sion, we tuned the laser to the low-frequency side of resonance and again observed conical emission, as was also observed in Ref. 7. The center frequency of this emission was again on the low-frequency side of resonance, in agreement with the model. Because of self-defocusing of the laser beam in this case, experimental conditions under which conical emission occurred were difficult to achieve. The occurrence of conical emission always on the low-frequency side of resonance and the agreement between Eq. (3) and the cone-angle data indicate that the creation of the cone can be explained as largely due to the model discussed above.

Finally, we have measured the temporal pulse shape of the off-axis emission, using a fast silicon photodiode. The pulse duration of this emission was found to be 2 to 3 ns, with no measurable time delay with respect to the incident laser pulse of duration 5 ns. This time dependence, which is independent of the laser detuning and sodium density, is consistent with the above-mentioned model based on four-wave parametric amplification.

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Oscillator Strength for Principal Series Transitions to the High Rydberg States of Potassium

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This Letter reports accurate values for the oscillator strength of potassium, and possible resolution of some outstanding discrepancies concerning the oscillator strengths and photoionization cross sections of this atom. Our results establish a value of $(5.2 \pm 0.5) \times 10^{-21}$ cm^2 for the photoionization cross section at the series limit.

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The oscillator strength of alkali atoms deviates significantly from that of the hydrogen atom because of deviations from a Coulomb potential. The effect of core polarization,[1,2] which was thought to be particularly important for transitions to higher Rydberg states and the continuum, has been dealt with in various semiempirical computations,[3-6] and apparent agreement with experimental values of the oscillator strength has been achieved. On the other hand, the quantum defect method[7] (or Coulomb approximation[8]), which has proved to be very powerful in predicting general features in the spectrum of photoionization, does not appear to be always accurate in calculating the value of oscillator strength for transitions between bound states. This difficulty is compounded further by lack of reliable experimental data, and many discrepancies exist concerning the oscillator strength and photoionization cross sections of alkali atoms. The purpose of this Letter is to report accurate values for the oscillator strength of the $4s$-np transitions (up to $n = 50$) of potassium determined with a single-frequency dye laser, and to discuss possible resolution of some outstanding discrepancies concerning oscillator strengths and photoionization cross sections of potassium. Our results establish a much improved value for the photoionization cross section at the series limit, and mark the first time when this cross section is determined by extrapolation from below the series limit.

The experimental setup used was similar to that reported previously.[9] Briefly, the frequency-doubled output from a single-frequency dye laser was used as the light source. This output was tuned to sweep across various $4s$-np transitions in the principal series of potassium, and traversed an absorption cell containing potassium. The absorption signal thus generated was normalized, passed through a precision logarithmic amplifier, and recorded on a chart recorder. In this manner, an absorption signal as small as one part in 10^3 could be detected. The line oscillator strength was obtained by numerically integrating the experimental line shape over an appropriate frequency interval.

For measurements of the absolute oscillator strengths $f_{3/2}$ and $f_{1/2}$ for $n = 9$, the absorption cell, which measured 30 cm in length, was kept at a uniform temperature some 20 °C higher than that of the potassium source. This latter temperature was varied between 490 and 527 °K, providing a vapor density[10] of 3×10^{14} to 1.3×10^{15} atoms/cm^3. Results averaged over the various densities used give a value of 1.2×10^{-5} for the sum $f_{3/2} + f_{1/2}$, with an absolute uncertainty of about 6%. This uncertainty reflects mainly uncertainties in vapor pressure data (4%) and temperature calibration (4%), with small contributions from data scatter and lack of reproducibility (2%).

Relative oscillator strengths for $n = 7$–50 were measured with the potassium cell operating in the heat-pipe mode, and results were generally reproducible within about 10%. For these relative measurements, the cell was filled with 4 Torr of helium buffer gas, and operated at an average temperature varying between 220 and 300 °C. Under these conditions, the absorption lines were found to be predominantly Doppler broadened. The homogeneous broadening was found to be due mainly to the He buffer gas, and appeared to be the same for all n values within the experimental uncertainty. The separation of the spin-orbit doublets was seen to decrease approximately as the inverse cubic power of the effective quantum number. These doublets were separated by more than one linewidth for $n < 17$, and were still resolvable for $n = 30$. Beyond $n = 30$, the doublet components were no longer distinguishable.

The intensity ratio $f_{3/2}/f_{1/2}$ of the doublet components was measured accurately for $n < 17$, and determined from deconvolution of the overlapping doublet components for $n > 17$. The observed intensity ratio approaches a value of 4.0 ± 0.8 at the series limit (Fig. 1), and agrees with the photoionization results of 3.85 ± 0.45 by Baum, Lubell, and Raith.[11]

Figure 2 shows our experimental results for the oscillator strength of the $4s$-np transitions of potassium as a function of the principal quantum number n. These oscillator strengths represent the sum of the two spin-orbit components for each p state. Here the ordinate represents the product $n^{*3} f (1 - n_s^{*2}/n^{*2})^{-1}$, in which n^* and n_s^* are, respectively, the effective quantum number for the states np and $4s$, derived from the experimental term values of potassium.[12] Also included for comparison are previous experimental results of Filippov[13] and Mazing and Serapinas,[14] and the values suggested by Marr and Creek,[15] which represent, in effect, interpolated results from the "smoothed" data of Filippov and the experimental photoionization cross sections of Marr and Creek.[16] For the purpose of later discussion, the results for lithium[15] are also included in Fig. 2.

For n values for which there are previous results, our values of the oscillator strength in

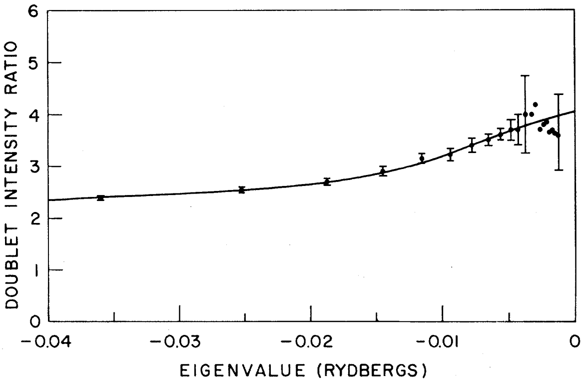

FIG. 1. Plot of doublet intensity ratio as a function of the eigenvalue of the upper p states.

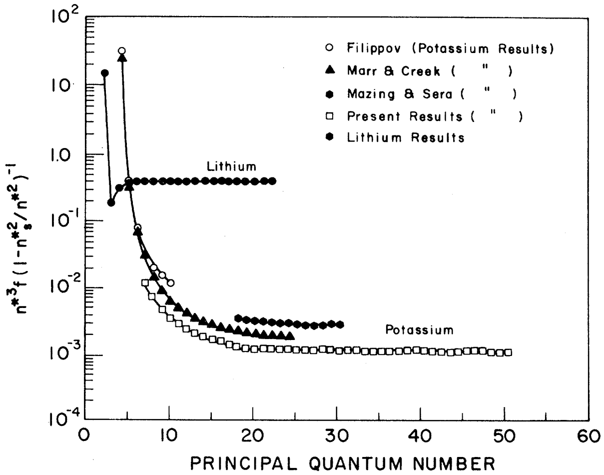

FIG. 2. Semilogarithmic plot of various experimental and interpolated values of $n^{*3}f(1-n_s^{*2}/n^{*2})^{-1}$ as a function of the principal quantum number for potassium. The interpolated values for lithium are also included for comparison.

Fig. 2 are about a factor of 2.5 lower. For $n > 30$ where no previous results exist, our results indicate a constant value of $(1.3 \pm 0.05) \times 10^{-3}$. Our results yield, when extrapolated to the series limit, a value of $(5.2 \pm 0.5) \times 10^{-21}$ cm^2 for the photoionization cross section. This value is somewhat smaller than the corresponding value of $(7 \pm 2) \times 10^{-21}$ cm^2 given by Marr and Creek.[16] Since measurements of discrete lines are, in general, much less susceptible to contaminants and other systematic errors, our determination of the photoionization cross section by extrapolation of the line oscillator strength to the series limit should be inherently more reliable. We should also like to point out that our measurements were made at potassium concentrations about four orders of magnitude lower than those used in measurements of photoionization cross sections. Whereas the presence of potassium dimer is known to account for the discrepancies among various previous measurements,[17] this presence should be of little concern to us on account of the much reduced dimer concentrations over the temperature range used in our measurements, and our use of a tunable, narrow-band laser source for excitation. It is perhaps no surprise that our value is also lower.

Weisheit and Dalgarno[4] were able to reproduce the suggested values of Marr and Creek[16] for the oscillator strength and report a value of 6×10^{-21} cm^2 for the photoionization cross section at the series limit. Examination of their numerical results indicate, however, that our results should be reproducible with a smaller value for the effective cutoff radius r_c used in their computations. We have also noted that the value of r_c chosen for potassium by these authors is higher than that for rubidium.[5] This is contrary to the general trend observed with other alkali ions, which indicates a progressively higher value of r_c for ions of larger sizes. This discrepancy could be resolved if a lower value such as that suggested by our results for potassium was chosen.[17] Whereas it would be interesting to repeat the semiempirical computations in the light of our experiments, it appears certain that our lower value for the photoionization cross section will help bring about a better accord among various parameters determined experimentally and computationally.

In Fig. 2, the quantity $n^{*3}f/(1-n_s^{*2}/n^{*2})^{-1}$ is seen to decrease rapidly but smoothly with increasing n, and tends to approach a constant value for large n. The latter tendency is the expected dependence for transitions to high-lying states of the hydrogen atom, and would imply that the oscillator strength for Rydberg states of potassium behave hydrogenically. It should be noted, however, that the proportionality constant (as deduced from Fig. 2) is 1.3×10^{-3} rather than 9.3 for the $4s$-np transitions[18] of the hydrogen atom, which is higher by nearly four orders of magnitude. It should also be noted that this approach to hydrogenic dependence for the oscillator strength is slower, and occurs at a higher n value, than is that for the quantum defect deduced from energy term values of potassium. The quantum defect is seen to reach its asymptotic value of 1.710 ± 0.005 at $n = 8$, whereas in Fig. 2 the asymptotic value is not reached until $n = 20$. It follows that the oscillator strength for these transitions with intermediate n values cannot be described quantitatively in terms of quantum defects alone, and that the empirical phase parameter introduced in the quantum defect method[7] will have to change rather rapidly for these transitions.

As is well known, the extremely low value for the asymptote noted in Fig. 2 is related to the occurrence of a Cooper minimum located somewhat beyond the series limit in the continuum. This may be contrasted with the case of lithium, for which the Cooper minimum occurs near $n = 3$ in the discrete spectrum. Since the precise location of the Cooper minimum also bears the influence of this rapidly changing phase parameter, it would be interesting to correlate with the experimental phase parameter values the location of the Cooper minimum and the shape of the photoionization cross section as a function of energy near this minimum.

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