1

Brief History of Superconductivity

1.1 Introduction

In 1911, the phenomenon of superconductivity was discovered by Heike Kammerlingh Onnes at the University of Leiden (The Netherlands). The centenary of this discovery was celebrated with a joint conference of the European Applied Superconductivity Conference (EUCAS 2011), the International Superconductive Electronics Conference (ISEC 2011), and the International Cryogenic Materials Conference (ICMC 2011) on September 2011 in The Hague (The Netherlands) [1]. Because of this anniversary, it seems to be worthwhile to briefly describe the history of superconductivity. This overview of the most important events in the history of superconductivity allows us to consider the high-temperature superconductors (HTSs) and their properties in the broader context of superconductivity in general. The most important milestones in the field of superconductivity are listed in Table 1.1.

1.2 Milestones in the Field of Superconductivity

1.2.1 Early Discoveries

In 1908, the successful development of helium liquefaction techniques in the laboratory of H. Kammerlingh Onnes at the University of Leiden made temperatures accessible down to about 1 K [2]. One of the first aspects to be investigated was the electrical resistance of pure metals at very low temperatures. At that time, no proper theory of the electrical resistivity of metals existed. It was already known that the electrical resistance of metals decreases with decreasing temperatures. At the lowest temperatures, a nearly temperature-independent residual resistivity was observed for platinum and gold. This residual resistivity was found to decrease with increasing purity of the investigated sample. Mercury was selected for the further investigations because this low melting-point metal

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Table 1.1 *Important milestones in the history of superconductivity*

Year	Milestone	Reference
1908	Successful liquefaction of He in the laboratory of Kammerlingh Onnes at the University of Leiden	[2]
1911	Onnes discovered superconductivity in Hg	[2]
1913	Onnes received the Nobel prize for the liquefaction of He	
1933	Meissner and Ochsenfeld found perfect diamagnetism for the superconducting state	[3]
1935	London theory of the superconductor electrodynamics	[4]
1941	Discovery of superconductivity in NbN ($T_c = 15$ K)	[5]
1950	Ginzburg and Landau developed a phenomenological theory of superconctivity	[6]
1953	Discovery of the first A15 superconductor V_3Si ($T_c = 17.1$ K)	[7]
1957	Bardeen, Cooper, and Schrieffer (nobel prize 1972) developed a quantum theory of superconductivity	[8, 9]
1960	Giaever measured the energy gap by electron tunneling	[10, 11]
1961	Experimental confirmation of flux quantization	[12, 13]
1962	Josephson predicted the possibility of Cooper pair tunneling between two superconductors separated by a thin insulating oxide layer	[14]
1973	Josephson received the Nobel prize for his theoretical prediction of Cooper pair tunneling through a thin insulating layer separating two superconductors	
1974	Record T_c-value of 23.2 K in Nb_3Ge	[15–17]
1980	Bechgaard and colleagues found the first organic superconductor	[18]
1986	Bednorz and Müller discovered superconductivity at 30 K in La–Ba–Cu–O	[19]
1987	Discovery of the first superconductor with a T_c well above the boiling point of liquid nitrogen ($YBa_2Cu_3O_{7-x}$, $T_c = 93$ K)	[20]
1987	Bednorz and Müller received the Nobel prize for the discovery of high-temperature superconductivity	
1991	Superconductivity in K_3C_{60} at 18 K	[21]
1993	Record T_c-value of 135 K in $HgBa_2Ca_2Cu_3O_{8+\delta}$	[22, 23]
2001	Akimitsu *et al*. found superconductivity at 39 K in MgB_2	[24]
2003	Abrikosov and Ginzburg received the Nobel prize for their contributions to the GLAG (Ginzburg–Landau–Abrikosov–Gor'kov) theory	
2008	Hosono and colleagues discovered superconductivity at 26 K in $La(O_{1-x}F_x)FeAs$	[25]

could be purified by repeated distillation. In 1911, Kammerlingh Onnes found that, at a temperature of approximately 4.2 K, the electrical resistivity of mercury suddenly drops to a value too small to be measured. The temperature at which the superconducting state is reached is called the transition or critical temperature (T_c). The resistance versus temperature data measured by Kammerlingh Onnes are shown in Figure 1.1. The loss of

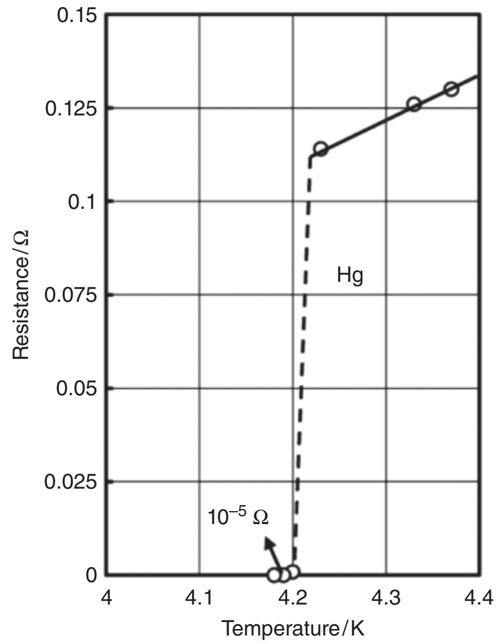

Figure 1.1 *Resistance vs. temperature plot obtained for mercury by Kammerlingh Onnes (adapted from [2])*

resistivity occurred within a temperature interval of 0.04 K. The unexpected phenomenon of superconductivity had been discovered [2]. Soon after this discovery, it was found in Leiden that tin (T_c < 3.8 K) and lead (T_c < 7.2 K) are also superconducting metals [2].

The most remarkable property of the superconducting state is field exclusion. In 1933, Meissner and Ochsenfeld observed that the magnetic field is expelled from the interior of a superconductor, which is cooled below the transition temperature in the presence of a small magnetic field [3]. This effect, nowadays known as the Meissner effect, cannot be explained by zero resistance and is in fact a second characteristic property of the superconducting state.

In 1935, the brothers Fritz London and Heinz London developed the first phenomenological theory of superconductivity [4]. The London equations (see Section 2.4) provide a theoretical description of the electrodynamics of superconductors, including the Meissner effect. In a thin surface layer, just inside the superconductor, screening currents flow without resistance, which cancel the applied magnetic field in the interior of the superconductor. The thickness of this layer, known as the London penetration depth, is a characteristic of the superconductor in question. In addition, London recognized that superconductivity is an example of a macroscopic quantum phenomenon. The behavior of a superconductor is governed by the laws of quantum mechanics like that of a single atom, but on a macroscopic scale [26, 27].

In 1941, Aschermann *et al.* found superconductivity in NbN with a transition temperature of 15 K [5]. Most superconductors discovered before 1941 were elemental metals or alloys. An interesting aspect is the relatively high critical temperature in a compound containing the nonmetallic element nitrogen.

1.2.2 Progress in the Understanding of Superconductivity

In 1950, Ginzburg and Landau developed a phenomenological theory of superconductivity [6], based on the principles of thermodynamics and empirical in nature. Abrikosov [28] solved the Ginzburg–Landau (GL) equations and found solutions explaining the penetration of magnetic flux into Type II superconductors (see Chapter 4), leading to the formation of a flux line lattice. The London theory of superconductivity is a special case of the GL theory [29].

In 1957, Bardeen, Cooper, and Schrieffer [8, 9] developed a microscopic quantum theory of superconductivity (BCS theory, see Section 3.2). The electron–phonon interaction leads to a weak attraction between two electrons. This leads to the formation of Cooper pairs consisting of two electrons of opposite spin and momentum. The total spin of a Cooper pair is therefore zero and as the consequence the paired electrons do no longer obey Pauli's exclusion principle. All the Cooper pairs condense into a single ground state.

Gor'kov demonstrated in 1959 [30] that the GL theory can be derived from the BCS theory, the microscopic theory of superconductivity. The GL theory is still a very useful tool in the field of applied superconductivity.

In 1960, Giaever used aluminum/aluminum oxide/lead sandwiches to perform electron tunneling experiments. The two metals were separated by an Al_2O_3 layer as thin as 1.5–2.0 nm. The tunneling current across the contact was measured in the temperature range of 1.6–4.2 K. Depending on the strength of the applied magnetic field, lead was in its normal or superconducting state, while aluminum (T_c = 1.18 K) was always in its normal state. The energy gap for the quasi-particles (unpaired electrons) in lead was determined from the current–voltage curves of the tunneling contact [10]. The existence of an energy gap for the quasi-particles in the superconducting state is one of the central predictions of the BCS theory.

Independent of each other, Deaver and Fairbank [12] and Doll and Näbauer [13] demonstrated experimentally in 1961 that the trapped flux in a superconducting hollow cylinder is quantized in units of $h/2e$, where h is Planck's constant and e is the charge of an electron. The fact that the flux is quantized in units of $h/2e$ instead of h/e indicates the existence of Cooper pairs in the superconductive state.

The possibility that Cooper pairs can tunnel through an insulating barrier separating two superconductors was predicted in 1962 by Josephson [14] (see Section 3.3).

1.2.3 Discovery of High-Temperature Superconductivity

The discovery of high-temperature superconductivity has to be considered in the context of the evolution of the record values of the critical temperature in different classes of superconductive materials (see Figure 1.2). In 1974, a record T_c-value of 23.2 K was reached in Nb_3Ge films [15–17]. This remained the highest known critical temperature until the discovery of superconductivity in the Ba–La–Cu–O system at a temperature as high as 30 K [19]. This discovery by Bednorz and Müller in 1986 started a race to higher and higher critical temperatures. In 1987, $YBa_2Cu_3O_{7-x}$, the first superconductor with a transition

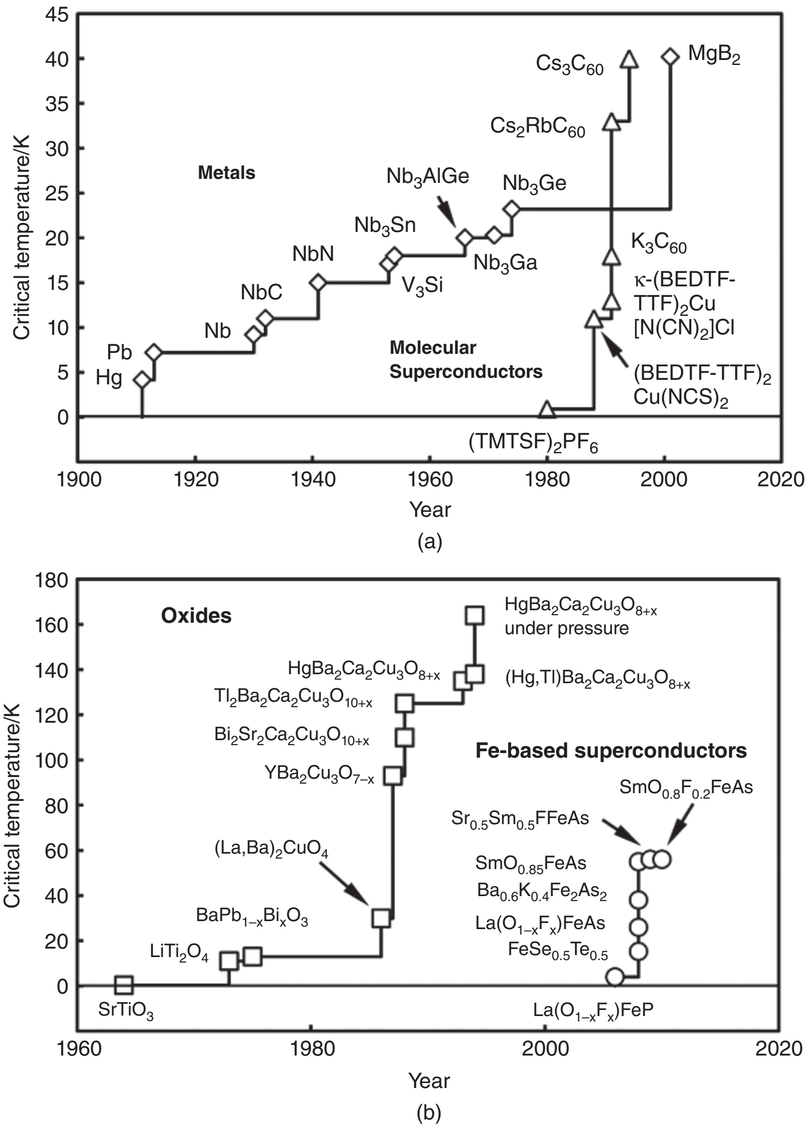

Figure 1.2 *Map of the highest known critical temperatures in different classes of superconducting materials. Panel (a): metals and molecular superconductors [2, 5, 7, 15–18, 21, 24, 29, 31–37]. Panel (b): HTSs (cuprates and iron-based superconductors) [19, 20, 22, 23, 25, 38–51]*

temperature well above the boiling point of liquid nitrogen (77 K), was discovered [20]. By 1993, the maximum critical temperature was pushed up to 135 K in $HgBa_2Ca_2Cu_3O_{8+x}$ (Hg-1223) [22, 23]. A slightly higher transition temperature of 138 K was reported for the partly thallium substituted $Hg_{0.8}Tl_{0.2}Ba_2Ca_2Cu_3O_{8+x}$ [44]. Under a large quasi-hydrostatic pressure of 31 GPa, a critical temperature as high as 164 K was measured in Hg-1223 [45].

A common feature of all HTSs is the presence of CuO_2 planes in the crystal structure; these materials are cuprates.

One of the merits of Bednorz and Müller was the search for higher transition temperatures in the oxides. It was already known that superconductivity can occur in oxides. However, the critical temperature of $SrTiO_3$, the first known oxide superconductor, was as low as 0.25 K [38]. In the seventies of the 20th century moderately high critical temperatures of 11 K [39] and 13 K [40] were found in the oxides $LiTi_2O_4$ and $BaPb_{1-x}Bi_xO_3$, respectively.

In 1980, Bechgaard and colleagues discovered the first organic superconductor $(TMTSF)_2PF_6$ (tetramethyl-tetraselenafulvalene) [18]. Other molecular superconductors are alkali-doped C_{60} molecules. In K_3C_{60}, superconductivity at 18 K [21] was found in 1991. The critical temperature of Cs_3C_{60} under a high pressure of 40 K [37] is considerably larger than that of Nb_3Ge and comparable to that of $La_{2-x}Ba_xCuO_4$, the first HTS discovered by Bednorz and Müller.

Akimitsu *et al*. found in 2001 that the simple binary compound MgB_2, a material known since the 1950s, is a superconductor with the extraordinarily high transition temperature of 39 K [24].

Hosono *et al*. discovered superconductivity in the iron pnictide $La(O_{1-x}F_x)FeAs$ at a temperature of 26 K in 2008 [25]. Very rapidly the critical temperature could be enhanced to a value as high as 55 K in the related iron pnictide $SmO_{0.85}FeAs$ [49]. Because they have transition temperatures well above 50 K, the iron pnictides are considered as the second family of HTSs. The discovery of the iron-based superconductors suggests that in addition to the cuprates and the iron pnictides their may exist other families of HTSs.

1.2.4 Importance of Higher Transition Temperatures for Applications

Most applications of superconductivity require critical current densities of at least $10^4\,\mathrm{A\,cm^{-2}}$ at the envisaged operation field and temperature [52]. Another aspect of importance is the cost of the cooling of the superconductive device. The efficiency of a refrigeration cycle is closely related to the operation temperature. Considering an ideal, reversible cooling cycle the maximum possible refrigeration efficiency is given by the Carnot efficiency

$$\eta_C = \frac{T_{\mathrm{op}}}{300\ \mathrm{K} - T_{\mathrm{op}}} \tag{1.1}$$

where T_{op} is the operating temperature. The input power (P_{in}), required by an ideal, reversible refrigerator, to remove the heat load dQ/dt is proportional to the reciprocal value of the Carnot efficiency:

$$P_{\mathrm{in}} = \frac{1}{\eta_C}\frac{dQ}{dt} \tag{1.2}$$

The reciprocal values of the Carnot efficiency as a function of operating temperature are presented in Figure 1.3. For example, the input power needed to remove a heat load of 1 W at operation temperatures of 4.2, 20, and 77 K is 70.4, 14, and 2.9 W, respectively. However, the efficiency of a real cooling cycle is considerably lower than the Carnot efficiency. In general, large refrigerators are more efficient than smaller ones [53, 54]. For large refrigerators with a heat removal capacity well above 10 kW, the efficiency can reach

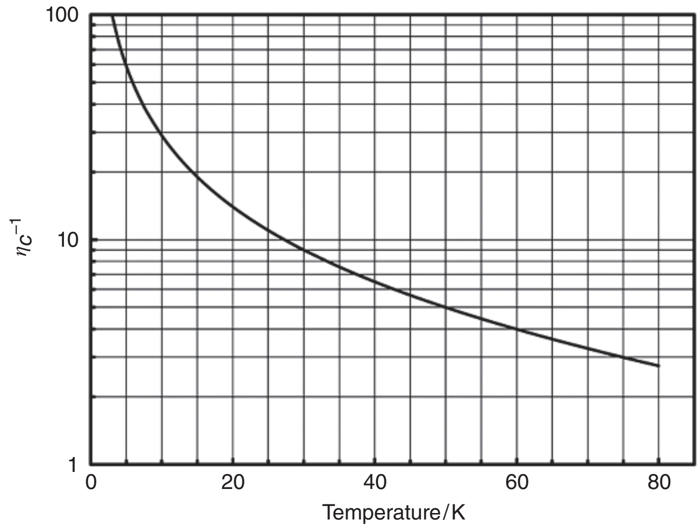

Figure 1.3 *The reciprocal value of the Carnot efficiency as a function of the operation temperature indicates that for the removal of a heat load of 1 W at an operation temperature of 4.2 K a power of ≈70 W is required, while this power is only 2.9 W at an operation temperature of 77 K*

≈30% of the Carnot efficiency. On the other hand, the efficiency of refrigerators with a heat removal capacity of 1 W is only a few per cent of the Carnot efficiency [53, 54].

The considerations of refrigeration efficiency clearly indicate that the ability to operate superconductive devices at 77 K instead of 4.2 K provides a drastic reduction in the cost of the cooling. The discovery of the cuprate HTSs therefore renewed interest in power applications of superconductivity, e.g. in power transmission cables, transformers, fault current limiters, and generators. For example, the technical feasibility of superconducting transmission cables, made of low-temperature superconductors requiring operation temperatures close to 4 K, was already demonstrated in the 1970s and the 1980s [55–59]. However, these cables were not economically competitive with conventional technology because of the high cost of cooling. As soon as industrially manufactured HTS tapes became available, several demonstrators of power transmission cables have been made of them [60–65]. Operation of HTS cables has been demonstrated with liquid nitrogen cooling; the cooling cost is considerably reduced as compared to operation at ≈4 K. In addition, high critical current density and sufficiently low superconductor cost are required if future HTS power transmission cables are to become economically competitive with conventional technology. The critical current density of HTS tapes is still increasing, and the cost is expected to be further reduced.

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