tum states J, M are not isolated in the close-coupling expansion.

[6]The strong-coupling region is recognized by the presence of interference terms which cause the cross sections in Fig. 1(a) to behave erratically with l and by the close proximity of these curves to the dashed curve which represents the upper bound where $\underline{T} = 2 \times \underline{1}$.

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Measurement of the Lifetime and the Electron-Impact Excitation Cross Section and Polarization of the 2^3P Term of Singly Ionized Lithium*

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The lifetime of the $2\ ^3P_j$ 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 $1s^2\ 2s\ ^2S \rightarrow 1s2p\ ^3P$ near threshold was measured to be $10^{-22\pm1}$ cm^2. Implications for the feasibility of rf resonance spectroscopy on the fine and hyperfine structure of the $2\ ^3P$ term will be discussed.

I. INTRODUCTION

An accurate measurement of the fine and hyperfine structure of Li^+ would be a very important addition to atomic spectroscopy. Two-electron atomic systems are of considerable theoretical interest because their relative simplicity allows very accurate calculations. In particular, comparisons of fine- and hyperfine-structure calculations for such systems with precision measurements of these quantities have provided a critical test of quantum electrodynamics and, until the recent transfer to the use of the ac Josephson-effect measurements, the accepted value for the electromagnetic coupling constant, $\alpha = e^2/hc$.

Probably the most intensively studied two-electron system has been that of the 2^3P multiplet in He. Hughes and his collaborators have used an atomic-beam double-resonance technique to measure the fine-structure splittings to ~ 1 ppm.[1] Recently, new theoretical values for the splittings have been obtained which give the 2^3P_0-2^3P_1 separation to ~ 6 ppm and the 2^3P_1-2^3P_2 separation to ~ 150 ppm.[2] The new level of accuracy achieved in this calculation is derived mainly from a preliminary evaluation of newly developed $\alpha^6 mc^2$ terms for the two-electron Hamiltonian, and an eventual accuracy of better than 1 ppm is expected from the continued refinement of this approach. Thus, there is the expectation that the He fine structure will provide a value for α better than 1 ppm.

Some of the reasons which make the intensive study of the 2^3P term in He so valuable also apply in the case of Li^+. The fact that the ratio of natural linewidth to fine-structure splitting in the 2^3P Li^+ term is about 1.2×10^{-4} means that a 1 ppm accuracy in the fine-structure measurement can be achieved by measuring the center of the resonance line to only $\frac{1}{100}$ of its width. Wave functions for this term in Li^+ have already been calculated to about the same accuracy as the He functions and have been used to provide the fine-structure splittings to order α^4 Ry.[3] The lithium study has two additional features.

(a) The fine structure of Li^+ is predominantly due to the spin-spin term; this fact means that the results in Li^+ can provide a more sensitive test for the correctness of the two-electron, spin-spin Hamiltonian than those in He, where the spin-orbit and spin-spin terms contribute about equally.

(b) In the course of the measurements the Li^+ hyperfine structure must also be determined to 2 parts in 10^5; the comparisons between experiment and the hfs calculations can advance our understanding of nuclear structure effects in hfs.

Our work on singly ionized lithium was initiated with the hope of eventually measuring the fine structure of the 2^3P term. Figure 1 is an energy-level diagram showing the fine and hyperfine structure of $^7Li^+$. rf or microwave resonance spectroscopy of the 2^3P states can be performed if the system is prepared having some alignment. Of the

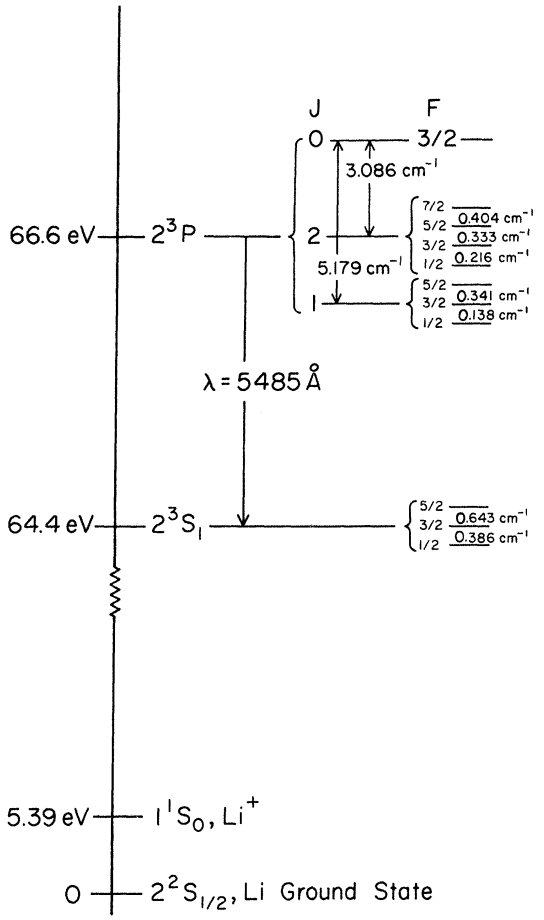

FIG. 1. The fine and hyperfine structure of $^7Li^+$ as measured optically by Hertzberg and Moore (Ref. 9).

two possible excitation methods, direct electron impact and optical pumping from ions prepared in the $2\,^3S$ state, we chose the former because signal-to-noise estimates indicated that, with the sources available at the time, the attainable intensity of $\lambda = 5485$-Å light and the density of $2\,^3S$ ions were inadequate to produce a measurable signal. (With the recent advent of moderate-power cw tunable lasers, this supposition may no longer be true.) Our goal was not achievable by the method chosen because, as our measurements show, the electron-impact excitation cross section and polarization associated with the $2\,^3P$ term are so small that a resonance experiment based on this mode of excitation is only marginally possible. The remainder of the paper describes these measurements and the measurement of the lifetime of the states from a rf resonance observed between the $F=\frac{7}{2}$ sublevels of the $2\,^3P_2$ level in $^7Li^+$.

II. EXPERIMENTAL METHOD AND MEASUREMENTS

The experimental apparatus is represented schematically in Fig. 2. Lithium atoms are discharged from the oven through a 1-cm-diam "crinkle foil" multichannel plug, which is essentially a stack of triangular tubes of length 1 cm and effective radius 0.027 mm.[4] The electron gun consisted of a 1-cm-diam oxide-coated cathode followed by a current-controlling grid and an energy-defining grid. Focussing of the electron current, which was typically a few mA, was provided by the Zeeman magnetic field produced by a pair of Helmholtz coils. For the Zeeman resonance part of the experiment an rf magnetic field, which was of the order of a few G at 50 MHz, was generated inside a "double loop" consisting of a wide copper strip bent into an elongated U shape which partially enclosed a narrower strip attached at one end to the base of the "U" and the other end to the center conductor of a coaxial line. This configuration provided good shielding for the relatively intense rf electric fields and minimized spurious effects associated with rf pickup and rf breakdown. The intensity of the radiation emitted in the transition $2\,^3P \rightarrow 2\,^3S$ ($\lambda = 5485$ Å) was measured with a photomultiplier having an S-11 response.

A. Electron-Impact Excitation Cross Section

For the measurements of the excitation function and polarization, the apparatus was arranged in a

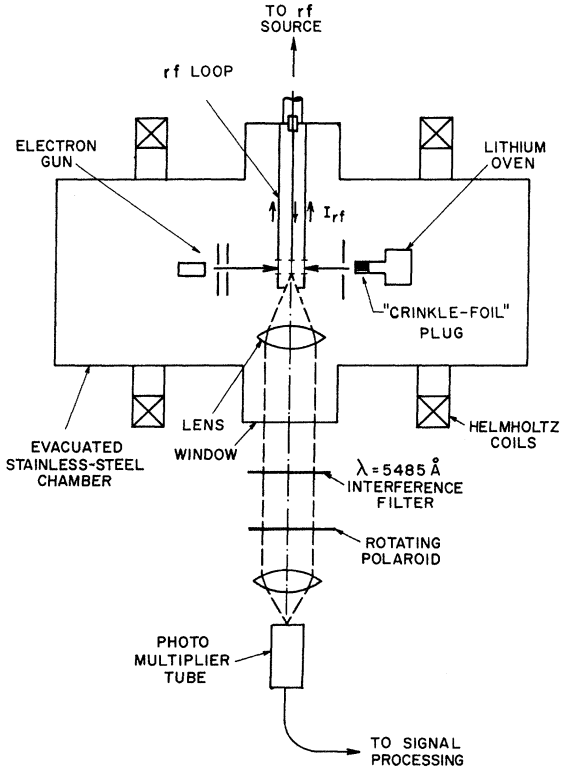

FIG. 2. Experimental setup. The rf loop was removed for the measurements of intensity and polarization as a function of excitation energy.

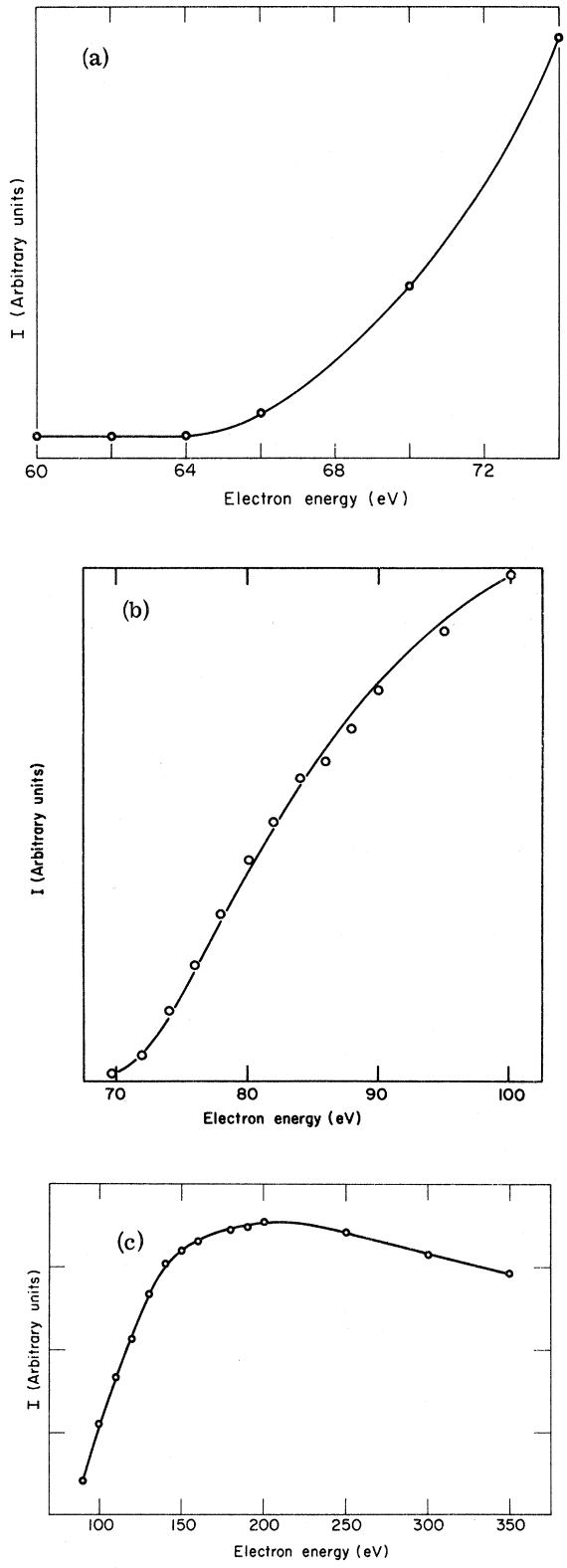

FIG. 3. (a) Excitation function 60–74 eV. The energy resolution of the electron gun was ~1 eV. (b) Excitation function 70–100 eV. (c) Excitation function 100–350 eV.

configuration somewhat different from that shown in Fig. 2. To make the interaction region well defined, the oven was placed so that its nozzle was 7 cm below the centerline of the electron beam in a crossed-beam configuration. The rf loop was removed and a Faraday cup was put in place of the oven. Also not shown in the figure are the optical shields which defined the amount of solid angle collected ($0.1\ \pi$ sr) and which eliminated stray light coming from the incandescent cathode and oven.

The measured excitation function $I(V)$ is shown in Figs. 3(a), 3(b), and 3(c). It is seen that the onset of 5485-Å emission occurs at about 66 eV, the threshold for the one-step process $1s^2\,2s\,{}^2S \rightarrow 1s2p\,{}^3P$. Emission from the two-step excitation $1s^2\,2s\,{}^2S \rightarrow 1s^2\,{}^1S \rightarrow 1s\,2p\,{}^3P$, which has a threshold at 61.2 eV and in which a second collision excites a $1s^2\,{}^1S$ ion trapped in the beam, was not observed in the region below 66 eV. The other two-step process, $1s\,{}^2s\,{}^2S \rightarrow 1s2s\,{}^3S \rightarrow 2s\,{}^3P$, has a threshold at 64.4 eV, which was too close to the 66-eV threshold for our system to clearly resolve. However, when we measured the relationship between the electron beam current and the light intensity we observed a linear dependence which implied that the contribution from both two-step excitations was smaller than the limits of our measurement (~2%). This was to be expected on the basis of estimates which indicated that the emission from such a two-step process would be a few orders of magnitude smaller than the minimum detectable signal.

The intensity of the $\lambda = 5485$ Å line was relatively small and at least several orders of magnitude less than some at the more prominent lines in Li I which we also observed. For instance, under optimum conditions the total measured intensity at 100 eV corresponds to a primary photocurrent of only 2×10^5 photoelectrons/sec. This intensity was close to the maximum achievable without creating conditions which would have interfered with our measurements. For example, attempts to increase the electron-beam current beyond a few mA at the low magnetic fields used in the experiment (~50 G) created problems in the polarization and resonance measurements, probably because of beam defocussing and beam instabilities associated with the high densities. The lithium flux through the crinkle-foil plug was close to a level beyond which there is no longer useful collimation and any increase in flux would have been achieved only at a cost of severely shortened running times.

In order to make a rough determination of the total electron excitation cross section, we estimate the atomic density in the following manner. Because the conditions for Knudsen flow, i.e., mean free path $\gg$ tube length $\gg$ tube radius, would have limited the intensity to an unacceptable low level,

we operated under conditions in which the mean free path was roughly the same as the tube diameter, and Knudsen flow only applied for a certain distance from the discharge end of the tube. In such a situation the tubes are no longer transparent (i.e, many collisions occur during an atomic transit to the discharge end), and the simple formula relating the intensity to the temperature and pressure behind the tubes is no longer applicable. The flow rates and beam profiles for crinkle-foil sources operated in the nontransparent mode were studied for the case of CO_2 by Giordmaine and Wang.[4] They found that the total flow rate N and the intensity in the forward direction $I(0)$ are related by the formula

$$I(0) = [(3r\bar{v})^{1/2}/16.9d]\sqrt{N},$$

where d is the molecular collision diameter, $\bar{v}$ the mean molecular velocity, and r the effective radius of the tube. The flow rate N, which was not found to be easily expressible in terms of the thermodynamic variables of the gas in the source, was measured at various pressures in the source. To convert these CO_2 results for the case of Li, we assumed that, for a given number density behind the source, the dependence of the total flow rate N on the mean velocity $\bar{v}$ is linear and hence

$$N_{\mathrm{Li}}/N_{\mathrm{CO_2}} = \bar{v}_{\mathrm{Li}}/\bar{v}_{\mathrm{CO_2}}.$$

Although this assumption ignores the effects of the molecular size difference and any nonlinear dependence on $\bar{v}$, it is probably more than adequate for an order-of-magnitude estimate. On this basis the calculated density in the collision region was 7×10^{12} atoms/cm^3.

The cross section was measured at 100 eV using electron-beam currents that were considerably below the point at which space-charge defocussing became noticeable. Other relevant factors in the determination were the quantum efficiency of the S-11 photocathode and the over-all gain for the multiplier. The phototube was not calibrated, but the specifications supplied by the manufacturer for that model were assumed to be within 50% of the actual values for the particular tube used. When the calculated density is combined with the other pertinent data to compute the cross section, the result is $3 \times 10^{-22 \pm 1}$ cm^2, with the estimated error mainly attributed to the uncertainty in the atomic density determination.

This measured cross section is several orders of magnitude smaller than the typical cross section associated with the more prominent lines of the Li I spectrum and also with the transitions in He[5] and Hg[6] studied by a similar rf resonance technique. The measured value is also about three orders of magnitude smaller than the value obtained by Wu and Yu with a Born approximation using screened hydrogenic functions for the wave functions of the ground state and the excited state.[7] The validity of their approximations is further questioned by the fact that they calculated a monotonic increase in cross section which extends to approximately 1000 eV, whereas the measured cross section peaks at about 200 eV.

B. Electron-Impact Polarization

The success of a resonance experiment depends on the existence of adequate population differences between the sublevels under study. In order to calculate the sublevel population distribution, one needs to know the excitation cross section for each individual sublevel. The calculation of these cross sections from theory is extremely difficult except in two cases: at the high-energy limit where the Born approximation is valid, and at threshold where conservation of angular momentum about the electron-beam direction may be applied to yield results independent of the details of the interaction (since the angular momentum of the two ejected, zero-energy electrons must be zero in this case).[8] A threshold calculation was performed for both the $2\,^3P$ of ^{6}Li$^+$ and ^{7}Li$^+$ and the threshold population distribution was determined in each case. The calculated polarization of light [defined as the ratio $(I_\parallel - I_\perp)(I_\parallel + I_\perp)^{-1}$, where $I_\parallel$ is the intensity parallel to the electron beam direction and $I_\perp$ perpendicular to it] emitted at threshold is 16.3% for ^{6}Li$^+$ and 9.9% for ^{7}Li$^+$. These threshold values represent the maximum degree of polarization and alignment.

Unfortunately, the extremely low intensity of emitted radiation near threshold prevented any polarization measurements in this region, and a comparison with the theoretical threshold values could not be made. The measured polarization functions $P(V)$ for the region from 70 to 100 eV are shown in Fig. 4. At 73 eV, the lowest energy for which reliable measurements were possible with our system, the polarization is somewhat less than half the calculated threshold value in both cases. It is interesting to note that the ratio $P(V)\ ^6\mathrm{Li}^+/P(V)\ ^7\mathrm{Li}^+$ is roughly constant and approximately equal to the ratio of the calculated values.

C. Observation of Resonance-Induced Transitions between Zeeman Sublevels in Small Magnetic Field

The preceding observations of the small cross section and polarization associated with electron-impact excitation of the $2\,^3P$ term indicated that any resonance measurement would be extremely difficult. The initial attempt involved the most favorable case for ^{7}Li$^+$: transitions induced simultaneously between the almost equispaced $J = 2$, $F = \frac{7}{2}$ sublevels in a small magnetic field. For this case the calculated change in polarization based on threshold conditions is from 9.9% to 7.8% for an

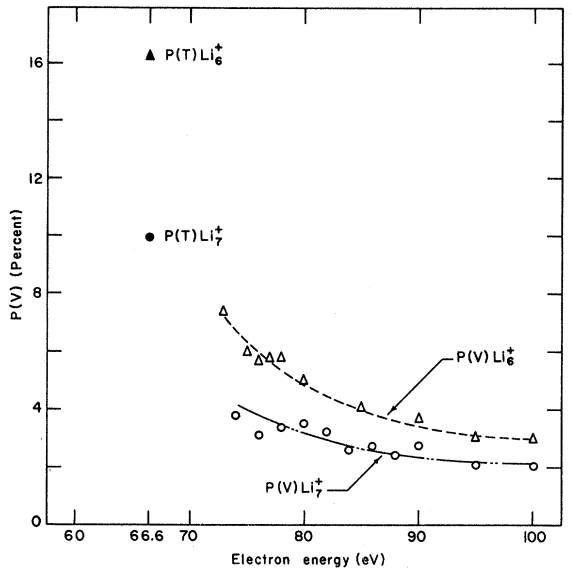

FIG. 4. Polarization function for $^6Li^+$ and $^7Li^+$ between 73 and 100 eV. The points labeled $P(T)$ are the calculated threshold values.

rf magnetic field of 3 G (close to half saturation). Of course, the actual change to be expected when the measured polarization is 3% would be considerably less.

In a small magnetic field H_z, the energy values of the magnetic sublevels are given to second order by

$$E_m(2^3P_J;\ F) = g_F\,\mu_B\,H_z m + \sum_{F'} \frac{|\langle JF'm|V|JFm\rangle|^2}{E_F(2^3P_J) - E_{F'}(2^3P_J)}\ ,$$

where V is the Zeeman Hamiltonian and $E_F(2^3P_J)$ is the zero-field energy of the array of sublevels with total angular momentum F, which were taken from the spectroscopic data of Herzberg and Moore.[9] To produce the maximum signal and simplify the line-shape analysis, we chose to work in a region where the second-order term was negligible compared to the natural linewidth. At $H = 40$ G (resonant frequency ~ 50 MHz) the second-order term for each of the $F=\frac{7}{2}$ sublevels was always less than 10^{-3} MHz. This value compared to a natural linewidth of ~ 7 MHz, and the effect of the term on the line shape was unobservable in our experiment.

A typical resonance signal representing the field-dependent decrease in polarization at 50 MHz is shown in Fig. 5. Because of the poor signal-to-noise ratio, the signal had to be averaged for 120 sweeps (~ 5 h) to produce this result. There are actually two overlapping resonances represented, one from the set of $F=\frac{7}{2}$ sublevels and the other, smaller one, from the set of $J=2$, $F=\frac{5}{2}$ states. Because these two resonances were unresolved, we could not accurately determine the g factors and the line shape simultaneously. Instead we used the factors calculated assuming Russell–Saunders coupling ($g_{7/2}=\frac{6}{7}$ and $g_{5/2}=\frac{33}{35}$) to locate the line centers and then constructed the "separated" Lorentzian indicated by the dashed line in the figure by enclosing an area that was symmetric about the calculated position for the $F=\frac{7}{2}$ resonance. The full width at half-maximum of the "separated" resonance is 6 ± 0.5 G which, under the conditions of a 3-G rf field including the resonances, allows us to determine that the lifetime is 45 ± 5 nsec. This value compares to the value of 43.9 nsec calculated by Weiss[10] and 32 ± 2 nsec measured by Bickel *et al.*[11] in a beam-foil experiment. It is felt that owing to unaccounted effects of cascade which produce difficulties in fitting the observed decay curves to exponentials, the beam-foil value is not as reliable as that indicated by the quoted uncertainty of ± 2 nsec. Calculated lifetimes for light elements such as Li^+ tend to be fairly accurate, and Weiss's value is probably the most accurate of three lifetime determinations. Our uncertainty arises from the difficulty in separating $F=\frac{7}{2}$ Lorentzian from the noisy overlapping signal of the $F=\frac{7}{2}$ and $J=2$, $F=\frac{5}{2}$ resonances. (Note that there are no other interfering resonances on the high-field side of the $F=\frac{7}{2}$ signal; the closest one, the $J=1$, $F=\frac{5}{2}$, occurs at ≈ 59.5 G.) The Russell–Saunders approximation is very good for the $2P$ terms in Li^+, and its use does not contribute any uncertainty at the quoted level.

III. FEASIBILITY OF FINE- AND HYPERFINE-STRUCTURE MEASUREMENTS

The detection of the resonance signal from transitions induced between the $F=\frac{7}{2}$ sublevels proved to be extremely difficult. Normally, a resonance signal may be detected by looking either for the induced intensity change in light polarized parallel to the z axis, $\Delta I_{\parallel}$, or perpendicular to the z axis, $\Delta I_{\perp}$. In our experiment there were spurious over-

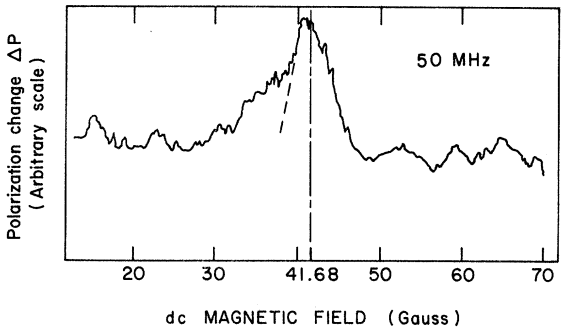

FIG. 5. Resonance signal 2^3P, $^7Li^+$ at 50 MHz. The calculated position of the $F=\frac{7}{2}$ resonance, 41.68 G, is indicated by the broken centerline. The overlapping $F=\frac{5}{2}$ resonance is centered at 37.89 G.

all intensity changes caused by the interaction of the rf electric field with the plasma. The component of the spurious changes which was transmitted in the bandpass of the $\lambda = 5485$ Å interference filter was several times larger than the inherently weak change caused by the resonance. To overcome this problem, we made use of the fact that the polarization of the spurious intensity changes could, with care, be made extremely small while in a resonance involving a small degree of inital alignment the intensity changes are related by $\Delta I_{\parallel} = 2\Delta I_{\perp}$.[12] This distinguishing characteristic was exploited in a "double-lock-in technique": (i) The rf power was square-wave modulated as the Zeeman magnetic field was swept through the resonances; (2) the output of the lock-in amplifier was recorded in a 400-channel signal averager; in alternate sweeps the signal from $I_{\parallel}$ was subtracted from that of $I_{\perp}$ by use of the subtract mode of the signal averager. (This system was later modified by the addition of a motor-driven polarizer and another lock-in detector. The rf power was synchronously square-wave modulated at twice the polarization modulation frequency, and the lock-in amplifiers were tuned to two sidebands: at the polarization modulation frequency and at three times this frequency.)

The signal detected in this manner was about 25 times smaller than that expected on the basis of the threshold calculation. An obvious reason for part of the decrease in signal is that at 90 or 100 eV, where the polarization is down by a factor of 4 from its threshold value, the population differences are considerably less than the threshold values. However, there also must have been considerable signal degradation owing to the noise produced by the rf electric field-plasma interaction. This problem became even more severe when we attempted to measure the hfs splitting in $^6Li^+$ with a 3-GHz field. The signal to be expected based on the threshold calculations was about $\frac{1}{10}$ that of the $F = \frac{7}{2}$ signal, and the magnitude of the spurious effect of the microwave field was greater than that at 50 MHz. As a result it was not possible to obtain reliably reproducible experimental results. In several instances apparent resonance signals have been observed, and further analysis is now being done to provide positive identification.

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