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of Simple Atomic and Ionic Systems

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This final technical report for NASA grant number NGL 33-008-009 covers the ten-year period from January 1, 1963, to December 31, 1972. It is believed that the time has been well spent and that the results which are summarized here more than justify the support that the National Aeronautics and Space Administration has given for research on the properties and interactions of simple atomic and ionic systems.

Most of the work undertaken has been brought to a successful conclusion. At least two of the projects, namely, work on the metastable autoionizing states of the helium negative ion and the lifetime measurements of the metastable $^2S_{1/2}$ state of singly ionized helium, are continuing with the support of the National Science Foundation's grant GP 13479.

The cooperation and help that we have received from the National Aeronautics and Space Administration for this program are deeply appreciated. It is hoped that NASA will see fit to provide additional support in the future for work in this area of research.

R. Novick  
Professor of Physics;  
Principal Investigator

August 5, 1974
I. PROPERTIES AND INTERACTIONS OF SIMPLE IONIC SYSTEMS

METASTABLE AUTOIONIZING STATES OF THE HELIUM NEGATIVE ION

Studies of the metastable autoionizing He⁻ ion were initiated in 1963 and are continuing at the present time under grant GP 13479 from the National Science Foundation with Professor R. Novick serving as Principal Investigator. Work accomplished under NASA grant NGL 33-008-009 is described in the publications listed below.


It is proposed to produce beams of polarized He³ negative ions by exploiting the differential metastability of the hyperfine and fine structure levels of the (ls2s2p) configuration of the ion. Such a source could be very compact and does not require deflecting magnets.


The decay of the differentially metastable autoionizing (ls2s2p)⁴P_J states of the He⁻ ion has been studied by time-of-flight techniques in an axial magnetic field, and Zeeman quenching has been observed. Two distinct lifetime components (11 µsec, 210 µsec) have been identified in a 100-eV beam with an axial field of 400 G. The zero-field lifetimes obtained from these and other measurements are 11 ± 5 µsec for J = 1, 3, and 345 ± 20 µsec for J = 3. The 3-2 fine-structure interval is estimated to be 0.050 ± 0.015 cm⁻¹.

3) R. Novick and D. Weinflash, "Precision Measurement of the Fine Structure and Lifetimes of the (ls2s2p)⁴P_J States of He⁻ and Li⁺," in Precision Measurement and Fundamental Constants, Proceedings of the International Conference Held at the National Bureau of Standards, Gaithersburg,
A series of measurements has been completed and others are in progress for the precise determination of the energies and lifetimes of the metastable autoionizing \(1s^22s2p \, ^4P_J\) states in He\(^-\) and Li\(^-\). Since these states have lifetimes in the range from 5 to 500 \(\mu s\) and since the fine and hyperfine structure intervals are of the order of a wave number, it will be possible to determine the splitting with a relative accuracy of one part in \(10^4\) or better. Present measurements in \(^4\)Li and \(^7\)Li are at the level of one part in \(10^3\) and a preliminary crude estimate has been made of the \(^4\)P\(_{1/2}\)-\(^4\)P\(_{3/2}\) interval in He\(^-\). Further work is in progress with radio-frequency techniques to obtain very much more precise results. Considerable theoretical effort must be made before these results can be interpreted in terms of fundamental constants. However, it is clear that if the necessary numerical wave functions can be obtained with sufficient accuracy and if the various radiative and relativistic corrections can be evaluated, then these results will provide a precise new independent value for the Sommerfeld fine structure constant.


Fine-structure transitions have been observed in the metastable \(1s^22s2p \, ^4P_J\) state of the negative helium ion \(\text{He}^-\) using an rf resonance technique. Preliminary determinations have been made of the energy separations: \(|E_{3/2} - E_{1/2}| = 825.23 \pm 0.92\) MHz and \(|E_{5/2} - E_{1/2}| = 863 \pm 56\) MHz. We have shown that the \(J = \frac{3}{2}\) level lies between the \(\frac{5}{2}\) and \(\frac{3}{2}\) levels. These results are to be compared with the current theoretical estimates of \(E_{3/2} - E_{1/2} = 2030\) MHz and \(E_{5/2} - E_{1/2} = 9410\) MHz.


Preliminary fine-structure and hyperfine-structure measurements have been made in the helium negative ion with the isotopes \(^4\)He and \(^3\)He. The measurements can be performed to high precision and provide an excellent test of the theory of three-electron atomic systems. If accurate wave functions become available, these measurements will lead to the possibility that our understanding of three-electron atomic systems can be tested on a fundamental level comparable to that obtained in hydrogen and in neutral helium.

All independent energy intervals have been measured in the metastable $1s2s2p^4P$ state of the helium negative ions $^4\text{He}^-$ and $^3\text{He}^-$. These results are of sufficient precision to provide a critical test of any proposed wave functions for this state and, when adequate wave functions become available, can be used to test relativistic and radiative corrections in three-electron atomic systems.
TWO-PHOTON DECAY SPECTRUM OF THE METASTABLE He$^+$ ION

Studies on the two-photon decay spectrum of the metastable $2^2S_{1/2}$ state of singly ionized helium were in progress from 1964 to 1969. A number of noteworthy publications resulted from this work.


The direct detection by coincidence counting techniques of the two-photon decay of the metastable $2^2S_{1/2}$ state of singly ionized helium was first reported.


Verification has been made of the theoretically predicted spectral distribution of the two-photon emission from the metastable $2^2S_{1/2}$ state of singly ionized helium by means of a broad-band spectroscopic coincidence counting technique.


A review of work to test the theory of the metastability of the hydrogenic 2S state is presented. It includes the history of the hydrogenic metastable state, the theory of the two-photon decay process, and the effect of perturbations on the metastable state. The apparatus is described in detail, and results and conclusions are presented. All of the tests performed indicate that the $2^2S_{1/2}$ state of He$^+$ decays by two-photon emission as predicted by Breit and Teller. It has been shown that photon coincidences are observed when and only when metastable ions are present in the ion beam. We have demonstrated that the angular correlation function for the coincidences agrees with the theoretically expected function and that the spectrum of the photons is consistent with the predicted function.


A review similar to that presented in paper 5).


It is noted in this Letter to the Editor that all the essential features of the theory of the decay of the metastable $2^2S_{1/2}$ state of hydrogenic atoms were verified by the present author and colleagues in a series of experiments on the helium ion and that this work culminated in the first observation of two-photon spontaneous emission.
LIFETIME MEASUREMENT OF THE METASTABLE $^{+} \text{He}$ ION

Upon completion of the two-photon coincidence observations of the metastable $2^2 S_{1/2}$ state of singly ionized helium, an experiment was undertaken to observe the decay in flight of metastable helium ions. The work resulted in the determination of the lifetime and is continuing at present under grant GP 13479 from the National Science Foundation. Professor R. Novick is the Principal Investigator.


The natural lifetime of the $2S$ state of He$^+$ has been measured in a decay-in-flight experiment. The result, $\tau = 2.04^{+0.31}_{-0.34} \text{ ns}$, is consistent with the theory of two-photon spontaneous emission and determines an upper bound $\lambda < 8 \times 10^{-11}$ for the coupling constant of the pseudoscalar "anapole" interaction.
OPTICAL EXCITATION WITH LOW-ENERGY IONS

Incident to the study of two-photon decay of the 2S state of He\(^+\), observation was made of radiation due to the impact of very slow helium ions on the rare gases and on some molecular gases. Reference is made to the following publications and theses which resulted from these studies.


Observation of radiation due to the impact of very slow helium ions on the rare gases and on some molecular gases is reported. The dependence of the cross section for this process on the kinetic energy of the He\(^+\) beam shows unexpected features down to the lowest energies studied (5 eV). The absolute cross sections are typically of the order of 10\(^{-16}\) cm\(^2\) and in some cases are almost an order of magnitude larger. In at least one case the radiation has been shown to result from charge exchange with simultaneous excitation.


Recent work on low-energy ion-atom and ion-molecule collisions indicates that these processes can be highly effective in producing optical excitation, in contradiction to the well-known "adiabatic criterion." Further evidence is presented for this conclusion in the important case of excitation of helium atoms by He\(^+\) ions.

A high current, electron bombardment ion source of simple design and suitable for low energy ion beam production is described. The source produces ion beams in the energy range from a few electron volts to several kiloelectron volts with an energy spread of a fraction of 1 eV, with high current density (5 μA/cm² at 25 eV), and without the problems associated with rf ion sources. The source has been used successfully to produce low energy beams of noble gas ions such as He⁺ and Ar⁺ without metastable contamination, as well as for H₂⁺ and N₂⁺.


Experimental and theoretical studies have been made of the excitation cross sections of various neutral excited states of He in low-energy He⁺-He collisions. Two striking results of the experimental observations are the large value of the cross section at low energies with thresholds slightly above the final state energies, and the strong oscillatory dependence of the cross sections on projectile energy. These phenomena can be interpreted in terms of the molecular-potential energy curves of the He₂⁺ system.


New data on the excitation of helium by low-energy helium ions have been obtained that provide strong support for the post-collision-interaction model proposed by Rosenthal and Foley in a companion paper. Certain details of the results that are not presently predicted by the theory are pointed out.
LIFETIME MEASUREMENTS OF SINGLY IONIZED LITHIUM (Li*)

WITH A RADIO-FREQUENCY MAGNETIC-RESONANCE TECHNIQUE

Studies were initiated with the hope of measuring the fine structure of the $2^3P$ term of Li*. While this objective was not attained, the lifetime, electron-impact excitation cross section, and polarization of the $2^3P$ term of this ion were determined, and our results are available in the following publications:


The lifetime of the $2^3P_1$ states in singly ionized lithium has been measured using an rf magnetic-resonance technique; the value is $\tau = 45 \pm 5$ nsec. Neutral lithium was ionized and excited by a unidirectional beam of electrons which produced an alignment in the excited state. The cross section for the electron-impact excitation $1s^22s^2S^+ \rightarrow 1s2p^1P$ near threshold was measured to be $10^{-17.1}$ cm$^2$. Implications for the feasibility of rf resonance spectroscopy on the fine and hyperfine structure of the $2^3P$ term will be discussed.
II. PROPERTIES OF SIMPLE ATOMIC SYSTEMS

METASTABLE AUTOIONIZING ATOMIC ENERGY LEVELS

IN THE ALKALI (GROUP I) ELEMENTS

A new class of long-lived autoionizing states in the alkali atoms was discovered and studied. The several publications which have appeared in the open literature as well as the dissertation of Dr. Paul D. Feldman fully describe the results of this work.


The existence of atomic energy levels in lithium, potassium, and rubidium which lie between the first and second ionization potentials and which are metastable against both radiation and autoionization with lifetimes in the microsecond region are first reported.


The \((1s2s2p)^4P_{5/2}\) configuration in He and Li is metastable against radiation and autoionization via the Coulomb interaction. Lithium atoms in this state can be produced by exchange excitation (energy = 56 eV) of ground state Li atoms. Experimentally, we bombard a neutral lithium beam with low energy electrons and collect ions resulting from the decay of metastable atoms 3 cm from the source. We have measured the excitation function, production cross section \(\sigma \sim 10^{-13} \text{ cm}^2\) and the lifetime \(\tau = 5.1 \pm 1 \mu\text{sec}\). A theoretical estimate of the probability of autoionization resulting from the spin-spin interaction gives a lifetime of 16 \(\mu\text{sec}\).

Discrete atomic energy levels lying between the first and second ionization potentials which are metastable against both auto-ionization and radiative decay have been observed in the alkali elements. These levels arise from the excitation of an electron from the outermost closed shell of the atom and have characteristic lifetimes in the microsecond region. A beam of metastable alkali atoms is produced by electron bombardment of neutral ground-state atoms and is detected by collecting the charged products of the decay. Stern-Gerlach magnetic-deflection experiments have been performed to show unambiguously that the signals arise from paramagnetic atoms and not from possible stray-photon effects in the apparatus. The excitation energies, electron-bombardment production cross sections, natural lifetimes, and the tentative spectroscopic assignments of these atoms are listed below:

<table>
<thead>
<tr>
<th>Element</th>
<th>Energy above ground state (eV)</th>
<th>Tentative assignment</th>
<th>Lifetime (μsec)</th>
<th>Electron-bombardment production cross section (cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Li</td>
<td>37.3±0.3</td>
<td>(3s2p3d)4P</td>
<td>5.1±1.0</td>
<td>10⁻¹⁰±10⁻⁹</td>
</tr>
<tr>
<td>Na</td>
<td>31.8±0.3</td>
<td>(3p3d)4D</td>
<td>90±20</td>
<td>10⁻¹⁰±10⁻⁹</td>
</tr>
<tr>
<td>K</td>
<td>19.9±0.3</td>
<td>(3p4d5s)5F</td>
<td>75±20</td>
<td>10⁻¹⁰±10⁻⁹</td>
</tr>
<tr>
<td>Rb</td>
<td>15.8±0.3</td>
<td>(5p6s7s)4P</td>
<td>40±15</td>
<td></td>
</tr>
<tr>
<td>Cs</td>
<td>12.6±0.3</td>
<td>(5p6s7s)4P</td>
<td>40±15</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>(5p6s7f)G</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The energy and lifetime of the metastable lithium atom are in good agreement with the available theoretical estimates. The metastable states in the heavier alkali atoms are classified in relation to the known 1S spectral terms. Magnetic and electric fields have been found to be effective in reducing the lifetime of the metastable atoms. The Zeeman quenching is discussed in a separate paper. The Stark quenching rate has been found to be proportional to the square of the electric field and to increase rapidly with atomic number Z. It is suggested that the Stark quenching results from a second-order interaction which involves the product of the electric field and the spin-orbit or other magnetic operators.
LIFETIME MEASUREMENTS OF THE ALKALI METAL ATOMS (GROUP I ELEMENTS) WITH LEVEL-CROSSING SPECTROSCOPY

This program was in progress under the Joint Services Electronics Program of the Columbia Radiation Laboratory at the time of the initiation of grant NGL 33-008-009 (formerly NeG-360). The program produced a number of publications enumerated below.


The method of level-crossing spectroscopy has been extended to a study of the Stark effect on the 3P term of lithium. Lithium atoms in a broad atomic beam are subjected to collinear electric and magnetic fields. The field values required to produce a level crossing or degeneracy are determined by observing the change in the angular distribution of the fluorescence resulting from the optical excitation of the 3P term. The magnetic field required to produce the crossing is found to shift to higher values as the electric field is applied. The shift increases as the square of the electric field and is given by \(\Delta H = -0.056 (11) + E^2\), where \(\Delta H\) is in gauss and \(E\) is in kilovolts per centimeter. The coefficient is in good agreement with the value 0.048 obtained from second-order perturbation theory and the Bates and Damgaard approximation.


The fine and hyperfine structures of the 3P term in Li have been studied by the technique of level-crossing spectroscopy. A preliminary value for the fine-structure separation is \(\Delta = 2883.9 \pm 1.2\) Mc/sec. It is noteworthy that the present technique can be extended to obtain a result that is reliable to a few parts per million. It is noteworthy that the fine-structure intervals in lithium are 8% and 12% smaller for the 2p and 3p levels, respectively, than the corresponding intervals in hydrogen. A theory is developed for interpreting the separation between hyperfine level crossings. Its application depends on a detailed knowledge of the core-polarization effects of the 3p electron. Evidence is presented indicating that the core-polarization effects in the np states (n=2, 3) scale as \((1/r)^n\), for the valence electron. Assuming this to be true, we estimate that \(g_3 (3P) = -3.05 (12)\) Mc/sec for Li. The limited precision achieved so far in this work has prevented observation of the Li quadrupole interaction.

4) A. Khadjavi, W. Happer, Jr., and A. Lurio, "Electric-Field Level-Crossing Spectroscopy," Phys. Rev. Letters 17, 463-465 (29 August 1966). The first observation of pure electric-field level crossings at finite electric field are reported, and the use of these
observations to determine the differential Stark effect in the second excited state of the alkali metal atoms is described. In previous work Stark parameters have been measured by level crossing in combined electric and magnetic fields. This work is the first experimental demonstration that the application of an electric field will cause certain hyperfine structure levels to intersect at other than zero electric field. Results are reported for Cs$^{133}$ and Rb$^{85}$ and compared with other experimental and theoretical work.


The fine-structure intervals of the 3^2P state of Li$^7$ and the 3^2P state of Li$^6$ have been measured by observing the level crossing signals from the $P_3/2$ and $P_1/2$ levels. Results for the magnetic fields at which the center of the crossing signals occur are the following, in terms of the proton NMR frequency in kHz for a mineral oil sample: 3^2P(Li$^7$), 3897.163(22); 3^2P(Li$^6$), 3856.88(20); 4^2P(Li$^7$), 1621.71(15). By using a theory of the Zeeman effect that includes estimates of the normal and specific mass effects and relativistic effects, we find the following values for the zero-field fine-structure intervals: $\Delta W(Li^7 - 3P) = 2882.903(18)$ MHz; $\Delta W(Li^6 - 3P) = 1199.65(11)$ MHz. The uncertainties are three times the standard error for the measurement.


The technique of level crossing spectroscopy has been used to study the hyperfine structure of the 3^2P and 4^2P states of Li$^7$ and the 3^2P state of Li$^6$. Both low-field (< 2 G) and high-field (> 215 G) level crossings were observed for the 3^2P state of Li$^7$. With the assumption that the ordinary magnetic-dipole coupling constants are related by $\tilde{g}_{5/2} = 5\tilde{g}_{3/2}$, values have been obtained for $\tilde{g}_{3/2}$ and the core-polarization constant $\tilde{a}_C$. In the 4^2P state of Li$^7$ and the 3^2P state of Li$^6$ we have inferred from our measurements individual values for $\tilde{g}_{3/2}$ and $\tilde{a}_C$ which are consistent with the assumptions that both constants scale as the ratio of nuclear $g$ factors among different isotopes and that they scale as $1/n^3$ among all the $n^2P$ states. The lifetime of the 3^2P state has also been obtained from the high-field level crossing data. The results for the hyperfine constants in MHz are the following: 3^2P Li$^7$: $\tilde{g}_{3/2} = 2.11(5)$, $\tilde{a}_C = -3.88(3)$, 4^2P Li$^7$: $\tilde{g}_{3/2} = 0.89(2)$, $\tilde{a}_C = -1.20(2)$, 3^2P Li$^6$: $\tilde{g}_{3/2} = 0.50(2)$, $\tilde{a}_C = -1.20(2)$. The lifetime of the 3^2P state is $t (3^2P Li^7) = 182(6)$ nsec.
LIFETIME MEASUREMENTS OF GROUP II-A ISOTOPES

WITH LEVEL-CROSSING SPECTROSCOPY

The technique of zero-field level-crossing spectroscopy was used to determine the lifetimes of the group II-A isotopes. The published results are listed below:


The lifetime of the first excited \( ^1P_1 \) state of zinc, calcium, and strontium has been measured by the zero-field level-crossing technique. The results are: zinc \( \tau(1P_1) = 1.41 \pm 0.03 \times 10^{-9} \) sec, calcium \( \tau(1P_1) = 4.48 \pm 0.15 \times 10^{-9} \) sec, and strontium \( \tau(1P_1) = 4.97 \pm 0.15 \times 10^{-9} \) sec. The results are compared to earlier experimental results and also to theoretical predictions. It is found that our results are in excellent agreement with the Bates and Damgaard Coulomb approximation method for estimating the oscillator strengths.

LIFETIME MEASUREMENTS OF CHROMIUM-53 ATOM (GROUP VI ELEMENT)
WITH OPTICAL DOUBLE RESONANCE AND LEVEL-CROSSING SPECTROSCOPY

The $^7P$ terms of two excited chromium configurations were studied with optical double resonance and level-crossing techniques, and theoretical and experimental values for the contribution of core polarization to the hyperfine structure of excited states of Cr$^{53}$ were determined.

The following results were published:


Theoretical and experimental values for the contribution of core polarization to the hyperfine structure of excited states of Cr$^{53}$ are compared. A theory is developed for extracting the core contributions to the magnetic field from the measured hyperfine constants. It is necessary to take account of configuration interaction with an actual configuration of unpaired $s$ electrons. A value of $-535000$ G is deduced for the magnetic field at the nucleus produced by core polarization. The minus sign indicates that this field is antiparallel to the net spin of the atom. This is compared with theory and with the results of other experiments. Corrections due to relativity, the second-order Zeeman effect, and the breakdown of $LS$ coupling are considered.
INSTRUMENTATION


The requirements on light sources for double resonance and level crossing spectroscopy are reviewed. A modern version of the Curie-Löchte-Holtgreven flow lamp is described and its performance for a number of elements is compared with that of sealed-off electrodeless discharge lamps and Schuler-type hollow cathode sources. None of these sources is useful under all circumstances. The electrodeless lamp is particularly effective in the case of the group IIIb intercombination lines. The high atomic density available in the flow lamp makes it useful for partially forbidden resonance lines for a number of chemical species including highly reactive metals, such as Ca. The hollow cathode lamp is well suited to the fully allowed resonance lines of a wide range of elements including highly refractory metals.
III. MISCELLANEOUS STUDIES


At very low atomic densities all Zeeman sublevels of an excited atomic state decay with the natural radiative lifetime of that state. Under these conditions the widths of the Hanle effect and optical double-resonance signals may be used to measure the lifetime of the state. However, at higher atomic densities the Zeeman sublevels can relax to each other through collision and radiation trapping so that, in general, \((2J + 1)^2\) parameters are necessary to specify the decay of the excited atoms. These processes result in a broadening or narrowing of the level crossing or optical double-resonance linewidths and can be ascribed to a change in the lifetime of the excited atomic state. These experiments demonstrate that under a wide range of experimental conditions the different multipole components of the density matrix describing excited atoms in a vapor relax with measurably different time constants. More detailed studies of this kind should yield considerable information about the mechanisms involved in depolarizing collisions.


Auroral spectra have been obtained in the wavelength region 1050-1550 Å over the altitude range 90-160 km in an Aerobee rocket at Fort Churchill, Manitoba. Spectral lines of atomic hydrogen, nitrogen, and oxygen, and 18 bands of the Lyman-Birge-Hopfield system of molecular nitrogen were identified. The primary electron flux and the 3914-Å emission of \(N_2^+\) were also measured throughout the flight. Altitude profiles for several spectral features and the electron flux are given, and correlations among the various observations are discussed. The hydrogen-Lyman-α radiation at 1216 Å is found to be independent of the aurora. Other origins for this radiation are suggested.

An analysis is given of the correlation in detection times of two successive radiations emitted spontaneously in a sequential decay. By a resolvent operator method, the time evolution of the coupled particle-field state is derived for a system consisting of a three-level atom coupled to the photon field. Photons A and B, emitted in the first and second stages of the cascade, are detected at distances $r_A$ and $r_B$ from the atom. When one of the photons is detected, the state vector for the system is assumed to be reduced, by projection onto the observed state. After this measurement, the system again evolves continuously, until the second photon is detected. The probability that a retarded time interval $T = (t_B - r_B/c) - (t_A - r_A/c)$ will elapse between the detection of photon A, at time $t_A$, and the detection of photon B, at time $t_B$, is found to be proportional to $\exp(-T)$ if $T > 0$ and to be zero if $T < 0$, where $1/T$ is the mean lifetime of the intermediate state. This result is consistent with a prequantum view of successive photon emission, but is obtained by a method which avoids the conceptual difficulties of a point-particle photon model.


Recently we reported the operation of an optically pumped $^{85}$Rb maser oscillating at 3.035734 GHz on the field-independent transition $5\,^2S_{1/2}(F = 3, m_F = 0 \rightarrow F = 2, m_F = 0)$. Apart from cavity pulling, the two major sources of frequency shifts are those due to the buffer gas and the pumping light. The short-term phase stability of the standard is not appreciably affected by these shifts. The buffer-gas shift can be used to advantage to select an output frequency that is different from the ground-state hyperfine frequency.


The vanishing of certain coupling matrix elements at level crossings is shown to follow from angular momentum commutation relations. A magnetic dipole transition having $\Delta M = 1$, induced near a crossing of the levels in a nonzero magnetic field, is found to have a dipole matrix element comparable to or smaller than the quotient of the level separation and the field. This result also applies in the analogous electric field electric dipole case.