

Contents

Preface	v
1 Introduction	1
1-1 What Is Optical Pumping?	1
1-2 The Early Experiments in Optical Pumping	1
1-3 Applications of Optical Pumping	2
2 The Optical Pumping Experiment	4
2-1 A Simple Example	4
2-2 Magnetic Resonances	7
2-3 A More General Example	8
3 The Dynamics of Optical Pumping	10
3-1 Pumping and Relaxation in Ground States	10
3-2 Orientation and Alignment	12
3-3 Optical Pumping of Excited States	14
4 Optical Resonance Radiation	18
4-1 Resonance Fluorescence and Line Fluorescence	18
4-2 Absorption of Optical Resonance Radiation	20
4-3 Broadening of Optical Resonance Lines	21
4-4 Self-reversal	22
5 Magnetic Interactions: Zeeman and Hyperfine Splitting	24
5-1 The Hamiltonian	24
5-2 Energy Levels in Weak, Strong, and Intermediate Magnetic Fields	25
5-3 Determination of Hyperfine Constants, g-Factors, and Nuclear Spins by Optical Pumping	26
6 Electric Interactions: Nuclear Electric Quadrupole and Stark Splitting	29
6-1 Nuclear Electric Quadrupole Coupling Constants	29
6-2 Electric Field Gradients in Atoms	30
6-3 Determination of Nuclear Electric Quadrupole Moments	30

6-4	Energy Levels	31
6-5	Stark Effect	32
7	Magnetic Resonance Detection	34
7-1	The Magnetic Resonance Condition	34
7-2	Optical Detection of Magnetic Resonances	36
7-3	Transmitted Radiation through Vapor Cells	37
7-4	Scattered Radiation from Vapor Cells; Change in Polarization	37
7-5	Scattered Radiation from Vapor Cells; Change in Frequency and Selective Reabsorption	37
7-6	Crossed Light Beams	38
7-7	Modulation of Optical Pumping Light	39
7-8	Spin-Exchange Method	39
7-9	Scattered Radiation from Atomic Beams	39
7-10	Deflection of Optically Pumped Atomic Beams	40
7-11	The Bloch-Siegert Shift	40
8	Phenomenological Description of Oriented Systems	42
8-1	Orientation As a Bulk Magnetization	42
8-2	Equations of Motion with Optical Pumping	43
9	Energy Level Crossing Experiments	47
9-1	Principle of the Experiment	47
9-2	Applications	49
10	Spin Exchange	50
10-1	Magnetic Resonance Detection by Spin Exchange	50
10-2	Mechanism for Spin Exchange	52
11	Pressure Shift of the Hyperfine Interaction	54
11-1	Origin of the Pressure Shift	54
11-2	Experimental Results	55
11-3	Theoretical Calculations of the Pressure Shift	56
12	Spin-Relaxation Times	58
12-1	Measurement of Spin-Relaxation Times	58
12-2	Diffusion Coefficients and Spin-Disorientation Cross Sections	59
12-3	The Spin-Relaxation Mechanism	60
12-4	Spin-Disorientation in Molecular Buffer Gases	61
12-5	Spin-Disorientation in Electric Dipolar Buffer Gases	61
13	Chemical Applications of Optical Pumping	63
13-1	Atomic Structure	63
13-2	Intermolecular Interactions	64

CONTENTS	ix
13-3 Chemical Kinetics	64
13-4 Molecular Structure	66
14 Optical Pumping in Solids	68
14-1 Fluorescence in Solids	68
14-2 Optical Detection of Magnetic Resonances in Solids	69
14-3 Application to Masers	70
Bibliography	71

Reprints

1. J. Brossel and A. Kastler, La Détection de la Résonance Magnétique des Niveaux Excités: L'effet de Dépolarisation des Radiations de Résonance Optique et de Fluorescence, *Compt. Rend.* **229**, 1213–1215 (1949) 87
2. A. Kastler, Quelques Suggestions concernant la Production Optique et la Détection Optique d'une Inégalité de Population des Niveaux de Quantification Spatiale des Atomes. Application à l'Expérience de Stern et Gerlach et à la Résonance Magnétique, *J. Phys. Radium* **11**, 255–265 (1950) 90
3. A. Kastler, Optical Methods of Atomic Orientation and of Magnetic Resonance, *J. Opt. Soc. Am.* **47**, 460–465 (1957) 101
4. J. Brossel and F. Bitter, A New 'Double Resonance' Method for Investigating Atomic Energy Levels. Application to Hg 3P_1 , *Phys. Rev.* **86**, 308–316 (1952) 107
5. W. B. Hawkins, Orientation and Alignment of Sodium Atoms by Means of Polarized Resonance Radiation, *Phys. Rev.* **98**, 478–486 (1955) 116
6. W. Franzen and A. G. Emslie, Atomic Orientation by Optical Pumping, *Phys. Rev.* **108**, 1453–1458 (1957) 125
7. R. H. Dicke, The Effect of Collisions upon the Doppler Width of Spectral Lines, *Phys. Rev.* **89**, 472–473 (1953) 131
8. H. G. Dehmelt, Modulation of a Light Beam by Precessing Absorbing Atoms, *Phys. Rev.* **105**, 1924–1925 (1957) 133
9. W. E. Bell and A. L. Bloom, Optical Detection of Magnetic Resonance in Alkali Metal Vapor, *Phys. Rev.* **107**, 1559–1565 (1957) 135
10. J. N. Dodd and G. W. Series, Theory of Modulation of Light in a Double Resonance Experiment, *Proc. Roy. Soc. (London) Ser. A* **263**, 353–370 (1961) 142

11. P. A. Franken, Interference Effects in the Resonance Fluorescence of 'Crossed' Excited Atomic States, *Phys. Rev.* **121**, 508–512 (1961) 161
12. P. Thaddeus and R. Novick, Optical Detection of Level Crossing in the (5s 5p) 3P_1 State of Cd¹¹¹ and Cd¹¹³, *Phys. Rev.* **126**, 1774–1780 (1962) 166
13. E. M. Purcell and G. B. Field, Influence of Collisions upon Population of Hyperfine States in Hydrogen, *Astrophys. J.* **124**, 542–549 (1956) 173
14. H. G. Dehmelt, Spin Resonance of Free Electrons Polarized by Exchange Collisions, *Phys. Rev.* **109**, 381–385 (1958) 181
15. L. W. Anderson, F. M. Pipkin, and J. C. Baird, Jr., Hyperfine Structure of Hydrogen, Deuterium, and Tritium, *Phys. Rev.* **120**, 1279–1289 (1960) 186
16. P. L. Bender, Effect of Hydrogen-Hydrogen Exchange Collisions, *Phys. Rev.* **132**, 2154–2158 (1963) 197
17. L. C. Balling and F. M. Pipkin, Spin Exchange in a Cesium-Electron System, *Phys. Rev.* **136**, A46–A53 (1964) 202
18. R. Herman, Theory of Spin Exchange between Optically Pumped Rubidium and Foreign Gas Nuclei, *Phys. Rev.* **137**, A1062–A1065 (1965) 210
19. M. Arditi and T. R. Carver, Pressure, Light, and Temperature Shifts in Optical Detection of 0–0 Hyperfine Resonance of Alkali Metals, *Phys. Rev.* **124**, 800–809 (1961) 214
20. G. A. Clarke, Effects of Helium Buffer Gas Atoms on the Atomic Hydrogen Hyperfine Frequency, *J. Chem. Phys.* **36**, 2211–2216 (1962) 224
21. H. G. Dehmelt, Slow Spin Relaxation of Optically Polarized Sodium Atoms, *Phys. Rev.* **105**, 1487–1489 (1957) 230
22. W. Franzen, Spin Relaxation of Optically Aligned Rubidium Vapor, *Phys. Rev.* **115**, 850–856 (1959) 233
23. A. L. Bloom, Spin Relaxation and Line Width in Alkali Metal Vapors, *Phys. Rev.* **118**, 664–667 (1960) 240
24. R. A. Bernheim, Spin Relaxation in Optical Pumping, *J. Chem. Phys.* **36**, 135–140 (1962) 244
25. R. J. McNeal, Disorientation Cross Sections in Optical Pumping, *J. Chem. Phys.* **37**, 2726–2727 (1962) 250
26. R. J. McNeal, R. A. Bernheim, R. Bersohn, and M. Dorfman, Optical Pumping and Chemical Reactions, *J. Chem. Phys.* **40**, 1678–1683 (1964) 252
27. H. G. Dehmelt and K. B. Jefferts, Alignment of the H₂⁺ Molecular Ion by

Selective Photodissociation. I., <i>Phys. Rev.</i> 125 , 1318–1322 (1962)	258
28. F. Varsanyi, D. L. Wood, and A. L. Schawlow, Self Absorption and Trapping of Sharp-Line Resonance Radiation in Ruby, <i>Phys. Rev. Letters</i> 3 , 544–545 (1959)	263
29. S. Geschwind, R. J. Collins, and A. L. Schawlow, Optical Detection of Paramagnetic Resonance in an Excited State of Cr^{3+} in Al_2O_3 , <i>Phys. Rev. Letters</i> 3 , 545–548 (1959)	264
30. J. Brossel, S. Geschwind, and A. L. Schawlow, Optical Detection of Paramagnetic Resonance in Crystals at Low Temperature, <i>Phys. Rev. Letters</i> 3 , 548–549 (1959)	267
31. N. A. Kurnit, I. D. Abella, and S. R. Hartmann, Observation of a Photon Echo, <i>Phys. Rev. Letters</i> 13 , 567–568 (1964)	269
Index	271