

EXPERIMENTAL
TECHNIQUES IN
LOW-TEMPERATURE
PHYSICS

BY
GUY K. WHITE

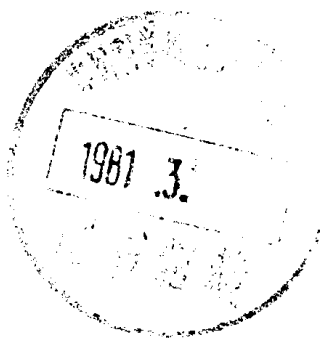
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PREFACE TO THIRD EDITION

SINCE the second edition of this book appeared in 1968, there has been a continued growth of large-scale cryogenics, concerned with liquefied natural gas, storage and transport of liquid helium on a large scale, applications of superconductivity, and space research. New books have been written to meet the needs of this cryogenic engineering industry, including *Cryogenic fundamentals* (edited by G. G. Haselden, Academic Press, London 1971). A regular series of International Cryogenic Engineering Conferences have been held in 1967, 1968, 1970–1976, 1978, and their proceedings have been published by IPC Science and Technology Press, Surrey, UK. These complement the annual conferences held in North America since 1954 which were published by Plenum Press as *Advances in cryogenic engineering*.

On the research laboratory scale, the millikelvin range of temperatures has become more accessible with developments in dilution refrigeration, Pomeranchuk cooling, nuclear cooling. Comprehensive books on these fields have been written by O. V. Lounasmaa (*Experimental principles and methods below 1 K*, Academic Press, London, 1974), D. S. Betts (*Refrigeration and thermometers below one kelvin*, Sussex University Press, 1976), and R. P. Hudson (*Magnetic cooling*, North-Holland, 1972). For more general coverage of laboratory techniques, there was a new edition in 1973 of *Low temperature laboratory techniques* by A. C. Rose-Innes (English Universities Press, London) and other books of use to the experimental physicist including *Cryogenic laboratory equipment* by A. J. Croft (Plenum Press, New York, 1970), *Éléments de cryogenie* by R. R. Conte (Masson et Cie, Paris, 1970), and *Advanced cryogenics* edited by C. A. Bailey (Plenum Press, New York, 1971).

Despite these additions to the library shelves, I have been persuaded that *Experimental techniques in low temperature physics* also fills a need and should reappear in a new edition. This edition has involved adding some new material on ^3He systems, temperature measurement, physical data, etc. At the same time the overall size has been reduced to keep costs down, by shortening some of the less-used sections on refrigeration, heat

exchange, magnetic cooling, and the background to physical properties while preserving some of the history of low-temperature physics.

My colleagues at the National Measurement Laboratory who have helped in offering suggestions include J. A. Birch, L. Besley, J. G. Collins, J. Dunlop, R. C. Kemp, W. R. G. Kemp, and R. B. Roberts. My old friend A. J. Croft has been a kind and persistent advocate for this edition which was partly stimulated also by a pleasant few months in 1976/77 spent at the Universities of Leeds and Oxford with help from the Science Research Council of the UK. Mrs. P. Riley has again done much of the secretarial work and Mrs. S. Williams has produced many new diagrams.

CSIRO National Measurement Laboratory
Sydney, Australia
February 1978

G. K. W.

PREFACE TO THE SECOND EDITION

SINCE the publication of the first edition of this monograph, there has been a continued growth of the study of low temperatures, largely because of the steady increase in the number of people and laboratories concerned with the properties of matter. This is partly a reflection of the need for cryogenic information among technically based industries concerned with communications, aerospace, gases, etc.

A number of new books have been written to meet the requirements of the times and it is pertinent to mention briefly their coverage *vis-à-vis* that of the present book. Among smaller texts which are of general interest to university students, both graduate and undergraduate, are the latest edition of *Low temperature physics* by L. C. Jackson (Methuen, London, 1962), *Cryophysics* by K. Mandelssohn (Interscience, New York, 1960), *The quest for absolute zero* by the same author (Wiedenfelf and Nicolson, London, 1966), and *Cryogenics* by M. McClin-tock (Reinhold, New York, 1964). These all give interesting accounts of the scope and applications of physics at low temperatures without any particular attention to the techniques or methods used.

The properties of solids (therefore not including liquid helium) at low temperatures have been surveyed by H. M. Rosenberg in *Low temperature solid state physics* (Clarendon Press, Oxford, 1963): this contains no treatment of either laboratory techniques or the principles of thermometry and refrigeration.

A larger compendium called *Cryogenic technology* (Wiley, New York, 1963) produced under the editorship of R. W. Vance includes chapters by some experienced contributors on the properties of solids, the principles of refrigeration, superconductivity, thermometry, as well as applications of cryogenics in biology, space research, cryopumping, etc.

Among the books devoted primarily to the techniques, methods, and properties of technical materials used at low temperatures in *Cryogenic engineering* by R. B. Scott (Van Nostrand, Princeton, 1959); as its name implies, this emphasizes large-scale engineering techniques and information rather than those used

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on the laboratory scale. On the other hand, *Low temperature techniques* by A. C. Rose Innes (Van Nostrand, Princeton, 1964) is a useful practical guide for the laboratory worker who needs to handle refrigerant liquids but is not concerned with making them or with cooling by demagnetization methods. A more comprehensive treatment is in *Experimental cryophysics* edited by F. E. Hoare, L. C. Jackson, and N. Kurti (Butterworths, London, 1961): various experienced physicists have contributed the different chapters which deal with all the aspects of producing and handling low temperatures.

The present monograph remains a compromise in size between the last two. The revision is intended to add new material, chiefly concerning the use of helium-3 as a refrigerant and superconducting magnets, and to bring chapters on liquefaction, temperature measurement, and the properties of materials up to date. Minor changes have been made to other chapters to include useful new information or to rectify omissions in the earlier edition. In the process some of the tables which are referred to most frequently have been gathered together in the Appendix but tables of limited or specific interest remain through the text. A short list of firms which presently supply low-temperature equipment is included in the Appendix.

Some preparatory work for this revised edition was done while I was enjoying the hospitality of the Bell Telephone Laboratories at Murray Hill and I am very grateful to a colleague there, Dr. Eric Fawcett, for his help. Here at the C.S.I.R.O. National Standards Laboratory I have discussed many cryogenic problems with my colleagues and I am most grateful to them, particularly W. R. G. Kemp, for their patience; Mrs. P. Riley has aided greatly with her secretarial help.

G. K. W.

C.S.I.R.O. Division of Physics
Sydney, Australia
January 1967

PREFACE TO THE FIRST EDITION

It is not very many years since most low-temperature physicists—those doing research on physical properties at the temperatures of liquid helium or liquid hydrogen—were trained in the techniques of this particular field at one of the comparatively few centres of low-temperature research such as Leiden, Berlin, Berkeley, Oxford, Cambridge, Toronto. Today, with the advent of increased research grants, defence contracts, and Collins helium liquefiers, many physicists in many laboratories around the world wish to carry out physical investigations in the low-temperature range and are faced with problems of designing cryostats, filling them with liquid helium, maintaining and measuring various temperatures. Often these problems are not very difficult, but nevertheless the technical information and published experience which may help to solve them are spread over a wide range of years and journals. There appears to be a need for a book which gives details of this information, including physical data for the technical materials used in cryostat design, methods of measuring and controlling temperatures, and associated problems. This book is an attempt to meet this need.

In a first flush of enthusiasm I hoped to include the full technical details of many operations, for example the winding of a countercurrent heat exchanger, but soon realized that lack of space made this impossible. Some chapters, notably those dealing with gas liquefaction and magnetic cooling, are merely brief discussions of the principles involved with examples and references to more detailed work. They are intended not only to give continuity to the subject-matter and to introduce those not familiar with the subject to the principles and the literature but also to act as a guide for anyone wishing to design a liquefier or adiabatic demagnetization cryostat; these are both subjects to which complete books could be devoted and recent reviews by Daunt, Collins, Ambler and Hudson, and de Klerk, and earlier books by Ruhemann, Keesom, Casimir, and Garrett cover these subjects far more competently than I hope to do. Any reader wishing for a more complete survey of low-temperature physics, its scope and its achievements rather than its techniques, should

prefer the texts of E. F. Burton, H. Grayson Smith, and J. O. Wilhelm (*Phenomena at the temperature of liquid helium*, Reinhold, 1940), L. C. Jackson (*Low temperature physics*, Methuen, 2nd edn, 1948), or C. F. Squire (*Low temperature physics*, McGraw-Hill, 1953).

I would like to acknowledge my debt of gratitude to the late Sir Francis Simon for his kindness and patience when I started the study of low-temperature physics; he asked me, as one of his students, to design and build a Linde helium liquefier. I hope this book is rather more successful than my first effort at solving the problems of helium liquefaction.

In preparing this book I have been helped considerably by various friends who have read and criticized individual chapters: these include my colleagues Drs. T. H. K. Barron, J. S. Dugdale, D. K. C. MacDonald, F. D. Manchester, and S. B. Woods of the Division of Pure Physics (National Research Council), Dr. J. A. Morrison of the Division of Pure Chemistry (National Research Council), Dr. H. Preston-Thomas of the Division of Applied Physics (National Research Council), and Dr. R. P. Hudson of the National Bureau of Standards in Washington.

As will appear throughout the text many publishers and learned societies have kindly granted permission for the reproduction of figures which originally appeared in their books and periodicals. The facilities and the co-operation of such sections of the National Research Council as the Central Drafting Office, Duplication, and the Typing Pool have been of great assistance.

Finally, I am happy to thank Dr. M. T. Elford for his help in the proof-reading of this book.

G. K. W.

Ottawa, Canada
July 1957

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CHAPTER I

PRODUCTION OF LOW TEMPERATURES

1. Isentropic cooling

Introduction

SINCE the entropy or degree of disorder of a system at constant volume or constant pressure is a monotonically increasing function of temperature, any process of cooling may be regarded as one of ordering or entropy reduction. In the words of Simon, a refrigerator is a form of 'entropy-squeezer'. This 'squeezing' is possible since entropy S is a function of other variable parameters as well as temperature, e.g. $S = S(T, X)$ where the parameter X is a physical property of the system which can be varied within limits so as to change the entropy.

Figure 1.1a shows that when X is altered isothermally from X_1 to X_2 the entropy is reduced. By further varying X from X_2 to X_1 under isentropic conditions, a lowering in temperature from a temperature T_B to temperature T_C is achieved; an isentropic change is adiabatic since $\Delta S = \int dQ/T$. The process is a reversible one and therefore by the second law of thermodynamics is the most efficient means of cooling, in terms of the external work required. From Fig. 1.1a it would seem possible, in principle at least, to cool the system to the absolute zero of temperature by a limited number of such steps as $A \rightarrow B \rightarrow C$. However, if the situation is as depicted in Fig. 1.1b, this is no longer the case; that 1b is correct, rather than 1a, is implied by the third law of thermodynamics, which in the form due to Simon (1930, 1956) states: 'at absolute zero the entropy differences disappear between all those states of a system which are in internal thermodynamic equilibrium'.

The equivalence of this statement to the alternative statement that 'it is impossible by any procedure, no matter how idealized to reduce the temperature of any system to the absolute zero in a finite number of operations', has been demonstrated by Guggenheim and is strongly suggested by Fig. 1.1b. However, we still have a means of lowering the temperature—even if not to

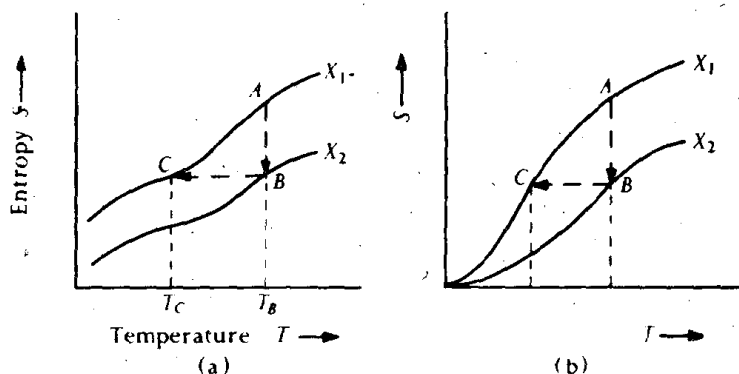


FIG. 1.1. A cooling process.

absolute zero—at our disposal and in practice such methods have been widely used. Associating the parameter X with the pressure p applied to a gas or with the magnetic field H applied to an assembly of magnetic dipoles, the principles of gas cooling by isothermal compression and adiabatic expansion, and of magnetic cooling by isothermal magnetization and adiabatic demagnetization are exemplified.

Examples

In the single-expansion helium liquefier (Simon 1932) shown schematically in Fig. 1.2, helium gas is compressed isothermally into chamber 1 to a pressure of 100 atm; a temperature of about 15 K is maintained, heat being transferred through the medium of helium exchange gas in the space 2 to a bath of liquid hydrogen boiling under reduced pressure in the dewar vessel 3. Thus the initial temperature $T_B \approx 15$ K; in practice T_B may be lowered to 10 K by reducing the pressure above the evaporating hydrogen to well below its triple-point pressure. Then the exchange gas is removed from 2 and the compressed helium in chamber 1 is expanded through the valve V to a pressure of 1 atm, so that the gas remaining in the chamber is adiabatically cooled to the final temperature T_C (equal to the liquefaction temperature); a substantial fraction of the chamber is left filled with liquid helium. The importance of the metal chamber 1 having a heat capacity small in relation to the gas is paramount, and is easily realized with helium gas at about 10 K, but would not be the case for compressed air at 200 K, for the heat capacity of the containing pressure vessel at this higher temperature would be considerable.

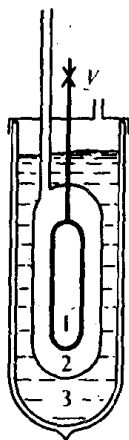


FIG. 1.2. Schematic diagram of the Simon expansion liquefier.

The desorption helium liquefier first discovered by Simon in 1926 and developed further by Mendelssohn (1931) is very similar. In this the inner container is partially filled with activated charcoal and the process $A \rightarrow B$ of Fig. 1.1 is simply the isothermal adsorption of helium gas to a pressure of 1–5 atm on the charcoal, the heat of adsorption being removed via exchange gas to the liquid-hydrogen bath. This is followed by an adiabatic desorption or pumping away of the adsorbed helium gas during which the temperature of the remaining gas, charcoal, and the container fall toward or to the liquefaction temperature.

The suggestion that a process of ordering by a magnetic field could be applied to an assembly of weakly interacting magnetic dipoles, as in a paramagnetic salt, and that a subsequent adiabatic demagnetization would cause cooling, was made independently in 1926 by Debye and Giauque. Within a few years Giauque and MacDougall (1933), de Haas, Wiersma, and Kramers (1933), and Kurti and Simon (1935) verified this experimentally; Fig. 1.3 illustrates the process schematically. A pill of a paramagnetic salt (gadolinium sulphate in Giauque's early experiments) is magnetized in a field of one tesla at a temperature of about 1 K, the heat of magnetization being transferred through helium exchange gas to the pumped liquid helium in the surrounding dewar vessel. After removal of the exchange gas, the magnetic field is reduced and the temperature of the salt pill falls. There is a further analogy with the expansion liquefier in that for appreciable cooling to occur, the lattice vibrational specific heat at 1 K of the

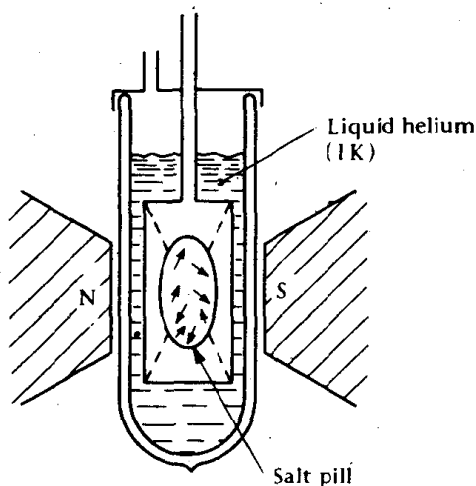


FIG. 1.3. Schematic diagram of the magnetic cooling process.

salt pill must be small in comparison with the 'magnetic heat capacity', i.e. with the thermal energy of the disoriented magnetic ions; if this were not so, the reduction in the entropy on magnetization would be small in comparison with the total entropy of the crystal lattice.

2. Isenthalpic cooling

Introduction

The discovery, in 1862, by Joule and Thomson that a gas undergoes a temperature change when it expands slowly through a porous plug, has been widely applied to gas refrigeration. The cooling on adiabatic expansion discussed in § 1 is a property of the perfect gas—in which attractive or repulsive forces are zero—and occurs for real gases at all temperatures by virtue of their performing 'external' work. However, the Joule-Thomson effect for any real gas depends both in magnitude and sign on the temperature and is zero for a perfect gas at all temperatures. This effect is sometimes called an 'internal work' process because the temperature change is determined by the change in energy of gas when the average separation between the gas molecules is increased.

In the Joule-Thomson process a gas undergoes a continuous throttling or expansion as it is driven by a constant pressure p_1 on one side of the expansion valve (or porous plug) and expands to a

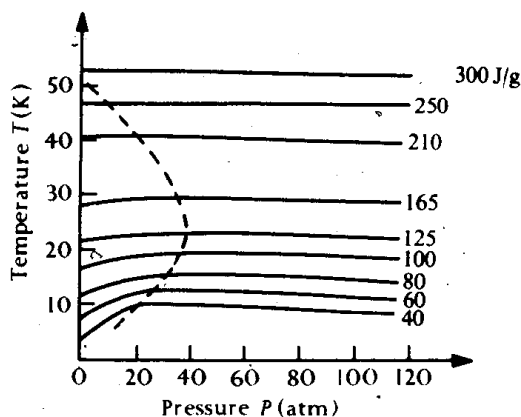


FIG. 1.4. Isenthalpic curves of helium.

lower pressure p_2 on the other. Considering a fixed mass of gas passing the valve, it can easily be shown that the total heat or enthalpy $H = U + pV$ is unchanged in passing from state 1 (pressure p_1) to state 2 (pressure p_2); U is the internal energy per unit mass and V is the volume.

Since $dH = 0$ such a process is called isenthalpic.

Performing such a throttling experiment for helium we should obtain a set of values of T_2 —the temperature after expansion—lying on a smooth curve. If T_1 —the temperature before expansion—is below the so-called inversion temperature, such curves have a maximum as seen in Fig. 1.4. The locus of the maxima encloses a region within which the differential Joule-Thomson coefficient

$$\mu = \left(\frac{\partial T}{\partial p} \right)_H$$

is positive and hence a cooling results on expansion.

We may show:

$$dH = \left(\frac{\partial H}{\partial p} \right)_T dp + \left(\frac{\partial H}{\partial T} \right)_p dT,$$

therefore

$$\begin{aligned} \left(\frac{\partial T}{\partial p} \right)_H &= - \left(\frac{\partial H}{\partial p} \right)_T / \left(\frac{\partial H}{\partial T} \right)_p \\ &= - \frac{1}{C_p} \left(\frac{\partial H}{\partial p} \right)_T \end{aligned}$$

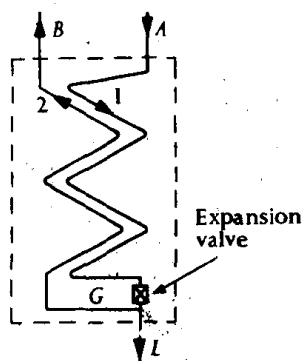


FIG. 1.5

or, since

$$dH = dU + p dV + V dp = T dS + V dp,$$

$$\begin{aligned} \left(\frac{\partial T}{\partial p}\right)_H &= -\frac{1}{C_p} \left\{ T \left(\frac{\partial S}{\partial p}\right)_T + V \right\} \\ &= \frac{1}{C_p} \left\{ T \left(\frac{\partial V}{\partial T}\right)_p - V \right\} \end{aligned}$$

and this vanishes for the perfect gas since $pV = RT$.

This isenthalpic process is important as it forms the final stage for nearly all 'circulation' liquefiers ('circulation' liquefiers exclude the Simon expansion and desorption liquefiers). Such a final stage, including expansion valve and heat interchanger, is shown schematically in Fig. 1.5, where compressed gas at pressure p_A , temperature T_A , and enthalpy H_A per gram enters at A. After expansion the gas (state p_B , T_B , H_B) leaves through the interchanger at B and liquid L (state p_L , T_L , H_L) collects at L.

If the liquefaction efficiency is denoted by \mathcal{E} , then since the process is isenthalpic

$$H_A = \mathcal{E}H_L + (1 - \mathcal{E})H_B,$$

therefore the efficiency $\mathcal{E} = (H_B - H_A)/(H_B - H_L)$.

Heat interchanger

So far the heat interchanger has not been mentioned, but it plays a vital role in determining the liquefaction efficiency as it determines T_B and therefore the enthalpy H_B . The exchanger efficiency, η , is usually defined as the ratio of the actual heat