MOLECULAR BEAMS

BY

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PREFACE

MOLECULAR-BEAM experiments have for many years been among the most fruitful sources of fundamental information about molecules, atoms, and nuclei. Some of the early experiments provided direct experimental evidence for spatial quantization and for the spin of the electron. More recent experiments have led to such important discoveries as the anomalous magnetic moments of protons and neutrons, the deuteron quadrupole moment with its implication of a nucleon tensor interaction, the Lamb shift in the fine structure of atomic hydrogen with its quantum electrodynamic implications, the anomalous magnetic moment of the electron, the existence of nuclear octupole moments, and many others. In addition molecular-beam experiments have produced a wealth of molecular, atomic, and nuclear data including an array of nuclear spin, magnetic moment, and quadrupole moment determinations which, among other things, have provided much of the initial evidence for nuclear shell models.

Despite the scientific importance of the subject, there have been remarkably few books on molecular beams. As discussed in Chapter I, a few review articles and brief monographs have been written but only one detailed book has been published. That book, by R. Fraser (FRA 31), was written twenty-five years ago, prior to almost all of the important discoveries listed in the preceding paragraph.

It is my hope that this book will satisfy the need for a detailed, consistent, and up-to-date discussion of the subject of molecular beams. Although the entire subject is discussed in the present book, experiments prior to 1930 are dealt with more briefly since they are described in greater detail in Fraser's book (FRA 31).

I wish to express my appreciation to the many workers with molecular beams who have supplied me with information and help in the preparation of this book. Special thanks are due to Professors I. I. Rabi, P. Kusch, and J. R. Zacharias and to Drs. H. Kolsky and H. Lew. I should also like to thank John Wiley & Sons for their permission to use some material, especially in Chapter III, from my book on *Nuclear Moments* published by them, and the John Simon Guggenheim Memorial Foundation for the fellowship I held at Oxford University during the completion of this book. Finally, I wish to express special appreciation to my wife, Elinor Ramsey, for her help in making available the time during which the book was written, and to my secretary, Phyllis Brown, for her extensive and excellent help in the preparation of the manuscript.

N. F. R.

Harvard University
Cambridge, Massachusetts
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CONTENTS

I. MOLECULAR AND ATOMIC BEAMS

II. GAS KINETICS

III. INTERACTION OF A NUCLEUS WITH ATOMIC AND MOLECULAR FIELDS

IV. NON-RESONANCE MEASUREMENTS OF ATOMIC AND NUCLEAR MAGNETIC MOMENTS

V. MOLECULAR-BEAM MAGNETIC RESONANCE METHODS

IX. ATOMIC MOMENTS AND HYPERFINE STRUCTURES

X. ELECTRIC DEFLEXION AND RESONANCE EXPERIMENTS

XI. NUCLEAR ELECTRIC QUADRUPOLE MOMENTS

XII. ATOMIC FINE STRUCTURE

XIII. MOLECULAR-BEAM DESIGN PRINCIPLES

XIV. MOLECULAR-BEAM TECHNIQUES

APPENDIXES

I

MOLECULAR AND ATOMIC BEAMS

I. 1. Introduction

THE subject of molecular beams is the study of directed beams of neutral molecules at such low pressures that the effects of molecular collisions are for most purposes negligible. The earliest molecular-beam experiments were those of Dunoyer (DUN 11). He used an apparatus illustrated in Fig. I. 1. A glass tube 20 cm long was divided into three separately evacuated compartments. Some sodium introduced into the first chamber was heated until it vaporized, whereupon a deposit of sodium appeared in the third chamber. The deposit was of the form to be expected on the assumption that the Na atoms travelled in straight lines in the evacuated tube.

Subsequent to Dunoyer's pioneering work, many different experiments based on the molecular-beam methods have been performed. Particularly significant from the point of view of the development of molecular-beam techniques have been the experiments of Stern and his collaborators at Hamburg in the nineteen-twenties and early thirties. The papers of Stern and Knauer in 1926 (STE 26, KNA 26, KNA 29) indeed laid down many of the principles of molecular-beam techniques which have been followed even to the present time.

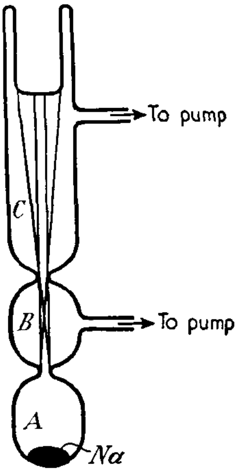

FIG. I. 1. Schematic diagram of Dunoyer's original atomic-beam apparatus. *A* is the source chamber, *B* the collimator chamber, and *C* the observation chamber (DUN 11).

Precision measurements of nuclear, molecular, and atomic properties began in 1938 with the introduction of the molecular-beam resonance method by Rabi and his associates (RAB 38, KEL 39). At first, the resonance method was applied to the measurement of nuclear magnetic moments only, but it was soon extended as a more general technique of radiofrequency spectroscopy initially by Kellogg, Rabi, Ramsey, and Zacharias (KEL 39*a*) in applications to molecules and later by Kusch and Millman (KUS 40) in the study of atomic states. Since 1938 the history of molecular beams has dominantly been the history of the resonance method. As discussed in subsequent chapters, important improvements and extensions of the molecular-beam resonance method have been made by Lamb (LAM 50*a*), Ramsey (RAM 50*c*), Kusch (KUS 51), Rabi (RAB 52), Zacharias (ZAC 53), Lew (WES 53), and others. However, since 1938, most molecular-beam researches have been applications of the resonance technique to physical measurements. The molecular-beam laboratories at Columbia University, Massachusetts Institute of Technology, and Harvard University have been particularly effective in measuring with the resonance method many varied properties of nuclei, atoms, and molecules.

The developments in molecular beams prior to 1937 have been described in detail by Fraser in two different books (FRA 31, FRA 37). Since that time no book devoted exclusively to molecular beams has been published. However, considerable discussion of molecular beams is given in the nuclear moment books of Kopfermann (KOP 40) and Ramsey (RAM 53*b*, RAM 53*e*) and in various review articles (HAM 41, BES 42, KEL 46, EST 46, KUS 50, RAM 50, RAM 52). In accordance with existing conventions as to the scope of the subject molecular beams, the present book will be limited to beams of electrically neutral molecules. Ion beam experiments have been discussed extensively by Massey (MAS 50, MAS 52), Burhop (MAS 52), and others.

I. 2. Typical molecular-beam experiment

Although molecular-beam experiments and apparatus vary greatly according to their applications, they also have many features in common. This is especially true of the experiments in recent years, which have almost exclusively been by the molecular-beam resonance method. For this reason a general description of a typical molecular-beam resonance apparatus is given in this section as an introduction to the more detailed considerations which follow in subsequent chapters.

A photograph of a typical apparatus is shown in Fig. I. 2. A schematic diagram of the apparatus is shown in Fig. I. 3. The molecules emerge from a source S into an evacuated source chamber. The source may be either a heated oven or a small chamber cooled with liquid nitrogen, depending on the vapour-pressure properties of the substance being investigated. The source shown in Fig. I. 3 is for the study of gaseous hydrogen and is cooled with liquid air. The pressure inside the source is about 6 mm Hg. A typical source slit width is 0·015 mm and height about 0·8 cm. Sometimes the slits are a few millimetres long to assure that a larger fraction of the emerging molecules goes in the direction of the detector. The molecules then pass through a separately pumped separating chamber and into the main chamber. The purpose of the separating chamber is to aid in the attainment of a better vacuum in the main chamber. Typical pressures are 2×10^{-5} mm in the separating chamber, and 5×10^{-7} mm in the main chamber. These pressures are sufficiently low for most of the molecules to travel the entire length of the apparatus without being subjected to collisions.

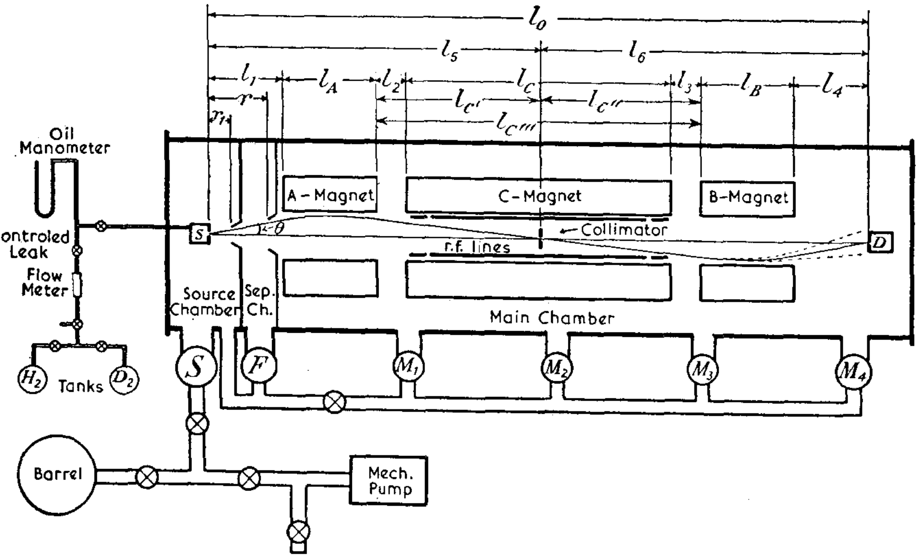

FIG. I. 3. Schematic diagram of a molecular-beam apparatus. The transverse beam displacements are much exaggerated (KOL 50).

The molecules are detected in the main chamber by any of several different kinds of detector described in Chapter XIV. Two of the most frequently used detectors are the surface ionization detector and the Pirani detector. The former consists of a heated wolfram (tungsten) wire of about 0·015 mm diameter. If an atom, such as potassium, with a low ionization potential (4·3 eV for K) strikes the heated tungsten wire, whose work function is 4·5 eV, the valence electron goes into the tungsten and the atom evaporates from the wire as a positive ion. The wire is surrounded by a negatively charged cylinder and the positive current to the cylinder is determined, thus providing a measurement of the beam intensity. The Pirani detector, used with non-condensable gases like H_2, consists of a small volume chamber (7·5 cm × 0·5 cm × 0·06 cm in a typical case) along whose length are stretched one or more thin platinum strips (typically 7·5 cm × 0·05 cm × 0·0001 cm). The beam is admitted to the Pirani cavity through a long narrow channel (2·5 cm long, 0·8 cm high, and 0·015 cm wide in a typical case).

The purpose of the long channel is to make it much easier for the directed beam molecules from the source to enter the cavity than for the randomly directed molecules inside the cavity to emerge; in this way a given number of incident molecules produces a much greater pressure inside the Pirani cavity than would be the case in the absence of the channel. A current is passed through the platinum strips to heat them about 100° C above the temperature of the cavity walls. The pressure changes in the cavity from the variations of beam intensity then lead to changes in the thermal conduction of the gas in the cavity and hence to variations of the temperature of the strips. These variations can be measured externally by noting the changes in the resistances of the wires. The resistances are usually measured by including the Pt strips as one or more arms of a Wheatstone bridge. To minimize effects from thermal and vacuum fluctuations two identical detectors are ordinarily used, their strips are placed in opposite arms of the bridge, and only one of the detectors is exposed to the beam. In this way most of the undesired fluctuations are partially cancelled. The detector in Fig. I. 3 is a Pirani detector.

The beam is defined by means of a collimating slit of about the same width as the source and detector. The collimator is usually placed approximately in the middle of the apparatus for reasons discussed in Chapter XIII. When the collimator is in place, the beam is fully defined and the full beam intensity will be measured by the detector only when the source, collimator, and detector are in line.

In Fig. I. 3 the molecules pass through two regions of inhomogeneous magnetic field produced by two magnets usually designated by the letters A and B and often called the deflecting and refocusing magnets. Fig. I. 4 is a photograph of a typical one of these magnets for producing an inhomogeneous magnetic field. Electrically neutral molecules or atoms which possess magnetic moments will be deflected by such inhomogeneous magnetic fields (in a uniform magnetic field there would be no net translational force since the forces on the north and south poles would be equal and opposite but in an inhomogeneous field one of these forces exceeds the other). Consequently, the molecules which in the absence of the field would have gone straight to the detector (as shown in the straight line of Fig. I. 3) will be deflected by the field so that they do not even pass through the collimator. On the other hand some molecules which in the absence of the field would have missed the detector will be deflected so as to pass through the collimator as shown by the curved line in Fig. I. 3; the deflexion shown in Fig. I. 3 is, of