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Stark broadening of potassium *ns*-4*p* and *nd*-4*p* lines in a wall-stabilized arc

J. P. Hohimer
Sandia National Laboratories, Albuquerque, New Mexico 87185

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Stark-width measurements are reported for lines in the *ns*-4*p* ($n=7-10$) and *nd*-4*p* ($n=5-8$) series in neutral potassium (K I). These measurements were made by observing the end-on emission from a low pressure (20 Torr) potassium-argon wall-stabilized arc source. The on-axis electron density and temperature in the 20-A arc were $(2.0\pm0.2)\times10^{15}$ cm^{-3} and 2955±100 K, respectively. The experimentally determined Stark widths were compared with the theoretical values calculated by Griem. The measured Stark widths agreed with theory to within 30% for lines in the *ns*-4*p* series; while the measured Stark widths of the *nd*-4*p* series lines were only one-third of the theoretical values.

I. INTRODUCTION

The alkali-metal atom emission spectra present a large number of isolated and hydrogenic lines that should provide a good test for Stark broadening theories and that would be of great use in determining electron densities in alkali-metal-seeded plasmas. Very few experimental measurements however have been reported for the Stark widths of excited-state lines of the alkalis.[1–6] Indeed, we are aware of only one instance[6] in which measurements have been made in a wall-stabilized arc source, although it is the preferred source for such measurements. This lack of experimental data is largely due to the difficulty of generating a stable, well-characterized alkali-metal-seeded plasma in which to make the measurements.

In the case of potassium, the only reported Stark linewidth measurements are for the 4*p*-4*s* resonance lines[7,8] which, due to their small Stark widths and large oscillator strengths, are of limited use for plasma diagnostics. The higher-lying excited-state transitions in the *ns*-4*p* and *nd*-4*p* series have larger Stark widths and smaller oscillator strengths thus making them better suited for diagnosing electron densities in plasmas. In addition, these higher-lying transitions will be in partial local thermodynamic equilibrium (PLTE) at a much lower electron density than the 4*p*-4*s* resonance lines.

In this paper, we report measurements of the Stark widths of excited-state transitions in the *ns*-4*p* ($n=7-10$) and *nd*-4*p* ($n=5-8$) series of potassium. These measurements, which to our knowledge are the first for potassium excited-state transitions, were made in emission in a potassium-seeded argon wall-stabilized arc source. This arc source operates at a reduced pressure (20 Torr) to minimize the large contribution of argon collisional broadening to the measured line shape.[9,10] The on-axis electron density ($2.0\pm0.2\times10^{15}$ cm^{-3} at 20 A current) in the potassium-argon plasma was independently determined by far-infrared laser interferometry; and the electron temperature (2955±100 K) was determined from Boltzmann plots of the relative emission intensities of several potassium lines in the *ns*-4*p* series.

II. EXPERIMENTAL APPARATUS

A schematic diagram of the experimental apparatus used to measure the Stark broadening of potassium excited-state transitions is shown in Fig. 1. The plasma source was a low pressure potassium-seeded argon wall-stabilized arc which is described in detail elsewhere.[11] The arc was constricted by an alumina channel (9.5 mm diameter by 7.6 cm long) and was free burning in the electrode regions at either end of the channel. A hollow cylindrical anode and an off-axis rod cathode (98% W, 2% Th) were used to provide an unobstructed optical path along the arc axis for end-on observation and laser interferometry. *Z*-cut crystalline quartz end windows were used to provide a high transmission in the visible and far-infrared regions of the spectrum.

Potassium metal (99.95% pure) was heated in a dual-stage oven to a temperature of 340 C, corresponding to a potassium partial pressure of about 1 Torr.[12] Pure argon flowed through the oven and carried the potassium vapor into the arc through a heated stainless-steel tube. The argon-entrained potassium seed entered the arc channel near the anode end, flowed down the channel, exited near the cathode end, and was trapped in a water-cooled chamber. Reverse flows of argon entered the arc near the end windows to blanket the electrode regions thus

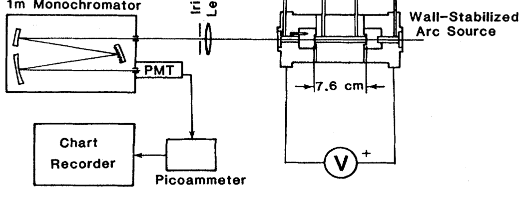

FIG. 1. Experimental apparatus for Stark broadening measurements.

preventing the diffusion of potassium into these cooler regions of the arc. During the experiment, the arc was operated at a current of 20 A and a pressure of 20 Torr.

The optical system consisted of a 1-m Czerny-Turner-type monochromator (Jarrell-Ash, model 75—150) equipped with a 1200 line/mm grating yielding a reciprocal dispersion of 8 Å/mm. The central region of the plasma was imaged end-on (1 to 1 magnification, beam aperture $f/100$) onto the entrance slit of the monochromator. The entrance and exit slits were set at 20 μm and the entrance slit height was set at 250 μm. The instrumental width was determined to be 0.42 Å by scanning across the $7S_{1/2}$-$4P_{1/2}$ line of potassium from a spectral lamp (Osram). The output current from the photomultiplier tube (RCA, C31034) was fed into a picoammeter (Keithley Instruments, model 410A) and subsequently recorded on a strip chart recorder for analysis. The wavelength markers from the monochromator were simultaneously recorded to provide a calibration for the linewidth measurements.

III. PLASMA DIAGNOSTICS

Accurate estimates or measurements of the electron temperature and density in the plasma are required for the interpretation of Stark broadening measurements. We used emission measurements of K I lines to determine the on-axis electron temperature[13] and far-infrared laser interferometry[14–17] to independently determine the on-axis electron density.

In order to determine the on-axis electron temperature in the arc source, the spectral response of the optical train (lens, monochromator, photomultiplier) was calibrated using a tungsten quartz iodine standard lamp (Eppley Laboratory). The electron temperature could then be inferred from a Boltzmann fit of the relative emission intensities of several K I lines in the ns-$4p$ series using the relative transmission probabilities of Aeschliman[18] and assuming LTE. From these measurements shown in Fig. 2, we determined the on-axis electron temperature to be 2955±100 K at 20 A current.

In order to determine the on-axis path-averaged electron density in the plasma, the arc was located in one arm of a dual-pass Michelson interferometer. A CO_2 laser pumped CH_3OH far-infrared laser (Advanced Kinetics, model FIRWL-50-300) operating at a wavelength of 118.8 μm was used as the source for these fringe shift measurements. A fast thyristor was used to short circuit the arc and the resultant electron decay fringe shift was detected and photographed for analysis. The arc was then automatically reignited after a time interval of 200 μs and the operating conditions reestablished.[11] The arc could also be completely extinguished in order to observe the entire electron decay fringe shift pattern.

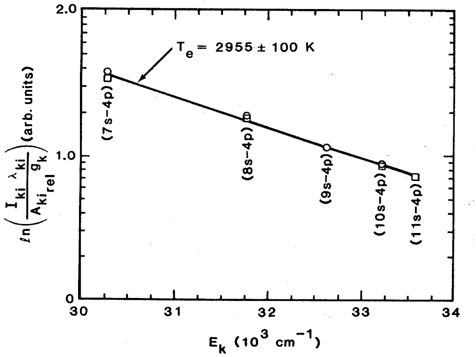

FIG. 2. Plot of $\ln(I\lambda/Ag)$ against excitation energy for excited-state lines of potassium. ○, $nS_{1/2}$-$4P_{1/2}$; □, $nS_{1/2}$-$4P_{3/2}$.

With potassium seeding of the arc, the on-axis electron density was measured for various operating currents in the range 4—16 A.[11] Measurements at higher arc currents were not possible with our dual-pass far-infrared laser interferometer because of the loss of fringe contrast due to inverse bremsstrahlung absorption[19,20] and/or refractive effects[14,20–22] in the plasma.

Three sets of electron density measurements were made in order to estimate the random error due to uncertainties in the fringe shift measurements and fluctuations in the potassium seed rate. In order to minimize this latter source of error, the potassium oven was cleaned and recharged prior to each measurement. The arc was then operated at 20 A current with the potassium oven switched on and the entraining gas flowing for approximately one hour before performing the electron density measurements. This same procedure was followed for the Stark linewidth measurements.

Figure 3 shows the results of these measurements which were used to determine the electron density at the current chosen for the Stark broadening measurements (20 A). From an extrapolation of our measured results to 20 A current, we estimated the on-axis electron density for our Stark broadening measurements to be $2.0\pm0.2\times10^{15}$ cm^{-3}. This electron density was about twice that measured at this same current for a pure argon flow ($9.3\pm0.5\times10^{14}$ cm^{-3}) due to the large fractional ionization of the potassium seed in the plasma.[11]

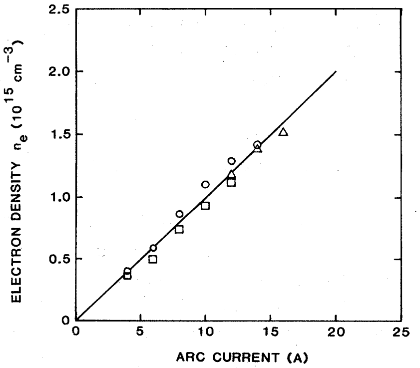

FIG. 3. Variation of the electron density with current in the potassium-seeded arc source as determined by far-infrared laser interferometry. From the three sets of electron density measurements (indicated by the different symbols), the electron density at 20 A current was estimated to be $2.0\pm0.2\times10^{15}$ cm^{-3}.

IV. RESULTS AND DISCUSSION

All emission line profile measurements were performed end-on along the arc axis. Although radial demixing or cataphoresis[6,11,23] occurred in the mixed gas plasma, this did not affect the line-shape measurements because of the small *f*-number (*f*/100) of the collection optics.

In extracting the Stark widths from the measured line profiles the contributions due to Doppler, collisional, and instrumental broadening were considered. The resonance broadening contribution to the line shapes was completely negligible. From the on-axis electron temperature (2955±100 K) an estimate of the Doppler broadening contribution to the total width of each line was made. The Doppler broadening contribution decreased with increasing upper state quantum number (from 5.2% for the $7S_{1/2}$-$4P_{1/2}$ line to 1.2% for the $10S_{1/2}$-$4P_{1/2}$ line and from 4.6% for the $5D_{5/2}$-$4P_{3/2}$ line to 0.8% for the $8D_{5/2}$-$4P_{3/2}$ line). Since the contribution of Doppler broadening amounted to only a small fraction of the total linewidth, it was neglected in arriving at the experimental Stark widths.

Because the argon collisional broadening coefficients were known to be very large for the potassium excited-state lines,[9,10] the arc source was designed to operate at a greatly reduced pressure to minimize this source of broadening. At the operating pressure of 20 Torr, the argon collisional broadening contribution was estimated to be less than 5% of the total linewidth for each of the measured transitions. Therefore, this source of broadening was also neglected. The instrumental width was determined by using the $7S_{1/2}$-$4P_{1/2}$ line from a low-current potassium spectral lamp thereby compensating in part for our neglect of Doppler and collisional broadening. The Stark (electrons and ions) linewidth was then simply obtained by subtracting the instrumental width from the measured linewidth.

The experimentally determined Stark widths (full widths at half maximum) for eight lines in the $nS_{1/2}$-$4P_{1/2}$ and $nD_{5/2}$-$4P_{3/2}$ series of potassium are listed in Table I. These lines were selected because they were free from overlaps and thus well suited for plasma diagnostics. For comparison, we have also listed in Table I the theoretical Stark widths of Griem[24] normalized to our on-axis electron density. Since the Stark widths are not strongly dependent on temperature, no attempt was made to normalize the theoretical widths to our measured electron temperature.

TABLE I. Experimental Stark widths of potassium excited-state transitions ($N_e = 2\times10^{15}$ cm^{-3}) compared with theory.

Transition	Wavelength (nm)	Full width (Å) Measured	Full width (Å) Calculated[a]
$7S_{1/2}$-$4P_{1/2}$	578.2	0.27±0.13	0.35
$8S_{1/2}$-$4P_{1/2}$	532.3	0.54±0.25	0.70
$9S_{1/2}$-$4P_{1/2}$	508.4	1.21±0.53	
$10S_{1/2}$-$4P_{1/2}$	494.2	2.25±0.95	
$5D_{5/2}$-$4P_{3/2}$	583.2	0.37±0.17	1.10
$6D_{5/2}$-$4P_{3/2}$	536.0	0.72±0.33	1.98
$7D_{5/2}$-$4P_{3/2}$	511.2	1.53±0.66	
$8D_{5/2}$-$4P_{3/2}$	496.5	3.25±1.35	

[a] From Ref. 24 with $T_e = 5000$ K.

The uncertainty in the measured Stark widths due to random errors in determining the electron density and the total linewidths is estimated to be about 20%. The uncertainty due to systematic errors is estimated to be in the range of 20—30% and arises from the following sources: uncertainty in the electron density due to variations in the potassium-seed rate over the course of the line-shape measurements (10%); uncertainty due to the neglect of Doppler and collisional broadening (0—10%); and uncertainty in determining the instrumental width (10%). Therefore, the total error for these Stark width measurements is estimated to be in the range 40—50%.

The experimentally measured Stark widths for the $7S_{1/2}$-$4P_{1/2}$ and $8S_{1/2}$-$4P_{1/2}$ lines of potassium are in good agreement with theory (see Table I). In the case of the higher-lying *ns*-4*p* lines there are no theoretical calculations with which to compare.[25] However, the measured Stark widths for these lines show a smooth dependence on the upper-state quantum number *n*′, as expected.[26] This is shown in Fig. 4 along with the theoretical widths calculated by Griem.

In the case of the $5D_{5/2}$-$4P_{3/2}$ and $6D_{5/2}$-$4P_{3/2}$ potassium lines, there is a substantial disagreement between the measured Stark widths and theory with the measured widths being a factor of three smaller than the theoretical values. We are unable to account for this large discrepancy since our experimental conditions satisfied all the usual criteria for the theoretical calculations: classical path treatment of electrons, neglect of Debye shielding, and isolation of the upper states.[4] The Stark widths of the measured lines for the *nd*-4*p* series again show a smooth dependence with *n*′ whereas this is not as evident for the theoretical widths (see Fig. 4). This may indicate a need

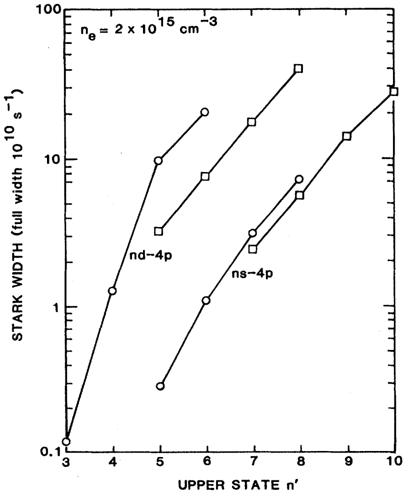

FIG. 4. Stark widths of the *ns*-4*p* and *nd*-4*p* lines of K I vs the upper-state quantum number *n*′; ○, theoretical widths, □, measured widths.

to reexamine the theoretical calculations for the higher-lying $5d$-$4p$ and $6d$-$4p$ lines.

V. CONCLUSION

We have reported the first experimental measurements of the Stark widths of excited-state transitions in potassium. The measured Stark widths for potassium lines in the ns-$4p$ series are in good agreement with theory. However, the measured Stark widths for lines in the nd-$4p$ series are a factor of 3 smaller than the theoretical values.

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[1] V. Kitaeva and I. Sobolev, Opt. Spektrosk. **1**, 302 (1955).

[2] P. M. Stone and L. Agnew, Phys. Rev. **127**, 1157 (1962).

[3] S. M. Gridneva and G. A. Kaslov, in *Proceedings of the Seventh International Conference on Phenomena in Ionized Gases, Belgrade, 1965,* edited by B. Perovic and D. Tosić (Gradjevinska Knjiga, Belgrade, Yugoslavia, 1966), Vol. II, pp. 581–584.

[4] R. F. Majkowski and R. J. Donohue, Phys. Rev. **173**, 177 (1968).

[5] P. E. Oettinger and J. Cooper, J. Quant. Spectrosc. Radiat. Transfer **9**, 591 (1969).

[6] J. Grumberg, G. Coulaud, and Nguyen-Hoe, Phys. Lett. **A57**, 227 (1976).

[7] J. Puric, J. Labat, S. Djenize, Lj. Cirkovic, and I. Lakicevic, Phys. Lett. **A56**, 83 (1976).

[8] J. Puric, J. Labat, Lj. Cirkovic, I. Lakicevic, and S. Djenize, J. Phys. B **10**, 2375 (1977).

[9] J. P. Hohimer, Phys. Rev. A **23**, 1110 (1981).

[10] J. P. Hohimer and J. Gee, Phys. Rev. A **25**, 1404 (1982).

[11] J. P. Hohimer (unpublished).

[12] B. Shirinzadeh and C. C. Wang, Appl. Opt. **22**, 3265 (1983).

[13] H. Griem, *Plasma Spectroscopy* (McGraw-Hill, New York, 1964).

[14] R. A. Alpher and D. R. White, in *Plasma Diagnostic Techniques,* edited by R. H. Huddelstone and S. L. Leonard (Academic, New York, 1965), pp. 431–475.

[15] K. R. Hearne and N. Konjevic, Z. Phys. **204**, 443 (1967).

[16] M. Brunger and M. Kock, Z. Naturforsch, Teil A **30**, 1560 (1975).

[17] C. O. Weiss and K. Teuber, Opt. Commun. **16**, 399 (1976).

[18] D. P. Aeschliman, J. Quant. Spectrosc. Radiat. Transfer **25**, 221 (1981).

[19] V. L. Berezhnyl and V. I. Kononenko, Sov. Phys. Tech. Phys. **20**, 1331 (1976).

[20] J. Glasser, R. Viladrosa, and J. Chapelle, J. Phys. D **11**, 1703 (1978).

[21] D. Baum, J. Hackmann, and J. Uhlenbusch, Plasma Phys. **17**, 79 (1975).

[22] P. Baessler and M. Kock, J. Phys. B **13**, 1351 (1980).

[23] L. M. Chanin, in *Geseous Electronics,* edited by M. N. Hirsh and H. J. Oskam (Academic, New York, 1978), Vol. I, pp. 133–171.

[24] H. Griem, *Spectral Line Broadening in Plasmas* (Academic, New York, 1974).

[25] H. Griem (private communication).

[26] W. L. Wiese and N. Konjevic, J. Quant. Spectrosc. Radiat. Transfer **28**, 185 (1982).