International Review of Science

Inorganic Chemistry Series Two

Volume 8
Radiochemistry

Edited by A. G. Maddock

International Review of Science

Inorganic Chemistry
Series Two

Consultant Editor
H. J. Emeléus, F.R.S.

Publisher's Note

The International Review of Science is an important venture in scientific publishing, which is presented by Butterworths in association with University Park Press, Baltimore. The basic concept of the Review is to provide regular authoritative reviews of entire disciplines. Chemistry was taken first as the problems of literature survey are probably more acute in this subject than in any other. Biochemistry and Physiology followed naturally. As a matter of policy, the authorship of the Review of Science is international and distinguished, the subject coverage is extensive, systematic and critical.

The Review has been conceived within a carefully organised editorial framework. The overall plan was drawn up, and the volume editors appointed by seven consultant editors. In turn, each volume editor planned the coverage of his field and appointed authors to write on subjects which were within the area of their own research experience. No geographical restriction was imposed. Hence the 500 or so contributions to the Review of Science come from many countries of the world and provide an authoritative account of progress.

The publication of Inorganic Chemistry Series One was completed in 1972 with ten text volumes and one index volume, and in accordance with the stated policy of issuing regular reviews to keep the series up to date, volumes of Series Two will be published between the autumn of 1974 and the spring of 1975. They will be followed by Series Two of Physical and Organic Chemistry in the period 1975–1976. Volume titles will generally be the same as in Series One but the articles themselves may either cover recent advances in the same subject or deal with a different aspect of the main theme of the volume. In Series Two an index will be incorporated in each volume and there will be no separate index volume.

Butterworth & Co. (Publishers) Ltd.

INORGANIC CHEMISTRY SERIES TWO

Consultant Editor
H. J. Emeléus, F.R.S.,
Department of Chemistry, University of Cambridge

Volume titles and Editors

1 **MAIN GROUP ELEMENTS—HYDROGEN AND GROUPS I–III**
Professor M. F. Lappert, *University of Sussex*

2 **MAIN GROUP ELEMENTS—GROUPS IV AND V**
Dr. D. B. Sowerby, *University of Nottingham*

3 **MAIN GROUP ELEMENTS—GROUPS VI AND VII**
Professor Viktor Gutmann, *Technical University of Vienna*

4 **ORGANOMETALLIC DERIVATIVES OF THE MAIN GROUP ELEMENTS**
Professor B. J. Aylett, *Westfield College, University of London*

5 **TRANSITION METALS—PART 1**
Professor D. W. A. Sharp, *University of Glasgow*

6 **TRANSITION METALS—PART 2**
Dr. M. J. Mays, *University of Cambridge*

7 **LANTHANIDES AND ACTINIDES**
Professor K. W. Bagnall, *University of Manchester*

8 **RADIOCHEMISTRY**
Dr. A. G. Maddock, *University of Cambridge*

9 **REACTION MECHANISMS IN INORGANIC CHEMISTRY**
Professor M. L. Tobe, *University College, University of London*

10 **SOLID STATE CHEMISTRY**
Dr. L. E. J. Roberts, *Atomic Energy Research Establishment, Harwell*

PHYSICAL CHEMISTRY SERIES TWO

Consultant Editor
A. D. Buckingham,
Department of Chemistry, University of Cambridge

Volume titles and Editors

1 **THEORETICAL CHEMISTRY**
Professor A. D. Buckingham, *University of Cambridge*

2 **MOLECULAR STRUCTURE AND PROPERTIES**
Professor A. D. Buckingham, *University of Cambridge*

3 **SPECTROSCOPY**
Dr. D. A. Ramsay, F.R.S.C., *National Research Council of Canada*

4 **MAGNETIC RESONANCE**
Professor C. A. McDowell, F.R.S.C., *University of British Columbia*

5 **MASS SPECTROMETRY**
Professor A. Maccoll, *University College, University of London*

6 **ELECTROCHEMISTRY**
Professor J. O'M Bockris, *The Flinders University of S. Australia*

7 **SURFACE CHEMISTRY AND COLLOIDS**
Professor M. Kerker, *Clarkson College of Technology, New York*

8 **MACROMOLECULAR SCIENCE**
Professor C.E.H. Bawn, C.B.E., F.R.S., *formerly of the University of Liverpool*

9 **CHEMICAL KINETICS**
Professor D. R. Herschbach, *Harvard University*

10 **THERMOCHEMISTRY AND THERMODYNAMICS**
Dr. H. A. Skinner, *University of Manchester*

11 **CHEMICAL CRYSTALLOGRAPHY**
Professor J. Monteath Robertson, C.B.E., F.R.S., *formerly of the University of Glasgow*

12 **ANALYTICAL CHEMISTRY—PART 1**
Professor T. S. West, *Imperial College, University of London*

13 **ANALYTICAL CHEMISTRY—PART 2**
Professor T. S. West, *Imperial College, University of London*

ORGANIC CHEMISTRY SERIES TWO

Consultant Editor
D. H. Hey, F.R.S.,
formerly of the Department of Chemistry, King's College, University of London

Volume titles and Editors

1 **STRUCTURE DETERMINATION IN ORGANIC CHEMISTRY**
Professor L. M. Jackman, *Pennsylvania State University*

2 **ALIPHATIC COMPOUNDS**
Professor N. B. Chapman, *Hull University*

3 **AROMATIC COMPOUNDS**
Professor H. Zollinger, *Eidgenossische Technische Hochschule, Zurich*

4 **HETEROCYCLIC COMPOUNDS**
Dr. K. Schofield, *University of Exeter*

5 **ALICYCLIC COMPOUNDS**
Professor D. Ginsburg, *Technion-Israel Institute of Technology, Haifa*

6 **AMINO ACIDS, PEPTIDES AND RELATED COMPOUNDS**
Professor N. H. Rydon, *University of Exeter*

7 **CARBOHYDRATES**
Professor G. O. Aspinall, *York University, Ontario*

8 **STEROIDS**
Dr. W. F. Johns, *G. D. Searle & Co., Chicago*

9 **ALKALOIDS**
Professor K. Wiesner, F.R.S., *University of New Brunswick*

10 **FREE RADICAL REACTIONS**
Professor W. A. Waters, F.R.S., *University of Oxford*

PHYSIOLOGY SERIES ONE

Consultant Editors
A. C. Guyton,
Department of Physiology and Biophysics, University of Mississippi Medical Center and
D. F. Horrobin,
Department of Physiology, University of Newcastle upon Tyne

Volumes titles and Editors

1 **CARDIOVASCULAR PHYSIOLOGY**
Professor A. C. Guyton and Dr. C. E. Jones, *University of Mississippi Medical Center*

2 **RESPIRATORY PHYSIOLOGY**
Professor J. G. Widdicombe, *St George's Hospital, London*

3 **NEUROPHYSIOLOGY**
Professor C. C. Hunt, *Washington University School of Medicine, St. Louis*

4 **GASTROINTESTINAL PHYSIOLOGY**
Professor E. D. Jacobson and Dr. L. L. Shanbour, *University of Texas Medical School*

5 **ENDOCRINE PHYSIOLOGY**
Professor S. M. McCann, *University of Texas*

6 **KIDNEY AND URINARY TRACT PHYSIOLOGY**
Professor K. Thurau, *University of Munich*

7 **ENVIRONMENTAL PHYSIOLOGY**
Professor D. Robertshaw, *University of Nairobi*

8 **REPRODUCTIVE PHYSIOLOGY**
Professor R. O. Greep, *Harvard Medical School*

BIOCHEMISTRY SERIES ONE

Consultant Editors
H. L. Kornberg, F.R.S.,
Department of Biochemistry University of Leicester and
D. C. Phillips, F.R.S., *Department of Zoology, University of Oxford*

Volume titles and Editors

1 **CHEMISTRY OF MACRO-MOLECULES**
Professor H. Gutfreund, *University of Bristol*

2 **BIOCHEMISTRY OF CELL WALLS AND MEMBRANES**
Dr. C. F. Fox, *University of California*

3 **ENERGY TRANSDUCING MECHANISMS**
Professor E. Racker, *Cornell University, New York*

4 **BIOCHEMISTRY OF LIPIDS**
Professor T. W. Goodwin, F.R.S., *University of Liverpool*

5 **BIOCHEMISTRY OF CARBO-HYDRATES**
Professor W. J. Whelan, *University of Miami*

6 **BIOCHEMISTRY OF NUCLEIC ACIDS**
Professor K. Burton, F.R.S., *University of Newcastle upon Tyne*

7 **SYNTHESIS OF AMINO ACIDS AND PROTEINS**
Professor H. R. V. Arnstein, *King's College, University of London*

8 **BIOCHEMISTRY OF HORMONES**
Professor H. V. Rickenberg, *National Jewish Hospital & Research Center, Colorado*

9 **BIOCHEMISTRY OF CELL DIFFER-ENTIATION**
Professor J. Paul, *The Beatson Institute for Cancer Research, Glasgow*

10 **DEFENCE AND RECOGNITION**
Professor R. R. Porter, F.R.S., *University of Oxford*

11 **PLANT BIOCHEMISTRY**
Professor D. H. Northcote, F.R.S., *University of Cambridge*

12 **PHYSIOLOGICAL AND PHARMACO-LOGICAL BIOCHEMISTRY**
Dr. H. K. F. Blaschko, F.R.S., *University of Oxford*

THE BUTTERWORTH GROUP

ENGLAND
Butterworth & Co (Publishers) Ltd
London: 88 Kingsway, WC2B 6AB

AUSTRALIA
Butterworths Pty Ltd
Sydney: 586 Pacific Highway 2067
Melbourne: 343 Little Collins Street, 3000
Brisbane: 240 Queen Street, 4000

NEW ZEALAND
Butterworths of New Zealand Ltd
Wellington: 26–28 Waring Taylor Street, 1

SOUTH AFRICA
Butterworth & Co (South Africa) (Pty) Ltd
Durban: 152–154 Gale Street

ISBN 0 408 70597 3

UNIVERSITY PARK PRESS

U.S.A. and CANADA
University Park Press
Chamber of Commerce Building
Baltimore, Maryland, 21202

Library of Congress Cataloging in Publication Data
Maddock, A. G
Radiochemistry
(Inorganic chemistry, series two, v.8) (MTP international review of science)
1. Radiochemistry. I. Title. II. Series. III. Series: MTP international review of science.
[DNLM: 1. Radiochemistry. QD601.2 R1324]
QD147.I56 vol. 8 [QD601.2] 546s [541'.38] 74-16212
ISBN 0-8391-0207-0

Typeset by Amos Typesetters, Hockley, Essex
Printed and bound in Great Britain by
REDWOOD BURN LIMITED
Trowbridge & Esher

Consultant Editor's Note

Following the successful production of the first series of ten volumes on inorganic chemistry in the International Review of Science, a second series has been prepared with the object of keeping inorganic chemists abreast of the many important advances that have taken place in the interim. The original plan has been adhered to in the main, though there are minor variations in the allocation of material to the volumes and in the space devoted to specific topics. The aim remains to provide a comprehensive critical survey of work published in the past few years in each of the main areas of inorganic chemistry. Many experts have collaborated in the production either as authors or as volume editors. I am deeply indebted to them all for the excellent work that they have done and for the way in which they have kept to production schedules. The high standard established in Series One has been fully maintained and I am confident that this publication will be of the utmost value to university teachers, research workers and advanced undergraduates.

Cambridge H. J. Emeléus

Preface

This volume, covering radiochemistry for the second series of reviews, generally follows the pattern of the first volume. In those areas developing fast enough to receive attention in both volumes, the relevant chapter only considers the work of the past two years; for the others a coverage of about five years has been chosen.

The problem of the superheavy elements is an example in the former category. A great deal of work, both theoretical and experimental, has appeared, but their identification appears as far away as ever. The chemistry of mesonic atoms is also included again. The increased facilities for such studies, using the 'meson factories' seems likely to lead to a rapid expansion of this area.

Amongst the topics not included in the last volume may be mentioned the chapter on techniques. This is almost never a static area and yet surprisingly few reviews have appeared in recent years. The length of this chapter shows the vitality of such studies. The chapter on hot atom chemistry concentrates its attention on those elements excluded from the related chapter in the previous volume. Nuclear chemistry is represented in a chapter covering the study of high-energy reactions appearing to proceed by simple mechanisms.

A great deal of work continues to appear concerning the chemical effect of nuclear transformations in solids. A small part of these investigations using Mössbauer emission spectroscopy was covered in the last volume. In this volume attention has been focused partly on the discrepancies between work in different laboratories and partly on the general conclusion to be drawn from all such studies.

Cambridge

A. G. Maddock

International Review of Science

Inorganic Chemistry
Series Two

Volume 8
Radiochemistry

Edited by **A. G. Maddock**
University of Cambridge

Butterworths · London
University Park Press · Baltimore

Contents

Muonium 1
V. G. Firsov, *Institute for Theoretical and Experimental Physics, Moscow*, and V. I. Goldanskii, *Institute for Chemical Physics, Moscow*

Chemical reactions of nuclear recoil particles in gases and liquids 49
D. S. Urch, *Queen Mary College, University of London*

Measurement techniques and instrumentation 105
J. D. Hemingway, *Universities Research Reactor, Lancashire*

Simple high-energy reactions on complex nuclei 197
L. B. Church, *State University of New York, Buffalo*

Superheavy elements 221
G. Herrmann, *Institut für Kernchemie, Mainz*

The chemical effects of nuclear transformations in solids 273
A. G. Maddock, *University of Cambridge*

Index 329

1
Muonium

V. G. FIRSOV
Institute for Theoretical and Experimental Physics, Moscow
and
V. I. GOLDANSKII
Institute for Chemical Physics, Moscow

1.1 INTRODUCTION 2

1.2 NUCLEAR PHYSICAL CHARACTERISTICS OF THE μ^+-MESON AND MUONIUM 2

1.3 THE PHYSICO-CHEMICAL BASIS OF MUONIUM INTERACTIONS 6
- 1.3.1 *Formation of muonium during μ^+-meson thermalisation in the medium* 6
- 1.3.2 *The hydrogen-like character of muonium* 8
- 1.3.3 *The main types of muonium interactions and possibilities for their study* 9

1.4 OBSERVATION AND INVESTIGATION OF TRIPLET ATOMIC MUONIUM 16

1.5 DETERMINATION OF ABSOLUTE RATE CONSTANTS FOR CHEMICAL REACTIONS INVOLVING MUONIUM 22
- 1.5.1 *Calculation of rate constants. The method of competing acceptors* 22
- 1.5.2 *Experimental data for some covalent compounds* 24
- 1.5.3 *Muonium reactions in the gas phase; comparison with atomic hydrogen reactions* 26

1.6 TEMPERATURE DEPENDENCE OF THE RATES OF MUONIUM REACTIONS 28
- 1.6.1 *Activation energies for muonium reactions with organic compounds* 28
- 1.6.2 *Phase transition ice–water* 30

1.7 REACTIONS OF MUONIUM WITH AROMATIC UNSATURATED COMPOUNDS 32
- 1.7.1 *The ratio of substitution to addition rate constants for aromatic halogen derivatives* 32
- 1.7.2 *Influence of the structure of organic compounds on their reactivity* 33

1.8 DEPENDENCE OF THE μ^+-MESON POLARISATION IN UNSATURATED COMPOUNDS ON THE LONGITUDINAL MAGNETIC FIELD STRENGTH 35

1.9 SOME PROBLEMS OF μ^+-MESON INTERACTION WITH THE CRYSTAL LATTICE 38

1.10 OTHER TYPES OF MUONIUM INTERACTIONS 42

1.1 INTRODUCTION

An increasing interest in problems spanning two research disciplines is characteristic of the present time. The use of elementary particles in chemical and physico-chemical investigations is an eloquent example of the interconnection between physics and chemistry. The nuclear physical parameters of particles, the specific experimental technique, the possibility of studying the behaviour of single atoms, the essentially short observation time—all these create favourable conditions for obtaining data inaccessible by other methods. The μ^+-meson, an elementary particle producing a single electron hydrogen-like atom, muonium, plays an essential role in such investigations.

Research on the interaction between muonium and matter was started only in recent years. It was triggered by the discovery of non-conservation of parity in π–μ–e decay (1957), by the study of the physical parameters characteristic of this process, as well as by the availability of intense meson beams.

1.2 NUCLEAR PHYSICAL CHARACTERISTICS OF THE μ^+-MESON AND MUONIUM

The μ^+-meson, an elementary antiparticle, is produced by π^+-meson decay in nuclear reactions above the threshold for meson production. The μ^+-meson mass is 206.76 $m_e \approx 1/9\ m_p$, the spin is $\frac{1}{2}$, and the magnetic moment is 8.89 magnetons. It is an essential feature that the μ^+-mesons, produced by π^+-meson decay in flight, because of non-conservation of parity in the $\pi \rightarrow \mu$ decay are polarised longitudinally, i.e. the spin of the emitted μ^+-meson is in the opposite direction to its motion. The μ^+-meson lifetime for decay to a positron, neutrino and antineutrino is 2.2×10^{-6} s. The positron produced by $\mu \rightarrow$ e decay ($\mu^+ \rightarrow e^+ + \nu + \bar{\nu}$) is emitted mainly in the direction of the meson spin, i.e. there is an asymmetry in the angular distribution of the positrons depending on the positron momentum. The energy spectrum of the emitted positrons ranges from zero up to $E_{max} = 53$ MeV/c (the maximum population is at ~ 35 MeV/c). Thus, the non-conservation of space parity results in a dependence of the probability of decay into a certain solid angle dΩ on the angle θ between the directions of motion of the μ^+-meson and the positron, and on the positron energy as well.

Using the theory of the two-component neutrino[1-3] the expression for the angular distribution of the $\mu \rightarrow e$ decay probability R (assuming a small rest mass for the positron compared to its kinetic energy) has the form:

$$R(x, \Omega)\, dx\, d\Omega \approx \frac{x^2}{2\pi} [(3 - 2x) + \xi(1 - 2x) \cos \theta]\, dx\, d\Omega \qquad (1.1)$$

where x is the positron energy given as a fraction of its maximum energy and ξ is a parameter characterising the degree of longitudinal polarisation of the μ^+-meson on $\pi \rightarrow \mu$ decay, i.e. the asymmetry of the angular distribution. In the universal Fermi interaction theory developed by Feynman and Gell-Mann[4], $\xi = -1$ if μ-meson decay is due only to vector and axial-vector interactions (the corresponding constants are equal in magnitude and opposite in sign).

The asymmetry coefficient for positrons with energy x is

$$C(x) = \frac{1 - 2x}{3 - 2x} \qquad (1.2)$$

At the maximum positron energy ($x = 1$) $C = -1$; as x decreases the asymmetry coefficient becomes zero (at $x = \frac{1}{2}$) and then changes its sign as it approaches $+1/3$ (at $x = 0$). The asymmetry coefficient averaged over all the energy spectrum (when expression (1.2) is integrated over x from 0 up to 1) has the value $C = -1/3$ for 100% initial polarisation of the μ^+-mesons ($P = 1$). Under conditions permitting depolarisation of the μ^+-meson spin, the absolute value of the asymmetry coefficient drops to zero, which corresponds to complete isotropy in the angular distribution of the positrons. Variations of the polarisation of the μ^+-mesons in the medium, depending on experimental conditions, reflect their interaction with the medium.

In accelerator experiments one should take into account the kinematic conditions of creation of the meson beam by $\pi \rightarrow \mu$ decays in flight, as these conditions may induce a change in the degree of polarisation of the initial beam. Normalisation of the asymmetry coefficients obtained is best made with a standard target which yields the maximum $| C |$ value, usually graphite or bromoform; this takes account of the difference in the energy spectrum of the recorded positrons. Absolute measurements of the μ^+-meson polarisation in bromoform ($CHBr_3$) have been made[5].

The hypothesis of an atomic species produced containing a μ^+-meson with an electron rotating around it, the system μ^+e^-, was proposed by Landau[2] and Friedman and Telegdi[6] in discussions of possible causes of μ^+-meson depolarisation in substances. By analogy with positronium this atom was called muonium (Mu).

Muonium is an analogue of the hydrogen atom, in a sense one of its lightest isotopes. The energy levels, the electron orbital radius, and the ionisation potential of an isolated muonium atom all coincide with those for atomic hydrogen after allowance for the difference in reduced masses. The difference between the proton and μ^+-meson magnetic moments leads to changes in hyperfine splitting (h.f.s.) levels. Figure 1.1 shows the scheme of the ground state levels of muonium $^2S_{\frac{1}{2}}$ and the energy dependence in a strong magnetic field when the interaction of the electron and the μ^+-meson magnetic

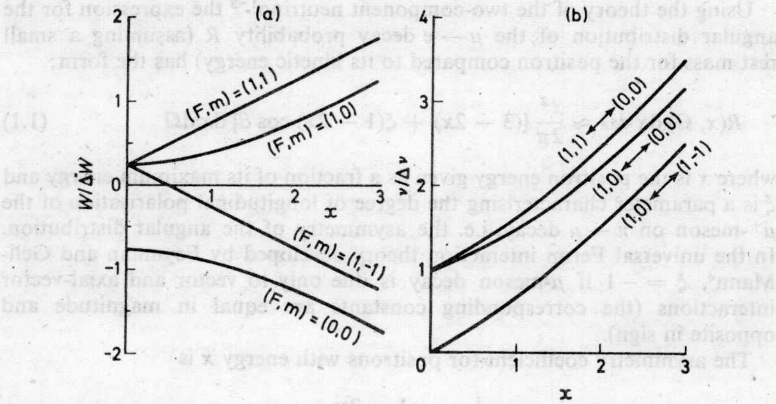

Figure 1.1. (a) Scheme of the h.f.s. energy levels for muonium in the ground state. Strength of the magnetic field created by the meson at the site of the electron ($x = H/H_0$) in units of the critical field H_0 versus the energy in units of the hyperfine splitting magnitude; (b) frequencies of transitions between levels with various quantum numbers m as a function of the magnetic field strength

moments with the external field is large relative to the h.f.s. energy ($x \gg 1$; Figure 1.1a). It also shows the frequencies of transitions between levels with different quantum numbers m as a function of the magnetic field strength (Figure 1.1b).

For longitudinally polarised μ^+-mesons two muonium states are possible, with equal probability, with parallel and antiparallel spin directions of meson and electron. The ratio 1 : 1 is because the spin direction of the μ^+-meson is fixed. The total momentum F of a muonium atom may be zero or 1 ($F = -1$ is absent), and the projection F on to the quantisation axis (m) is 0 and +1, respectively (Table 1.1).

Table 1.1 States of the muonium atom in magnetic fields

	Weak magnetic field ($H \ll H_0$)		*Strong magnetic field* ($H \gg H_0$)		
F	m	*Population*	m_μ	m_e	*Population*
1	-1	0	$-\frac{1}{2}$	$-\frac{1}{2}$	0
1	0	$\frac{1}{4}$	$-\frac{1}{2}$	$+\frac{1}{2}$	0
0	0	$\frac{1}{4}$	$+\frac{1}{2}$	$-\frac{1}{2}$	$\frac{1}{2}$
1	$+1$	$\frac{1}{2}$	$+\frac{1}{2}$	$+\frac{1}{2}$	$\frac{1}{2}$

In the state (1, +1) which is a purely triplet state of muonium $^3S_1$, $m = 1$ with a fifty per cent population, the μ^+-meson spin in the absence of external perturbations retains its initial polarisation; on the other hand harmonic oscillations with frequency $\Delta\nu$, inducing a disturbance of the initial μ^+-meson spin orientation, i.e. depolarisation, occur between states (1,0 and 0,0) (superposition of states $^1S_0$, $m = 0$ and $^3S_1$, $m = 0$ with $\frac{1}{4}$ and $\frac{1}{4}$ populations).

In strong magnetic fields ($H \gg H_0$) when the interaction between the magnetic moments of the meson and electron disappears, the magnetic quantum numbers of the meson (m_μ) and the electron (m_e) are the motion integrals. In this case, corresponding to the Paschen-Back effect, both populated states ($+\frac{1}{2}$, $-\frac{1}{2}$) and ($+\frac{1}{2}$, $+\frac{1}{2}$) retain the initial μ^+-meson polarisation.

The transition (1,0) $\leftrightarrow$ (0,0) is a specific quantum-mechanical phenomenon ensuring the loss of a half of the μ^+-meson polarisation upon formation of muonium. The transition frequency is a function of the h.f.s. interaction energy ΔW (for an isolated atom $\Delta W \approx 1.85 \times 10^{-5}$ eV). The theoretical transition frequency, taking account of the reduced mass of muonium, the radiation corrections due to the μ^+-meson and electron magnetic moments and also the Breit relativistic correction, is[7]

$$\omega_0 = \Delta W/h = 4463.16 \text{ MHz} = 2.804 \times 10^{10} \text{ rad s}^{-1} \quad (1.3)$$

(where ω_0 is the vacuum value).

Thus the characteristic time of spin flip for muonium with antiparallel meson and electron spins is $\tau = 1/\omega_0 \approx 3.6 \times 10^{-11}$ s and the only polarisation retained for a period of $\geqslant 10^{-10}$ s is that connected with the triplet muonium state (parallel spin directions of μ^+-meson and electron).

The role of the external magnetic field is in decoupling of the magnetic moments of the μ^+-meson (μ_μ) and the electron (μ_e) when the magnetic energy of muonium, given by ($\mu_e H - \mu_\mu H$), in the H field is essentially higher than ΔW. The critical condition when the longitudinal magnetic field, relative to the meson spin, is equivalent to that created by the μ^+-meson at the site of the electron, is determined for an individual muonium atom by

$$H_0 = \frac{\Delta W}{2(\mu_e - \mu_\mu)} = 1585 \text{ gauss} \quad (1.4)$$

In this way the concepts of strong and weak magnetic fields acquire a real physical meaning.

Both the method of Larmor precession in a magnetic field and observation of induced transitions between h.f.s. levels in weak and strong magnetic fields can be used for identification, observation and study of muonium. Certainly, other muonium interactions, for instance exchange with the electrons of the environment with opposite spins (conversion), chemical reaction of the muonium when it ceases to be an atomic system, etc., must always be taken into account.

The electron magnetic moment is essential for studies using the Larmor precession of triplet muonium. As the triplet muonium state spin is unity and its magnetic moment is controlled by the vector sum of the electron and meson magnetic moments ($\sim 206\ \mu_\mu$) the Larmor precession frequency is higher than that for a free μ^+-meson by a factor of *ca.* 103. This enables one to distinguish the state of a μ^+-meson in a substance.

The application of a longitudinal magnetic field and the relevant Bohr frequency induces muonium transitions to states with the opposite μ^+-meson spin. Alteration of the static field strength, or of the microwave frequency, gives a typical resonance curve with a maximum corresponding to the transition frequency and an amplitude characteristic of the intensity of the process. Such measurements are possible both with strong and weak magnetic fields, transitions between states $+\frac{1}{2}, +\frac{1}{2}$ and $-\frac{1}{2}, +\frac{1}{2}$ and between $1, +1$, and $0,0$, respectively.

A review[7] gives a very extensive discussion of theoretical and experimental research on the physics of muonium.

1.3 THE PHYSICO-CHEMICAL BASIS OF MUONIUM INTERACTIONS

1.3.1 Formation of muonium during μ^+-meson thermalisation in the medium

In order to clarify the depolarisation processes one must consider the main features of μ^+-meson slowing down. Under the experimental conditions the μ^+-mesons enter the target with energies of tens of MeV and are moderated in the sample to thermal velocities. At high energies ionisation losses predominate for the μ^+-mesons, as is common for other particles of high energies. The contribution from elastic collisions, and also from energy losses due to the excitation of the orbital electrons of atoms and molecules, increases at lower energies. A charge exchange stage[8, 9], involving capture and loss of electrons in successive μ^+-meson collisions, sets in when the velocity of a moderated positively charged ion becomes comparable with that of orbital electrons in the environment ($v \sim v_{at}$). It is essential, owing to the low μ^+-meson mass, that the number of charge exchanges in the Bohr region is large enough. As a moderated μ^+-meson is a deep trap for electrons ($\sim$ 13.5 eV), neutral muonium atoms, which rapidly attain thermal velocities, will be predominant at velocities lower than v_{at}[10].

Detailed analysis of the probability of muonium formation during the slowing down of μ^+-mesons in the medium is possible only provided the cross sections of a number of elementary processes and their energy dependence are known. It is reasonable to consider the limiting case with a low-energy maximum of the cross section for neutral atom production. The 'Öre gap'[11] may be an accepted approximate theoretical model. The lowest energy (E_{min}) of the μ^+-meson at which the muonium production is possible equals the difference between ionisation potentials of the medium molecules and muonium atoms ($\Delta I = I_{med} - I_{Mu}$) since below this threshold electron abstraction is energetically forbidden. Naturally, $E_{min} = 0$, when $I_{med} \leqslant I_{Mu}$. The effective upper boundary (E_{max}) may lie between I_{med} and the first excitation level of the molecules of the medium (E^*). The probability of energy transfer with excitation to higher levels is predominant at higher μ^+-meson energies. Assuming a rectangular energy spectrum of moderated μ^+-mesons the probability of muonium production is

$$P = \frac{E_{max} - E_{min}}{E_{max}} \tag{1.5}$$

Thus, in terms of the Öre model the probability of neutral atom production lies within

$$\frac{I_{Mu}}{I_{med}} > P > \frac{E^* - \Delta I}{E^*}, \quad (1.6)$$

when $I_{med} > I_{Mu}$, and $P \equiv 1$ if $I_{med} \leqslant I_{Mu}$.

The latter condition is fulfilled for most chemical compounds; experimental verification of the model is possible using compounds with high ionisation potential. Table 4.2 lists the calculated values of the probability of muonium production, making use of the Öre model, for inert gases. As the probability of muonium production is minimum for helium, it is most suitable for comparison between the model and experiment; the residual μ^+-mesons can attain thermal energy as positive ions.

Table 1.2 Energies of the first electron excitation level (E^*), ionisation potentials (I), limiting probabilities of muonium production (P) using the Öre gap model for inert gases

Element	E^*(eV)	I(eV)	P_{max}	P_{min}
He	19.82	24.58	0.55	0.44
Ne	16.62	21.56	0.63	0.51
Ar	11.55	15.76	0.86	0.81
Kr	9.91	14.00	0.97	0.95
Xe	8.31	12.13	1	1

Naturally, chemical interactions involving muonium, making the process more complicated, must be absent. According to Hughes *et al.*[12–17], the polarisation of μ^+-mesons in oxygen-free argon is 0.08 ± 0.15, and the probability of muonium production is close to unity. This is not at variance with the model but the accuracy is insufficient for an unambiguous conclusion. Extensive theoretical and experimental research has been reported for the similar case of proton moderation in argon[18–21], showing that the probability of neutral hydrogen production is ~ 0.9. As to the μ^+-mesons, the experimental data reported are too sparse for precise determination of the energy boundaries for muonium production.

The Öre gap model is approximate as it does not take into account the production of neutral atoms above the gap boundary, as well as particle injection into the low-energy region in the form of an ion. However, in many cases this model gives plausible results. Detailed analysis of the Öre gap model and its application to positronium production and comparison with the extensive experimental data can be found in the book by Goldanskii[22].

The time of slowing down of μ^+-mesons and muonium to thermal energies (thermalisation) naturally depends on the masses of the atoms of the medium and on the medium density. For a condensed phase, as well as for gases at pressures above 50–100 atm, it is found to be 10^{-12}–10^{-13} s[23–25]. This can be compared to the time of positronium moderation in the medium (10^{-11}–10^{-12} s)[22,26–28]. As would be expected the moderation time is longer for the lighter positronium. It is essential that the time of muonium moderation is shorter than that of its characteristic depolarisation time ($\tau = 1/\omega_0 \approx 3.6 \times 10^{-11}$ s). Thus, since before the μ^+-meson is stopped, its depolarisation is negligible; the multiple charge exchanges during moderation do not change the μ^+-meson spin. Consequently, the formation of an ortho- or