their discussions Abrikosov and Zavaritzkii considered for the first time the possibility that the length difference $\xi - \lambda_m$ can also become negative ($\xi < \lambda_m$). Based on the Ginzburg–Landau theory, Abrikosov calculated the critical magnetic field also in the case of $\xi < \lambda_m$. He was able to show that only in this case was a good agreement with Zavaritzkii’s experiments achieved. Now, Abrikosov and Zavaritzkii were convinced that they had found a new type of superconductors, which they called “the second group”. Subsequently, these materials were named type-II superconductors, whereas the materials with positive wall energy ($\xi > \lambda_m$) are called type-I superconductors.

When Abrikosov then applied the Ginzburg–Landau theory for analyzing type-II superconductivity more exactly, he found that in the presence of an external magnetic field a new superconducting state is possible: the superconductor is penetrated by a lattice of magnetic flux quanta. He had discovered the famous Abrikosov vortex lattice and the superconducting mixed state. On several occasions, Abrikosov described how he was led to this discovery, which earned him the Nobel Prize in 2003 [16–18]. Experimentally, the Abrikosov vortex lattice was observed for the first time in 1964 by small-angle neutron diffraction [19]. An important demonstration, which finally convinced most skeptical people, was presented in 1967 based on the Bitter decoration technique [20]. In Figure 3, we schematically show a lattice of nine magnetic flux lines and the Bitter pattern obtained in the case of a superconducting niobium plate.

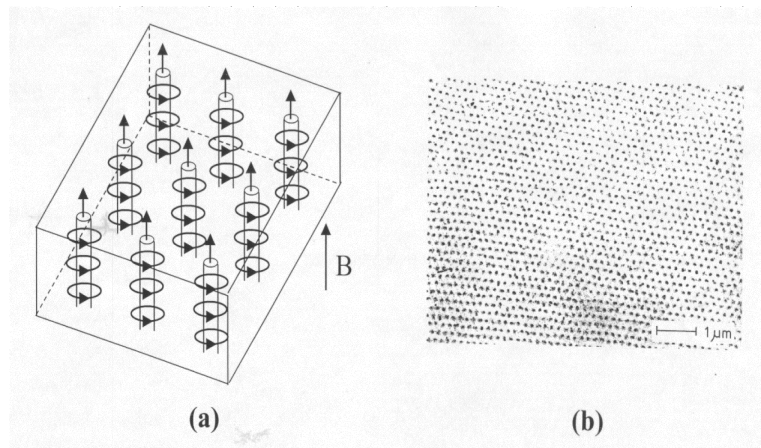


Figure 3. Abrikosov vortex lattice: (a) Schematics of nine magnetic flux lines generated by circular supercurrents around them. (b) Bitter pattern of the vortex lattice obtained in the case of a superconducting niobium plate of 0.5 mm thickness. The dark spots mark the locations at which the individual magnetic flux quanta reach the surface of the niobium plate. (U. Essmann).

5. Flux Creep and Flux-Flow Resistance

Abrikosov vortices generate important effects in many areas of physics, and in particular in the case of superconducting microelectronic devices. In the following, we briefly discuss the appearance of electric resistance in superconductors due to the motion of magnetic flux quanta because of the Lorentz force generated by an applied electric current.

In the early 1960s, **Bernd Matthias**, working together with **Theodore Geballe** at Bell Laboratories discovered the new superconducting alloys Nb₃Sn and NbZr, showing promising high values for the critical electric current density and for the critical magnetic field. For the first time, technical applications of superconductivity in power electronics became an interesting possibility. It was found that these technically promising materials can develop a “critical state” depending on the electric current load. Above a “critical current density”, electric resistance appeared within a wide range between zero and its normal value. Together, Matthias and Geballe discovered the A15-compounds, leading to the record-high T_c of 23.2 K [21].

At the time, **Philip W. Anderson** recognized that a new process must be involved: dissipation because of the current-induced motion of magnetic flux quanta [22]. Previously, **C. J. Gorter** had already discussed the issue of flux flow as a resistive mechanism in a type-I superconductor [23]. The motion of magnetic flux quanta in a superconductor with velocity $\vec{v}_\phi$ causes the electric field $\vec{E}$, given by

$$\vec{E} = -\vec{v}_\phi \times \vec{B}. \quad (12)$$

The magnetic flux density is $\vec{B} = n\vec{\phi}_0$ with the magnetic flux quantum $\vec{\phi}_0$ and the areal density n of flux quanta. If the vortex motion is caused by the Lorentz force $\vec{f}_L = \vec{j} \times \vec{\phi}_0$ of an electric current of density $\vec{j}$, electric power is dissipated and electric losses appear, since $\vec{E}$ and $\vec{j}$ are oriented parallel to each other. The underlying process is described by the (simplified) force equation

$$\vec{j} \times \vec{\phi}_0 - \eta \vec{v}_\phi = 0, \quad (13)$$

where η is a damping coefficient. In (14), the forces are given per unit length of flux line. From (13) and (14), one obtains the flux-flow resistivity

$$\rho_f = \phi_0 B / \eta. \quad (14)$$

In Figure 4, we show a typical example of the flux-flow resistance. The flux-flow voltage is plotted versus the electric current in the case of a niobium foil of 18 μm thickness. We see that the voltage

sets in at a certain finite critical current, above which the curve turns upward and then turns into a branch increasing linearly with increasing current. The slopes of the linear branches represent the flux-flow resistance. With increasing magnetic field, the critical current decreases and the flux-flow resistance increases. The finite critical current is due to the important process of flux pinning and prevents magnetic flux motion and the generation of electric losses. In recent years, flux pinning and the introduction of suitable pinning centers have developed into an important subject of the materials science of superconductors.

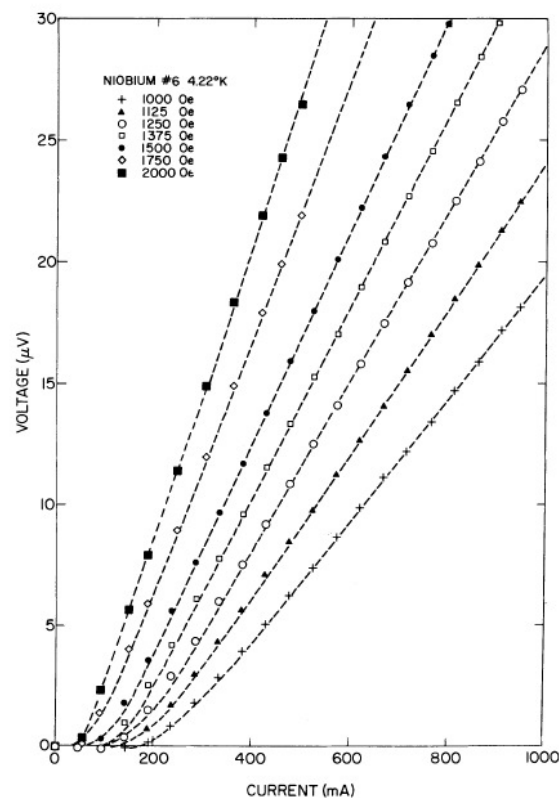


Figure 4. Flux-flow voltage plotted versus the electric current in a niobium foil of 18 μm thickness and 4 mm width for different perpendicularly oriented magnetic fields. $T = 4.22\text{ K}$; $T_c = 9.2\text{ K}$ [24].

An intuitively attractive model of flux-flow resistivity has been proposed by Bardeen and Stephen [25]. Noting that the volume of the vortex core of radius ξ resides in the normal state, and that its volume fraction is given by H/H_{c2} , at low magnetic fields the model yields the flux-flow resistivity ρ_f

$$\rho_f = \rho_n H/H_{c2} \quad (15)$$

(H_{c2} = upper critical field; ρ_n = normal-state resistivity). Further details about the many aspects of the motion of magnetic flux quanta can be found in the monograph by the author [26].

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