TECHNOLOGY OF PHYSICAL EXPERIMENTS

R. König
Physikalisches Institut, Universität Bayreuth, Germany

Keywords: Cryogenics, Ultralow temperatures, Superconductivity, Superfluidity, Magnetic fields, Magnetometry, Permanent magnets, Mass spectroscopy, Vacuum technology, High pressure technology

Content

Summary

Scientific research at the extreme limits in temperature, pressure and magnetic fields provides important information on the fundamental physical properties of matter. All these parameters have something in common, that they are nowadays accessible in wide ranges extending over several orders of magnitude. This chapter describes the historical development of basic principles and techniques in each of these fields, achieved mainly during the 20th century with certain rather astonishing similarities in their chronology, and it comprises the progress made in each of the subjects. It deals with the success in achieving refrigeration of matter to ultralow temperatures far into the sub-millikelvin temperature range, and introduces two of the most stunning low temperatures properties

of matter: superconductivity and superfluidity. The discussion on the generation of magnetic fields focuses on high field technology and summarizes the performance of superconducting magnets, Bitter magnets and hybrid magnets. The treatment is complemented by a compilation of recent developments in research on materials suitable for their application as permanent magnets. The introduction to the methods of magnetic field measurements concentrates on devices developed for extremely low magnetic fields with emphasis on the by far most sensitive technology known today, based on the properties of a Superconducting QUantum Interference Device (SQUID). A survey of vacuum technology, mass spectroscopy and the generation and measurement of high pressures concludes the chapter.

1. Matter and Techniques at Low Temperatures

1.1. Introduction

The refrigeration of matter into the milli- or even micro-kelvin temperature range opens up the fascinating and unique possibility to study physical properties of matter in an experimental environment where most of the unavoidable disturbances present at higher temperatures are almost completely frozen out. At the end of the first quarter of the twentieth century, when scientists became aware of the insurmountable limitations of the sole cooling technique at that time, the gas liquefaction and evaporation of the vapor above the liquid, there was a strong demand for the possibility to investigate fundamental questions of solid state physics at still lower temperatures. This demand led, step by step, to the development of a variety of cooling techniques during the remaining part of the twentieth century which extended the accessible temperature range for experimental research by more than five orders of magnitude: from the minimum temperature of about 0.7 K achieved in 1926 by means of vapor pressure reduction above liquid helium, to the present lowest (electronic) temperature of 1.5 μK obtained by nuclear demagnetization of platinum in 1997 (see Section 1.2).

Some of these newly developed techniques, like, e.g., the Pomeranchuk cooling and the adiabatic demagnetization of paramagnetic salts, played an important role only during a rather restricted period of time, until they have been replaced by superior and much more advanced techniques, like, e.g., the ^{3}He-^{4}He dilution refrigeration. Today, the temperature range down to about 4 mK is "commercially available", and refrigeration into the range between about 4 mK and 1 K is strongly dominated by the ^{3}He-^{4}He dilution refrigeration. The combination of dilution refrigeration with a magnetic cooling technique, the so-called adiabatic demagnetization of nuclear magnetic moments, provides access to temperatures of a few millionth of a degree Kelvin close to absolute zero. This technology for getting access to the microkelvin temperature range is nowadays fully developed, too. However, in contrast to ^{3}He-^{4}He dilution cryostats available on the market in numerous different types, nuclear demagnetization stages are still unique, home-made constituents of a cryostat of an incomparably greater complexity. As a consequence of the development in cryogenics, to date, scientists are in the position to perform experimental studies of liquid and solid matter at temperatures several orders of magnitude below the minimum temperature existing in nature which is the background temperature of the universe of T = 2.7 K (see Section 1.3).

The thrust towards lower and lower temperatures gives us the possibility to study matter in a completely new experimental environment and under conditions which have been unknown so far. This is certainly one of the major attractions of low temperature physics, and, of course, this applies to every single progress made in the development of cooling techniques in the course of the 19th century. However, it still might have been quite a different situation at the beginning of the twentieth century, when the experimental access to temperatures of only a few Kelvin revealed results which had at that time an extraordinary impact on the development of fundamental concepts of physics with far-reaching consequences until today. The discovery of superconductivity in 1911 by Heike Kamerlingh-Onnes and the discovery of superfluidity in liquid helium by Piotr Kapitza in 1938 are just two of the most important milestones to mention (see Section 1.4). The importance of these - sometimes fully unexpected or, at first, even disregarded - observations of properties of matter, is not just given by their uniqueness: More than that, the development of low temperature physics at the beginning of the twentieth century led to significant contributions and confirming evidences to predictions of the fast developing fields of quantum physics, statistical physics and thermodynamics, and to the corresponding revolutionary departure from classical thinking.

The discussion in the following subsections is restricted to "classical" low temperature physics and techniques. Recent developments, like, e.g., the outstanding achievements in the field of the Bose-Einstein condensation of atomic gases, or, on a more industrial level, the progress in the development of cryocoolers, will not be subject of this chapter.

1.2. Historical Remarks on the Development of Low Temperature Physics and Techniques

The economical and social interest in developing new cooling techniques in the second half of the nineteenth century was determined by the fundamental need to preserve food on the long-distance sea voyages to remote countries and continents. For this purpose, engineers developed cooling machines the cooling process of which was based on the expansion of a previously compressed gas against a movable piston. These cooling devices were able to provide temperatures several decades below zero degree Celsius at that time. From the scientific point of view, the main research concerned with the study of the properties of gases was focused on the observation that many gases could have been liquefied under great pressures; with some gases, however, liquefaction seemed to be impossible. The idea of so-called "permanent" gases arose, and the general question whether all gases finally become liquids when they are just cooled to low enough temperatures and/or brought to large enough pressures was one motivation to take on further efforts to approach lower and lower temperatures.

In 1877, Louis Cataillet and Raoult Pictet independently succeeded in liquefying tiny amounts of oxygen, and shortly afterwards, the liquefaction of nitrogen was also announced by Cataillet. However, in these early experiments it was not possible to obtain a reasonable quantity of the liquid necessary for further investigations of its properties. It took about six more years, until, in 1883, liquid oxygen was obtained, in an apparatus under stable conditions, by Karol Olszewski and Zygmunt Wroblewski in

Cracow. The successful liquefaction of oxygen and nitrogen was followed by the liquefaction of hydrogen by James Dewar in London in 1898.

At the beginning of the 20th century, helium remained the final gas to be liquefied. At the time when Dewar succeeded to liquefy hydrogen, the existence of helium has only been known for less than thirty years: it was discovered as part of the optical spectrum of the sun during the total eclipse in 1869. On earth, helium was not found until 1895 when it was discovered in mineral springs and gas wells (in both cases as product of radioactive alpha decay). It is also part of the earth's atmosphere, but only at a minute concentration of less than 0.1 ppm. There was a strong competition as to the liquefaction of helium between James Dewar in London and the Dutch scientist Heike Kamerlingh-Onnes from Leiden, who, finally, in 1908, was the first to liquefy the noble gas: this year of the first liquefaction of helium is nowadays often referred to as the beginning of the era of low temperature physics. The boiling point of helium (to be more precise: of ^{4}He) was found to be at a temperature of 4.21 K under normal conditions, and despite its low boiling point and its small heat of evaporation, liquid helium soon turned out to become the most important material indispensable for all further attempts to approach absolute zero as closely as possible. In today's low temperature cryostats for the studies in the subkelvin temperature range, liquid ^{4}He constitutes the most important pre-cooling stage. In addition to its inestimable value for refrigeration of matter to lower and lower temperatures, both stable helium isotopes known today - ^{3}He and ^{4}He - turned out to exhibit an outstanding variety of exotic and fundamental properties on their own, in their liquid as well as in their solid phases. The physical importance of the properties of both helium isotopes which have been extensively studied during the 19th century is reflected by the fact that they serve as excellent model systems of, for example, theoretical considerations of phase transitions (superfluid transitions) or the Landau Fermi liquid theory.

Soon after the successful liquefaction of helium, a straightforward but effective technique was introduced for a further lowering of the temperature of the liquid: the reduction of its vapor pressure by pumping on it, resulted in a minimum temperature of around 1 K. Further reduction of the vapor pressure was only possible by using much stronger pumps, and, finally, in 1926, a minimum temperature of 0.7 K was obtained at the Leiden laboratory. Surprisingly, no indication for solidification of helium was observed, and the question remained whether helium might even stay liquid down to absolute zero.

Within about half a century, from 1877 until 1926, scientists have approached absolute zero by almost two orders of magnitude in temperature. Figure 1 shows that with this final attempt it became obvious that in order to achieve a further reduction in temperature, a new and different cooling technique had to be developed. The technique of lowering the temperature by means of gas liquefaction and subsequent reduction of its vapor pressure had been pushed to its limits, and the lowest possible temperature with helium as coolant has been reached. At that time, the existence of the lighter helium isotope, ^{3}He, was yet unknown. It became only available in sufficient amounts at the end of the 1950's as a by-product of the tritium decay in nuclear power plants. Although, owing to this complicated production process, ^{3}He was - and still is - very expensive, ^{3}He gas evaporation cryostats provided then an easy way to obtain minimum

temperatures of typically 0.3 K. Nowadays, they are to a large extent replaced by ^{3}He-^{4}He dilution refrigerators which have been developed approximately one decade later.

In 1926 when H. Kamerlingh-Onnes reported the minimum temperature of 0.7 K, an entirely new cooling technique was proposed by Peter Debye and William F. Giauque, the so-called adiabatic demagnetization of paramagnetic salts, based on a magnetic cooling effect and expected to make the millikelvin temperature range accessible for research. Indeed, its realization in 1933 led to a minimum temperature of 0.27 K. The cooling process of this method is based on the fact that the magnetic disorder entropy of electronic magnetic moments in a paramagnetic salt, like, e.g., CMN (Cerium Magnesium Nitrate) should remain constant during a demagnetization of the sample in an adiabatic process, i.e. when the sample is thermally isolated from its environment. Adiabatic demagnetization of a paramagnetic salt was the first technique to provide temperatures significantly below 1 K, and finally turned out to be applicable in the temperature range between about 2 mK and 1 K. Today, this technique is almost completely replaced by the ^{3}He-^{4}He dilution refrigeration technique, too.

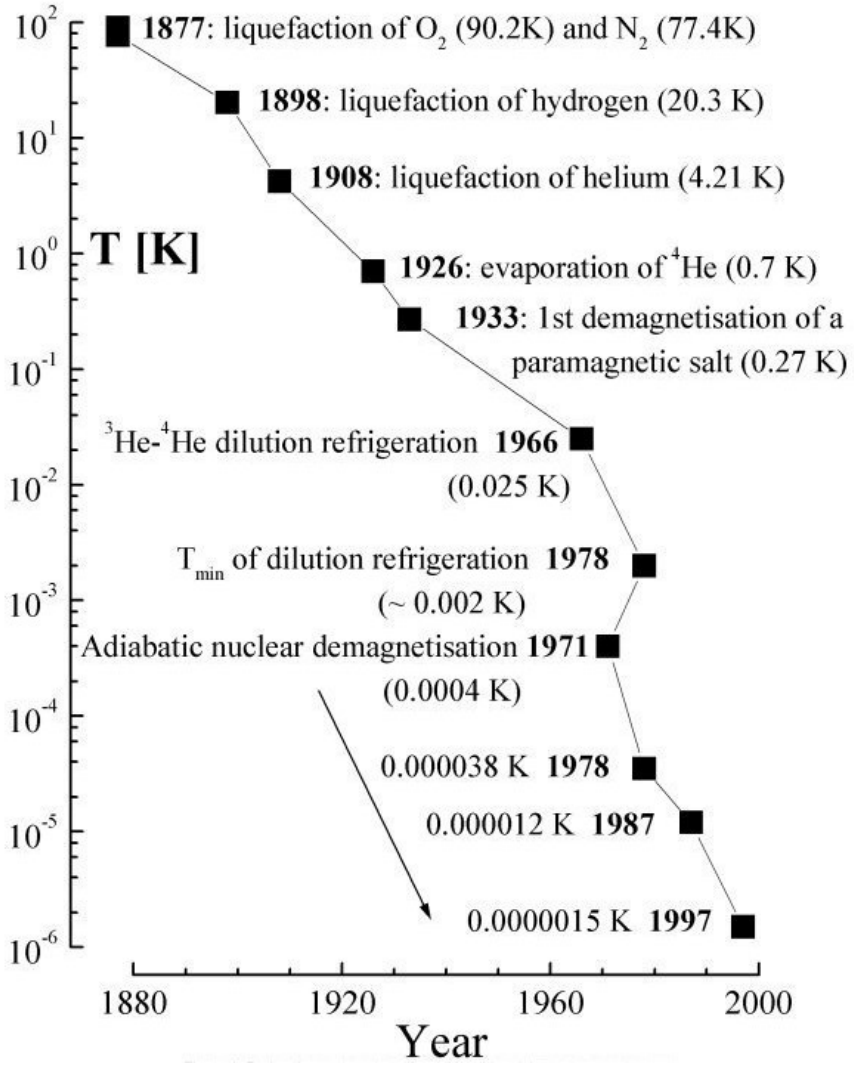

Figure 1. During the past century, the achievable minimum temperature was lowered by about eight orders of magnitude. For the sake of clarity, this figure presents only a

selection of the most important achievements on the way towards absolute zero. In particular, temperatures of about 2 mK have been reached long before the 1978 record of the ^{3}He-^{4}He dilution refrigeration, by means of adiabatic demagnetization of paramagnetic salts and of the Pomeranchuk cooling. (Modified from Pobell, F. (1996): Matter and methods at low temperatures. Berlin: Springer-Verlag.)

The underlying principle of the cooling technique which made the microkelvin temperature range accessible to research is the same as the adiabatic demagnetization of paramagnetic salts. The difference between both methods is essentially determined by the different magnitude of the magnetic moments involved. This technique, the adiabatic demagnetization of nuclear magnetic moments, is the only method known today which enables refrigeration far into the microkelvin temperature range. Whereas the demagnetization of electronic moments enables refrigeration only down to about 2 mK, the much smaller nuclear magnetic moment provides access to temperatures almost three orders of magnitude below the minimum temperature obtained by the demagnetization of paramagnetic salts. Nuclear refrigeration was independently proposed in 1934 by Cornelius J. Gorter and 1935 by Nicholas Kurti and Francis E. Simon, soon after the first successful attempt of the magnetic refrigeration of a paramagnetic salt has proven the applicability of the principle of magnetic cooling in 1933. The first experimental realization of nuclear magnetic refrigeration by N. Kurti at Oxford dates back to 1956, and in the following 40 years it was gradually improved, especially by the research group of Olli Lounasmaa at the Helsinki University of Technology during the 1960s and 1970s, and by Frank Pobell and his co-workers in Jülich and Bayreuth in the 1980s and 1990s. Whereas in the first attempt, in 1956, the minimum temperature could be maintained only for a very short time due to the strong coupling between the cold nuclei and the "hot" electrons of the demagnetized sample, the nuclear demagnetization stages are nowadays so much advanced that they can provide hold times below 1 mK for more than a month (see Section 1.3).

At the time of the first attempts to get nuclear magnetic refrigeration to work, a major breakthrough was achieved in the 1960s with the development of the ^{3}He-^{4}He dilution refrigeration, the first continuous cooling technique for the millikelvin temperature range. This technique was proposed by H. London, G.R. Clarke and E. Mendoza in 1962 and takes into account the unique property of the finite solubility of ^{3}He in liquid ^{4}He even at T = 0 K. The cooling process is based on the different enthalpies of pure ^{3}He and a diluted ^{3}He-^{4}He solution with a ^{3}He concentration of about 6.5%, and cooling occurs by forcing ^{3}He atoms to move across a phase boundary between the "concentrated" (pure) and the "diluted" ^{3}He phase. In 1965, ^{3}He-^{4}He dilution refrigeration reached a temperature of 0.22 K, and only one year later a minimum temperature of 0.025 K has been achieved at the laboratories in Dubna and Manchester. Dilution refrigeration soon became and still is the most important and most powerful technique for cooling into the millikelvin temperature range. Besides ^{3}He-^{4}He dilution refrigeration and the demagnetization of paramagnetic salts, the so-called Pomeranchuk cooling is another technique which, based on the particular properties of the ^{3}He melting curve, is able to provide temperatures around 2 mK. This technique, however, was of great importance only in the 1970s; today, the whole millikelvin temperature range is almost exclusively covered by the ^{3}He-^{4}He dilution refrigeration.

1.3. Combining Dilution Refrigeration with Adiabatic Nuclear Demagnetization: The Way to Microkelvin Physics

The development of the ^{3}He-^{4}He dilution refrigeration technique in the 1960s marks an important turning point in low temperature physics: for the first time, a continuous cooling technique for the lower millikelvin temperature range was available. Nowadays, a temperature as low as about 5 mK is the typical base temperature of a dilution refrigerator, and it can be practically maintained without any temporal limitations. ^{3}He-^{4}He dilution units are commercially available in almost all configurations tailored to the particular needs and the experimental requirements of the scientist.

The distinct properties which make ^{3}He-^{4}He dilution refrigeration superior to all other cooling techniques for the millikelvin range can be summarized as follows: it is a continuously working cooling method, it is insensitive to large magnetic fields, and, dependent on the particular configuration and design of a ^{3}He-^{4}He dilution cryostat, it may provide a large cooling power even at very low temperatures. All these properties turned out to be indispensable prerequisites for the development of the adiabatic nuclear demagnetization technique.

In contrast to ^{3}He-^{4}He dilution refrigeration, nuclear magnetic refrigeration is a "one way" cooling processes. In an ideal adiabatic demagnetization process, the starting conditions - i.e. the starting temperature T_i to which the refrigerant has been pre-cooled in a static magnetic field B_i by means of a ^{3}He-^{4}He dilution refrigerator - determine the final temperature T_f of the nuclear stage achievable in the final field of demagnetization: $T_f / B_f = T_i / B_i$. Typical initial values for nuclear demagnetization are $T_i \sim 10$ mK and $B_i \sim 8$ T (tesla), and these conditions are reached after approximately 2-3 days of pre-cooling. In order to reach these starting conditions it is obvious that the above mentioned properties of a dilution refrigerator - its insensitivity to large magnetic fields as well as its large cooling power - are of crucial importance for a successful pre-cooling process. Subsequently, the nuclear stage is decoupled from the ^{3}He-^{4}He dilution refrigerator by opening a superconducting heat switch which makes use of the very different thermal conductivities of a superconductor like, e.g., aluminum or indium, in its normal- and superconducting phase (see Section 1.4.1). The switch is operated by a magnetic field coil which enables the suppression of superconductivity by applying a magnetic field larger than the critical magnetic field of superconductivity of the heat switch material (see Sections 2 and 2.2.1). In an ideal process, demagnetization from the above given starting conditions to a final field of 8 mT would result in a minimum temperature $T_f = 10$ μK. However, losses during the demagnetization, e.g. due to eddy current heating, and internal as well as external heat leaks lead to deviations from an ideal adiabatic behavior especially at small magnetic fields which eventually results in higher final temperatures.

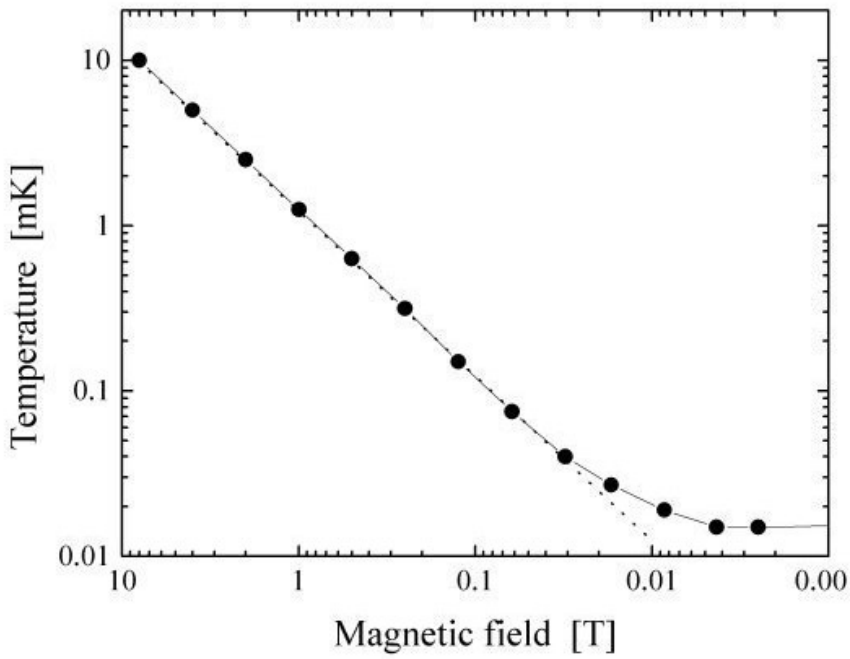

Figure 2. Typical temperature development of the Bayreuth nuclear demagnetization cryostat during a demagnetization from the initial conditions of T = 10 mK and B = 8 T to a final field of 4 mT. The achieved minimum temperature after demagnetization was about 15 μK. The typical time scale for the demagnetization to the final field is of the order of 2-3 days. The adiabatic behavior of the demagnetization, which can be expressed by the condition B / T = constant, is observed to temperatures clearly below 0.1 mK and is indicated by the dotted line. (Modified from Pobell, F. (1996): Matter and methods at low temperatures. Berlin: Springer-Verlag.)

Figure 2 shows the typical correlation of the reduction of the magnetic field and the lowering of the temperature during a demagnetization process. The time available for the scientist to perform experiments at the final temperature after demagnetization is basically determined by the magnitude of the final magnetic field and the nuclear magnetic properties of the refrigerant (as well as its mass, i.e. its heat capacity in the magnetic field). Copper is the material which is almost exclusively used as coolant in nuclear refrigeration, not because it is the ideal material, but because it is considered to be the best possible compromise among all metals in question. Its properties fulfill a number of important prerequisites necessary for a successful cooling into the microkelvin temperature range: it has a large nuclear Curie constant and a non-zero nuclear spin ($I > 0$), internal fields and intrinsic heat release (caused, e.g., by relaxation of so-called tunneling systems or due to ortho-para conversion of hydrogen) are small, it has a good thermal conductivity, and neither a superconducting transition nor magnetic order has been observed. The latter point does only apply to the electrons in copper; magnetic order of the nuclei has been observed in experiments where only the nuclei of the sample had been cooled and the lattice and the electrons stayed at a "much higher" temperature: magnetic order in copper has been found at a temperature of the nuclei of T_N = 60 nK, with lattice and electrons at T ~ 100 μK. This decoupling of the nuclei from the rest of the sample is a result of the poor coupling between electrons and nuclei in copper. The occurrence of nuclear magnetic order in copper far in the sub-microkelvin

temperature range implies that nuclear demagnetization should not be affected by this magnetic phase transition at microkelvin temperatures.

To date, the lowest electronic temperature has been obtained in a double stage process by nuclear adiabatic demagnetization of a (much smaller) platinum sample which has been pre-cooled to its starting conditions by a copper nuclear stage. A minimum temperature of T_f = 1.5 μK in a final magnetic field of B_f = 2.5 mT has been obtained. Under these extreme conditions, the smallest heat input into the sample immediately leads to a warm-up from the base temperature; in the case of the platinum experiment, the sample remained at the base temperature for a few hours only due to an external heat load of about 10 pW.

-
-
-

Bibliography

Andrä, W., Nowak, H., (Eds.) (1998): Magnetism in Medicine. A Handbook. Berlin: Wiley-VCH. [This book is a survey of the most important developments in the field of magnetism in medicine, with an emphasis on modern imaging techniques like magnetic resonance tomography and magnetoencephalography].

Asner, F.M. (1999): *High field superconducting magnets.* Oxford: Clarendon Press. [Compilation of the current state-of-the-art in superconducting magnet technology]

Buschow, K.H.J., (1997): Magnetism and Processing of Permanent Magnet Materials. *Handbook of Magnetic Materials.* Vol. 10 (ed. K.H.J. Buschow), p. 463-593. Amsterdam: Elsevier. [This review follows a former review by Buschow in 1988 (Ferromagnetic Materials, Vol. 4, E.P. Wohlfahrt, K.H.J. Buschow, ed.) and concentrates on the progress in permanent magnet materials research during the past two decades of the twentieth century.]

Dahl, P.F., (1992): *Superconductivity. Its Historical Roots and Development from Mercury to the Ceramic Oxides.* New York: American Institute of Physics. [A survey of the historical development of almost one century of research on superconductivity.]

Duckworth, H.E., Barber, R.C., Venkatasubramanian, V.S., (1990): *Mass spectroscopy.* Cambridge: University Press. [In-depth compilation of methods and applications of mass spectroscopy]

Eremets, M., (1996): *High pressure experimental methods.* Oxford: Oxford university press. [Introduces the technique of high pressure experiments and includes many drawings of high pressure cells]

Hablanian, M., (1997): *High-vacuum technology. A practical guide.* New York: Marcel Dekker, Inc. [This book provides a profound introduction to high vacuum technology with particular emphasis on the achievement of very low gas densities and a detailed discussion of a wide range of different pumping systems.]

Kapusta, Cz., Riedi, P.C., Tomka, G.J. (1998): Magnetism of permanent magnet materials and related compounds as studied by NMR. *Handbook of Magnetic Materials.* Vol. 11 (ed. K.H.J. Buschow), p. 407-

491. Amsterdam: Elsevier. [This review contains a recent compilation of nuclear magnetic resonance data of modern materials used for permanent magnets.]

Lounasmaa, O.V. (1974): *Experimental principles and methods below 1K*. London: Academic. [The first detailed compilation of low and ultralow temperature physics and techniques, and still today a book of inestimable value, suitable for the newcomer in the field as well as for the expert.]

Mendelssohn, K. (1977): *The quest for absolute zero*. London: Taylor and Francis Ltd. [The description of the history of low temperature physics, from the liquefaction of gases at the end of the 20th century until the realization of the magnetic cooling techniques, and of the role which low temperature physics played in the development of thermodynamics, statistics and quantum mechanics, makes this book an outstanding contribution to the "meaning of low temperature physics" as its subtitle reads.]

Pobell, F. (1996): *Matter and methods at low temperatures.* Berlin: Springer-Verlag. [This book is a summary of the state of the art in low and ultralow temperature physics and techniques. It is particularly valuable because of its numerous tables and graphs of materials' properties at low temperatures.]

Roth, A., (1990): *Vacuum technology*. Amsterdam: Elsevier. [Includes theoretical as well as technical aspects of modern vacuum technology]

Tilley, D.R., Tilley, J. (1987): *Superfluidity and superconductivity.* Bristol: Adam Hilger Ltd. [This book covers all fundamental aspects of superconductivity and superfluidity.]

Tsuneto, T., (1998): *Superconductivity and superfluidity*. Cambridge: University Press. [Tsuneto's monograph is a rather compressed treatise of both phenomena. It also deals with high temperature superconductivity and is addressed to a reader who already disposes of some basic knowledge in the field.]

Vollhardt, D., Wölfle, P., (1990). *The superfluid phases of ^{3}He.* London: Taylor and Francis. [An extraordinarily comprehensive compilation of today's knowledge about this unique quantum liquid]

Biographical Sketch

Reinhard König

Education:

Physikalisches Institut, Universität Bayreuth, D-95440 Bayreuth, Germany
1993: Doctoral thesis, Universität Bayreuth, Germany
1994/1995: Department of Physics, University of Nottingham, Nottingham, England
1996-2001: Physikalisches Institut, Universität Bayreuth, Germany
2000: Habilitation, Universität Bayreuth, Germany

Research activities and interests.

Quantum fluids and solids at very low temperatures: transport properties, thermal boundary resistance, nuclear magnetic properties, quantum liquids in high magnetic fields: effects of spin-polarization
Acoustic properties of amorphous and polycrystalline materials at low temperatures: sound velocity and internal friction
Magnetic properties of mesoscopic metal particles
Superconductivity of granular platinum
Electronic and magnetic properties of La-Sr-Mn-O perovskites (giant magneto-resistance)
Cooling techniques and low temperature thermometry