APPLIED ATOMIC COLLISION PHYSICS

Editors: H. S. W. MASSEY, E. W. McDANIEL, and B. BEDERSON

Volume 3

Gas Lasers

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E. W. McDANIEL and W. L. NIGHAN

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Treatise Preface

Research in atomic physics and especially in the physics of atomic collisions has developed at an explosive rate since the Second World War. The high rate of increase of knowledge of atomic collision processes has been of great value in many applications to pure and applied physics and chemistry. For the full understanding of the physics of planetary and stellar atmospheres, including those of the earth and the sun, detailed knowledge is required of the rates of a great variety of atomic and molecular reactions. Gas lasers depend for their operation on atomic collision processes of many kinds, and a knowledge of the corresponding reaction rates is important for laser design. The release of energy by controlled nuclear fusion offers a possibility of an effectively infinite source of power in the future. Many aspects of the complex techniques involved are affected by atomic reactions. Again there are many applications of collision physics to the study of condensed matter.

These major activities have expanded rapidly at a rate which has been accelerated by the availability of data and understanding from atomic collision physics. There are many smaller areas which depend on this subject.

In these five volumes we planned to give an account of the wide range of applications which are now being made, as well as the additional requirements for further applications. Volume 1 deals with applications to atmospheric and astrophysics, Volume 2 to controlled fusion, Volume 3 to laser physics, and Volume 4 to condensed matter. Volume 5 includes various special applications.

In all cases the emphasis is on the discussion of these applications and the atomic physics involved therein. However, sufficient background is provided to make clear what has been achieved and what remains to be done through further research in collision physics.

We are much indebted to Academic Press for the ready assistance they have afforded us at all times.

H. S. W. MASSEY
E. W. MCDANIEL
B. BEDERSON

Preface

This volume of "Applied Atomic Collision Physics" deals with gas lasers, a subject of research activity that has significantly influenced the direction and scope of atomic and molecular physics in recent years. Presented in Chapter 1 is a historical summary of gas laser developments and a description of the basic operating principles of major gas laser types. This introductory chapter provides an effective background and overview for the content of this volume. Fundamental processes are treated in Chapters 2–7, including various aspects of charged particle kinetics and vibrational and rotational energy exchange, subjects of considerable importance to the operation of gas lasers. Specific topics relevant to the operation of IR molecular lasers and UV/visible excimer lasers are covered in Chapters 8–14, with particular emphasis placed on electrically excited laser media.

The topics covered in this volume complement one another unusually well, as is evidenced by considerable cross-referencing between chapters of the works of our contributing authors. This factor, along with the diligent efforts of the authors to update their already comprehensive references to the last possible moment in production, should make our volume on gas lasers especially useful to the reader interested in this exciting and important application of applied atomic collision physics.

WILLIAM L. NIGHAN

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Introduction and Overview

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I. Introduction to Gas Lasers

A. The Laser Revolution

Over the past 25 years the laser has developed from a laboratory curiosity into a practical tool for addressing a variety of problems and applications. The study of molecules and their interactions has already been transformed in a revolutionary fashion. At the same time, our increasing knowledge of the spectroscopic and collisional properties of excited atoms and molecules has led to the discovery and development of many new types of lasers.

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In many ways the laser is an intrinsically new technological development. No other source of ultraviolet, visible, or infrared radiation combines the properties of high energy density, directivity, and spectral purity. These properties are of value for many of the applications we now have in mind, but the laser is so young that little progress has been made in identifying new processes and phenomena for which no analogies exist in previous technology. Some important applications have already been realized, including communication, laboratory research, microsurgery, microfabrication, and, on a macroscopic scale, laser welding. Future applications include laser isotope separation, laser fusion, and laser controlled chemistry. In addition, numerous military applications of lasers, both realized and prospective, have been an effective spur to laser development.

B. Simplified Laser Concepts

The foundation of the laser, the concept of stimulated emission of radiation, was provided by Einstein (1917) in his description of the statistical mechanics of blackbody radiation at thermal equilibrium. As a simple example, consider the well-used case of a two-level atom. Let $|1\rangle$ and $|2\rangle$ be the nondegenerate ground and excited states of our hypothetical atom. We consider three types of processes involving these two states and photons with energy ($h\nu$) equal to the energy separation ($\Delta\mathscr{E}$) between the two states: absorption, fluorescence, and stimulated emission. These three processes are illustrated in Fig. 1a and in the following equations:

$$|1\rangle + h\nu \rightarrow |2\rangle, \tag{1}$$

$$|2\rangle \rightarrow |1\rangle + h\nu, \tag{2}$$

and

$$|2\rangle + h\nu \rightarrow |1\rangle + 2h\nu. \tag{3}$$

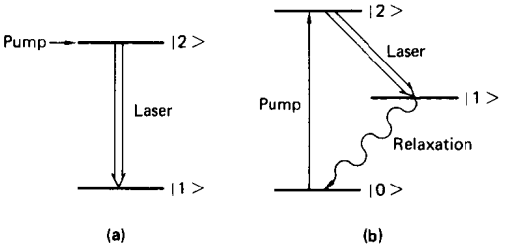

Fig. 1. Simplified laser level diagram: (a) two-level atom, (b) three-level atom.

In a sample containing both ground and excited states the processes of absorption, fluorescence, and stimulated emission will all be occurring. The most common case is where the populations of the ground and excited states are in thermal equilibrium, which we express using the Boltzmann relation

$$N_2 = N_1 \exp(-\Delta\mathscr{E}/kT), \tag{4}$$

where N_1 and N_2 are the populations (per unit volume) of the ground and excited states, respectively, k is the Boltzmann constant, and T is the temperature. Because the cross sections for absorption and stimulated emission are the same, the probability that a photon will be amplified, compared to the probability that it will be absorbed, is determined by the relative values of the populations of the excited and ground states. This is used to calculate the net gain, or absorption, per unit length through the equation

$$\text{gain} = \sigma_{se}(N_2 - N_1), \tag{5}$$

where σ_{se} is the stimulated emission (or absorption) cross section. Thus to make an amplifier (gain >0) it is necessary to devise a means to place the majority of the atoms in the excited state ($N_2 > N_1$). Such a situation appears to violate the requirements of statistical mechanics as expressed in Eq. (4), if the temperature is restricted to be positive. This violation is removed if we remember that Eq. (4) assumes that thermal equilibrium has been achieved in the sample, which need not be the case. Thus we must seek to identify schemes by which this population inversion can be achieved. This search forms the basis of the majority of the contributions to this volume.

For two-level systems the most usual means for preparing a population inversion is to begin with a laser medium in which the lasing species is initially absent. The excited state is to be selectively produced by a scheme such as chemical reaction or photodissociation. Such a population inversion is necessarily of finite duration, unless the lower level $|1\rangle$ is rapidly and selectively removed by chemical reaction. Typically, we obtain a "self-terminating" laser in which the process of stimulated emission depletes the population inversion, eliminating the amplification. Gordon *et al.* (1954) used this type of scheme to demonstrate the first laser, by physically separating the upper and lower levels in a magnetic field.

Most lasers utilize three or more levels in the lasing species. This situation is illustrated in Fig. 1b. In this case a population inversion can be much easier to achieve. We need only find some collisional processes that excite $|2\rangle \leftarrow |0\rangle$ with a larger cross section than that for $|1\rangle \leftarrow |0\rangle$. We can be further aided if the rates of radiation or collisional depopulation of level $|1\rangle$ to level $|0\rangle$ are much faster than those depopulating level $|2\rangle$. In the latter case the radiative and collisional processes do not limit the temporal

duration of laser action, and we can achieve "continuous wave" or cw laser operation. Thermodynamically, we may describe laser excitation mechanisms as energy transfer between two systems with different temperatures: the lasing species initially at low temperature and the pumping species initially at high temperature. Clearly, we are not just interested in static (equilibrium) properties. The essential knowledge we require is of rate processes: excitation, radiation, and relaxation.

C. Overview of the Volume

This volume reviews the results of many years of research on the collisional process of importance in gas lasers. Several of the chapters focus on the details of certain classes of collisions, the techniques that can be used to investigate them, and their impact generally on laser operation. Other chapters describe specific classes of lasers and outline the dominant kinetic and optical processes involved.

The common emphasis is on how the remarkable progress of gas lasers development has encouraged investigation of basic collision processes and how the building on the foundations of atomic collision physics has enabled the rapid discovery of many new types of gas lasers and has guided the attempts to develop these lasers to their maximum potential.

II. Historical Summary

In this section we will summarize the historical progress of gas laser development, concentrating on the advances that have been made in our understanding of the mechanisms that can be used for producing the upper laser level and achieving a population inversion. A more inclusive listing of gas lasers demonstrated through 1972, with a similar emphasis on excitation mechanisms, is provided by Willett (1974).

A. Energy Transfer Lasers

The first gas laser, helium–neon, was proposed by Javan (1959) on the basis of a careful study of possible excitation, deexcitation, and quenching mechanisms that might lead to population inversions in discharge excited gas mixtures. The concept, the experimental demonstration of which he and his co-workers reported two years later (Javan *et al.*, 1961), is based on excitation by the discharge electrons of a small fraction of the helium atoms from the ground state to the metastable 2^1S and 2^3S levels. These excited

states may collide with neon atoms, transferring the energy stored, to selectively produce two groups of excited neon levels, $2p^55s$ near 20.6 eV and $2p^54s$ near 19.8 eV. This produces local population inversions between the $2p^55s$ levels and the $2p^54p$ levels (between 20.2 and 20.4 eV) and the $2p^53p$ levels (between 18.5 and 19 eV), and between the $2p^54s$ and $2p^53p$ levels, because the lower levels are not produced in the energy transfer reaction. Laser action is observed on the population inversions referred to above. The evaluation of the performance of such a laser, and consideration of the other kinetic processes that must be understood [to maximize the energy efficiency or achieve continuous wave (cw) oscillation, as examples] will be the subject of most of this chapter and of the subsequent chapters of this volume. Such studies have led to the demonstration of numerous gas lasers based on energy transfer from excited rare-gas atoms.

A second type of energy transfer laser is illustrated by the CO_2 laser. The lowest excited vibrational level of N_2 lies in near resonance with the 00^01 vibrational level of CO_2. Thus we might expect that energy transfer collisions of $N_2(v = 1)$ with CO_2 should lead to population inversions in the selectively populated 00^01 level, relative to the unpopulated lower levels. Laser action using this principle was demonstrated independently by Patel (1964) and by Legay and Legay-Sommaire (1964), using discharge excitation of the N_2. Numerous alternate schemes have been developed for exciting the CO_2, including electron-beam, chemical, gas dynamic, and optical pumping. The CO_2 laser, because of its high efficiency (about 10%) and capability for high power cw operation (more than 100 kW), has become the best studied and most developed of all gas lasers (see Leland, Chapter 8 of this volume). It has provided the impetus for the investigation of a wide variety of atomic collision problems and has served as a training ground for the development of laser concepts and laser physicists.

Subsequent developments along this line include the demonstration of the Ar/N_2 energy transfer laser by Searles and Hart (1974) and by Ault *et al.* (1974). This laser is based on energy transfer from excited argon atoms, $Ar^*(3p^54s)$, to the near-resonant $N_2(C^3\Pi_u)$ state as illustrated in Fig. 2. The excited argon atoms are created by electron-beam excitation, causing this system to have some similarities with the excimer laser systems to be further discussed later. The intrinsic efficiency of the Ar/N_2 laser has been limited to about 1% because of the less than unity branching ratio for N_2 C-state production, and rapid quenching of the upper laser level, as compared to the rate of removal of the lower laser level, in the high pressure mixture (Hill *et al.*, 1974).

A fourth important energy transfer laser is the O_2^*/I chemical laser. The reaction of Cl_2 with H_2O_2 in basic solution, although still poorly understood, is known to produce $O_2(a^1\Delta_g)$ in high yield. Energy transfer can be used to

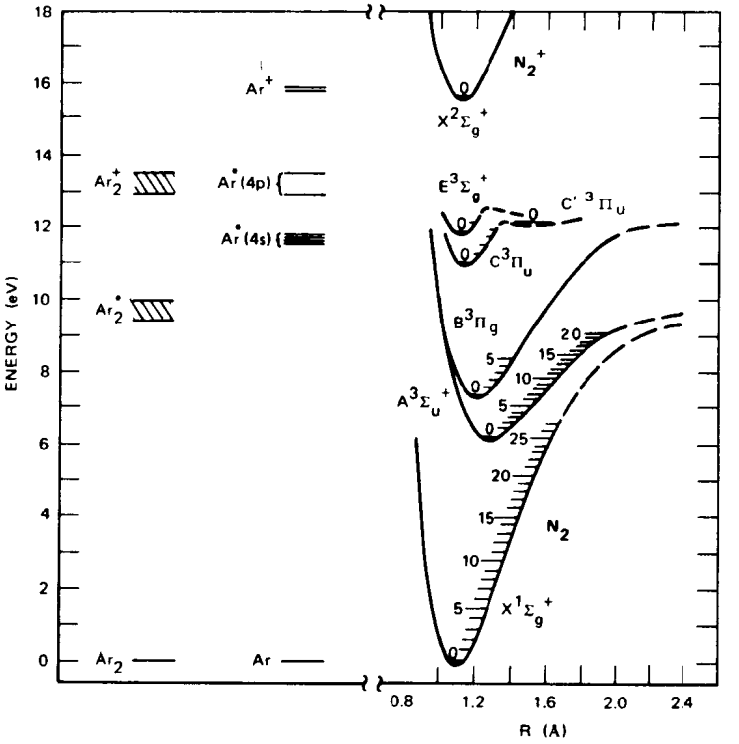

Fig. 2. Selected energy levels for Ar and N_a.

produce an absolute inversion on the $I(^2P_{1/2} \rightarrow {}^2P_{3/2})$ transition at 1.315 μm, provided that the ratio $[O_2(^1\Delta_g)]/[O_2(^3\Sigma_g^-)]$ is sufficiently large. This chemical, electronic-transition, energy transfer laser was first demonstrated by McDermott *et al.* (1978) and is now the subject of active investigation.

B. Direct Excitation Lasers

Lasers based on direct excitation of the upper laser level, by electron impact or other means, appear superficially simple in concept, but in practice may be difficult to devise. The requirement is to find a species and excitation process in which the upper laser level is selectively populated, in preference to the lower energy states that would predominate if the various excitation cross sections were equal. Failing that, one might be able to find a means to selectively depopulate the lower laser level.

The argon ion laser operates on transitions between excited states of Ar^+. It was demonstrated independently by Bridges (1964), Convert *et al.* (1964a,b),

Bennett *et al.* (1964), and Gordon *et al.* (1964). The upper laser levels, in the vicinity of 19–20 eV above the ground state of the ion, can be populated by single-step excitation/ionization from the ground state of neutral Ar and by two-step processes: (1) ionization:

$$Ar + e \longrightarrow Ar^+ + 2e, \tag{6}$$

followed by (2) excitation:

$$Ar^+ + e \longrightarrow Ar^{+*} + e. \tag{7}$$

The argon, krypton, and other rare-gas ion lasers have been of particular use in laboratory investigations, usually for pumping dye lasers. They offer high output powers (up to 1000 W) in the visible and ultraviolet under cw operation. Unfortunately, their efficiencies tend to be low, because of the very high energy of the upper laser levels (35 eV in Ar) and because of the multitude of lower energy excitations that may occur in electron rare-gas collisions.

Direct electron impact of excitation of electronic states of diatomic molecules can lead to gas lasers at a wide variety of wavelengths. Molecular nitrogen forms the basis for such a laser, as first demonstrated by Mathias and Parker (1963a). They observed laser action on the $B^3\Pi_g \rightarrow A^3\Sigma_u^+$ transition in the near infrared (see Fig. 2). Laser action on the ultraviolet transition $C^3\Pi_u \rightarrow B^3\Pi_g$ was observed in the same year by Heard (1963). Subsequently, lasers between 3 and 4 μm were demonstrated on the $a^1\Pi_g \rightarrow a'^1\Sigma_u$ and $w^1\Delta_u \rightarrow a^1\Pi_g$ transitions by McFarlane [(1965) and (1966), respectively]. Of these the N_2(C–B) laser has received the most investigation. As suggested by Gerry (1965), the upper laser level, $N_2(C^3\Pi_u)$, is excited in electron collisions with the ground state $N_2(X^1\Sigma_g^+)$, with a larger cross section than the lower vibrational levels of the lower laser level, $N_2(B^3\Pi_g)$. Laser action occurs predominantly on the 0–0 transition of the C–B (or second positive) system at 337.1 nm. This laser has also been of importance in laboratory research, largely for pumping high power pulsed dye lasers. The overall performance of the N_2 laser is limited to low energy pulses (a few millijoules in a few nanoseconds) and low efficiency (about 0.1%) by the fact that population accumulates in the lower laser level and the inversion is quickly lost (bottlenecking).

Simultaneous to the development of the N_2 laser described earlier, considerable work has been performed on lasers utilizing various transitions in the CO molecule. The first report of laser action in this molecule was made by Mathias and Parker (1963b), on the Ångstrom band ($B^1\Sigma^+ \rightarrow A^1\Pi$) in the visible. Infrared laser action on vibrational transitions in the ground state was first observed by Patel and Kerl (1964). This was followed by