Final Report on
Spectroscopic Diagnostics of Electron-Atom Collisions

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Progress from May 1, 1987 to April 30, 1991 is summarized in two Progress Reports that are reproduced in Appendix A, and in attached publications. Since then, we have completed manuscript preparations and publications of earlier observations, while carrying out a high energy-resolution measurement of electron collisional excitations in sodium. The results of the latter experiment have now been prepared for publication, and the manuscript is included as Appendix B. An additional manuscript, describing the unique high-current electron monochromator developed for this experiment, is in preparation and not enclosed. All additional results and conclusions of our work under the contract are now available in four publications that are attached at the back of this report. Consequently, we will elaborate on these only to note that we have achieved our proposed goals, with the full detail proposed but at a slightly slower pace than we had hoped.

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Progress Report on "Spectroscopic Diagnostics of Electron-Atom Collisions"

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January 1, 1990 - April 30, 1990

The primary objective of this three-year contract was to complete measurements, and analysis of these measurements, of highly differential electron excitation of the Na 3P state. This was completed, and the publication reporting this will appear in the Physical Review in approximately August, 1990. During the course of this experiment other aspects of atomic beam optical pumping and fluorescence were, of necessity, also addressed. A publication regarding the optical pumping has now also been prepared, and is scheduled for publication in the Journal of the Optical Society of America in the fall of 1990. The title and abstract of this paper are reproduced below. A publication regarding the fluorescence study has also been prepared and will be submitted to the Physical Review in 1990. A copy of this title and abstract is reproduced below.

Experimental work during the current period has concentrated on testing and improving a cylindrical, trochoidal electron monochromator. This has been designed to provide optimum currents for a range of electron-energy resolutions between 0.10 and 0.005 eV. The combination of retardation plus trochoidal motion, used by Schulz to achieve this goal, is highly susceptible to space charge in the barrier region, hence not a very high current device. Our design spreads out the charge by using a constant beam energy in a long cylinder, and in principle should achieve much higher currents for a given resolution. However, in practice our approach exposes the beam to a large surface area and a long trochoidal path, hence it is very sensitive to any surface-potential variations and to slight misalignments. Relatively high beam currents at ~ 0.05 eV resolution have now been achieved, but not yet with day to day reproducibility and reliability. As soon as this is achieved, we will study resonances in the electron excitation of metal atoms.

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The primary objective of this three-year contract was to complete measurements, and analysis of these measurements, of electron excitation of the Na 3P state. A secondary objective was to demonstrate the feasibility of a powerful new measurement technique. We had proposed to measure this cross section differentially with respect to the atomic spin change (ΔM_S), angular momentum change (ΔM_L), and electron scattering angle θ . This was to be done in the threshold-energy region where measurement and theory are generally most difficult. The proposed method combined the diagnostic strengths of atomic-recoil analysis¹ with atomic spin alignment² and radiative-excitation analysis³ to achieve a uniquely detailed separation of the partial components of the cross section. In essence, four of seven independent excitation parameters were to be measured by this technique, differentially with respect to electron scattering angle. These measurements and preliminary analysis were completed in early 1989. A manuscript describing these results has now (December 28, 1989) been completed and submitted for publication. This, plus three earlier manuscripts that describe significant experimental advances or partial results (without θ dependence), are enclosed with this report. Two theses based on this study provide additional details, but are not enclosed.

This experiment involved a complicated combination of laser and electron spectroscopy, which can be justified by the unique diagnostic power of the method but which placed some doubt on its feasibility with realistic effort. We thus believe that reaching or surpassing all of our goals, plus demonstrating the excellent S/N and power of this method, has been a

significant achievement. This involved 4 1/2 years of effort by each of two graduate students, starting without an apparatus. In order to reach these goals we developed, amongst other things, an extremely effective recirculating atomic-beam oven, an atomic-beam spin-alignment that greatly surpassed anything previously achieved, and methods for saturated-absorption locking and scan-control of the laser-frequencies which are at the forefront of dye-laser technology. The data reduction also required extremely careful and detailed analysis in order to achieve optimum reduction to θ dependencies. In essence, each θ dependence produced a recoil Doppler-shift distribution of typically ~ 50 MHz width in this Na case, whereas natural level widths and residual atomic-beam Doppler spread were ~ 25 MHz. Nevertheless, our data reduction was able to reliably determine the important aspects of these angular dependencies. As many other species and excitations have a more favorable ratio of these widths, we believe this is an important demonstration of the overall power of this new method.

The data from our experiment provides four partial cross sections for Na $3S \rightarrow 3P$ excitation, but not their three relative phases. During the course of our work the NIST group of McClelland, Kelley and Celotta have succeeded in measuring several other partial components of this excitation cross section, using one laser and a spin-polarized electron beam.⁴ Since neither experiment has yet detected the full set of seven cross-section parameters, and the quantization axes are rotated by 90° , it is not possible to directly intercompare these experimental results. The two experiments thus obtain complementary data, and each has provided independent tests of the predictions of the best available theories.

A new graduate student joined the program early in 1989, and a post-doctoral student joined a few months later. In addition to gaining

familiarity with the laser and beam equipment, they have designed and started testing a trochoidal electron monochrometer. The purpose is to obtain higher energy resolution in order to study the patterns of resonances that are expected to occur in electron excitation of weakly-bound excited states. Here the feasibility of some of the planned experiments depends on obtaining much higher electron-beam current, at a given resolution, than has normally been achieved with electrostatic or trochoidal monochrometers. We have therefore designed a cylindrical (first-order energy-spread compensated) monochrometer with aperture designs assisted by computer simulations. Since transverse, spiraling energy is not easily measured we plan to study the monochrometer resolution using the excitation resonances under study.

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Appendix B

Threshold electron excitation of Na

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Abstract

Electron collisional excitation of the 4D, 5D, 4P and 6S states of Na has been measured with about 30 meV energy resolution. Very rapid, unresolved threshold onsets are seen for all but the 4P state, but only weak undulations in the cross sections are observed above threshold. Some of these undulations are identified with higher excited state thresholds, while the causes of others are not identified.

I. INTRODUCTION

As described in the classic review paper of G. Schultz [1], Feshbach resonances often occur slightly below excited states, and core-excited shape resonances can occur slightly above. The former signal excited negative ion states with net binding of the extra electron, and the latter signify quasi-bound states. These are seen most clearly in angularly differential elastic scattering and excitation of states below the resonance. Thus, it is most common to study these resonances in differential scattering, but they also influence total excitation. Measurements of scattering resonances have been concentrated on volatile gases and on atomic hydrogen, due to the obvious experimental advantages in the former case and the essential physics in the latter. The metal vapors are much less amenable to experiment, and relatively few groups have studied these, particularly with high energy resolution. Due to their relatively simple yet regularly varying atomic structure, resonances for metal vapors and particularly the alkalis are of considerable interest.

As the binding of an excited state decreases, its polarizability and its ability to bind another electron increases. All excited states of alkali atoms above the first excited P state have very weak binding (< 2 eV) and can be expected to strongly bind an electron and yield resonances in electron scattering. However, the strength and observability of these resonances are not apparent, as they depend on resonance widths and coupling strengths to the observed scattering channels.

Current knowledge of electron scattering resonances in the alkalis is very sketchy. Eyb and Hoffman [2,3] measured differential elastic scattering by Na and K in the energy region of the first excitation, and K elastic and 4P excitations up to 3.1 eV, slightly above the

fourth excitation threshold. At their ~ 60 meV resolution three resonances, at and slightly below the second and third K excited states, were seen versus scattering angle. However, it is not clear how observable these might be in total excitation, and measurements of total Na 3P state excitation by Gould [4], Hafner [5], and Zapesochnyi et al. [6] with ~ 50 meV resolution did not detect any equivalent resonances. Gehenn and Reichert [7,8] measured Na and Cs elastic scattering and Cs(6P) total excitation in the region of the 6P excitation, also with ~ 60 meV resolution. In Cs they found a strong resonance at 1.49 eV, just above the 1.39 and 1.46 eV excitation thresholds of the $6P_{1/2}$ and $6P_{3/2}$ states. In contrast to the Na case, this resonance was also prominent in the total $6P_J$ state excitations, producing a very rapid rise to a peak at ~ 1.5 eV. Johnston and Burrow [9,10] have measured dT/dE with 100-200 meV resolution, where T is electron transmission through Na, K, Rb or Cs, under conditions alternately sensitive to elastic, total and resonance-line excitation cross sections. They observed highly undulating energy dependences in dT/dE , where the undulations result from several causes of transmission change, including scattering resonances and excitation onsets. Johnston [10] suggested identifications of patterns of resonances; it is not known if these will be visible in total (optical) excitations. With the above-mentioned exceptions of the Na (3P) and Cs (6P) states, total (optical) excitations have only been carried out with ≥ 300 meV electron energy resolution. However, even at 300 meV FWHM, Stumpf and Gallagher [11] saw apparent consequences of resonances, in that the threshold onset of Na (3D) excitation was indistinguishable from a (step-function) energy-analyzer signal.

This suggestive but very incomplete picture of alkali electron scattering resonances has prompted the present work. An additional motivation of our Na excitation study is the recent

development, by Zhou et al. [12], of a very exacting R-matrix, close-coupling code for electron-Na collisions. The present experiment extends high resolution excitation measurements to the higher states, as well as achieving the highest energy resolution yet reported for electron collisions with metal atoms.

Several difficulties are unique to high resolution experiments involving electron collisions with metal vapors. Foremost is the problem of obtaining sufficient target vapor density without poisoning the monochromator and interaction chamber surfaces with adsorbed metal, which ultimately becomes metal oxide. Such deposits act as insulators and also produce major work function changes, both disastrous to energy resolution. Other problems include the difficulty of manipulating low energy electron beams and of producing dense metal vapor beams, or of confining the vapor if a vapor filled interaction region is used. Space charge effects are also more limiting at the low electron-beam energies appropriate to metal atom excitation, and achieving adequate S/N in optical excitation function measurements becomes quite difficult. In the present experiment these problems were attacked with a combination of a high-current monochromator, an efficient optical-emission collector, and a high-flux, highly-collimated atomic beam.

We have made high energy-resolution measurements of the threshold behavior of several optical excitation functions of Na. These 4P, 4D, 5D, and 6S state excitation functions have already been measured with low energy resolution by Phelps and Lin [13], so we have only investigated the threshold region where resonances are expected to play a role. These states are bound by 0.5-1.5 eV, as shown in the Fig. 1.

II. EXPERIMENT

The experiment uses orthogonal electron and atom beams, and fluorescence is detected along the third axis. The electron beam propagates in a 35 G magnetic field, produced in a 40 cm gap between 30 cm square iron plates by permanent magnets. Except for the size of this field and the addition of the electron monochromator, the apparatus and arraignment is virtually identical to that described in Ref. [14], and will not be described in detail here. A mirror reflects $\sim 16\%$ of the fluorescence into a quartz lens, and then through a vacuum window and interference filter to the photomultiplier. The supersonic sodium beam from a recirculating oven was collimated by cooled slits to yield a 3 mm thick, 5 mm wide target of typical density $5 \times 10^{10} \text{ cm}^{-3}$ in the interaction region.

A cylindrical trochoidal monochromator, with 1 mm diameter entrance and exit apertures and 250 mm length, provided energy selection for the electron beam. The apertures were separated by 180 degrees on a 6.0 mm radius, midway between the 4.5 and 7.5 mm radius inner and outer cylinder surfaces. The beam energy transmitted by the monochromator was typically 0.3 eV, and an electric field of $\sim 1.5 \text{ V/cm}$ yielded trochoidal motion to the exit aperture. These parameters of magnetic field, electric field, and beam energy were chosen to yield compensation of a beam-shape distortion that normally occurs in planar trochoidal monochromators [15]. This distortion results from the energy difference, hence axial velocity difference, between electrons traveling at different heights in the electric field direction. In a cylindrical monochromator this extra velocity can be compensated by a shorter circumferential path around the 180 degrees to the exit aperture. This effect is observed in our cylindrical monochromator as a large difference in transmitted current when

the sign of the electric field is reversed. This form of monochromator was also chosen to optimize shielding of space charge fields, by spreading out the electron beam between nearby metal surfaces. Our optimum results were 30-40 meV FWHM at 30 nA current, and ~ 70 meV resolution at 150 nA. The disadvantage of the design is that the low-energy beam is exposed to a very large surface area, and contact potential variations can easily cause problems; gold plated copper cylinders were used. The entrance and exit apertures were 1 cm long molybdenum tubes, to limit the transverse energy of the beam by cyclotron motion into the tube walls. In the 35 G field this yields a transverse energy spread of ~ 13 meV [15,16].

A theoretical monochromator longitudinal energy spread is given by the convolved size of the apertures as a fraction of the 1.9 cm electron beam circumferential path between the cylinders, times the mean energy E_z of the transmitted beam. For the typical $E_z \sim 0.3$ eV used here this is ~ 30 meV. However, this fails to account for the fact that the aperture transmission is higher at the center, where electrons with larger cyclotron orbits are transmitted. This causes the effective aperture size to be $\sim 60\%$ of the physical dimension, and a correspondingly smaller longitudinal energy spread should occur. The transverse (spiraling) energy spread of 13 meV must be added to this, yielding ~ 30 meV total spread. The energy spread of the collisions includes any additional contact-potential spread in the interaction volume, plus residual Doppler spread due to the atomic beam angular spread. The last factor is negligible for the low interaction energies and ~ 0.1 radian atomic-beam angular spread used here.

Although the interaction chamber was heated to 250°C to minimize Na coating of surfaces, the Na beam was highly collimated and the vacuum was maintained below 10^{-9} Pa to minimize oxidation of temporarily adsorbed Na, electric fields clearly did build up and deteriorate the actual energy resolution over a period of hours. The presence of a molybdenum screen in front of the (insulating) mirror, only a few millimeters from the atomic beam, was probably the primary cause of this problem. A cylindrical retardation analyzer of the electron energy spread provided a useful monitor but not a true measure of the actual spread in beam or collision energy. The experimental signals are taken to be the only real indicator of the effective energy resolution.

Excitation functions were measured for the 4P-3S, 4D-3P, 5D-3P and 6S-3P transitions, at 330, 569, 498 and 515 nm wavelengths respectively, using interference filters and a bialkali photomultiplier. Attempts to measure the 5P-3S, 285 nm transition were unsuccessful, as were attempts to quiet a red-sensitive photomultiplier sufficiently to detect longer wavelength transitions from the 3D and 5S states. In essence, the photoelectron count rates were only 100-2000/s for the transitions reported, leaving little leeway for additional losses in S/N. The measured excitation functions are given in Figures 2-5; they have been normalized there to the data of Phelps and Lin for presentation as excitation cross sections.

As already noted, the energy resolution normally deteriorated rapidly with Na beam time. In addition, the monochromator resolution and current varied with each apparatus cleaning and reassembly, presumably due to different internal surface conditions. As a result, the data presented here were taken with varying resolution. As can be seen from the rapidity of the threshold rise in the figures, the highest resolution was obtained for the 4D and 5D

state excitations. This is due primarily to the fact that most tests and resolution studies were done on these two transitions. The threshold rate of rise of the 6S state data in Fig. 4 is considerably slower than for the 4D and 5D data in Figs. 2 and 3, but this is entirely due to a lower energy resolution on the particular day when these best 6S data were obtained. Data for the D states, taken essentially simultaneously with the reported 6S data, had the same threshold rate of rise as is seen in the 6S data. In contrast, the much slower rise of the 4P state threshold in Fig. 5 is not due to experimental resolution, as essentially simultaneous data for the 4D state was very similar to that in Fig. 2. Bars in the figures indicate these apparent experimental resolutions.

III. DISCUSSION

The threshold excitation in Figs. 2 and 3 rises from 25 to 75% in ~ 25 meV, and several data sets yielded similar behavior. This can be compared to the convolution of a hypothetical step-function cross section with a Gaussian electron energy distribution, in which case the FWHM of the Gaussian would be 43 meV. This is close to the theoretical characteristics of the monochromator for the conditions used, particularly as threshold laws suggest that the cross sections should not rise this abruptly. Thus, this strongly suggests that the monochromator was operating near its theoretical limit, and that the cross section rose too abruptly at threshold to be resolved with that resolution. Such rapid threshold onsets are expected and known for atomic hydrogen, due to its excited state degeneracies and resultant extreme polarizabilities, but the alkalis do not share this structural characteristic.

Except for H, very few atomic excitation cross sections have been measured with high resolution, and as already noted H has unique properties. Na (3P) and Cs (6P) measurements were discussed in the Introduction; the former shows no resonances and a slow threshold onset, while a strong resonance just above threshold dominates the latter. Perhaps the other data that is most closely related to that presented here are in a study of Kr excitation functions, studied in 45° scattering with 30-50 meV resolution by Swanson et al. [17]. This is not the same as a total excitation cross section, but the energy dependence should be indicative. These data for exciting nine different states almost all show very rapid threshold onsets, with most rising to a very prominent resonance 0.1 eV or so above threshold. (The same resonances are seen in Kr elastic scattering.) The present, Na data do not show any similarly pronounced resonances slightly above threshold, so this obvious cause of some of the rapid Kr threshold onsets does not appear to be applicable here. On the other hand, two of the nine Kr states studied show rapid (unresolved) onsets without an accompanying strong resonance, similar to the behavior seen here. These Kr states are 2.5-4.0 eV below the ionization threshold, compared to 0.5-1.4 eV for the Na states studied here, so it is not clear if the cause is related. In the present Na case, one plausible explanation of these very rapid threshold onsets is the existence of many different scattering-channel resonances near the threshold, so that a single resonance is not observable at our experimental resolution.

Whereas the 4D, 5D and 6S state excitation onsets were faster than our ~ 30 meV experimental resolution could observe, the 4P state excitation onset was about 200 meV wide and well resolved. The binding of the 4P state is only 60% greater than that of the 4D state, so this alone does not appear to be a likely explanation of this difference. Also, recall that

Ref. 11 inferred that the 3D state excitation cross section had a very sharp threshold onset. Thus, although we were unsuccessful in our attempt to observe 5P state excitation to confirm this distinction, this appears to be a characteristic of the P state, presumably due to a weak but significant dipole- allowed, long-range component to its excitation.

Acknowledgment

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Figure captions

- Fig. 1. Na energy levels and excitation energies, in parentheses. Fluorescence transitions observed here are indicated by arrows, with wavelengths and branching into these transitions adjacent.
- Fig. 2. Excitation cross section for the Na 4D state, obtained with ~ 30 meV electron energy resolution. The structures near 4.35 eV and the 6S state are reproducible.
- Fig. 3. Excitation cross section for the Na 5D state, obtained with ~ 30 meV electron energy resolution. The structures near 4.68 eV and 4.9 eV are reproducible.
- Fig. 4. Excitation cross section for the Na 6S state, obtained with ~ 70 meV energy resolution.
- Fig. 5. Excitation cross section for the Na 4P state, obtained with ~ 30 meV energy resolution. The cross section calculated by Zhou et al. [Ref. 12] is indicated by + symbols.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

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