Processing of High-Temperature Superconductors at High Strain Rates

A. G. Mamalis
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FOREWORD

The discovery of superconductivity at high temperature in 1986 has set fire to the research on cuprates. Thousands of articles have been published these last 10 years on high-T_c superconducting cuprates. In a similar way, many books have been written on the physics of these compounds. In contrast, only few lines have been written on the applied side of these materials.

At this point of the investigations, it is time to take stock about the processing of these materials in view of their applications. There is no doubt that several of these numerous superconductors, such as the famous "123" YBCO or the Bi "2212" or "2223" cuprates will be used in devices in the next years.

Specialists of the processing of these materials, A. G. Mamalis and his coworkers and A. Szalay, have performed a very efficient work in this field. Through this book, the reader benefits from the high experience of the authors in the field of the processing of bulk superconductors at high strain rates. Not only do the authors describe conventional methods of fabrication of bulk superconductors and thin films, but they also consider the impact loading of solid and porous media from the theoretical and experimental viewpoint.

Besides these methods, which are of capital importance for the elaboration of optimized high-T_c superconductors, this book relates the main methods of characterization of these materials and gives an excellent overview of their field of applications.

This book is well structured and conceived in such a way that it can be used by beginners in the elaboration of materials and by specialists who need details and references in this area. There is no doubt that this work will be used as a reference by students and scientists who are interested in the processing of HTS.

BERNARD RAVEAU
Caen

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PREFACE

The introduction that follows sufficiently describes our aims in presenting this book on the net-shape processing of high-temperature advanced superconducting ceramics, which are subjected to elevated, and in particular, high-strain rates, relevant to design and manufacture of electrical/electronic, transportation and bioengineering equipment. Essentially, it comprises the results of our extensive theoretical and experimental work on the topic and the material of a series of lectures on net-shape manufacturing of advanced materials given to undergraduates and graduates in Mechanical, Material Science and Electrical/Electronic Engineering. The book is intended to illustrate and indicate the engineering design outlets and applications of the analytical and experimental work, mainly on macro- and micromechanics, static and dynamic powder consolidation and processing, stress-wave propagation into solid and porous materials, as well as applications of bulk and thin-film superconductors and the new notions and considerations in applied engineering applications. We hope that the contents of our work will be of value to students, teachers and many kinds of professional engineers.

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CHAPTER 1

Introduction

1.1 BRIEF HISTORY OF SUPERCONDUCTIVITY

The historical evolution of the superconducting materials is presented diagramatically in Figure 1.1.

On July 10, 1908, Heike Kamerlingh Onnes at the University of Leiden in the Netherlands successfully operated the first liquefier of helium, which exhibited a boiling point of approximately 4.2 K at atmospheric pressure. Since then, Onnes and other researchers tried to determine the electrical properties of matter at very low temperatures. In some metallic materials, such as copper (Cu), platinum (Pt), gold (Au), showing a continuous decrease of electrical resistance with decreasing temperature, a finite value of resistance at liquid helium temperature was finally obtained. Only mercury (Hg) revealed a rapid decrease in the resistance at 4.25 K, which was vanished at about 4.2 K (boiling point of He), see Figure 1.2. The mercury-work was first reported by Onnes in the 1st Solvay Congress in Brussels, during October 1891 and received the Nobel Prize in Physics in 1913 [1].

Onnes followed this work showing that indium (In), tin (Sn) and lead (Pb) became superconducting at 3.4, 3.7 and 7.2 K, respectively. These materials, that present zero resistance at a certain temperature above absolute zero, are named *superconductors* and the related phenomenon *superconductivity*. A generalized comparison of resistivity for normal metal and superconductors as a function of temperature is shown in Figure 1.3.

The highest *critical temperature* of pure superconductors was shown in the case of niobium (Nb), possessing superconductivity below 9.5 K. Note that the research in superconductivity has led to the formation of superconducting alloys and compounds based on Nb. Characteristic Nb-alloy systems are the Nb-Ti and the Nb-Zr, but the highest critical temperatures, i.e., 18 and 23 K,

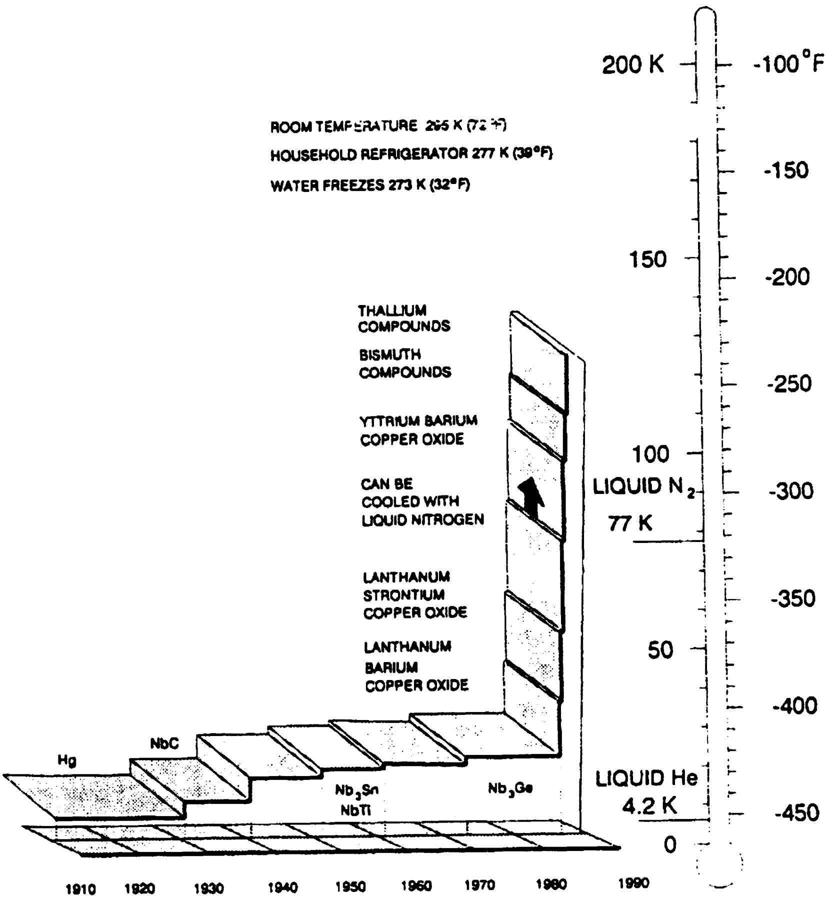

FIGURE 1.1. Evolution of superconductors.

are indicated by the intermetallic compounds Nb_3Sn and Nb_3Ge, respectively, which possess a characteristic β-W type lattice (A_3B).

In 1986 a *high*-T_c or *high-temperature superconductivity* was invented by Bednorz and Müller at IBM in Zürich [2]. The new superconductor was a mixed oxide of lanthanum, barium and copper (La-Ba-Cu-O) of perovskitic crystal structure; the critical temperature of this ceramic material was higher than 30 K. It has to be noted, that, after several false starts and disappointments, the turning point for high-T_c superconductivity come in late 1985 when Bednorz was alerted to a publication by the French team of Michel and Raveau in Caen [3], who reported metallic-like conductivity in La-Ba-Cu-O in the range from −150°C to 300°C. The French team had not been investigating superconduc-

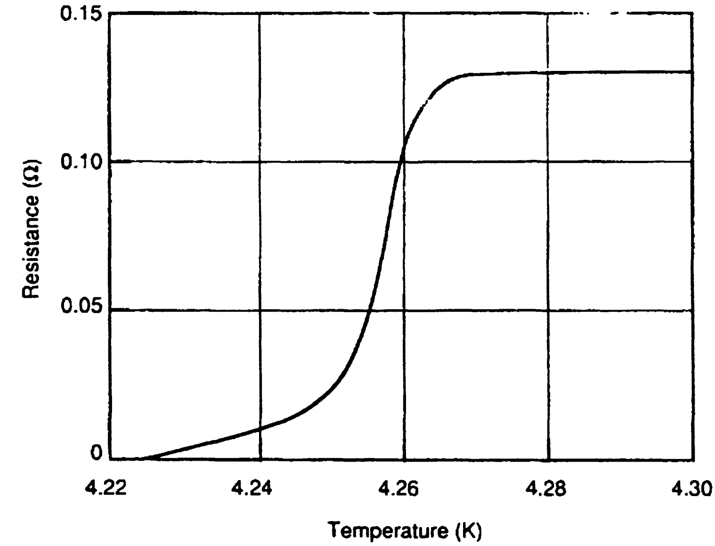

FIGURE 1.2. Resistivity-temperature diagram for mercury (Hg).

tivity at that time but had shown several years before, see Reference [4], the possibility to stabilize the mixed valent state copper at normal pressure, in 2D cuprate in connection with the Jahn Teller effect of copper. Bednorz and Müller received the Nobel Prize in Physics, for their invention, in 1987.

The main high-T_c superconductors, from a practical application point of view, were invented afterward. Chu and Wu and their group at Texas Center of High-T_c Superconductivity (TCSUH) in Houston, Texas, discovered the superconducting ceramic $YBa_2Cu_3O_{7-x}$ (1-2-3 material) with critical tempera-

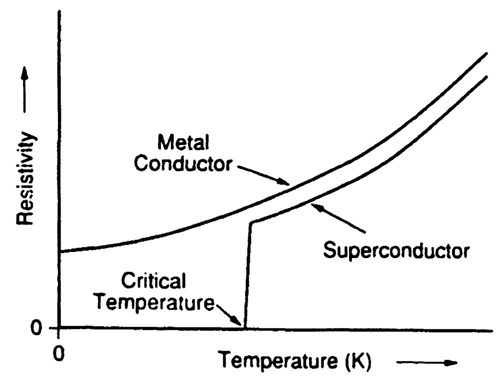

FIGURE 1.3. Resistivity-temperature diagram for a superconductor and a metal-conductor.

ture up to 93 K above liquid nitrogen temperature [5]. This fact makes the use of superconductivity very profitable because liquid nitrogen as the cooling medium is much cheaper and more easily handled than helium.

Maeda's group at Tsukuba Labs in Japan synthesized the Bi-Sr-Ca-Cu-O oxide, becoming superconductor at 110 K [6]. More ductile and stable than the orthorombic 1-2-3 compound, it exhibits several superconductive or normal phases that are not trivial to be separated.

Another characteristic Cu-O superconductor is the Tl-Ba-Ca-Cu-O, invented by Sheng and Hermann and their group from the University of Arkansas [7]. Similarly to Bi-Sr-Ca-Cu-O oxide, the Tl-based material is a multiphase system; the highest critical temperature possessed is approaching 125 K.

1.2 THE STRUCTURE OF THE MONOGRAPH

The discovery of high-temperature superconductivity by Bednorz and Müller in the La-Ba-Cu-O system resulted in very extensive research work about the discovery and synthesis of other high-temperature superconductors, such as Y-Ba-Cu-O and Bi-Sr-Ca-Cu-O. These new superconducting materials, possessing superconductivity above liquid nitrogen boiling point, are used in many engineering applications, from electronic sensors to rotating electrical generators and from nanometer-scale thin films to kilometer-long wires and coils. Therefore, design and net-shape manufacturing of superconducting components, starting from the initial synthesized powders, is nowadays of utmost industrial importance.

This book is primarily focused on the bulk-fabrication techniques of high-temperature ceramic superconducting components, especially on the combination of dynamic powder-consolidation and subsequent deformation processing. The properties of these ceramics, which are difficult-to-be-formed materials by applying conventional techniques, are combined for the net-shape manufacturing of such components for the construction of HTS devices; this is the core of the superconductor research work that has taken place through the collaboration of the Manufacturing Technology Division of the National Technical University of Athens, Greece, and the Metalltech Ltd. of Budapest, Hungary, according to the fabrication sequence, from the raw material to industrial applications as shown in Figure 1.4. However, very important topics, such as superconducting structures, chemical synthesis, film fabrication and characterization techniques, are also reviewed throughout this book, to provide a complete comprehensive view of superconductors engineering.

In Chapter 2 the fundamental aspects of superconductivity are outlined, whereas Chapter 3 deals with the most important chemical synthesis methods of most of the high-temperature superconducting compounds.

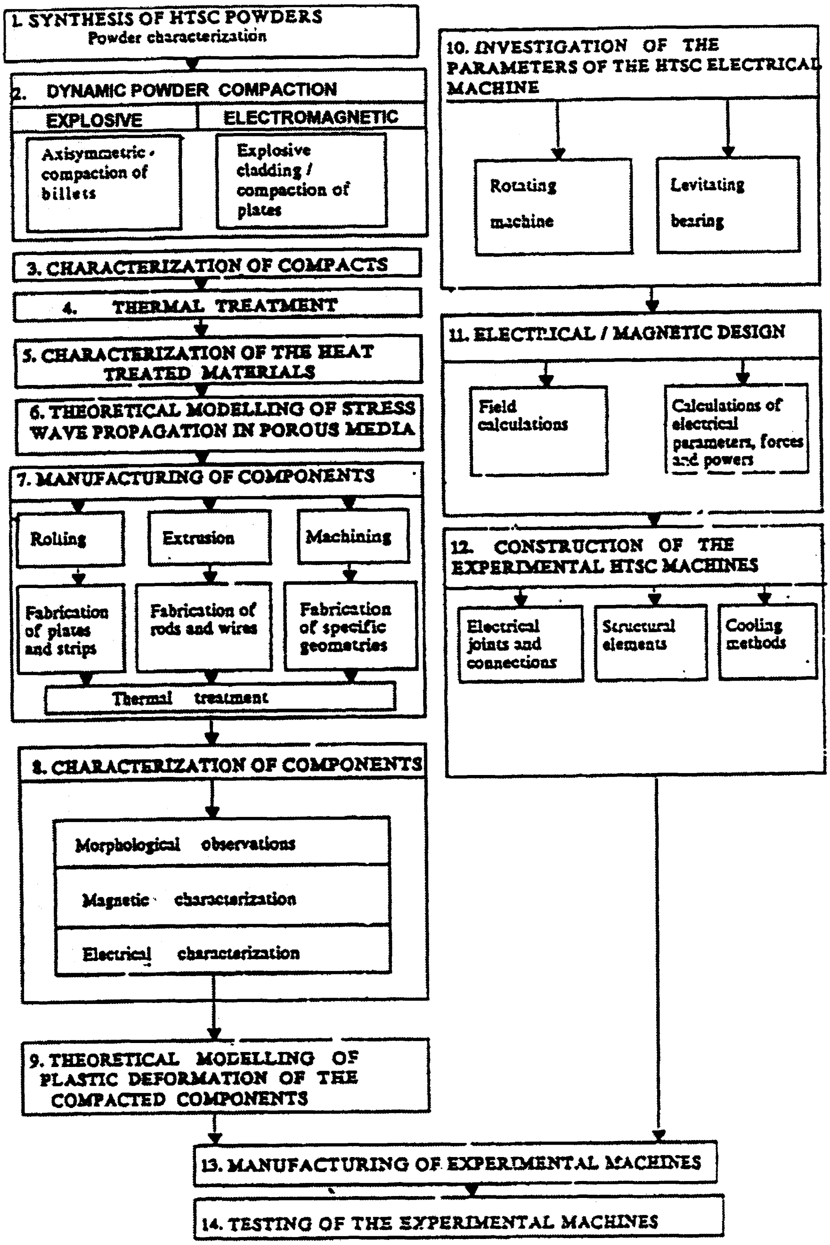

FIGURE 1.4. Process flow diagram from the powder synthesis to the final stages of fabrication and applications of high-T_c superconductors.

In Chapter 4 the dynamic powder compaction techniques and extensive experimental pertaining to the effect of shock waves on microstructure, properties and defect formation of impacted superconductive components of the YBCO and BSCCO compounds are discussed in detail. In addition, the theoretical modeling of stress wave propagation through the porous media, resulting in the consolidation of the powders, is attempted, considering also the Hugoniot curves and equation-of-state (EOS) of the materials.

The deformation processing of HTS compacts in various geometries in plastic deformation-microstructure-processing-properties relationships, is presented in detail in Chapter 5. The defects and the deformation mechanisms for the forming techniques used are also considered. Furthermore, other bulk-processing techniques, such as melt-texturing, composite reaction texturing and mechanical texturing, are briefly discussed.

In Chapter 6 the processes used for the fabrication of HTS films are discussed in detail.

Chapter 7 deals with the physicochemical techniques used for the characterization of HTS powders and components, whereas in Chapter 8 the most important HTS applications are presented. Finally, in Chapter 9 some new classes of superconducting materials and their applications are suggested, whereas future perspectives of high-T_c superconductivity and its techniques toward higher critical temperatures are outlined.

This book is addressed to scientists, researchers and engineers involved in the multidisciplinary field of high-temperature superconductivity approaching the subject more from an engineering point of view. It is our hope that the contents of this monograph will be of value to students, teachers and many kinds of professional engineers and, thus, enhance interest in superconducting materials and the related technology.

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CHAPTER 2

Fundamentals of Superconductors

2.1 NOTATION

B = magnetic induction
c = heat capacity
e = charge of electron
F_L = Laplace force
h = Planck's constant
H = magnetic field intensity
H_c = critical magnetic field intensity
H_{c1} = lower critical field
H_{c2} = upper critical field
H_o = critical field intensity at zero Kelvin temperature
I = supercurrent flow
J = current density
J_c = critical current density
k_B = Boltzmann's constant
M = magnetization
n = integer
$N(E_F)$ = energy state density below Fermi level
T = temperature
T_c = critical temperature
U = voltage
V = correlation factor
α = fitting constant
Δ = semiwidth of energy gap
κ = parameter ($= \lambda/\xi$)
λ = penetration depth (London) of magnetic field

M = isotopic mass
ξ = coherence length
ρ_o = resistive transition
Φ = magnetic flux
Φ_o = flux quantum
$\psi(x)$ = wave function
ω = frequency
ω_D = Debye's frequency

2.2 BCS THEORY

The explanation of the superconductivity theory, mainly in metallic systems, originated from the American researchers Bardeen, Cooper and Schrieffer in 1957 [1], who won the Nobel Prize in 1972. The Schrieffer corresponding theory is abbreviated as *BCS theory*.

The basic characteristic effects of the superconductive state is the persistence of the electric currents, the diamagnetic effect and the specific heat discontinuity. From electrical measurements, the elementary charge of a material in the superconducting state was found equal to $2e$, where e is the charge of the electron (1.6×10^{-19} Cb). This result yields to the conclusion that, the current flowing in a superconducting material is composed of electron pairs, which are called *Cooper pairs,* and this pairing occurs despite the repulsive interaction between the two electrons. This attraction may be simply explained by the "mattress effect," i.e., when a heavy ball rolls fast on a soft mattress, the mattress will bend downward or sink where the ball is, whereas in the case of the ball rolling very fast, the time for back relaxation is not enough for the springs to return to the starting position immediately after the passage of the ball. If, now, a second ball is traveling on the same mattress and comes closer to the first ball, the mattress will sink, bringing the two balls together. Substituting electrons for balls and the solid composed of sluggish ions for the mattress, an attractive interaction between electrons is, thus, obtained.

The electron-pairing mechanism is based on the so-called *electron-phonon interaction.* An electron moving in a solid lattice can be affected by another electron via acoustic quanta (i.e., phonons), which are originated from the vibrations of the lattice atoms. The electron-electron interactions are realized through phonon exchange, leading to electron condensation, being responsible for the neutralization of Coulomb repulsion. The electronic pair consists of two electrons possessing opposite momentum and spin, and the spacing between them is known as coherence length ξ. The coherence length is of the order of $100 \div 1000$ nm in pure superconducting materials. At temperatures higher than the critical temperature, the thermal energy obtained results in breaking the Cooper pairs and stopping the superconduction. The BCS theory may be, finally, summarized as

$$T_c = \frac{1.14h\omega_D \exp[-1/N(E_F)V]}{2\pi k_B} \tag{2.1}$$

where, following the Notation section, h is the Planck's constant, ω_D the Debye's frequency, $N(E_F)$ the energy state density below Fermi level, V a correlation factor and k_B the Boltzmann's constant.

2.3 BASIC NOTIONS

The fundamental phenomena related to the superconductivity may be listed as [2]:

- zero resistivity
- diamagnetic or Meissner effect
- energy gap
- specific heat discontinuity
- magnetic flux quantization
- isotopic effect
- tunneling effect

2.3.1 Zero Resistivity

The curve representing the variation of resistivity as a function of temperature is shown in Figure 2.1. By convention, the *onset temperature,* $T_{c,\mathrm{onset}}$, is defined as the temperature value at which the slope changes entering from the normal to the transition behavior and the *critical temperature,* T_c, as the temperature corresponding to *zero resistivity point.*

Measurements of resistivity or magnetic susceptibility may lead to the determination of T_c. For narrow transitions, the transition temperature corresponds to the zero resistivity. However, because the resistivity slope near T_c may widely vary between ac and dc measurements, a corresponding difference in the measured value of T_c may be shown. Long resistive tails indicating the presence of secondary phases, superconducting or not, make the determination of T_c difficult. By convention, T_c is found with respect to the half of the resistive transition corresponding to $\rho_o/2$, see Figure 2.1. T_{co} is the onset and T_{cf} is the offset of the transition (end or zero resistivity point).

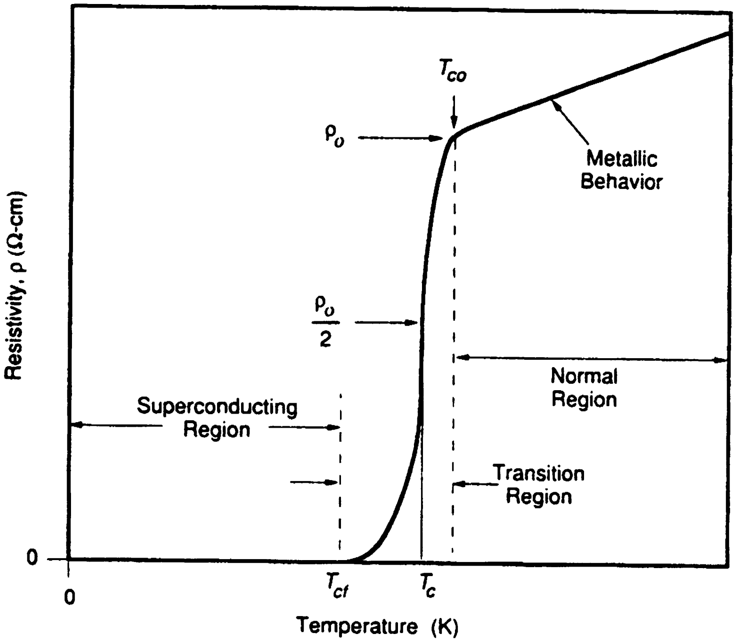

FIGURE 2.1. Characteristic resistivity-temperature curve for a superconductor.

2.3.2 Diamagnetic or Meissner Effect

Apart from the outstanding properties, the superconductive materials possess exceptional magnetic properties. When a magnetic field of intensity H is applied to a superconductive material ($T < T_c$), exclusion of magnetic field lines from the interior of the material has been observed. This is valid when the field intensity H is lower than a critical intensity H_c. The ideal superconducting material behavior is then called *pure diamagnetic behavior,* while the magnetic susceptibility is equal to -1. This phenomenon is known as *Meissner-Ochsenfeld* or *diamagnetic effect.*

For temperatures higher than T_c, the diamagnetic effect is eliminated, and the magnetic field penetrates in the interior of the material.

A schematic representation of the diamagnetic effect is illustrated in Figure 2.2(a), while the variation of H_c as a function of temperature is shown in Figure 2.2(b), given by the equation

$$H_c = H_o\left[1 - \left(\frac{T}{T_c}\right)^2\right] \tag{2.2}$$

where H_o is the critical intensity at zero Kelvin temperature.

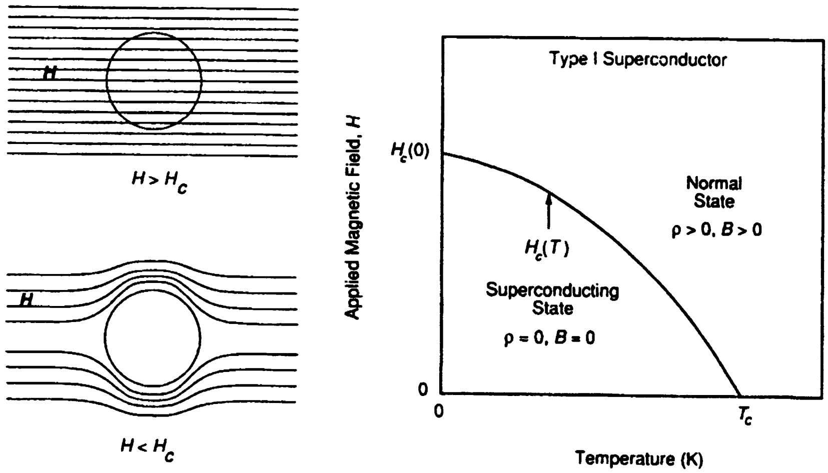

FIGURE 2.2. (a) Schematic representation of the Meissner-Ochsenfeld effect; (b) evolution of the critical magnetic field as a function of temperature for a Type I superconductor.

The magnetization, M, defined by the relation (see also Notation section) $B = \mu_o(M + H)$, for a certain temperature $T < T_c$ varies with magnetic field according to the curve of Figure 2.3(a).

LONDON EQUATION—PENETRATION DEPTH

The electrodynamic behavior of a superconducting material is described by London's equations. By combining with Maxwell's equation a fundamental relation is obtained

$$\nabla^2 H = \frac{H}{\lambda^2} \tag{2.3}$$

where λ is the characteristic penetration depth (London) of the magnetic field in the interior of the material.

Under the semi-infinite solid assumption, the solution of the differential equation yields the following spatial distribution of the magnetic field

$$H(x) = H(0) \cdot \exp(-x/\lambda) \tag{2.4}$$

The value of penetration depth is of the order of magnitude of 50 nm for pure metals superconductors.

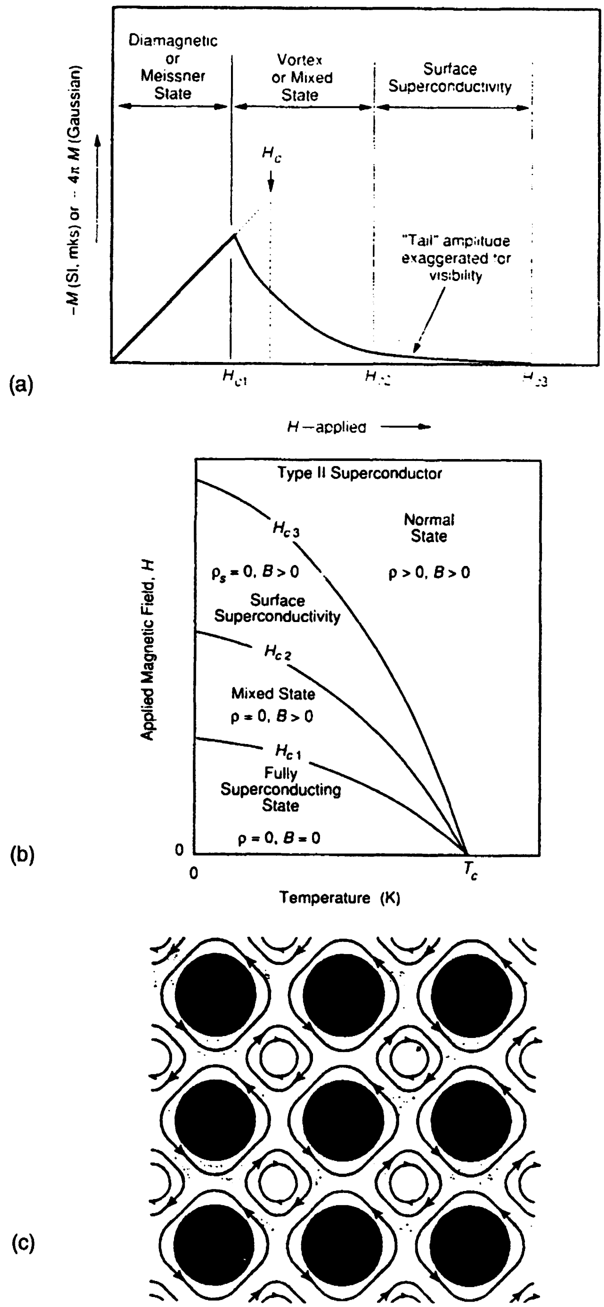

FIGURE 2.3. (a) Evolution of magnetization as a function of the applied magnetic field for Type I and II superconductors; (b) critical magnetic field vs. temperature curve for a Type II superconductor; (c) schematic diagram of the intermediate state for a Type II superconductor.

GINSBURG-LANDAU THEORY

The phenomenology of the superconductivity has been thoroughly studied by Ginsburg-Landau in 1957. Assuming that the characteristic wave function $\psi(x)$ is describing the electronic fluid state, then the expression $[\psi(x)]^2$ is a measure of the density of the superconductive electrons. The wave function ψ in the case of the Ginsburg-Landau superconductivity theory is called *order parameter.*

Ginsburg and Landau have also introduced the κ-parameter, which represents the ratio of the characteristic lengths λ and ξ, i.e.,

$$\kappa = \frac{\lambda}{\xi} \tag{2.5}$$

TYPES OF SUPERCONDUCTORS

Type I Superconductors ($\kappa < 1/\sqrt{2}$)

This type of superconductor possesses all the diamagnetic characteristics that have been already mentioned. All pure metals, except niobium (Nb), belong in this category.

Type II Superconductors ($\kappa > 1/\sqrt{2}$)

In this class of superconductors two different values of the critical magnetic field, a lower critical field H_{c1} and an upper critical field H_{c2}, are present. When the applied field $H < H_{c1}$, then the diamagnetic effect (exclusion of magnetic field lines from the interior of the material) is observed. In the case of $H_{c1} < H < H_{c2}$, progressive penetration of the magnetic field begins forming an intermediate or mixed or vortex state. Finally, when H exceeds H_{c2}, the magnetic field totally penetrates into the interior of the material, destroying, therefore, the superconducting state in the interior of the material that remains in a surface superconducting state for $H_{c2} < H_c < H_{c3}$, see Figure 2.3(b). Type II superconductors are all the alloys (including Nb), intermetallic components and ceramic oxides.

INTERMEDIATE STATE

The *intermediate state,* denoted also as *mixed state, vortex state* or *Schubnicov phase,* appears in Type II superconductors when the applied magnetic field H lies between the values of lower and upper critical field, H_{c1} and H_{c2}, respectively. In this case, the progressive penetration of magnetic field lines into the interior of the material forms normal, nonsuperconducting regions, adjacent to superconducting ones. The field penetration creates, therefore, the so-called vortices or fluxoids in an ordered array, which is known as *Abrikosov lattice,* see Figure 2.3(c). Every fluxoid is a closed loop characterized by a supercurrent flow I. The increase of the external field gives rise to the magnetic induction, B, in the vortex interior causing Laplace forces ($F_L \propto B \cdot I$) tending to move the vortices (characterized as flux flow or flux creep). The spreading of the vortices results in the decrease or even the total elimination of the superconducting phase.

FLUX PINNING

Considering that the flow of the electric current causes magnetic field (Ampere's law), every superconductor of Type I may hold electric current, having a density lower than a critical one, J_c. Above this *critical current density* J_c the induced field exceeds the critical field value, H_c resulting in the destruction of the superconducting state.

For the Type II superconductors the critical density, J_c corresponds to the current value at which the vortex flow starts to occur. The degree of difficulty of the fluxoid motion defines the ability of the material to stay at the diamagnetic (superconductive) state. This characteristic material property is known as *flux pinning*. Increased flux pinning results in high values of J_c. Therefore, vortex stability and J_c can increase by the introduction of impurities or defects that act as flux trappers, stabilizing, therefore, the vortices. It is important that this array of fluxoids remain stable, because their motion leads to energy dissipation by normal currents induced in the vortex cores.

Metallurgical defects, including dislocations, point defects, precipitates, secondary phases and grain boundaries, resulted from processing techniques or from radiation damage, for example neutron irradation, may serve to pin the vortices found in Type II materials. Chemical doping results also in enhancement of flux pinning in novel oxide superconductors [3]. Effective pinning is achieved when the pinning defects and their intermediate spacings are of the order $(1 \div 5)\xi$, where ξ is the coherence length.

It is noteworthy that, although T_c and H_c may be generally considered as intrinsic material properties, J_c is a function of microstructure and can be varied several orders of magnitude, depending on the material processing technique.

2.3.3 Energy Gap

During the normal to superconducting transition, the electronic structure is reconstructed to allow the developed electron pairs to construct the superconducting current. This new electronic structure results in an anomaly in the continuity of the permitted energy levels, creating, therefore, near the Fermi surface, a forbidden *energy gap* of width equal to 2Δ. When the energy gap is exceeded, the breaking of electron pairs takes place, leading to the destruction of the superconductivity. Taking into account the BCS theory, the width of the energy gap 2Δ increases with increasing critical temperature according to

$$2\Delta = 3.528k_BT_c \tag{2.6}$$

where k_B is the Bolzmann's constant.

2.3.4 Specific Heat Discontinuity

An abrupt change of heat capacity c is observed during the normal to superconductive transition, which may be explained by the presence of the energy gap. The change of the specific heat as a function of temperature is presented in Figure 2.4.

2.3.5 Magnetic Flux Quantization

The magnetic flux, passing through a superconductive ring, is an integer multiple of the flux quantum Φ_o, passing through a single fluxoid, i.e.,

$$\Phi = n\Phi_o = n\frac{h}{2e} \tag{2.7}$$

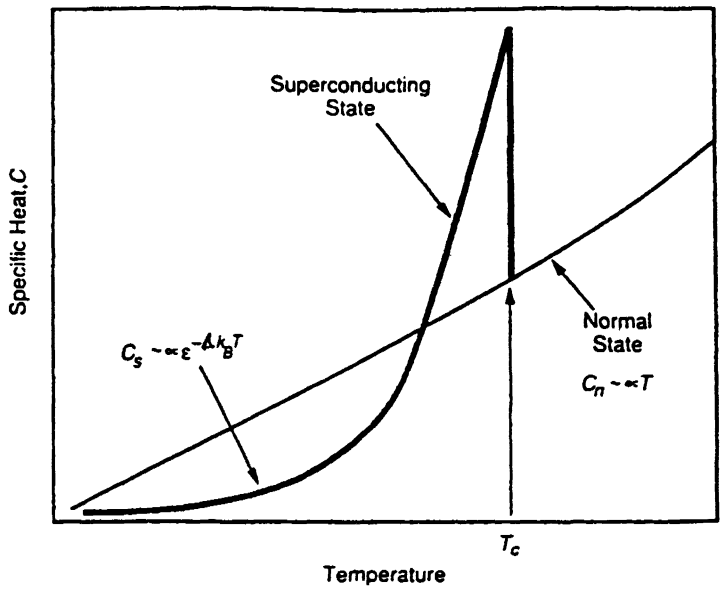

FIGURE 2.4. Evolution of specific heat as a function of temperature (from Reference [2]).