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Fine structure energies for the $1s2s\ ^3S-1s2p\ ^3P$ transition in helium-like ions

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Abstract

We have measured the $1s2s\ ^3S_1-1s2p\ ^3P_0$ and $1s2s\ ^3S_1-1s2p\ ^3P_2$ fine structure transition wavelengths in the helium-like Ar^{16+} ion, using the beam-foil spectroscopy technique with position-sensitive photon detection. The wavelength results have precisions of <30 ppm and are sensitive to the total QED corrections to $<0.5\%$. The measurements confirm the significance of $(Z\alpha)^4$ -dependent relativistic corrections to the $n=2$ state fine structures suggested by recent calculations. The small remaining discrepancies between these measurements and the accurate theoretical values suggest that the magnitude of uncalculated higher order QED terms is about $0.15(Z\alpha)^4$ a.u. The status of our experimental search for the $^3S_1-^3P_0$ transition in Ni^{26+} is discussed.

1. Introduction

Helium-like atomic systems represent a fundamental testing ground for *ab initio* calculations of the fully relativistic electromagnetic interaction between charged particles in a many-body system. Precise variational calculations of the energies of low-lying S and P states in helium-like ions with $Z \leq 10$, including the dominant relativistic corrections, were carried out by Accad, Pekeris and Schiff in 1971 [1]. An extension of these calculations for the $n=1$ and 2 states for $Z=2$ to 100 was formulated by Drake [2], whose benchmark unified method combines variational techniques and the relativistic $1/Z$ expansion. A similar perturbation-expansion technique was applied by DeSerio et al. [3] for the $2\ ^3S-2\ ^3P$ transitions for $Z \leq 26$. Multiconfiguration Dirac–Fock (MCDF) calculations have been reported by Hata and Grant [4] and by Indelicato et al. [5] for selected ions up to $Z=54$.

The unified method of Drake omitted some terms of order $(Z\alpha)^4$ a.u. and higher in both the relativistic and the QED corrections to the energies. The estimated uncertainty due to these uncalculated terms was $\pm 0.2(Z\alpha)^4$ a.u. or $\pm 1.2(Z/10)^4\text{ cm}^{-1}$ [2].

We have recently presented [6] a comparison of experiment with theory for the $1s2s\ ^3S_1-1s2p\ ^3P_{0,2}$ transition

energies in all helium-like ions for which accurate experiments exist from $Z=2$ to 92. We concluded that although no significant discrepancy appears between experiment and the unified calculations for the $^3S_1-^3P_2$ transition, a systematic deviation from the calculations of approximately $-2.3(Z/10)^4\text{ cm}^{-1}$ exists for the $^3S_1-^3P_0$ transition. This conclusion is consistent with the results of a new generation of accurate relativistic calculations of the helium-like $n=2$ state structure. These theoretical approaches include the many-body perturbation theory (MBPT) technique of Johnson and Sapirstein [7] for $Z=10-36$, the configuration interaction (CI) method of Chen, Cheng and Johnson [8] for $Z=5-100$, and the many-body all order (MBAO) procedure of Plante, Johnson and Sapirstein [9] for $Z=3-100$.

In the present work, we have chosen to measure the $1s2s\ ^3S_1-1s2p\ ^3P_{0,2}$ transition energies in the helium-like ion Ar^{16+} in order to provide a test of the new calculations at the level of $<0.5(Z/10)^4\text{ cm}^{-1}$, or $<0.1(Z\alpha)^4$ a.u., as well as to resolve a discrepancy between the calculations and a previous measurement. The two previous measurements of the $^3S-^3P$ transition wavelengths in helium-like argon are an early beam-foil spectroscopy study by Davis and Marrus [10] and a recoil-ion spectroscopy study by Beyer et al. [11]. The beam-foil measurement was a pioneering investigation of the $2s-2p$ transitions at high Z , but with modest instrumentation and low precision. The recoil-ion study was based upon identification of weak features in a complex spectrum produced using collisional

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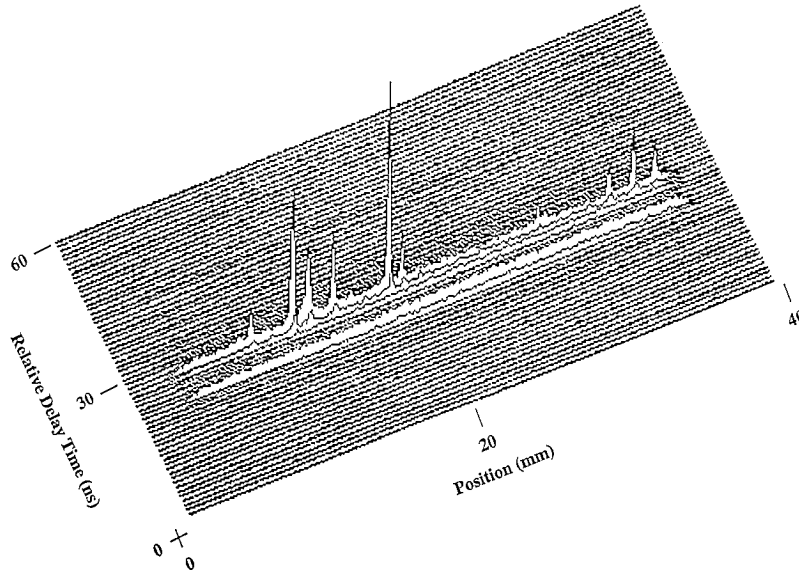


Fig. 1. Time-resolved spectra obtained using the position-sensitive detector. The spectrum of highly-ionized argon is isolated in a 2 ns wide ridge.

excitation of argon gas by energetic heavy ions. The recoil-ion result for the ${}^3S_1-{}^3P_0$ transition wavelength disagrees by several standard deviations with the value expected from the recent calculations and from our isoelectronic predictions [6].

The present study utilized the beam-foil fluorescence technique with position-sensitive photoelectric detection to provide the first unambiguous high resolution spectroscopic observation of the $1s2s {}^3S_1-1s2p {}^3P_{0,2}$ fine structure transition wavelengths in the helium-like Ar^{16+} ion.

Our experimental results determine the ${}^3S_1-{}^3P_{0,2}$ fine structure transition energies to < 30 ppm, and they are sensitive to the total QED contributions to $< 0.5\%$. We confirm the importance of new $(Z\alpha)^4$ -dependent relativistic corrections to the $n = 2$ state fine structure provided by recent calculations, and our results establish a limit to the size of uncalculated higher order QED corrections for $Z = 18$.

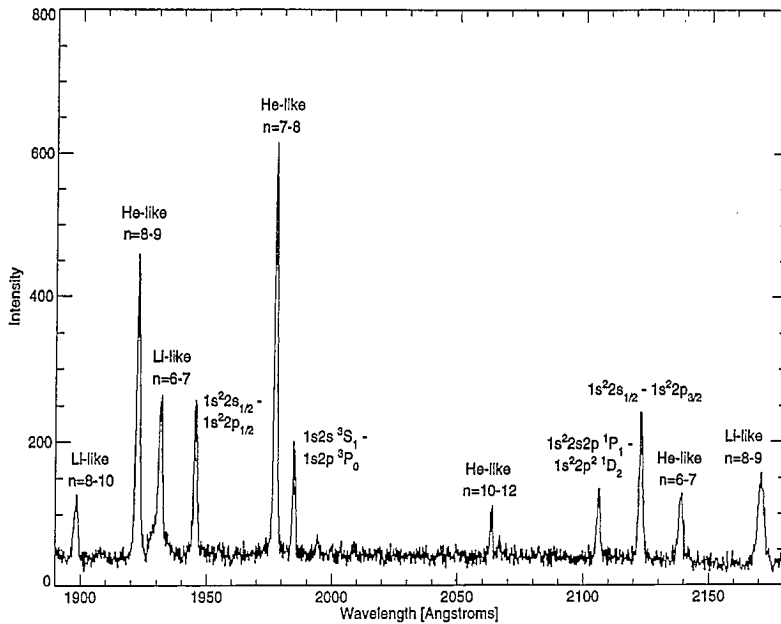


Fig. 2. Spectrum of highly-ionized argon showing the $1s2s {}^3S_1-1s2p {}^3P_0$ transition in the third order of diffraction.

2. Experiment

The emission spectrum of highly-ionized argon was produced using excitation of a 160 MeV beam of Ar^{9+} ions by passage through a $100 \mu\text{g}/\text{cm}^2$ carbon foil target at the ATLAS accelerator facility. The fluorescence spectrum of highly-ionized argon was observed perpendicular to the beam direction using a 1 m vacuum ultraviolet (VUV) spectrometer. Transitions in Ar^{14+} to Ar^{17+} ions were observed in high orders of diffraction with linewidths of $0.2\text{--}0.5 \text{ \AA}$ using a position-sensitive detector (PSD). Timing signals from the pulsed ion beam allowed suppression of background in the data acquisition (see Fig. 1). Further experimental details are described elsewhere [12].

A spectrum of highly-ionized argon is shown in Fig. 2. The $^3\text{S}_1\text{--}^3\text{P}_0$ transition in helium-like Ar^{16+} appears in the third order of diffraction. Rydberg transitions and the lithium-like $2s\text{--}2p$ transitions are also observed.

3. Results

The determination of an accurate wavelength scale for the helium-like $2^3\text{S}\text{--}2^3\text{P}$ transitions required two procedures: the establishment of the wavelength dispersion across the PSD, and the determination of sufficiently accurate reference transition wavelengths in the beam spectra. The wavelength dispersion was established by recording a series of spectra of ArI and ArII transitions produced in a gas discharge lamp. The in-beam reference lines were determined primarily by accurate calculation of the structures of Rydberg transitions excited in the ion–foil interaction [12].

The final results for our measured transition wavelengths in helium-like Ar^{16+} are $559.944(16) \text{ \AA}$ for $1s2s^3\text{S}_1\text{--}1s2p^3\text{P}_2$ and $661.533(18) \text{ \AA}$ for $1s2s^3\text{S}_1\text{--}$

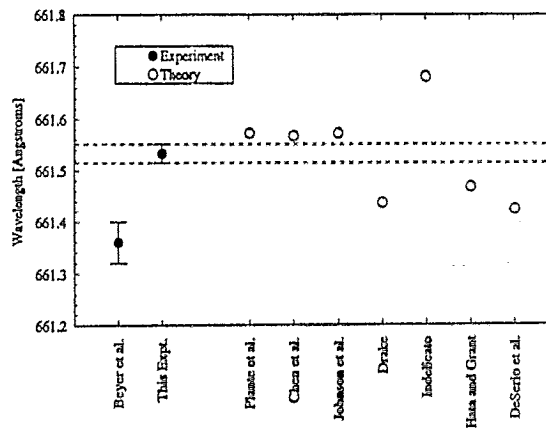


Fig. 3. Comparison of our wavelength result for the $1s2s^3\text{S}_1\text{--}1s2p^3\text{P}_0$ transition in Ar^{16+} with the previous measurement and with calculations.

$1s2p^3\text{P}_0$. The wavelength uncertainty is dominated by uncertainties in the accurately calculated Rydberg transition reference wavelengths. We compare our results with calculations and with previous measurements for Ar^{16+} in Table 1 and in Figs. 3 and 4. Our result for the $^3\text{S}_1\text{--}^3\text{P}_0$ transition disagrees substantially with that for the recoil-ion measurement [11]. Our result for the $^3\text{S}_1\text{--}^3\text{P}_2$ transition improves upon the previous precision [11] by a factor of 6. The measured values are sensitive to the relativistic contributions to 140 ppm and 460 ppm for the $^3\text{P}_2$ and $^3\text{P}_0$ transitions, respectively, and they test the total QED corrections to 0.45% and 0.35%, using the designations of Drake [2]. The $^3\text{P}_2\text{--}^3\text{P}_0$ fine structure interval is determined to 230 ppm.

Our experimental results show best agreement with the new many-body and CI calculations [7–9] for both of the fine structure transitions in Ar^{16+} . Note that the new relativistic many-body and CI results include QED correc-

Table 1

Calculated and measured energies (in cm^{-1}) for the $1s2s^3\text{S}_1\text{--}1s2p^3\text{P}_{0,2}$ transitions and the $2p^3\text{P}_{2-0}$ fine structure interval in helium-like argon

	Ref.	$^3\text{S}_1\text{--}^3\text{P}_0$	$^3\text{S}_1\text{--}^3\text{P}_2$	$^3\text{P}_2\text{--}^3\text{P}_0$
<i>Theory</i>				
DeSerio et al.	[3]	151 189	178 574	27 385
Hata and Grant	[4]	151 179	178 558	27 379
Indelicato	[5]	151 130	178 546	27 416
Drake	[2]	151 186	178 577	27 391
Johnson and Sapirstein	[7]	151 155	178 576	27 421
Chen et al.	[8]	151 156	178 578	27 422
Plante et al.	[9]	151 155	178 576	27 421
<i>Experiment</i>				
Davis and Marrus	[10]	151 350(250)	178 510(290)	27 160
Beyer et al.	[11]	151 203.6(9.1)	178 591(32)	27 387
This work		151 164.0(4.1)	178 589.3(5.1)	27 425.3(6.5)

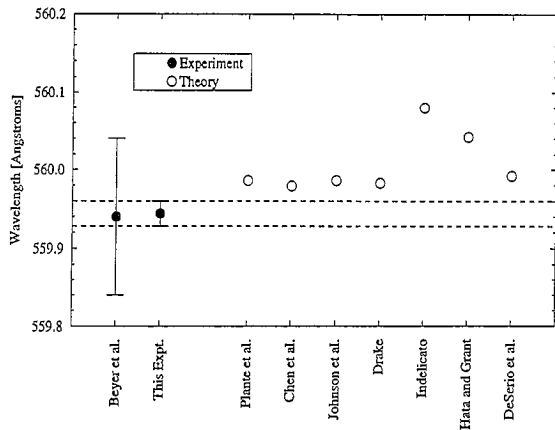


Fig. 4. Comparison of our wavelength result for the $1s2s^3S_1 - 1s2p^3P_2$ transition in Ar^{16+} with the previous measurement and with calculations.

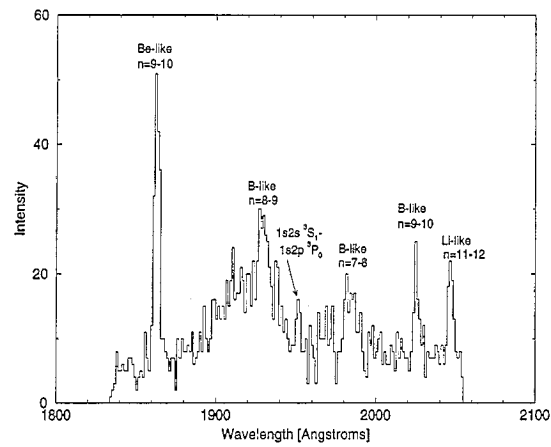


Fig. 6. Observation of the $1s2s^3S_1 - 1s2p^3P_0$ transition in helium-like Ni^{26+} in the fifth order of diffraction, along with nearby Rydberg transition structures in highly-ionized nickel. The spectrum was produced by excitation of 6.2 MeV/u Ni^{26+} ions in a $10 \mu\text{g}/\text{cm}^2$ carbon foil target.

tions taken from Drake [2], to enable comparison of the accurate non-radiative calculations with experiment.

We have also measured the wavelength value of the $2s2p^1P_1 - 2p^2D_2$ transition in beryllium-like argon (see Fig. 2) to be $421.29 \pm 0.02 \text{ \AA}$. This result may be compared with the interpolated semi-empirical value by Edlén [13] of 421.21 \AA .

We have initiated spectroscopic measurements in highly-ionized nickel in order to provide the first measurement of the helium-like $1s2s^3S_1 - 1s2p^3P_0$ transition wave-

length in Ni^{26+} . A preliminary observation of this transition at 390 \AA is shown in Fig. 6 in the fifth order of diffraction. We have performed MCDF (GRASP [14]) calculations of the complex neighboring Rydberg transition structures from boron-like and beryllium-like nickel and have confirmed that these structures are not blended with the helium-like line. Improved measurements are underway.

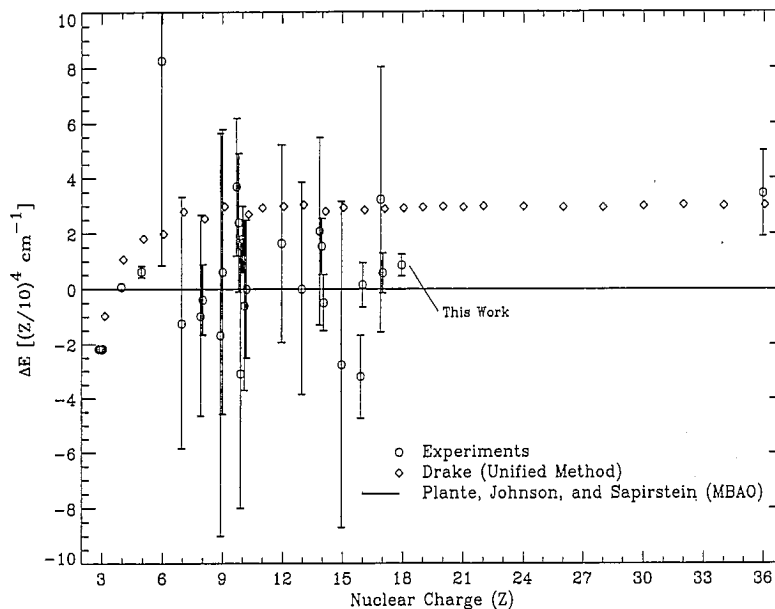


Fig. 5. Comparison of experiment and theory with respect to the MBAO calculations of Plante, Johnson and Sapirstein [9] for the scaled $1s2s^3S_1 - 1s2p^3P_0$ transition energy in helium-like ions. The unified calculations of Drake [2] are shown as diamonds. The references to other measurements are found in Ref. [12].

4. Discussion

The new generation of relativistic calculations of $n = 2$ state energies in helium-like ions that has appeared over the past two years [7–9] has established significantly improved benchmarks of theoretical accuracy in our understanding of the two-electron atomic system. The most dramatic aspect of these calculations is the removal of the previously dominant energy uncertainty [2] that was due to uncalculated relativistic corrections of order $(Z\alpha)^4$ a.u., and the identification of the strong fine-structure dependence of these corrections. Our experimental result for the $1s2s^3S_1-1s2p^3P_0$ transition energy in Ar^{16+} reveals a discrepancy of $(-1.0 \pm 0.2) \times 10^{-4}$ a.u. from the earlier unified calculations [2] that accounts for about 70% of the new order- $(Z\alpha)^4$ relativistic corrections for $Z = 18$ in the recent calculations [7–9]. The remaining differences in transition energies between our measurements and the three new calculations are about $10 \pm 4 \text{ cm}^{-1}$. Since the new calculations are expected to account for non-radiative contributions to an accuracy of $\leq 0.01(Z\alpha)^4$ a.u., our results suggest the need for a reduction in the QED contributions to the $1s2s^3S_1$ energy for $Z = 18$ by about $0.15(Z\alpha)^4$ a.u., a 1% correction to the QED effects. This magnitude of correction is consistent with the results of new QED calculations [15] for singlet states in helium-like ions. Such determinations of uncalculated QED terms of order $(Z\alpha)^4$ a.u. represent the most interesting theoretical problem remaining for the helium isoelectronic sequence [9].

We compare our result for the $1s2s^3S_1-1s2p^3P_0$ fine structure transition energy in Ar^{16+} with previous measurements and with calculations for the isoelectronic sequence of helium-like ions in Fig. 5. The data are plotted with respect to the most recent of the accurate calculations, the MBO results of Plante, Johnson and Sapirstein [9]. These calculations show excellent agreement with the recent MBPT [7] ($Z \geq 10$) and CI [8] ($Z \geq 5$) calculations. The earlier unified results of Drake [2] are included in Fig. 5 to emphasize the nearly constant Z^4 -dependent magnitudes of these new relativistic corrections for $Z > 10$.

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Note added

Following submission of this paper, a study of recoil-ion spectra of argon was published [Hallett, Howie, Silver and Dietrich, Phys. Lett. A 192 (1994) 43] in which wavelength values of $559.95(2) \text{ \AA}$ and $661.58(2) \text{ \AA}$ have been suggested for the $1s2s^3S_1-1s2p^3P_{2,0}$ transitions, respectively, in Ar^{16+} . The identifications of the weak Ar^{16+} lines amid many unidentified lines in the recoil-ion spectra were based upon approximate agreements with theoretical wavelengths. The $^3P_2-^3P_0$ fine structure interval of $27434(7) \text{ cm}^{-1}$ suggested by these new studies disagrees by 2σ with the accurately established theoretical interval of 27421 cm^{-1} [7–9] (see Table 1), which is expected to be weakly sensitive to uncalculated QED contributions for $n = 2$ triplet states.

References

- [1] Y. Accad, C.L. Pekeris and B. Schiff, Phys. Rev. A 4 (1971) 516.
- [2] G.W.F. Drake, Can. J. Phys. 66 (1988) 586.
- [3] R. DeSerio, H.G. Berry, R.L. Brooks, J. Hardis, A.E. Livingston and S.J. Hinterlong, Phys. Rev. A 24 (1981) 1872.
- [4] J. Hata and I.P. Grant, J. Phys. B 16 (1983) 523; 17 (1984) 931.
- [5] P. Indelicato, Nucl. Instr. and Meth. B 31 (1988) 14; P. Indelicato, O. Gorcex and J.P. Desclaux, J. Phys. B 20 (1987) 651; P. Indelicato, F. Parente and R. Marrus, Phys. Rev. A 40 (1989) 3505.
- [6] H.G. Berry, R.W. Dunford and A.E. Livingston, Phys. Rev. A 47 (1993) 698.
- [7] W.R. Johnson and J. Sapirstein, Phys. Rev. A 46 (1992) R2197.
- [8] M.H. Chen, K.T. Cheng and W.R. Johnson, Phys. Rev. A 47 (1993) 3692.
- [9] D.R. Plante, W.R. Johnson and J. Sapirstein, Phys. Rev. A 49 (1994) 3519.
- [10] W.A. Davis and R. Marrus, Phys. Rev. A 15 (1977) 1963.
- [11] H.F. Beyer, F. Folkmann, and K.-H. Scharfner, Z. Phys. D 1 (1986) 65.
- [12] K.W. Kukla, A.E. Livingston, J. Sulciman, H.G. Berry, R.W. Dunford, D.S. Gemmell, E.P. Kanter, S. Cheng and L.J. Curtis, to be published in Phys. Rev. A.
- [13] B. Edlén, Phys. Scripta 28 (1983) 51.
- [14] F.A. Parpia, C.F. Fischer and I.P. Grant, private communication.
- [15] K.T. Cheng, M.H. Chen, W.R. Johnson and J. Sapirstein, Phys. Rev. A 50 (1994) 247.