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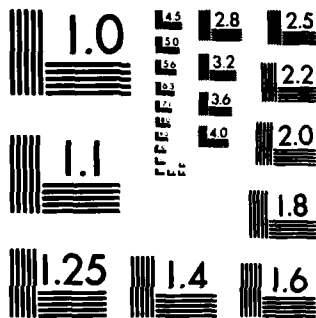
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KINETICS AND STRUCTURE OF EXCITED STATES

By: T. F. Gallagher

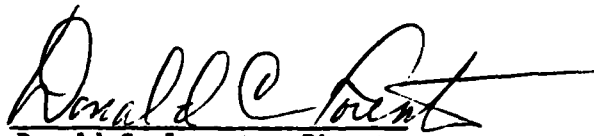
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I INTRODUCTION

The objective of this research program was to reach an understanding of excited atom processes by a systematic experimental study of their spectroscopy, interactions with strong fields, and collisions. The method used is laser excitation of excited atoms in conjunction with a variety of state selective detection techniques developed in our laboratory. Although it is unusual to study spectroscopy and collisions in the same research program, it has proved to be a very enlightening approach.

The following section of this report summarizes the accomplishments. For reference, a list of the scientific papers published under this contract is included. These papers embody the main conclusions of our study.

II ACCOMPLISHMENTS

Under this contract, we have investigated problems ranging from purely spectroscopic to purely collisional. For continuity in this section, we discuss similar projects together, irrespective of when they were conducted.

High resolution radio frequency and microwave spectroscopy is an excellent tool for the study of subtle atomic effects, and the development of such a technique may lead to a sensitive infrared detector.¹ Under this contract, we have demonstrated a new technique, delayed field ionization for the detection of microwave resonances in Cs.² In Cs, in two sets of measurements we were able to observe the f-g-h-i intervals and ascertain that the g fine structure intervals are no larger than hydrogenic in magnitude.^{2,3} In the Na, though, we have been able to extend the measurements of the nonhydrogenic f states down to $n = 9$, and the deviation from hydrogenic behavior, 5%, is in good agreement with the existing theory.⁴ Finally, in Ba, we have observed the rf transitions from the Ba 5d7d state to other nearby states and have measured the Stark shifts as a means of measuring the Rydberg character of the perturber.⁵

We have studied two types of collision processes. The first is the collision of excited Na atoms with ground state atoms and molecules. Examples of this are the investigations of the quenching of the Na ns states by CO and H₂⁶ and the angular momentum mixing of the Na nd states with the nearly degenerate higher angular momentum states by CH₄ and C₃H₈.⁷ These measurements were undertaken to explore the effect of larger collision partners on the quenching and angular momentum mixing processes. The angular momentum mixing process is a fundamentally interesting process because of the near degeneracy of the Na l states. Thus, we have studied this process with Xe in electric fields that distort the atomic wave functions.⁸ It was found that an electric field reduced the l cross section even at low fields, but we were not able to discern any visible alteration in the selection rules, which would be expected under these conditions.⁹ Unfortunately, in Na, the large Stark energy shifts complicate the

interpretation of the experimental results. Thus, it would be interesting to do such an experiment using Li.

The second type of collision process studied is the dipole-dipole collision of two Rydberg atoms in strong microwave fields. Using a very crude arrangement, we first observed the process, $\text{Na } 22s + \text{Na } 22s \rightarrow \text{Na } 22p + \text{Na } 21p + h\nu$, ν being 12-15 GHz, with only a few watts of microwave power.¹⁰ This was followed by a more refined experiment in which we were able to apply much stronger microwave fields to the atoms. This allowed us to observe collisions accompanied by the stimulated emission of up to four microwave photons. This is well into the strong field regime in which the field cannot be treated as a perturbation.¹¹ Although this problem had not previously been treated theoretically, we were able to develop a model to describe our results. These experiments should prove to be the beginning of a very important area of research.

The above study involved colliding pairs of atoms in strong microwave fields. It is only logical to pose the simpler question, "What happens to one atom in a strong field?" Accordingly, we began by investigating the microwave ionization of Na atoms by 15 GHz microwave fields. This immediately produced immediately the surprising result that a field strength, in atomic units, of $1/3n^5$ was required to ionize the atoms.¹² This is in marked contrast to all previous measurements in which a field scaling as $1/16n^4$ was required. Further investigation showed that the $1/3n^5$ behavior was restricted to the $|m| = 0$ states and l states; the $|m| = 2$ states behave like hydrogen and exhibit a $1/9n^4$ scaling of the ionizing field.¹³ These observations have led to a good phenomenological understanding of the ionization of atoms by non static electric fields.

To begin to understand the dynamics of the atoms in strong microwave fields, we considered this problem in terms of a simple hydrogenic picture, introducing the nonhydrogenic effects as a perturbation. This picture predicted that each n state would develop sidebands in a microwave field and that when the sidebands of n and $n + 1$ overlapped, ionization via the $1/3n^5$ mechanism would occur. To test this suggestion, we observed the spectrum of the Na atom in a strong microwave field, and in fact, our notions regarding the onset of ionization were verified.¹⁴ The field at which ionization of state n occurred was the field at which the sidebands of n and $n + 1$ over-

lapped. Further work in this area will no doubt lead to a different perspective on strong field, or equivalently, multiphoton processes.

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