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Lamb Shift and Fine Structure of $n=2$ in ^{35}Cl XVI

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We have measured the wavelengths of the $2s\ ^3S_1-2p\ ^3P_2$ and $2s\ ^3S_1-2p\ ^3P_0$ transitions in Cl XVI to be 613.825±0.013 Å and 705.854±0.076 Å. Our precision is sufficient to provide measurements of the $2s_{1/2}-2p_{1/2}$ and $2s_{1/2}-2p_{3/2}$ Lamb shifts to an accuracy of ±0.3% and to test quantum electrodynamics (QED) theory in the strong-field region. We compare our results with the one-electron QED theories of Mohr and Erickson and discuss the accuracy of calculations of electron correlation in two-electron atoms.

We wish to emphasize in this paper that Lamb-shift measurements in high-Z atoms need not be confined to the one-electron hydrogenic ions, but higher precision may be attainable in ions with a few (two or three) electrons. We present a measurement in two-electron chlorine to justify this suggestion and discuss briefly the limitations in the precision of such measurements due to the accuracy of calculations of relativistic energies in many-electron atoms.

In two-electron ions of low Z ($Z \lesssim 10$), there have been several experimental tests[1] of quantum electrodynamics, but of lower precision than measurements in the corresponding one-electron atoms. Electron correlation effects have been calculated approximately to first order in the Lamb-shift terms[2] and less accurately than the experimental values.[3] However, such effects become less important at high Z since the correlations can be expressed as part of a $1/Z$ expansion.

Tests of quantum electrodynamics by $2s$-$2p$ Lamb-shift measurements in one-electron atoms have been reviewed recently by Kugel and Murnick[4] and by Mohr.[5] They point out that although the highest-precision measurements are in hydrogen (20 ppm),[6] the higher-order terms of the Lamb shift, which is usually expressed as a power series expansion in $Z\alpha$, are more easily tested in higher-Z ions. The present most precise measurements are those in $^{19}F^{8+}$ ($Z=9$)[7] and $^{40}Ar^{17+}$ ($Z=18$)[8] of ±2% and ±4% accuracy, respectively, both of which test the higher-order terms of the Lamb shift to approximately the same accuracy as the work in hydrogen.[6] The higher-order terms in $Z\alpha$ probe quantum electrodynamic (QED) theory in strong fields where the perturbative theory must eventually break down. Comparable tests occur only in the comparisons between theory and experiment of the binding energy of K-shell electrons in high-Z atoms,[9] where an accuracy of about 10% has been achieved. Davis and Marrus[10] measured the two-electron Lamb shift in Ar XVII, but only with low precision (~±30%).

We accelerated chlorine ions in the Argonne FN tandem accelerator to an energy of 80 MeV and further stripped and excited the ion beam in a thin 20-μg-cm^{-2} carbon foil. The observation angle (close to 90°) was deduced from the relative Doppler shifts at beam energies between 56 and 88 MeV. Thus, the first- and second-order Doppler shifts are ~3 Å/deg and ~3 Å, respectively, at 1200 Å for 80 MeV ion beam energy. As we were able to use internal calibration lines in the beam-foil spectra, such Doppler shifts had very little effect on the values of our measured wavelengths. The monochromator was refocused for the fast-moving light source ($v/c=\beta\sim 0.07$) by adjustment of both the entrance slit and the grating.[11]

In Fig. 1, we show wavelength scans including

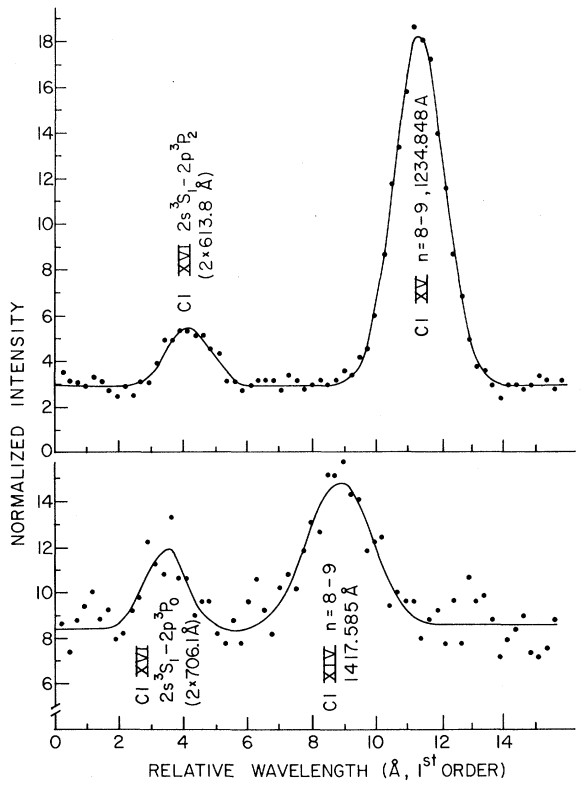

FIG. 1. Wavelength scans including the Cl XVI $2s\ ^3S_1$–$2p\ ^3P_2$ transition near 2×614 Å (upper) and the Cl XVI $2s\ ^3S_1$–$2p\ ^3P_0$ transition near 2×706 Å (lower). The wavelength was stepped in increments of 0.12 Å (second order), and the profiles are nonlinear least-squares Gaussian fits (solid line).

the $2s\ ^3S_1$–$2p\ ^3P_2$ and $2s\ ^3S_1$–$2p\ ^3P_0$ transitions in second order. Our precision depends primarily on the relative and absolute wavelength measurements. The relative wavelength measurement consists of an accurate determination of the separation between the two wavelengths shown in each part of Fig. 1. The reproducibility of this measurement presently limits our precision to ± 0.012 Å for the line from $^3P_2$. Improved statistics would allow determination of line centers to better than the present 1/50 of the linewidth and a study of possible profile asymmetries. Hyperfine structure of the two-electron transitions is small and produces a symmetric broadening of the lines. The absolute wavelengths of the calibration lines $n = 8$–9 in Cl XVI and Cl XV have been calculated directly from Dirac theory for the $8k$-$9l$, $8i$-$9k$, and $8h$-$9i$ transitions. A small core polarization is included in the Cl XIV transition from direct measurement of the fine structure of the $n = 5$–6 transition. The mean wavelength $\bar{\lambda}$ was then found using hydrogenic transition probabilities and assuming statistical $(2l+1)$ population distributions. Such distributions have been observed in high-(n,l) state excitation[12]; however, $\bar{\lambda}$ is not very sensitive to distribution. Thus, an l^2 distribution[13] would increase $\bar{\lambda}$ by 0.008 Å for both transitions. This possible error has been included in our result. The small 0.1-Å separation of the fine structure does not lead to measurably asymmetric profiles.

Assuming wavelengths of 1234.848 and 1417.133 Å for the Cl XV and Cl XIV $n = 8$–9 transitions, respectively, we deduce wavelengths of 613.825 ± 0.013 Å for the $2s\ ^3S_1$–$2p\ ^3P_2$ transition, and 705.854 ± 0.076 Å for the $2s\ ^3S_1$–$2p\ ^3P_0$ transition.

The error limits represent 1 standard deviation in the fits of profiles to eight different sets of data similar to those of Fig. 1 for the first transition and in the fitting uncertainty of the one set of data for the second weaker transition, plus half the shift due to core polarizability. Improved statistics in future work will require a more detailed analysis of the above systematic errors.

In Table I we compare our results with theory. The transition energy consists of three main parts: the nonrelativistic, the relativistic, and the QED contributions. The nonrelativistic energy has been calculated using Z-dependent variational perturbation theory[14] and is expressible as a power series in $1/Z$. The series rapidly converges for $Z = 17$ with the Z^{-4} term giving a contribution of 0.52 cm^{-1}. Relativistic energy corrections can be developed as a power-series expansion in $(\alpha Z)^2$ with further electron-correlation corrections providing similar terms to powers of $1/Z$. Thus, the Dirac equation gives the $2s_{1/2}$-$2p_{3/2}$ fine-structure contribution of the $^3S_1$-$^2P_2$ transition with no structure for the transition from $^3P_0$. The first electron-correlation terms are the relativistic part of the terms for one-photon exchange plus the Breit interaction terms given by Doyle,[15] while Mohr[16] and Cheng[17] have extrapolated the two next higher terms as given in Table I. The mass polarization is calculated by Ermolaev and Jones.[18]

The QED corrections are taken from Mohr[5] for the $2p_{1/2}$ state ($^3P_0$), while the j-dependent corrections for the $2p_{3/2}$ state ($^3P_2$) are taken from Erickson and Yennie.[19] It should be noted that our results are insensitive at our level of accuracy to the fourth-order self-energy term $S_{SE}^{(4)}$ and the relativistic mass and recoil terms, S_{RM} and S_{RR}, and scarcely sensitive to the nuclear structure shift S_{NS}. However, our measurements

TABLE I. Transition energy of $2s$-$2p$ in Cl XVI (cm^{-1}). See text for explanation of each contribution.

Contribution		$2s\ ^3S_1 - 2p\ ^3P_2$	$2s\ ^3S_1 - 2p\ ^3P_0$
Non-relativistic	ΣZ^{-n}	135,256.3	135,256.3
Dirac Fine Structure	$\Sigma(\alpha Z)^n$	30,800.2	0.0
Breit plus photon exchange	$Z^{-1}(\alpha Z)^4$	-2084.0	8201.1
Extrapolated Breit	$Z^{-2}(\alpha Z)^4$	1.0	-892.
plus photon exchange	$Z^{-1}(\alpha Z)^6$	-27.0[a]	90.9
Mass Polarization		-69.4	-69.4
Total		163,877.1	142,586.9
		(610.213 Å)	(701.327 Å)
QED Terms			
$S^{(2)}_{sE}$	$\alpha(\alpha Z)^4\,\Sigma(\alpha Z)^n$	-1040.7	-1106.9
$S^{(2)}_{vP}$	$\alpha(\alpha Z)^4\,\Sigma(\alpha Z)^n$	69.2	69.2
$S^{(4)}_{sE}$	$\alpha^2\ (\alpha Z)^4$	-0.28	-0.28
S_{RM}	$\alpha(\alpha Z)^4\ m/M$	0.03	0.03
S_{RR}	$(\alpha Z)^5\ m/M$	-0.71	-0.71
S_{NS}	$[(\alpha Z)^4+(\alpha Z)^6]$	-6.93	-6.93
Total QED		-979.4	-1045.6
Total Transition Energy			
From above		162,897.7	141,541.3
This experiment		162,913 ± 3	141,672 ± 13
Johnson - rel. RPA		164,186	143,212

[a] P. Mohr had not calculated this number in Ref. 16.

test the second-order self-energy $S_{SE}^{(2)}$ at the $(\alpha Z)^6$ term (21.5 cm^{-1} for $^3P_0$, 26.1 cm^{-1} for $^3P_2$) and the residuals $(\alpha Z)^6\ G_{SE}$ (−106.6 cm^{-1}) and the second-order vacuum polarization at the $(\alpha Z)^5$ term (11.5 cm^{-1}). None of these terms has been previously tested by Lamb-shift measurements. In addition, the one-electron QED correction of Erickson and Yennie[19] is 19.4 cm^{-1} greater than that of Mohr[5] and thus our measurement provides the first possibility to distinguish between these two calculations.

Table I shows that our experimental result for the $^3S_1$-$^3P_2$ energy is close to the "one-electron" theory which is 5 standard deviations (15.3 cm^{-1}) away, while the $^3S_1$-$^3P_0$ experimental energy differs considerably from theory (by 107 cm^{-1}). We also compare our results with a less accurate relativistic random-phase approximation (RRPA) of Johnson[20] to which we have added the mass-polarization and QED corrections.

TABLE II. Fine structure $2p\ ^3P_0$–$2p\ ^3P_2$ in Cl XVI.

Method	Separation (cm^{-1})
This experiment	21 241 ± 13
One-electron theory (Table I)	21356.4
Johnson — RRPA[a]	20 974
Two-electron theory[b]	21 198

[a] Ref. 20. [b] Ref. 21.

In Table II we compare the fine-structure separation $^3P_0$-$^3P_2$ with the one-electron Dirac theory of Table I, Johnson's RRPA calculation,[20] and the two-electron calculation of Ermolaev and Jones.[21] These comparisons suggest that electron-correlation effects must be small, and that their evaluation by $1/Z$ expansion techniques should be accurate.

The nonrelativistic interaction between the two electrons $(1/r_{12})$ has been evaluated "exactly" by the variational perturbation technique. However, the relativistic part, which is represented approximately by the Breit operator, is given as a double expansion in $1/Z$ and $(\alpha Z)^2$, and only its leading terms have been calculated. It can be seen from Table I that to our experimental accuracy, only the leading term is significant for the $^3S_1$-$^3P_2$ transition, while for the $^3S_1$-$^3P_0$ transition the first three terms are significant, and that further uncalculated terms may also contribute. Hence, we conclude that uncertainties in electron-correlation effects are important in Cl XVI only for the $^3S_1$-$^3P_0$ transition, and that our measurement of the $^3S_1$-$^3P_2$ transition can be used to check the quantum electrodynamic terms.

Thus, we obtain the following Lamb-shift values (E-Y denotes Erickson-Yennie):

$$S_{\text{expt}}(2s_{1/2}\text{-}2p_{3/2}) = 964 \pm 3\ \text{cm}^{-1}, \quad (1)$$

$$S_{\text{Mohr}}(2s_{1/2}\text{-}2p_{3/2}) = 979.4\ \text{cm}^{-1}. \quad (2)$$

$$S_{\text{E-Y}}(2s_{1/2}\text{-}2p_{3/2}) = 998.8\ \text{cm}^{-1}. \quad (3)$$

A two-electron Lamb-shift calculation by Ermolaev[2] gives 832 cm^{-1} for $n=2$ Cl XVI and is in clear disagreement. His calculations show better agreement at low Z, where the one-electron theories disagree with experiment (e.g., in lithium, Ref. 3). Further experiments and calculations are necessary to study the reduction of electron-correlation effects in the Lamb shifts as a function of increasing nuclear charge Z.

We summarize our results by drawing the fol-

lowing conclusions: For the two-electron system, with a nuclear charge of $Z=17$, the relativistic part of the electron-electron interaction is known accurately for the $^3P_2$ state, but not for the $^3P_0$ state; the $2s$-$2p$ Lamb-shift measurement favors the calculations of Mohr over those of Erickson and Yennie.

We conclude that the level of accuracy of our measurements of two-electron–atom excitation energies is sufficient to allow accurate tests of QED at $Z=17$ and that similar measurements can be expected to become more accurate for higher Z. Thus, we are able to differentiate for the first time between the Mohr and Erickson-Yennie one-electron Lamb-shift theories. We note the lack of agreement between our fine-structure measurements and the calculations of Ermolaev, and suggest that the discrepancy is due to the higher-order relativistic corrections of the photon exchange and relativistic interactions. Our results emphasize the need for a consistent and complete relativistic theory of atomic structure. The discrepancy between experiment and theory may lie in the perturbative treatment of either the Lamb shift or the relativistic correction to the transition energy. Preliminary analysis of the resonance transitions $2s\,^2S-2p\,^2P$ of the three-electron system suggests that equivalent accuracy may be feasible for similar Lamb-shift measurements, particularly for $Z>17$ where Z-expansion techniques can accurately account for non-QED effects.

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