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Hyperfine splittings and isotope shifts in eight transitions from the metastable $4p^5\,5s$ $J=2$ and $J=0$ states of Kr I

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We report isotope shifts for eight transitions in Kr I for all the stable isotopes and hyperfine constants for the $1s_5$, $2p_9$, $2p_8$, $2p_7$, $2p_6$, $2p_4$, $2p_3$, $3p_9$, and $4d'_4$ states in ^{83}Kr. We used continuous-wave single-frequency lasers to perform resonance ionization on an atomic beam of krypton atoms in the metastable $1s_5$ and $1s_3$ states of the $4p^5\,5s$ configuration. Our results agree well with previous work and with calculations of the hyperfine constants for ^{83}Kr in the $4p^5\,5p$ states.

I. INTRODUCTION

The spectroscopy of krypton has been studied at very high resolution by many authors, who reported energy levels,[1] hyperfine splittings,[2-9] and isotope shifts.[7-14] In spite of all this work, there are important gaps in the available data on the stable isotopes of krypton. Data on the isotope shifts and the hyperfine splittings of all the stable krypton isotopes are required for evaluating the feasibility of proposed ultratrace isotope analysis schemes[15] for ^{81}Kr and ^{85}Kr. Complete isotope shift data are known for only two of the seven near-infrared transitions from the metastable $1s_3$ and $1s_5$ states. These transitions are to states with the configuration $4p^5\,5p$. While isotope shifts for six of these transitions have been reported for the even krypton isotopes,[7,11,12] the ^{83}Kr shifts are missing for five of them. Of the eight $4p^5\,5p$ states with $J\neq 0$, the ^{83}Kr hyperfine constants, A and B, are known for only five of them.[3-8] In an effort to fill in the gaps in the spectroscopic data on krypton, we have used resonance ionization with cw lasers to measure the isotope shifts and hyperfine spectra of a number of transitions originating from the metastable $1s_5$ and $1s_3$ states. We report four of these missing ^{83}Kr shifts and more precise even isotope shifts for six of these near-infrared transitions. Table I shows the transitions we have studied in both Racah and Paschen notation and the transition wavelengths.

TABLE I. Paschen and Racah notation for the transitions studied in this work and their vacuum wavelengths. The $1s_5$-$4d'_4$ transition is a two-photon transition.

Paschen	Racah	Wavelength (nm)
$1s_3$-$2p_3$	$5s'[1/2]_0$-$5p'[1/2]_1$	785.7
$1s_3$-$2p_4$	$5s'[1/2]_0$-$5p'[3/2]_1$	806.2
$1s_5$-$2p_6$	$5s[3/2]_2$-$5p[3/2]_2$	760.4
$1s_5$-$2p_7$	$5s[3/2]_2$-$5p[3/2]_1$	769.7
$1s_5$-$2p_8$	$5s[3/2]_2$-$5p[5/2]_2$	810.7
$1s_5$-$2p_9$	$5s[3/2]_2$-$5p[5/2]_3$	811.5
$1s_5$-$3p_9$	$5s[3/2]_2$-$6p[5/2]_3$	432.0
$1s_5$-$4d'_4$	$5s[3/2]_2$-$5d[7/2]_4$	2×811.1

It is interesting to compare the hyperfine constants of the $4p^5\,5p$ states of ^{83}Kr with the theoretical calculations[4] of Husson *et al.* Brandenberger[6] recently used diode lasers to measure previously unknown hyperfine constants for three states. These results are an independent test of the semiempirical theory of Husson *et al.* The theoretical predictions agree with Brandenberger's results to within his error estimates. Our results provide a more stringent test of this theory by determining three previously unmeasured hyperfine constants and four of the constants Brandenberger reported to a factor of 10 higher precision.

We also studied the two-photon transition $1s_5$-$4d'_4$. This transition has an enormous cross section for one-color two-photon excitation because the virtual level is only 7 cm^{-1} from the $2p_9$ level. Excitation to the $4d'_4$ level is potentially a very efficient and selective path for resonance ionization of krypton $1s_5$ metastables.[16]

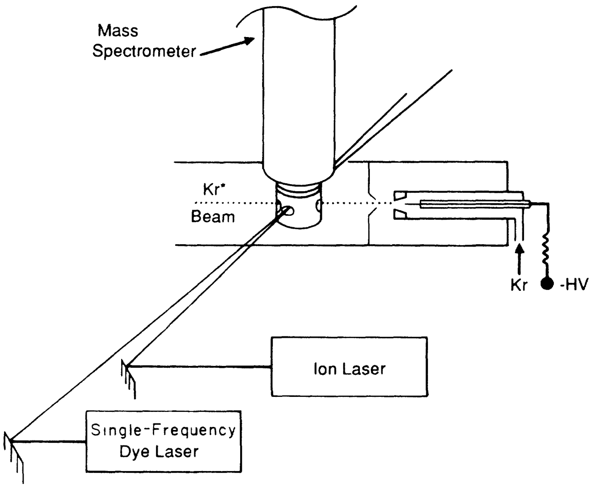

FIG. 1. Experimental schematic of cw resonance ionization mass spectroscopy apparatus for spectroscopic studies on krypton metastables Kr* in a supersonic atomic beam.

II. EXPERIMENT

Figure 1 is a schematic of our experimental apparatus. The source of kyrpton metastables ($1s_5$ and $1s_3$ states) is similar to that of Verheijen *et al.*[17] A discharge is struck from a cathode through a dielectric nozzle to a grounded skimmer cone, with typical discharge currents of 7 mA. The supersonic beam of krypton metastables entered the ionizer of a quadrupole mass spectrometer where it perpendicularly intersected the dye and ionizing laser beams. Electrons from the krypton metastable source were excluded from the ionizer by an electrode (not shown) between the skimmer and the ionizer entrance. The ions were filtered by a mass spectrometer, detected by a channeltron, and counted by a multichannel scaler. Counting times per channel were either 0.1 or 1 s depending on the signal levels.

The atomic-beam divergence was 8 mrad [full width at half maximum (FWHM)] and a standing-wave single-frequency dye laser (Coherent 599-21) was used except for the $1s_5$-$3p_9$ transition. We collected the data on this transition during our measurements of isotopically selective optical deflection of metastable krypton $1s_5$ atoms,[18] when our atomic-beam system had a different configuration. The beam collimation during those experiments was 0.8 mrad and a ring dye laser (Coherent 699-21) was used for excitation. The dye-laser frequency was controlled by a fringe offset lock technique described in detail elsewhere.[19] This control system reduces drift and gives a calibrated linear frequency scan. We offset scans made with different locks on the dye-laser control system to minimize the variance between all overlapped peaks. The ionizing laser beam was typically 1–5 W from an argon-ion or krypton-ion laser. For some two-photon excitation spectra of the $1s_5$-$4d'_4$ transition, the focused dye laser (70 mW) produced sufficient ionization. The laser beams were unfocused except for the $1s_5$-$3p_9$ and $1s_5$-$4d'_4$ transitions and the dye-laser beams were attenuated to the point that power broadening was small. The linewidths, typically 10–15 MHz (FWHM) were due to lifetime broadening (≈ 5 MHz), residual Doppler broadening (≈ 2 MHz), laser jitter (≈ 5 MHz), power broadening, and optical pumping.[20] Peaks were fit to Lorentzian line shapes using a nonlinear least-squares program to find the line centers.

III. RESULTS AND DISCUSSION

The hyperfine energies in each level are given by

$$\nu_F=\nu_J+A(C/2)+B\frac{\frac{3}{4}C(C+1)-I(I+1)J(J+1)}{2I(2I-1)J(2J-1)} \qquad (1)$$

where F is the total angular momentum quantum number, J is the electronic angular momentum quantum number, I is the nuclear spin quantum number ($\frac{9}{2}$ for ^{83}Kr), ν_J is the unperturbed energy of the state, $C=F(F+1)-I(I+1)-J(J+1)$, A is the magnetic hyperfine constant, and B is the electric quadrupole hyperfine constant. Using Eq. (1), peak positions relative to the ^{86}Kr peak can be expressed in terms of five constants (three if one of the states has $J=0$). We determined these constants by fitting the observed peak positions using a weighted linear least-squares fit. The peak positions were the average of typically four to six scans of each resolved line and the weighting was the one standard deviation error estimate of the average peak positions. These typically were about 1 MHz.

Figure 2 shows one of our hyperfine spectra for the $1s_5$-$4d'_4$ transition of ^{83}Kr. The abscissa in this graph is the relative transition frequency, that is, twice the change in dye-laser frequency. This two-photon spectrum was taken with a focused 70-mW dye-laser beam; only the dye-laser beam was used for photoionizing the upper level. These data were taken on an isotope with 11.5% abundance and on atoms in a metastable state 9.9 eV above the ground state with a fractional excitation of $\approx 10^{-4}$. The oscillator strength of this transition is split among 25 allowed lines whose relative strengths span 3 orders of magnitude. They are all observable in this data set, but one is too weak to be seen in this plot. In addition, the weak peak at $\nu=0$ is from 0.05% of the ^{84}Kr ions leaking through the mass spectrometer. These data clearly demonstrate the power and sensitivity of cw resonance ionization when combined with a mass spectrometer.

Table II shows our results for the hyperfine constants in ^{83}Kr for the nine states we measured. The error estimates are from propagation of the peak position error estimates through the weighted linear least-squares fit. Table II also shows previously published results for these hyperfine constants. The general agreement of our results with previous results is good with only six cases out of 23 where the error estimates do not overlap; all agree to within twice the error estimates.

We determined the $1s_5$ state's hyperfine constants from six different transitions. The error estimate for those constants is the one standard deviation error estimate from the weighted average of the six determinations. The entries for the $1s_5$ state in the "nonlaser" column in

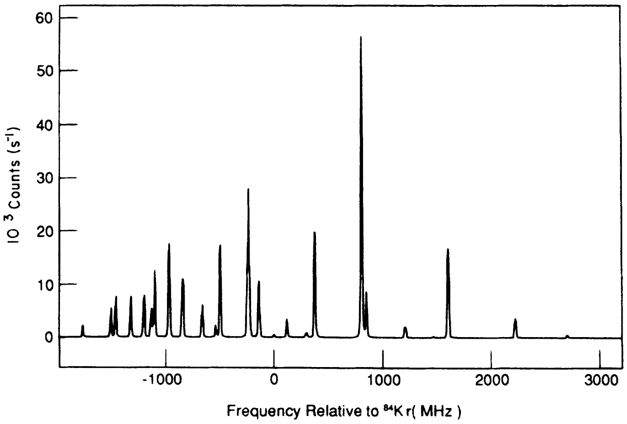

FIG. 2. Hyperfine spectrum of the two-photon $1s_5$-$4d_4$ transition in ^{83}Kr taken with 70 mW of dye-laser power for two-photon excitation and one-photon ionization. The small peak at 0 MHz is ^{84}Kr leaking through the mass spectrometer.

TABLE II. Hyperfine constants (in MHz) for ^{83}Kr from this work and other reports in the literature. Values in parentheses denote one standard deviation error estimates based on the last digits shown.

State		This work	Nonlaser	Ref. 4	Ref. 8	Ref. 6	Ref. 7	Theory[a]
$1s_5$	A	−243.93(4)	−243.96900[b]		−243.99(4)[c]			−239.54
	B	−452.93(60)	−452.157(2)		−452.0(5)			−449.69
$2p_3$	A	226.47(16)	226.6(6)[d]	226.85(20)	226.0(3)	226.8(4)	228.7(15)	228.44
	B	26.5(12)	21.0(30)		25.4(20)	22.0(20)	27(6)	20.69
$2p_4$	A	−576.68(16)	−576.8(5)[d]	−576.9(12)				−571.11
	B	18.6(12)	22.8(30)					24.28
$2p_6$	A	−108.61(11)						−108.53
	B	−88.6(15)						−80.94
$2p_7$	A	−176.80(18)		−177.03(24)				−179.58
	B	−66.7(11)						−68.05
$2p_8$	A	−156.49(8)				−156.0(10)		−158.99
	B	−407.7(13)				−410(20)		−407.72
$2p_9$	A	−104.02(6)				−103.0(10)		−103.13
	B	−436.9(17)				−430(30)		−431.70
$3p_9$	A	−100.62(7)			−100.65(3)			
	B	−452.0(17)			−451.6(5)			
$4d'_4$	A	−73.62(12)						
	B	−459.1(40)						

[a] Reference 4.
[b] Reference 2.
[c] Weighted average of two values in Ref. 8.
[d] Reference 3.

Table II are from atomic-beam microwave experiments[2] that have a precision more than 100 times better than our work. Our A value agrees with the atomic-beam microwave result to within our 0.02% error estimate. Our B value for this state disagrees with the atomic-beam microwave result by 0.2%, which exceeds our error estimate by 30%. The precision of our measurements and the agreement of our results with the atomic-beam microwave results demonstrate the good linearity and accuracy of our laser control system.

It is interesting that the $2p_3$ state has been such a favorite of spectroscopists with five previously reported values for A and four for B. The agreement of all the A values for this state is excellent except for the value from Champeau and Keller,[7] which disagrees by 1.4 times its estimated error from the weighted average value of 226.56(11) MHz. The small B value for this state seems difficult to determine based on the range of reported values. Neglecting Champeau and Keller's value, whose large error bars encompass all the results, the measured B values cluster around two values: Jackson[3] and Brandenberger[6] report values near 22 MHz while our work and that of Gerhardt *et al.*[8] find a value near 26 MHz. This discrepancy may be related to Jackson's and Brandenberger's studies using the $1s_3$-$2p_3$ transition, which has only three hyperfine lines for determining A and B for the $2p_3$ state. Assuming a +3-MHz error in determining the position of the central peak in the spectra of Jackson and Brandenberger would resolve this disagreement. This would not significantly change their A's ($\approx$0.05 MHz) and would give B's of 25 MHz. A 3-MHz error is within Jackson's error estimates for his measured splittings. In Brandenbergers's work, this central peak with its 100-MHz linewidth is only partially resolved from the slightly stronger peak due to the even isotopes in his isotopically enriched sample. The assumed error is in the expected direction, towards the even isotope peak, and 50% larger than his error estimate on the splitting. Our measurement is not subject to this systematic error because of our narrower linewidths and the mass spectrometer's high rejection ($>10^3$) of the even masses. The work of Gerhardt *et al.*[8] used the $2p_3$-$1s_5$ transition, which has seven of its nine lines well separated from the even isotopes. We think the best estimate of B for this state is 26.2(10) MHz, the weighted average of our result with that of Gerhardt *et al.*

The results of the semiempirical study of Husson *et al.*[4] of the hyperfine constants in the $5s$ and $5p$ manifolds in krypton are also in Table II. Husson *et al.* used experimental A and B constants from the $5s$ states with $J\neq0$ and the $2p_7$, $2p_4$, and $2p_3$ states to fit radial monoelectronic hyperfine parameters from which they predicted the A and B constants for the states in the $5s$ and $5p$ manifolds. Their predictions agree with our results to within ±1% for the A's except in the $2p_7$ state, which disagrees by 1.6%. It is curious that this is one of the states that was used in fitting the parameters of the theory and that the experimental value of Husson *et al.*

agrees well with ours. The agreement on the *B*'s is not as good. The agreement is good ($<2\%$) for the three lowest-energy $5p$ states we examined, fair ($<10\%$) for the next state, the poor (20–30 %) for the upper two $5p'$ states. The two *B* values that have the largest disagreement with our results are the only two *B* values from the $5p$ manifold that went into the fit of the radial monoelectronic hyperfine parameters. This may reflect the disagreement between our experimental *B* values and Jackson's, which Husson *et al.* used. If errors in Jackson's *B* values caused the errors in the theoretical *B* values, then it is surprising that the other *B* values agree as well as they do. Another way of looking at the errors is to look at absolute rather than the fractional differences. Looked at this way, the 5.8- and −5.7-MHz disagreements for, respectively, the $2p_3$ and $2p_4$ states are consistent with the −7.7- to +1.3-MHz disagreements for the other four states. Thus it is probably the small magnitudes of the *B*'s for the two $5p'$ states that are responsible for the large relative error.

Table III shows our results on the isotope shifts of the transitions we measured along with previous results in the literature. Our error estimates represent the one standard deviation error estimate from the average of typically four to six measurements for the even isotopes. For the 86-83 isotope shift, the error estimate comes from the linear least-squares fit of the hyperfine structure described above. The normal mass effect is responsible for 80–90 % of the shifts for these transitions except for the $1s_5$-$4d'_4$ transition where it represents 70–80 % of the observed shift. The residual shifts, i.e., the difference between the observed shift and the normal mass shift, are small. If estimates of the specific mass and the nuclear field contributions to the residual isotope shifts are to be obtained then high precision is needed in the measurement of the isotope shifts.

Jackson[11,13] exhaustively studied the isotope shifts of the even isotopes of krypton using a Fabry-Perot interferometer and a krypton-helium discharge. The average of the absolute value of the difference between our measurements and those of Jackson is 3.5 MHz with a range of 0.0–7.1 MHz. The difference exceeds the combined error bars in 13 of 28 cases but there is no systematic trend. Increasing the combined error bars by 50% would resolve all these disagreements. The ^{83}Kr-^{86}Kr disagreements with Jackson are 10.3 and 0.2 MHz. We believe our data are more accurate for three reasons; (1) the difficulty of Jackson's experiment, which measured the shift between a pair of Doppler broadened lines to $\approx 0.3\%$ of the linewidths, (2) the good agreement of our data on isotope shifts and hyperfine spectra with other laser experiments, and (3) the fact that our data fit a straight line on a King plot better than Jackson's data (see below).

Others have made limited studies of the isotope shifts of the even isotopes of krypton. Most of these studies measured only one[10] or two[8,12,14] transitions for all the stable even isotopes; other transitions[7,10] have been measured for only some of the stable even isotopes. Of the four transitions for which all the even isotope shifts have been reported, only the work of Gerhardt *et al.*[8,12] on the $1s_5$-$3p_9$ transition is on a transition we also measured. As can be seen in Table III, the agreement is fair with two of the shifts disagreeing by slightly more than combined error bars.

We made King plots of our data using the $1s_5$-$2p_8$ transition as the reference. We selected this transition because it gave the best straight lines in the King plots with no systematic distortions. For comparison with Jackson's work, we also made King plots using Jackson's data on the $1s_2$-$2p_4$ transition as the reference. Jackson used this transition as the reference for his work.[11,13]

TABLE III. Isotope shifts (in MHz) in the krypton transitions examined in this work. Our results are on the same line as the notation for the transition.

Transition	86-84	84-82	82-80	80-78	86-83
$1s_3$-$2p_3$	69.3(16)	67.0(8)	78.2(6)	81.7(6)	93.3(16)
Ref. 11	66.0(12)	66.3(21)	74.4(15)	82.2(18)	83.0(18)
$1s_3$-$2p_4$	68.6(23)	64.2(17)	76.9(8)	78.4(9)	84.9(19)
Ref. 11	69.3(15)	68.7(21)	79.8(15)	83.1(18)	85.1(21)
$1s_5$-$2p_6$	73.4(11)	69.3(11)	78.6(8)	85.5(7)	94.1(14)
Ref. 11	69.0(12)	69.3(27)	79.5(36)	87.6(24)	
$1s_5$-$2p_7$	74.2(12)	66.9(16)	81.4(17)	82.6(12)	90.4(15)
Ref. 11	68.1(15)	74.7(36)	78.0(18)	87.6(30)	
$1s_5$-$2p_8$	70.0(13)	66.3(10)	77.4(10)	79.1(10)	87.2(13)
Ref. 11	71.1(24)	61.5(18)	77.4(18)	84.0(15)	
$1s_5$-$2p_9$	65.9(13)	63.9(9)	74.0(9)	78.8(9)	80.7(14)
Ref. 11	65.7(18)	63.6(30)	76.2(15)	72.9(18)	
$1s_5$-$3p_9$	114.7(9)	114.0(14)	128.5(11)	132.8(4)	153.6(10)
Ref. 13	117.0(24)	110.1(38)	127.8(38)	137.1(38)	
Refs. 8 and 12	115.8(9)	112.9(9)	126.0(9)	135.0(9)	160.0
$1s_5$-$4d'_4$	159.8(25)	145.0(40)	174.9(37)	170.1(28)	216.9(23)

The quality of fit from the two sets of King plots is not significantly different. The slope for a transition is the same for the two different references to within 7% except for the $1s_5$-$2p_7$ transition where the difference is 15%.

Figure 3 shows a typical King plot using the $1s_5$-$2p_8$ transition as reference. The error bars on the points come from our error estimates on our measured shifts. The straight line is the weighted least-squares fit. The weighting includes the scaling for each splitting and the standard error estimate for each splitting in the King plot. If the error estimates for all the splittings for a transition were equal, this procedure finds the line that minimizes the sum of the squares of the difference between the observed and predicted splittings. For krypton, this biases the line towards fitting the 78-86 and 80-86 splittings; the center two on the plot.

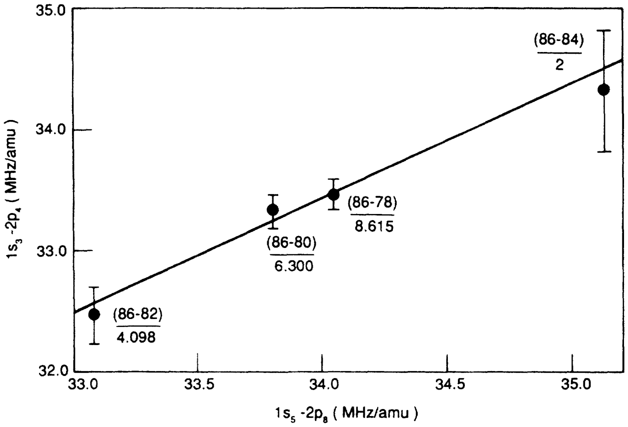

FIG. 3. King plot for the $1s_3$-$2p_4$ transition using the $1s_5$-$2p_8$ transition as the reference transition. Each data point is labeled as to which isotope shift and the mass scaling used for that point (see Ref. 11).

Table IV shows the changes of the observed splittings that are needed to place the splittings on the line from the King plot fits. These numbers are smaller than the estimated errors in the splittings for all but two points and are frequently much smaller than the estimated errors. Table IV also contains the slopes that we find from our King plots and the slopes reported by Jackson. The only difference between his slopes and ours that is not within the combined error bars is for the $1s_5$-$3p_9$ transition, which is slightly outside of the combined error bars. There is a significant disagreement, however, in our data for the $1s_5$-$2p_8$ transition. Plotted against Jackson's reference transition data, his data give a slope of 1.8 while our data give a slope of 0.96. If this were due to an error in our data, all our other slopes should differ from Jackson's by about a factor of 2. Since the other slopes agree quite well, the errors must be in Jackson' data. This is consistent with Jackson's slope of 1.8 for this transition being substantially different from his results on all the other transitions in this manifold that he studied.

The slopes from our King plots confirm Jackson's finding that the field shifts in krypton do not vary much among the transitions originating in the metastable states. Gerhardt *et al.*[8,12] extracted from their data the change in the mean-square nuclear charge radii with increasing neutron number by using unpublished muonic x-ray data on a single pair of krypton isotopes from Fricke and co-workers. Gerhardt *et al.* find a decrease in the size of the krypton nuclei with increasing neutron number and a negative specific mass shift (opposite direction of the normal mass shift). Unfortunately the error bars for the change in the mean-square nuclear charge radii are very large (≈ ±70%). This is due to the smallness of the residual shifts and the large errors bars (≈ ±100%) on the ^{86}Kr-^{84}Kr specific mass shift determined from the muonic x-ray data. With these large errors, there is no point in reporting the separation of the residual isotope shifts into the specific mass shifts and field shifts for the transitions we studied. It is a simple algebraic exercise for anyone interested.

IV. CONCLUSION

These are the results of our extensive study of the isotope shifts and hyperfine splittings for all the stable isotopes of Kr I. We studied all but one of the near-infrared transitions originating from the metastable $1s_5$ and $1s_3$ levels of krypton as well as two transitions to higher levels. These data will be of use in evaluating possible ultratrace isotope analysis schemes for krypton. They also may provide an incentive for the reexamination of the parametric analysis of the krypton hyperfine constants

TABLE IV. Changes needed (in MHz) to put data points on the straight line found from the King plot fits and its slope.

	86-78	86-80	86-82	86-84	Slope	Jackson's slope[a]
$1s_3$-$2p_3$	−0.42	0.48	0.23	0.58	0.76(10)	1.1
$1s_3$-$2p_4$	0.15	−0.45	0.42	0.37	0.95(8)	1.1
$1s_5$-$2p_6$	−0.69	1.10	−0.80	−0.36	0.93(16)	0.9
$1s_5$-$2p_7$	0.79	−0.92	−0.30	−0.80	1.14(16)	0.9
$1s_5$-$2p_9$	−0.55	0.89	0.68	1.53	0.90(22)	1.0
$1s_5$-$3p_9$	−0.38	−0.22	1.42	1.01	0.70(30)	1.4
$1s_5$-$4d'_4$	0.45	−0.62	0.12	2.5	1.4(5)	

[a] References 11 and 13, estimated error is ±0.3 except for $1s_3$-$2p_4$ which is larger but it is not said how much larger.

and test electronic structure calculations of krypton. In addition, this study shows the power and sensitivity of combining cw single-frequency lasers for resonance ionization with a mass spectrometer. This is highlighted by the high signal levels shown in Fig. 2.

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