ALAN CORNEY

ATOMIC AND LASER SPECTROSCOPY

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Preface

It is now over forty years since *Resonance radiation and excited atoms* by A.C.G. Mitchell and M.W. Zemansky, (Cambridge University Press, 1934), first appeared. Since then there have been many advances, and in teaching for the Final Honour School of Physics at Oxford I have often felt the need of an up-to-date account of the progress that has been made in the field of optical physics, particularly during the last quarter of a century. This volume is an attempt to fill that need. The first five chapters of the book prepare the foundations of atomic physics, electromagnetism, and quantum mechanics which are necessary for an understanding of the interaction of electromagnetic radiation with free atoms. The application of these concepts to processes involving the spontaneous emission of radiation is then developed in Chapters 6, 7, and 8 while stimulated transitions and the properties of gas and tunable dye lasers form the subject matter of Chapters 9 to 14. The last four chapters are concerned with the physics and applications of resonance fluorescence, optical double-resonance, optical pumping and atomic beam magnetic resonance experiments. It proved remarkably difficult, once the manuscript had been completed, to find a title which accurately described the content of the book. *Atomic and laser spectroscopy* is intended to indicate the wide range of the material treated within, but it is slightly misleading in the sense that I have been concerned not so much with an explanation of the gross spectra of atoms but rather with demonstrating how the techniques of atomic spectroscopy and lasers have been applied to a wide range of problems in atomic and molecular physics. In a sense therefore this text is complementary to those well-known books *Atomic spectra* by H.G. Kuhn (2nd edn Longmans 1970) and *Elementary atomic structure* by G.K. Woodgate (McGraw-Hill 1970).

This volume is an extended version of a set of lectures on atomic physics given to third year undergraduates at the

University of Oxford. By this stage these undergraduates have already completed introductory courses in electromagnetism, atomic physics, and quantum mechanics, courses which I regard as prerequisite for a full understanding of this book. This text should then provide an excellent basis for an advanced undergraduate course in atomic physics. However, the later sections of many chapters are more suitable for work at the graduate level and for this reason the book should also prove valuable to those engaged in research in the fields of atomic physics, lasers, astrophysics, and physical chemistry. Since this book is intended mainly for experimentalists, I have not attempted to develop the theory of the interaction of atoms and radiation in a rigorous manner but have made use of classical or semi-classical calculations whenever possible. This approach has the advantage of simplicity and moreover it often yields great insight into the physics of the problems under discussion. For those students who wish to progress to a more rigorous development of the theory I strongly recommend *The quantum theory of light* by R. Loudon, (Clarendon Press, Oxford 1973).

Throughout the book the equations are given in SI units although readers who prefer to work in the c.g.s system can make the transposition in most cases by removing factors of $(1/4\pi\varepsilon_0)$ or $(\mu_0/4\pi)$ from the expressions given. The problems at the ends of the chapters form an important part of the book since they are designed to develop a feel for the order of magnitude of the quantities involved and to illustrate and extend the topics discussed in the main text. The bibliography at the end of each chapter includes only those papers which I consider most important for the particular topics under discussion. However, these references, together with the suggestions for further reading, should enable the student to progress easily to the current literature in most fields.

In spite of the length of the book some important topics had to be omitted; these include photodissociation, photoionization, and free-free transitions. Perhaps more important still it was not possible to include a discussion of the non-resonant scattering of light, the refractive index of gases, and the non-linear susceptibility of atomic vapours. Similarly, although tensor operators are briefly mentioned, it was felt that a complete discussion of radiation processes in general and resonance experiments in particular using irreducible tensor operators was beyond the level of this book.

Over the years my understanding of atomic and laser spectroscopy has benefited considerably from discussions with my colleagues and students in the Clarendon Laboratory. I am especially grateful to Professor G.W. Series who first aroused my interest in atomic physics and under whose expert and enthusiastic guidance I performed my first research. I wish to thank Dr C.E. Webb for advice on laser population mechanisms and for his constructive criticism of Chapters 11-13, and Dr G.K. Woodgate who read the complete manuscript. Many of the improvements in the final draft are due to his invaluable assistance. I also want to thank Professor J.N. Dodd and the staff members of the Department of Physics, University of Otago, New Zealand for providing the hospitality and seclusion which enabled me to complete a substantial part of this book. Of my many antipodean friends I am particularly indebted to Drs C.G. Carrington and D.M. Warrington who read the manuscript in an incomplete state and made many helpful suggestions.

Finally I thank my wife not only for her patience and encouragement but also for computer programming and curve plotting, for tracing the line drawings of most of the figures, and for a true labour of love in typing the whole manuscript.

A.C.

Clarendon Laboratory, Oxford.
July 1976.

Contents

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Chapter 15 continued.....

1
Introduction

The purpose of this book is to present a unified account of the exciting developments in the field of quantum electronics which have occurred during the last two decades. Following introductory chapters on electromagnetism and quantum mechanics, the basic processes involving the interaction of radiation and atoms are discussed in some detail. We believe that a thorough understanding of this material is essential for later chapters.

However, in order that the reader shall not lose sight of our ultimate objective, we present here a brief historical introduction showing how our understanding of light and matter has developed since the end of the nineteenth century. It is hoped that this will allow the student to set the material which follows in the proper context and will give him or her an appreciation of the beauty and fascination of the field of optical physics.

1.1. Planck's radiation law

By the end of the nineteenth century the development of the wave theory of light, commenced by Young and Fresnel, seemed to have reached its culmination in the brilliant theoretical work of Maxwell. In another branch of physics the theory of heat leading on to the kinetic theory of gases and to statistical mechanics as developed by Clausius, Boltzmann, Maxwell, and Gibbs also seemed to be complete in most of its essential points. This encouraged Michelson to write in 1899:

'The more important fundamental laws and facts of physical science have all been discovered, and these are so firmly established that the possibility of their ever being supplanted in consequence of new discoveries is exceedingly remote.'

There was, however, an unresolved discrepancy in the theory of black-body radiation. In fact the solution of this apparently minor problem by Planck (1901) was to be the starting point of the quantum theory and of our present understanding of the structure of atoms and molecules.

Following the work of Kirchhoff on the connection between emission and absorption coefficients, it had been proved that the radiation inside a totally enclosed cavity maintained at a uniform temperature was a function of the temperature alone and was identical with the radiation which would be emitted by a perfectly black body at the same temperature. The spectral distribution of the radiation had been investigated experimentally and it was found that the intensity increased slowly with decreasing frequency until it reached a peak, after which it decreased very rapidly (Fig.1.1). However, all attempts to derive an equation giving the intensity as a function of frequency had failed. The most convincing approach made by Rayleigh and Jeans on the basis of classical thermodynamics gave the result

$$\rho(\omega) = \frac{\omega^2}{\pi^2 c^3} kT , \tag{1.1}$$

where $\rho(\omega)\, d\omega$ represents the energy of radiation per unit volume in the angular frequency range between ω and $\omega + d\omega$. The term kT in equation (1.1) comes from the equipartition of energy applied to a linear oscillator which is assumed to act as the emitter or absorber of radiation of frequency $\omega/2\pi$. We see from equation (1.1) and Fig. 1.1 that the intensity predicted by the Rayleigh-Jeans law increases indefinitely at higher frequencies, the well-known ultraviolet catastrophe, in complete contradiction with the experimental results.

Planck realized that the difficulty lay in the principle of equipartition which could only be circumvented by a complete departure from classical mechanics. He postulated that an oscillator of frequency $\omega/2\pi$, instead of being able to assume all possible energy values, could exist only in one of a set of equally spaced energy levels having the values 0, $\hbar\omega$, $2\hbar\omega$, ... , $m\hbar\omega$, ... m is an integer and $\hbar = h/2\pi$, where h is a constant now known as Planck's constant.