

Precision spectroscopy of trapped highly charged heavy elements: pushing the limits of theory and experiment

J D Gillaspy

NIST, Atomic Spectroscopy Group, Gaithersburg, MD 20899-8422, USA

E-mail: john.gillaspy@nist.gov

Received 16 July 2014

Accepted for publication 27 August 2014

Published 3 November 2014

Abstract

Atomic spectroscopy results from the electron beam ion trap at the National Institute of Standards and Technology have generally agreed with the predictions of theory extremely well. An interesting exception is our recent result on the helium isoelectronic sequence at $Z=22$, which agrees instead with a meta-analysis of all prior measurements above $Z=15$, but disagrees with both theory and a contemporaneous report of an independent measurement at $Z=18$ which claims to validate theory to high accuracy. Here, a potential systematic shift involving high- n satellite lines induced by double charge exchange is quantitatively estimated and shown to be potentially significant in experiments involving gasses. Suggestions for further refinements in estimating the magnitude of this systematic shift are given.

Keywords: charge exchange, precision spectroscopy, satellite line shifts, highly charged ions

(Some figures may appear in colour only in the online journal)

1. Introduction

Since the previous (August 2010) International Colloquium on Atomic Spectra and Oscillator Strengths for Astrophysical and Laboratory Plasmas, 14 papers [1–14] have appeared in the literature detailing atomic spectroscopy results from the National Institute of Standards and Technology (NIST) electron beam ion trap (EBIT) facility. Cumulatively, these papers report measurement of hundreds of spectral lines from a wide variety of ions. In cases where more accurate previous measurements or reliable theoretical results are available, our results generally agree with them. For example, our work on the D-lines of Na-like Xe, Ba, Sm, Gd, Dy, Er, W, Pt, and Bi [2] validates the *ab initio* calculations of Blundell [2] and tests the sum of a variety of quantum electrodynamics (QED) effects (including the exchange of up to three virtual photons) at the level of 0.4%, providing one of the most stringent high- Z tests of QED currently available.

Our work on He-like Ti^{20+} [9], however, is an exception to this general conclusion. This work shows a marked (3σ) discrepancy with the predictions of Artemyev *et al* [15], in stark contrast to the findings of Kubicek *et al* [16] which

validates QED to 0.5% in Ar^{15+} . When a weighted average of all previous experiments for the w-line ($1s2p\ ^1P_1-1s^2\ ^1S_0$) published prior to Kubicek *et al* [16] is calculated at each value of Z for which Artemyev *et al* [15] have predicted values, the trend is more consistent with our findings than with the findings of Kubicek *et al* [16]. Specifically, this meta-analysis reveals a discrepancy with Artemyev *et al* [15] which grows approximately as Z^3 (with an uncertainty of approximately 1 on the exponent) at the statistical significance level of 5σ .

This is reminiscent of the 5σ discrepancy between theory and experiment found in the spectroscopy of muonic hydrogen [17]. In fact, there are some intriguing correspondences between that work and our work [9] which are interesting to consider, although they may eventually turn out to be coincidental. Both test theory in an exotic type of atom for which the effective Bohr radius of the bound lepton is reduced by over an order of magnitude from the nominal value of $0.5\ \text{\AA}$ which is characteristic of ordinary neutral atoms (thus compressing the density of the electron wavefunction by over a thousand-fold and increasing substantially the overlap of the lepton with the nucleus or any other real or virtual particles

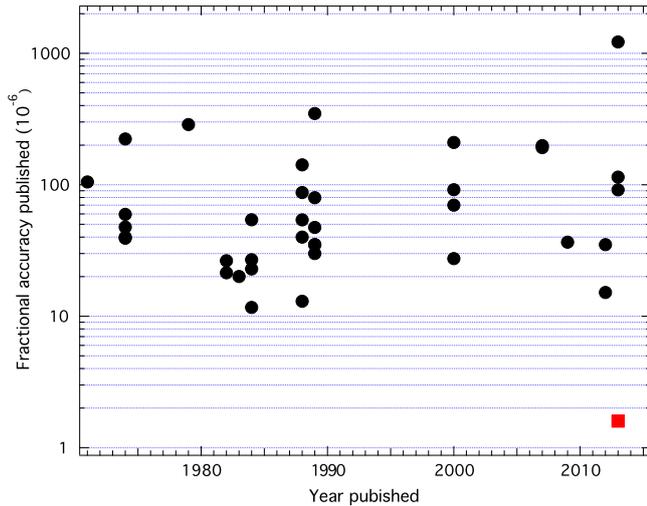


Figure 1. Fractional accuracy of measurements of the w-line in He-like ions as a function of publication year. Black dots are cited in [9]. The red square is the result of Kubicek *et al* [16].

near the nucleus). Both experiments are wavelength measurements with a fractional uncertainty of 15 meV keV^{-1} (15 parts-per-million). Both use QED to tie together disparate measurements (proton scattering and muonic hydrogen spectroscopy in one case, and ultraviolet and x-ray spectroscopy of a wide variety of ions in the other case), which then reveals the 5σ global discrepancy. In both cases, the uncertainty in the calculations is believed to be much smaller than the uncertainty in the experiments, suggesting either a neglected systematic error in one or more of the experiments, or the discovery of new physics beyond that represented by the fundamental theory.

Beyond these similarities, there are significant differences between the two sets of experiments. In one case, the orbital radius of the bound lepton is reduced by increasing the mass of the electron (replacing it with a muon, which is supposed to be identical to the electron in every way except mass) and in the other case orbital radius is reduced by increasing the nuclear charge. One experiment probes transitions from the *ground state* and the other probes transitions *between excited states*. In one experiment, the atom is 50 times smaller than hydrogen (a factor of 200 smaller due to the mass, but then a factor of 4 times larger because the muon is in the $n=2$ state), whereas in the other experiment the ion is smaller than the ordinary Bohr radius by approximately a factor of $Z=22$. One experiment involves only *one* bound lepton, and the other involves *two* (one of which partially screens the nucleus, increasing the bound state radius somewhat further—neutral helium, for example, is approximately 20% larger than hydrogen). One is less sensitive to uncertainties in the nuclear size (besides the fact that the electron is orbiting a factor of two or more farther away than the muon, the radius of the Ti nucleus is also more accurately known than the proton radius).

The relative accuracy (claimed uncertainty in wavelength divided by the wavelength) for all previous measurements of He-like ions reported in [9] is shown in figure 1. The

groundbreaking work of Kubicek *et al* [16], the red square on lower right, has such an extraordinarily small uncertainty that it stimulates the consideration of new types of systematic errors which have been presumed to be safe to neglect in all previous measurements. Below, one possible fundamental systematic error involving satellite shifts mediated by double charge exchange (DCX) is considered. This effect should apply to a wide range of experiments and does not appear to have been generally considered previously.

2. DCX-mediated satellite shifts in an EBIT

If a significant population of H-like ions is also present in the trap at the time that spectroscopy is performed on He-like ions, then DCX into those H-like ions can populate doubly excited Li-like states, with at least one electron in a high- n level (n =principal quantum number). If the high- n satellite electron remains relatively high as the second captured electron cascades to the ground state, the last emitted cascade photon will be only slightly shifted from the value that corresponds to the transition in a He-like ion. If the energy difference is less than or equal to approximately one half of the instrument-broadened linewidth, even a strong satellite line can be masked by the shot-noise in the total signal, so the residuals to a fit to a single line may not reveal the presence of the satellite, yet the composite line may fit to a significantly different center value. The magnitude of this effect is estimated in detail below. The uncertainties associated with this estimate are sufficiently large that it cannot be construed as proof that such an effect is actually significant in any particular experiment, but it does suggest that further work (both experimental and theoretical) is warranted to investigate and better quantify this possibility. The estimates presented below are meant to be a first step in that direction.

Much of the data necessary for a quantitative estimate are available for the case of argon ($Z=18$), so I use that ion as the prototype. In the following four sections, results from the literature are used to estimate the maximum intensity of the satellite line, the magnitude of the shift of the satellite line, and the magnitude of the systematic error that would result from neglecting the presence of such a satellite.

2.1. Relative intensity of DCX-mediated satellite lines

Under typical EBIT conditions for producing He-like ions from a gas injected into the trap, the excitation of a resonance line can proceed via two channels: electron impact excitation of the He-like ions and single charge exchange into the H-like ions which are naturally present simultaneously. An estimate for the ratio of the number of event per second for these two channels, $R=3.3$ (with electron impact excitation typically occurring several times more frequently than charge exchange), is given in the appendix under a rather general set of assumptions that are satisfied by typical EBIT conditions. This ratio, R , is found to be remarkably *independent* of gas injection pressure over a wide range of typical values. This is because as the gas injection pressure is reduced, the reduction

Table 1. Satellite shifts to the w-line from [21].

| n | Odd shift (eV) | Even shift (eV) |
|-----|----------------|-----------------|
| 5 | 1.26 | 0.71 |
| 6 | 0.71 | 0.39 |
| 7 | 0.47 | 0.23 |
| 8 | 0.31 | 0.15 |
| 9 | 0.23 | 0.15 |
| 10 | 0.15 | 0.07 |

in projectile density is compensated by a corresponding increase in the relative target density of H-like ions. This increase in the ratio of H-like to He-like ions with decreasing pressure occurs because, in the absence of charge exchange, there would be virtually no He-like ions present in the trap if the electron beam energy is set well above threshold for the production of both H-like and He-like ions (as it typically is in actual experiments).

For high charge states, double charge capture (in which two electrons are captured in a single collision with a neutral gas atom) occurs nearly as often as single charge capture, and hence if R is of order 1, the satellite line intensity may be nearly the same order of magnitude as the unperturbed resonance line intensity, even in the complete absence of dielectronic recombination. The appendix estimates the ratio of the satellite line intensity to the unperturbed line intensity to be $1/R3=8\%$. In the following sections, it is shown by direct construction that the energy shift of such a satellite line can be sufficiently small that it remains hidden in the residuals to the fit yet sufficiently large that it leads to a significant systematic error.

2.2. Principal quantum number of the spectator electron

The analysis of independent experiments shows that single electron capture into H-like Ar is dominantly into levels with principal quantum numbers $n=7-10$, with the most probable level $n=8$ [18]. In multiple electron capture, Trassinelli *et al* [18] find evidence for preferential capture into states starting around $n=6$. Ali *et al* [19, 20] argue that the dominant process involves ‘asymmetric capture’ in which one electron is at relatively high principle quantum number and the other is relatively low. Since the decay rates (Einstein A-coefficients) scale strongly with n (roughly as n^{-3}), asymmetric capture ensures that the electron at lower- n cascades down to the ground state while the electron captured into high- n remains at high- n for a reasonably large fraction of the decays. In the model developed by the Kansas State group [19], capture of multiple electrons into $n=7-9$ for H-like Ar, which then stabilize with one electron cascading to the ground state while one electron remains in the initial $n=7-9$ state and any additional electrons are ejected during the cascade, is explicitly discussed.

Thus, it seems reasonable to assume that the principal quantum number of the spectator electron is in the range $n=5-10$. The magnitude of the shift for spectators in these levels is considered below.

2.3. Magnitude of the wavelength shift of the satellite line

From table 3 of the work of the TFR group *et al* [21], the calculated wavelength shifts for high- n satellites to the w-line fall into two groups, depending on whether the upper state is even or odd. Table 1 lists the shifts [21] for both sets. These shifts are taken from the calculated unperturbed w-line obtained by extrapolating the odd series to infinity using $1/n^3$ scaling. The even series extrapolates to the same limit to better than 13 meV. The satellite lines are on the long wavelength (low energy) side of the primary line. Independent studies (see e.g. [22], p 170) suggest that capture may predominantly take place into one particular sublevel, although it can be difficult to reliably predict which one for capture from multi-electron atoms. Hence it is assumed here that either the even or odd shift dominates, but no assumption is made about which of the two actually dominates.

2.4. Magnitude of the systematic error

The measured w-line profile shown in figure 6 of Kubicek *et al* [16] is taken as a prototype and simulated for the current purposes by two Gaussians, one with 8% of the intensity of the strongest line but shifted by the range of values Δ listed in table 2. A more complete analysis would use a Voigt profile, but near the line center the difference between the two is small. The Gaussian width parameter was chosen to correspond to 41 channels FWHM ($=1.14$ eV) and the peak intensity was chosen to be 900 counts (at zero shift) to match that in the figure reporting the experimental results. Random shot noise was added from a Poisson distribution and a constant background of 50 photons/channel was added. When the simulated profile was fit to a single Gaussian, the fit center deviated from the actual center of the primary line by the amounts given by S in table 2 (the values tabulated are the average of seven simulations and fits). For satellite line shifts that are within three-quarters of a FWHM of the primary line ($\Delta < 0.75$), the residuals typically appear normal to the eye due to the obscuring nature of the shot noise. This is shown explicitly in figures 2–4. Even at relative intensities approaching 100%, the residuals appear normal for $\Delta \leq 0.5$. Figure 5 shows the individual components of the profile simulated in figure 3. The systematic error has a maximum near the point where the residuals begin to show a systematic deviation (around $\Delta = 0.75$); beyond this point the systematic error begins to decrease because the fit can distinguish between the two lines and more accurately hone in on the center of the primary line.

Although I do not claim that the present simulation corresponds to the actual experiment of Kubicek *et al* [16], I note for comparison that if the sort of correction estimated here were applied to that work, it would move their published value for the line center, 3139 581(5) meV, closer to the deviation curve obtained in the meta-analysis of Chantler *et al* [9], 3139 640(12) meV (68% confidence level), by the amounts shown in figure 6. For Δ near 0.5 and 8% relative intensity, the systematic error is in the range 30–37 meV. The estimated 8% relative intensity presented here could easily be

Table 2. Systematic shifts (fourth column) in the apparent energy of the w-line of He-like Ar, induced by satellite lines populated at an intensity 8% of the primary line and shifted by the amount $\Delta = \text{satellite shift}/\text{FWHM}$, for $\text{FWHM} = 1.14 \text{ eV}$. The fifth column indicates whether or not a systematic error appears visually evident in the residuals to a single Gaussian profile fit to the composite line (see figures 2–4).

| Capture level | $\Delta = \text{satellite shift}/\text{FWHM}$ | $S = \text{fit shift}/\text{FWHM}$ | Systematic error (meV) | Visible? | Figure |
|-------------------------------------|---|------------------------------------|------------------------|----------|--------|
| $n = 5$ (odd) | 1.1 | 1.9% | 22 | Yes | 4 |
| $n = 6$ (odd) or $n = 5$ (even) | 0.62 | 3.3% | 37 | Rarely | 3 |
| $n = 7$ (odd) | 0.41 | 2.6% | 30 | No | |
| $n = 6$ (even) | 0.34 | 2.1% | 24 | No | |
| $n = 8$ (odd) | 0.27 | 1.8% | 20 | No | |
| $n = 9$ (odd) or $n = 7$ (even) | 0.20 | 1.2% | 14 | No | |
| $n = 8, 9$ (even) or $n = 10$ (odd) | 0.13 | 0.8% | 9.2 | No | |
| $n = 10$ (even) | 0.061 | 0.4% | 5.0 | No | |
| $n = \text{infinity}$ | 0 | 0.04% | 0.5 | No | 2 |

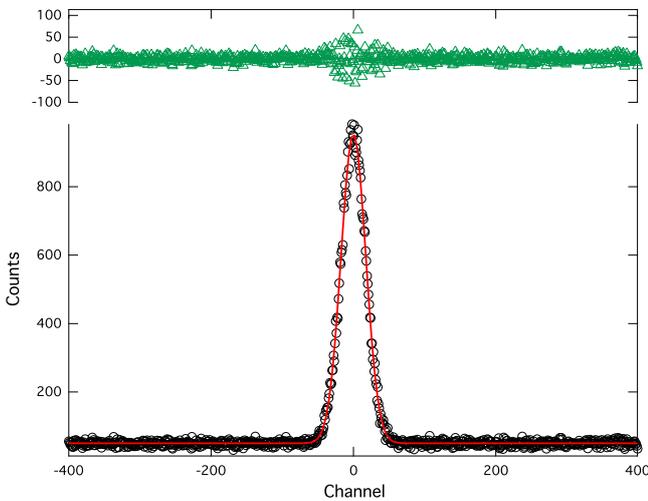


Figure 2. Fit to simulation with zero satellite energy shift.

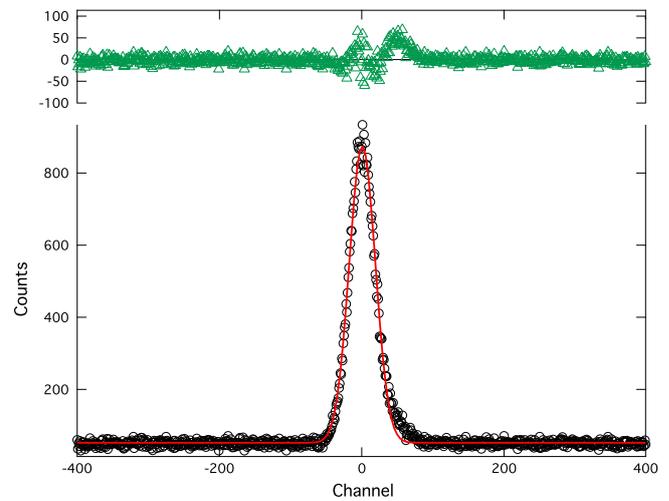


Figure 4. Fit to simulation with 8% satellite intensity at energy shift of 1.1 FWHM.

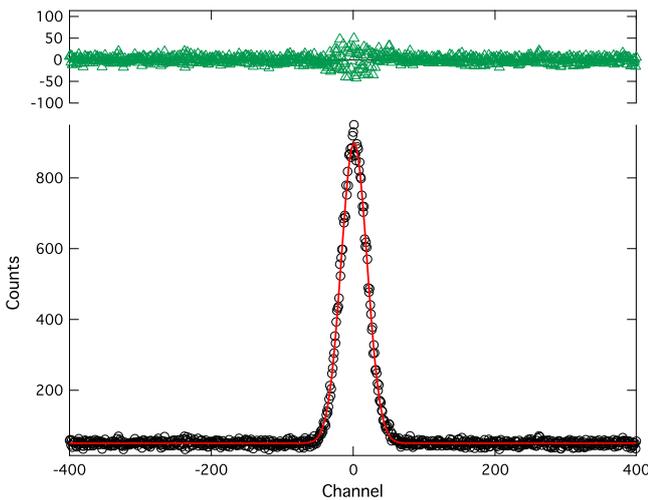


Figure 3. Fit to simulation with 8% satellite intensity at energy shift of 0.62 FWHM.

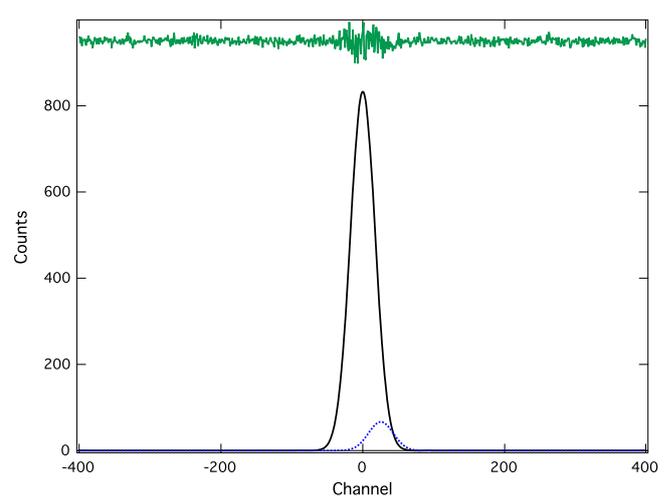


Figure 5. The two Gaussian lines and the shot noise for the case of the profile simulated in figure 3. The shot noise is symmetric about $y = 0$, but is offset in the plot for clarity of presentation.

off by a factor of 2, so also shown in figure 6 is the case for 16% relative intensity, in which case the results overlap with the 1σ error bars of the deviation curve of Chantler *et al* [9] for spectator electrons in the $n = 6$, $n = 7$, or $n = 8$ shell.

The larger corrections scale roughly linearly with the intensity of the satellite line, and thus the $n = 7$ (odd) correction remains larger than the total uncertainty published by Kubicek *et al* [16] down to satellite intensities as low as

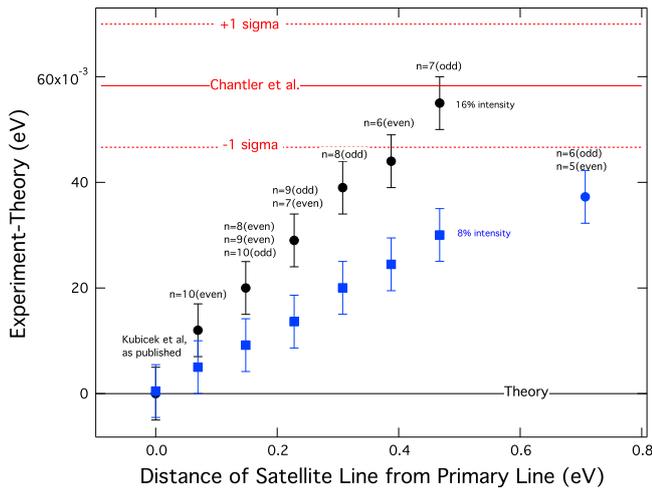


Figure 6. The extent to which the corrections simulated here would shift the measurements of Kubicek *et al* [16] away from the theory of Artemyev *et al* [15] towards the deviation curve of Chantler *et al* [9] for assumptions of 8% satellite intensity (listed in table 2, and plotted as blue squares) and for 16% satellite intensity (black dots). The principal quantum number of the assumed position of the satellite electron from table 1 is indicated in the text above the points. The error bars are 5 meV from Kubicek *et al* [16].

approximately 2% of the primary line. Alternatively, for satellite intensities higher than 20%, even satellites with shifts equal to those predicted here for $n > 10$ can produce a significant systematic error (the difference between the satellite line center and the primary line center scales roughly as $1/n^3$). The simulations indicate that 5–10 times more intensity (photon counts) would be required to visually discern a systematic error in the residuals for the predicted $n = 7$ (odd) satellite at 16% relative intensity.

A similar analysis for potential systematics in the results of Chantler *et al* [9] for the case of He-like Ti is limited by the lack of available data for the corresponding satellite shifts and charge exchange for this ion, but since the titanium experiment [9] has an error bar 14 times larger than that of the argon experiment [16] (70 meV instead of 5 meV) and involved loading the trap in a fundamentally different way which does not lend itself to significant charge exchange (the ions are loaded as low density ions rather than higher density neutral atoms), it seems reasonable to assume that the DCX-mediated satellite shifts considered here are negligible in the experiment of Chantler *et al* [9]. Even if they were not negligible, the expected sign of the shift is such that it would increase the discrepancy with the calculation of Artemyev *et al* [15], rather than reduce it. Note that evaporative cooling gas (typically nitrogen) is sometimes introduced into EBITs during spectroscopy experiments to enhance the signal strengths, and this can enhance the magnitude of charge exchange.

3. More recent experiments

After the work of Kubicek *et al* [16] was published, an independent experiment of similar claimed accuracy was

published by the Paris group and collaborators [23], who measured the separations of w-, x-, y-, and z-lines. That paper reports deviations from the calculation of Artemyev *et al* [15] of up to 45.4 meV (3.8 experimental uncertainties) for He-like Ar. Although this deviation was attributed primarily to the y-line, with the w-line being off by only 15.1 meV (1.4σ), the experiment only measured separations not absolute values, so attributing the discrepancy to the y-line hinges on assuming that the calculation for the z-line is exactly correct (while in fact they reported an absolute measurement of the z-line separately [24] which deviated from the same calculation [15] by 1.6σ). In any case, this result provides additional evidence supporting our [9] general conclusion that there is a deviation between experiment and theory at $Z = 18$ which is approximately equal to 58(12) meV, far above the claimed accuracy of either the experiments or the theory.

The paper by Schlessler *et al* [23] assumes that satellite line shifts are negligible because (1) the residuals did not show evidence of this and (2) previously published calculations [25, 26] predicted that the satellite lines were either too far away in energy or too weak to matter. The first of these reasons seems worth revisiting in light of the results of section 2.4 which explicitly demonstrates that the residuals can be a poor predictor of systematic errors due to satellite shifts in He-like Ar. The second of these reasons is limited to a consideration of shells $n = 4$ and lower (and only for the case of sulfur), whereas the above consideration suggests that it is $n > 5$ that matters in Ar. Because the experiment from the Paris group was performed in an ECR ion source (where the electron energy distribution is wide), satellites can also be populated by dielectronic recombination, which would tend to increase the chance of a significant satellite line shift, but on the other hand that experiment has both higher spectrometer resolution and higher signal/noise than the experiment of Kubicek *et al* [16] so any systematics should be less hidden and thus more easily avoided.

4. Evolution of a discrepancy

Experiment and theory on the He isoelectronic sequence appear to have been teetering on the verge of disagreement for many years, although typically the discrepancy has been on the edge of the error bars and the claims have not been sustained over time. Some of the specific claims from the papers cited by Chantler *et al* [9] are reviewed below.

Beiersdorfer and co-workers first reported a hint of a discrepancy in results from a series of ions published in 1989 [27], noting that ‘In most cases the differences between the measured and calculated wavelengths lie within or just outside the experimental error limits. Therefore we had to rely on a large number of data to demonstrate systematic differences’. This claim was strengthened when they extended their measurements to higher Z in 1992 [28], finding ‘a significant difference between our experiment and the theoretical wavelengths’, but then later, at even higher Z (Kr), a different conclusion was arrived at: ‘unlike earlier measurements, our results are in good agreement with recent theoretical

predictions' [29]. The same conclusion was found in 2009 [30] at even higher Z (Xe): 'the two calculations that include *ab initio* QED contributions...fall within the experimental error'.

The Heidelberg EBIT group began by reporting 'a slight discrepancy to different theoretical approaches' in argon in 2005 [31], but then in 2007, in both Cl and Ar, reported 'we now establish excellent agreement with the predictions of BSQED' [32]. This conclusion was strengthened in 2009 with the study of S ('excellent agreement with all theoretical predictions' [33]) and with their work on Ar in 2012 ('comparing our present result with predictions, it can be stated that it agrees excellently' [16]).

Aglitsky and coworkers, in a study from $Z=16$ to 39 in 1988 [34], claimed 'a slight difference between the theory and the experiment in the region $Z=38, 39$ has been found' but noted that 'in most cases the discrepancy ... is within the limits of the experimental error' and only 'at $Z=29$, Δ exceeds the estimated error of the measurements.' They concluded that '...the analysis of possible errors in the theoretical calculations...does not allow one either to find a definite source of the discrepancy between the theory and the experiment.'

Briand and coworkers have had consistent conclusions in their study of Xe in 1983 ('calculations are compared to the experimental results, and a good agreement between both...is found' [35]) and of U in 1990 ('the absolute energies...are well-fitted with theory' [36]). This conclusion also agreed with the conclusion of Deslattes and coworkers for Ar in 1984 ('our results are in agreement with very recent theoretical approaches employing the most refined evaluations of the two-body QED corrections' [37]).

Chantler and co-workers found no discrepancies in V at the level of accuracy reported in 2000 ('the results are in accord with...current theories' [38]), and only with substantially reduced uncertainty in 2012 for Ti was it reported that: 'our measurement...results in one of the most statistically significant discrepancies from theory' [9].

5. Results for $Z < 12$

Comparing the results of Engstrom and Litzen [39] at $Z=7$ and $Z=8$ to the predictions of Drake [40], one sees a deviation of nearly 3σ and 1.5σ but of the opposite sign compared to the deviation discussed above at higher- Z (in comparison to the predictions of Artemyev *et al* [15]). Unfortunately a direct comparison to Artemyev *et al* [15] is not possible in the lower- Z region because those calculations stop below $Z=12$; moreover, extrapolating their differences from Drake [40] one finds a reversal of sign around $Z=10$. Additional calculations would be useful below $Z=12$ in order to determine whether or not the deviation of experiment from Artemyev *et al* [15] actually passes through zero around $Z=18$, as suggested by a comparison to the results of Drake [40] (see, e.g., Beiersdorfer [27]), or rises monotonically from the origin following a perturbative expansion of the theory as assumed by Chantler *et al* [9].

6. Future refinements of the DCX-mediated satellite model

The considerable limitations of the simple estimates presented here do not make a definitive case for the existence of significant DCX-mediated satellite shifts in the Ar results, but suggest that more detailed modeling and experimental investigation of this systematic is warranted. Particular issues that might be addressed include detailed predictions/measurements of state-dependent (including angular momentum quantum number) electron capture distribution, inclusion of higher order terms in the rate equations (coupling ions separated by two units of charge), calculation of electron cascade to ground state from initial capture distribution, electron impact excitation to $n>2$ followed by cascade to the ground state, more accurate forms for the cross sections, including the effect of double and triple ionization (Santos *et al* [26] were unable to accurately model the observed x-ray emission without this), independent determinations of the neutral gas density in the trap, independent determinations of the velocity and spatial distribution of the various ions and of the electrons in the trap, search for beam-energy dependence and residual pressure dependence of measured wavelengths, the effect of radiative recombination at lower pressures, predictions for beam energies near thresholds, dependence on neutral gas species, velocity dependence of cross sections, more accurate modeling of the lineshape, polarization effects, and detailed analysis of the z -dependence of the overall shift.

7. Conclusion

The bulk of evidence from two sets of spectroscopy experiments [9, 17] in the simplest atomic systems (one- and two-electron atoms and ions) currently show striking disagreements with the predictions of the most advanced atomic theory currently available. The aggregate deviation in both cases rises to the level of 5σ or greater. The disagreement in the two-electron ion case is contradicted by the findings of Kubicek *et al* [16], but the analysis presented here suggests that a previously neglected effect (DCX-mediated high- n satellites) might, within the uncertainty of the present analysis, shift that result into good agreement with the 1σ confidence interval of the deviation curve suggested by the other experiments [9].

There is one more common aspect of the muonic hydrogen and He-like ion experiments that may be worth noting in conclusion: both experiments revealed results that surprised the original investigators and were only published after many years of scrutiny and analysis (the muonic hydrogen experiment first began in 2003 and the He-like Ti experiment took place in 2005; both were published seven years after these dates), and both have triggered considerable debate and controversy which remains unresolved as of the present.

Table A1. Definitions of subscripts used in the equations in this appendix.

| Subscript | Abbreviation |
|-----------|---|
| H | H-like ion |
| He | He-like ion |
| e | electron (from EBIT electron beam) |
| i | ion |
| o | neutral gas (e.g. Ar) |
| rr | radiative recombination |
| cx | total (n electrons captured) charge exchange |
| scx | single (1 electron captured) charge exchange |
| dcx | double (2 electrons captured) charge exchange |
| ioniz | electron impact ionization |
| excite | electron impact excitation |

Acknowledgements

Discussions with Yuri Ralchenko, Paul Indelicato, Csilla Szabo, Yuri Podpaly, Larry Hudson, Chris Chantler, and Joseph Smiga are gratefully acknowledged.

Appendix. DCX-mediated production of doubly excited states

In equilibrium, the time rate of change of the number of ions (dN/dt) in a given charge state is zero. It is known from experiments that the time scale for ion escape from an EBIT is of order hours, whereas the time scale to achieve charge state equilibrium is of order seconds [41], hence for an approximate calculation one can consider only ionization and recombination terms in the rate equations and neglect trap escape. If charge exchange terms are retained only up to first order (i.e. neglecting double electron capture into bare ions), then bare ions are coupled only to H-like ions, and the number of recombination events per second of bare into H-like must equal the number ionization events per second of H-like to bare.

This can then be used to simplify the rate equation for H-like ions: because the number of ionization and recombination events per unit time connecting H-like to bare cancel (previous paragraph), the number of ionization and recombination events connecting H-like to He-like must also cancel (and so on, up the chain of charge states). Thus, the equilibrium condition for each charge state can be written in terms of only one other charge state, allowing a simple expression to be obtained for the ratio. For beam energies away from dielectronic recombination resonances, the rate equation can be solved for the ratio of the number of He-like to H-like ions in terms of the cross sections (σ), the collision velocities (v), and the number of projectile electrons (N_e) or neutral atoms (N_o) inside the ion cloud as follows [1]

$$N_{\text{He}}/N_{\text{H}} = \left[(N_e \sigma_{\text{rr}} v_e) + (N_o \sigma_{\text{cx}} v_i) \right] / (N_e \sigma_{\text{ioniz}} v_e). \quad (\text{A.1})$$

Subscripts are defined in table A1. It is assumed that the velocities are such that $v_o \ll v_i \ll v_e$, and that the He-like and

H-like ions have approximately the same velocity distribution. To the extent that multi-electron processes are not stronger than single-electron processes, the above expression should give the right order of magnitude of the ratio of the number of H-like and He-like ions in the trap.

Trap pressure

The following inequality defines the ‘strong charge exchange regime’ in which radiative recombination can be neglected in (A.1)

$$(N_e/N_o)(\sigma_{\text{rr}}/\sigma_{\text{cx}})(v_e/v_i) \ll 1. \quad (\text{A.2})$$

It will be shown below that this regime is typical for EBIT experiments in which ions are loaded into the trap via a gas injector.

In the strong charge exchange regime, equation (A.1) can be used to determine the approximate density of neutral atoms (n_o) in the trap. In the limit that the H-like and He-like ions occupy approximately the same volume of space, V

$$n_o = (N_{\text{He}}/N_{\text{H}})(n_e F)(\sigma_{\text{ioniz}}/\sigma_{\text{cx}})(v_e/v_i). \quad (\text{A.3})$$

Because it has been previously shown that the ion cloud may be significantly larger than the electron cloud [42], the effective electron density that the ions experience ($n_e F$) may be somewhat different than the actual electron density (n_e) due to the factor F , which is defined to be

$$F = V_{\text{ie}}/V_{\text{io}}, \quad (\text{A.4})$$

the ratio of the volume of the ion cloud that overlaps with the electron beam (V_{ie}) to the volume of the ion cloud that overlaps with the neutral gas (V_{io}). For uniform illumination of the ion cloud with background gas, F is equal to the square of the ratio of the radii of the electron beam and the ion cloud, $V_{\text{ie}}/V_{\text{io}} = (r_e/r_i)^2$, which is typically less than unity. Uniform densities are assumed throughout. Note that in the case of highly collimated gas injection perpendicular to the electron beam [43], F could be greater than unity. The ion motion into and out of the electron beam and/or region of neutral gas is rapid compared to the charge state equilibrium times.

Simple formulas for some relevant cross sections are given below.

Radiative recombination cross section

The total radiative recombination cross section (sum of partial cross sections into all open shells with principal quantum number n) is given by Kim and Pratt [44]

$$\sigma_{\text{rr}} = 5.26 \times 10^{-23} \chi \ln \left[\left(1 + \chi \right) / \left(2n_{\text{ve}}^2 \right) \right] \text{cm}^2 \quad (\text{A.5a})$$

with,

$$\chi = \left[Z + (Z - N') \right]^2 (6.8/E_e), \quad (\text{A.5b})$$

where Z is the number of protons in the nucleus, N' is the number of electrons on the ion (before the radiative recombination takes place), E_e is the electron beam energy in eV, and the effective valence quantum number is given

approximately by

$$n_{ve} = n + 0.7 - W_n \quad (\text{A.5c})$$

with W_n equal to the fraction of available states (ratio of the number of unoccupied states to the total number of states) in the valence shell n . Here, the equations of Kim and Pratt [44] have been converted into formulas involving physical units following Penetrante *et al* [41] (in the latter, $\lambda_c = 3.861 \times 10^{-11}$ cm is the *reduced* Compton wavelength, the Compton wavelength divided by 2π). For a H-like ion, $n=1$ and $W_n=0.5$. For H-like Ar at 9500 eV electron beam energy, equations (A.5a)–(A.5c) give $\sigma_{tr} = 1.2 \times 10^{-23}$ cm². This is approximately a factor of 2 lower than the relativistic Dirac–Fock value calculated by Trzhaskovskaya *et al* [45].

Electron impact ionization cross section

The ionization cross section is given by the Lotz formula [46], which simplifies to a single term for atoms with N electrons in the $n=1$ shell

$$\sigma_{\text{ioniz}} = 4.5 \times 10^{-14} \left[N / (E_c E_{\text{IP}}) \right] \ln (E_c / E_{\text{IP}}) \text{ cm}^2, \quad (\text{A.6})$$

where E_{IP} is the ionization potential of the ion in its initial state, and all energies are expressed in eV. For He-like Ar at 9500 eV beam energy, and $E_{\text{IP}} = 4121$ eV [47], $\sigma_{\text{ioniz}} = 1.9 \times 10^{-21}$ cm².

Electron impact excitation cross section

The electron impact excitation cross section is given by the Van Regemorter formula [48]

$$\sigma_{\text{excite}} = 2.36 \times 10^{-13} f G(E_c) \left(\frac{1}{E_t E_c} \right) \text{ cm}^2, \quad (\text{A.7})$$

with the electron beam energy E_c and transition energy E_t given in electron volts (eV). For He-like ions, the electric dipole oscillator strength (f -value) for the w-line varies from $f=0.73$ to $f=0.75$ for $Z=11$ – 19 [47]. When the electron beam energy is much larger than the transition energy, the Gaunt factor is given simply by (see, e.g., [49])

$$G(E_c) = \left(\frac{\sqrt{3}}{2\pi} \right) \ln (E_c / E_t). \quad (\text{A.8})$$

For the He-like Ar w-line ($E_t = 3140$ eV), equation (A.8) gives $G=0.3$ for a beam energy of 9500 eV. With these values for the w-line, equation (A.7) then gives 1.76×10^{-21} cm².

Charge exchange cross sections

For the case of H-like Ar colliding with neutral Ar, the total charge capture cross section has been measured to be [19]

$$\sigma_{\text{cx}} = 2.35 \times 10^{-14} \text{ cm}^2 \quad (\text{A.9})$$

and the ratio of the double charge capture to single charge capture cross sections has been measured to be approximately

[19]

$$D = \sigma_{\text{dcx}} / \sigma_{\text{scx}} = 0.35. \quad (\text{A.10})$$

The higher order charge capture processes are negligible (contribute only about 4% to the total capture cross section). These measurements (like EBIT experiments) were performed in the low-energy regime where the cross sections are approximately independent of energy [50].

It is important to keep in mind that the cross sections defined here and measured in the experiments [19] are for charge *capture* (in which two electrons are retained on the ion) not merely charge *transfer* (which is often followed by autoionization). Double charge transfer followed by autoionization thereby increases the effective (measured) single capture cross section. The distinction will be important in the considerations below.

Electron to ion velocity ratio

In the non-relativistic limit, the electron velocity is

$$v_e = \sqrt{2E_e / m_e}. \quad (\text{A.11})$$

At 9500 eV beam energy, this expression gives a velocity that is 19% of the speed of light.

The ion velocity is taken to be the characteristic velocity for a thermal distribution with the temperature set by the trap depth (V_t), the Spitzer heating from the electron collisions, and the degree of evaporative cooling. Measurements of the Doppler broadening of the Zeeman splitting of a UV line in Ti-like Ba³⁴⁺ in the NIST EBIT indicate that under ‘deep trap’ (500 V) operating conditions the ion temperature is approximately 750 eV for 2250 eV beam energy and 50 mA beam current, $C=23$ times lower than the value given by equating the electrostatic energy of a minimally trapped ion to kT [51]. On the LLNL EBIT [52], x-ray spectroscopy was performed on a line in He-like Ti²⁰⁺ for typical EBIT operating conditions of 130 mA beam current, 5000 eV beam energy and 300 V trap depth, and the resulting ion temperature was measured to be 550 eV, $C=11$ times lower than the value for an ion minimally trapped by the axial trap potential in that experiment. Thus the ion velocity is taken to be the most probable energy of a Maxwell–Boltzman distribution with a temperature reduced from that set by the energy of the trap depth

$$v_i = \sqrt{2q e V_t / (C m_i)}, \quad (\text{A.12})$$

where the trap depth is of order $V_t = 220$ V, the ion charge is qe with $q=Z-N$, and the evaporative cooling factor $C=10$. For He-like Ar at 220 eV (typical of experiments at the NIST EBIT), this expression gives an ion velocity of 0.014% of the speed of light.

The ratio of the two numerical examples given above is $v_e/v_i = 1400$.

Pressure for strong charge exchange regime

From equation (A.2) and the numerical values above, it can be seen that for H-like and He-like Ar exposed to a 9500 eV

electron beam of effective current density $n_e F = 10^{12} \text{ cm}^{-3}$ [53, 54], and $v_e/v_i = 1400$ one is in the strong charge exchange regime when $n_o \gg 1.4 \times 10^6 \text{ cm}^{-3}$ which corresponds to a room temperature ideal gas pressure of $p \gg n_o kT = 5.7 \times 10^{-11} \text{ hPa}$ ($4 \times 10^{-11} \text{ torr}$). This is on the order of typical EBIT base pressures (without any gas injection) and suggests that one is on the threshold of the strong charge exchange regime even when no additional gas is injected for spectroscopy. In EBIT experiments at NIST using injected argon to study H-like and He-like ions, the estimated pressure at trap center is on the order of $1 \times 10^{-9} \text{ hPa}$ (the pressure measured in the periphery when the trap is at liquid nitrogen temperature and argon gas is injected at the same rate as in the actual experiment); this is approximately 20 times higher than the estimated pressure needed to enter the strong charge exchange regime.

Once it is clear that the EBIT is in the strong charge exchange regime, one can use equation (A.3) and the ideal gas law to determine the actual pressure from the observed relative strengths of lines from He-like and H-like ions. For equal numbers of H-like and He-like Ar in the trap, for example, the intensity of the He-like w-line compared to the sum of the H-like Ly-alpha1 and Ly-alpha2 intensities should be approximately equal to the ratio of the f -values: $\text{He}(w)/\text{H}(1+2) = 0.7/0.4 = 1.75$. Time normalizing the spectra shown by Braun *et al* [31] gives $N_{\text{He}}/N_{\text{H}} = 2$. Using this value together with the cross section ratios and velocity ratios given above, the result is $P = 9.4 \times 10^{-9} \text{ hPa}$ ($7 \times 10^{-9} \text{ torr}$) for $n_e F = 10^{12} \text{ cm}^{-3}$ and lower for lower effective electron densities.

Relative magnitude of charge exchange excitation of a spectral line

From the above (again assuming that the H-like and He-like ions occupy approximately the same volume of space), it can be seen that the ratio of the population flux of producing excited He-like ions (into the upper level of a particular line) by electron impact excitation (R_{ei}) to the population flux of producing the same excited state by single charge capture into H-like ions ($W1 * R_{scx}$) is

$$R = \frac{R_{ei}}{W1 * R_{scx}} = (n_e F / n_o) (n_{\text{He}} / n_{\text{H}}) \left(\frac{\sigma_{\text{excite}}}{W1 * \sigma_{\text{scx}}} \right) (v_e / v_i), \quad (\text{A.13})$$

where $W1$ is the fraction of all single charge capture (not transfer) events that eventually produce photons in the line under question (e.g. the w-line). Note that even though the effective electron density is approximately 4000 times larger than the neutral gas density for $P < 10^{-9} \text{ hPa}$ (first term in (A.13)), and the ion-electron collision velocity is approximately 1000 times higher than the ion-gas collision velocity (last term in (A.13)), the ratio of the excitation cross section to the charge exchange cross section nearly makes up for the product of these other two large numbers.

Removing the neutral gas density from equation (A.13) using equation (A.3) gives

$$R = (\sigma_{\text{cx}} / \sigma_{\text{scx}}) (\sigma_{\text{excite}} / \sigma_{\text{ioniz}}) (1/W1), \quad (\text{A.14a})$$

$$R = 1.45 (\sigma_{\text{cx}} / \sigma_{\text{scx}}) (f/N) (E_{\text{IP}} / E_t) \left\{ \ln(E_e / E_t) / \ln(E_e / E_{\text{IP}}) \right\} (1/W1). \quad (\text{A.14b})$$

Note that in the limit that the total charge exchange cross section is well represented by the single charge exchange cross section (first factor of cross section ratios in (A.14a) and (A.14b) approximately 1), the expression for R is independent of the magnitude of the total charge exchange cross section, and the relevant ratio of cross sections is excitation to ionization (A.14a) rather than excitation to CX (A.13), due to cancellation of terms from the neutral gas density. For the conditions assumed above (beam energy $E_e = 9500 \text{ eV}$, He-like Ar with ionization potential $E_{\text{IP}} = 4121 \text{ eV}$, w-line with transition energy $E_t = 3140 \text{ eV}$, $N = 2$ and $f = 0.74$), $R = 1.3$ for $W1 = 1$. A more realistic estimate for $W1$ can be informed by the observed results of Trassinelli *et al* [18] who have measured the x-ray emission from charge exchange of neutral argon onto H-like Ar over a sufficiently broad range of photon energies that they can observe the relative intensities of Rydberg cascade ($n = 10$ to $n = 1$ emission, $n = 9$ to $n = 1$ emission, ... $n = 2$ to $n = 1$ emission). Their results indicate that the fraction of photons that are emitted into the w-line from excited He-like ions (and their unresolved satellites) created by all orders of charge exchange into H-like ions is approximately $W = 0.4$ (including the fact that the x- and y-lines are blended into an unresolved line of broadened width). Assuming that $W1 = W = 0.4$, then $R = 3.3$.

Remarkably, this ratio is of order unity and does not depend on any of the trap details other than the electron beam energy. It does not depend on pressure (in the regime that it is valid) because raising the P gives fewer H-like target ions in exactly the right amount to cancel the greater number of projectiles. A similar argument explains why this expression is also independent of electron density. The simple calculation below suggests that for reasonable EBIT operating conditions, photon emission mediated by charge exchange may be comparable to (or even larger than) that due to electron impact excitation.

DCX-mediated satellite line intensity ratio

The ratio of the intensity of the unperturbed line to the intensity of the satellite line ($R3$) is approximated by the following

$$R3 = [R_{ei}(\text{He}) + W1 * R_{scx}(\text{H})] / W2 * R_{dcx}(\text{H}). \quad (\text{A.15a})$$

The first term in the numerator is the number of photons per second produced by direct electron impact excitation of the unperturbed line from the ground state of the He-like ions in the trap. The second term in the numerator is the number of photons per second in the unperturbed line produced by single charge exchange from the neutral gas colliding with the H-like ions in the trap. $R_{scx}(\text{H})$ is the number/second of ground

state He-like ions produced by single charge capture into H-like ions (computed using the experimental single capture cross section, as discussed above, thus including all orders of multiple electron transfer and subsequent autoionization that stabilize the excited ion into the ground state with only one net electron captured). The factor $W1$ is the fraction of those single charge exchange processes just described ($R_{scx}(H)$) which emit a photon into the unperturbed line; $W1$ is taken to be approximately 0.4, from the observed results of Trassinelli *et al* [18] discussed below equation (A.14) above. The denominator is the number of photons per second emitted into satellite lines that are within one experimental linewidth of the unperturbed line. Analogous to the second term in the numerator, the denominator contains a factor ($R_{dcx}(H)$) that is the number/second of ground state Li-like ions produced by DCX in collisions of H-like ions with neutral gas, and the factor $W2$ that is the probability that such a ground state Li ion emitted a photon into a nearby satellite of the unperturbed line during its stabilization into the ground state.

By direct substitution from the expressions above, equation (A.15a) becomes

$$R3 = \frac{(\sigma_{cx}/W2*\sigma_{dcx})(\sigma_{excite}/\sigma_{ioniz})}{(W1*\sigma_{scx}/W2*\sigma_{dcx})} \quad (A.15b)$$

or simply,

$$R3 = (R + 1)(W1/W2)/D. \quad (A.15c)$$

For high Rydberg spectators ($n \gg 1$), the second excited electron in the Li-like ion is assumed to cascade to the ground state as it would in a He-like ion, so $W1$ and $W2$ are of similar magnitudes. For simplicity, it is assumed here that $W1 = W2$. Taking, as discussed above, $W1 = 0.4$, $D = 0.35$ and $R = 3.3$, one finds $R3 = 12$, and thus the relative satellite intensity $1/R3 = 8\%$. Note that this is independent of pressure in the range over which the above approximations hold.

References

- [1] Ralchenko Y and Gillaspay J D 2013 *Phys. Rev. A* **88** 012506
- [2] Gillaspay J D, Osin D, Ralchenko Y, Reader J and Blundell S A 2013 *Phys. Rev. A* **87** 062503
- [3] Chantler C T *et al* 2013 *Phys. Rev. Lett.* **110** 159302
- [4] Reader J, Gillaspay J D, Osin D and Ralchenko Y 2012 *AIP Conf. Proc.* **1438** p 86
- [5] Osin D, Reader J, Gillaspay J D and Ralchenko Y 2012 *J. Phys. B: At. Mol. Opt. Phys.* **45** 245001
- [6] Osin D, Gillaspay J D, Reader J and Ralchenko Y 2012 *Eur. Phys. J. D* **66** 286
- [7] Kilbane D, O'Sullivan G, Gillaspay J D, Ralchenko Y and Reader J 2012 *Phys. Rev. A* **86** 042503
- [8] Gillaspay J D 2012 *AIP Conf. Proc.* **1438** p 97
- [9] Chantler C T *et al* 2012 *Phys. Rev. Lett.* **109** 153001
- [10] Silver E *et al* 2011 *AIP Conf. Proc.* **1336** p 146
- [11] Ralchenko Y, Draganic I N, Osin D, Gillaspay J D and Reader J 2011 *Phys. Rev. A* **83** 032517
- [12] Gillaspay J D, Lin T, Tedesco L, Tan J N, Pomeroy J M, Laming J M, Brickhouse N, Chen G X and Silver E 2011 *Astrophys. J.* **728** 132
- [13] Draganic I N, Ralchenko Y, Reader J, Gillaspay J D, Tan J N, Pomeroy J M, Brewer S M and Osin D 2011 *J. Phys. B: At. Mol. Opt. Phys.* **44** 025001
- [14] Gillaspay J D 2010 *J. Instrum.* **5** C10005
- [15] Artemyev A N, Shabaev V M, Yerokhin V A, Plunien G and Soff G 2005 *Phys. Rev. A* **71** 062104
- [16] Kubicek K, Braun J, Bruhns H, Lopez-Urrutia J R C, Mokler P H and Ullrich J 2012 *Rev. Sci. Instrum.* **83** 013102
- [17] Pohl R *et al* 2010 *Nature* **466** 213
- [18] Trassinelli M, Prigent C, Lamour E, Mezdari F, Merot J, Reuschl R, Rozet J P, Steydli S and Vernhet D 2012 *J. Phys. B: At. Mol. Opt. Phys.* **45** 085202
- [19] Ali R, Cocke C L, Raphaelian M L A and Stockli M 1994 *Phys. Rev. A* **49** 3586
- [20] Ali R, Cocke C L, Raphaelian M L A and Stockli M 1993 *J. Phys. B: At. Mol. Opt. Phys.* **26** L177
- [21] Group T F R, Bombarda F, Belydubau F, Faucher P, Cornille M, Dubau J and Loulergue M 1985 *Phys. Rev. A* **32** 2374
- [22] Palchikov V G and Shevelko V P 1995 *Reference Data on Multicharged Ions* (Heidelberg: Springer)
- [23] Schlessler S *et al* 2013 *Phys. Rev. A* **88** 022503
- [24] Amaro P, Schlessler S, Guerra M, Le Bigot E O, Isac J M, Travers P, Santos J P, Szabo C I, Gumberidze A and Indelicato P 2012 *Phys. Rev. Lett.* **109** 043005
- [25] Martins M C, Marques J P, Costa A M, Santos J P, Parente F, Schlessler S, Le Bigot E O and Indelicato P 2009 *Phys. Rev. A* **80** 032501
- [26] Santos J P, Costa A M, Marques J P, Martins M C, Indelicato P and Parente F 2010 *Phys. Rev. A* **82** 062516
- [27] Beiersdorfer P, Bitter M, Vongoeler S and Hill K W 1989 *Phys. Rev. A* **40** 150
- [28] Maclaren S, Beiersdorfer P, Vogel D A, Knapp D, Marrs R E, Wong K and Zasadzinski R 1992 *Phys. Rev. A* **45** 329
- [29] Widmann K, Beiersdorfer P, Decaux V and Bitter M 1996 *Phys. Rev. A* **53** 2200
- [30] Thorn D B, Gu M F, Brown G V, Beiersdorfer P, Porter F S, Kilbourne C A and Kelley R L 2009 *Phys. Rev. Lett.* **103** 163001
- [31] Braun J, Bruhns H, Trinczek M, Lopez-Urrutia J R C and Ullrich J 2005 *Rev. Sci. Instrum.* **76** 073105
- [32] Bruhns H, Braun J, Kubicek K, Lopez-Urrutia J R C and Ullrich J 2007 *Phys. Rev. Lett.* **99** 113001
- [33] Kubicek K, Bruhns H, Braun J, Lopez-Urrutia J R C and Ullrich J 2009 *J. Phys.: Conf. Ser.* **163** 012007
- [34] Aglitsky E V, Antsiferov P S, Mandelstam S L, Panin A M, Safronova U I, Ulitin S A and Vainshtein L A 1988 *Phys. Scr.* **38** 136
- [35] Briand J P, Mosse J P, Indelicato P, Chevallier P, Girardvernhet D, Chetoui A, Ramos M T and Desclaux J P 1983 *Phys. Rev. A* **28** 1413
- [36] Briand J P, Chevallier P, Indelicato P, Ziocck K P and Dietrich D D 1990 *Phys. Rev. Lett.* **65** 2761
- [37] Deslattes R D, Beyer H F and Folkmann F 1984 *J. Phys. B: At. Mol. Opt. Phys.* **17** L689
- [38] Chantler C T, Paterson D, Hudson L T, Serpa F G, Gillaspay J D and Takacs E 2000 *Phys. Rev. A* **62** 042501
- [39] Engstrom L and Litzen U 1995 *J. Phys. B: At. Mol. Opt. Phys.* **28** 2565
- [40] Drake G W 1988 *Can. J. Phys.* **66** 586
- [41] Penetrante B M, Bardsley J N, Dewitt D, Clark M and Schneider D 1991 *Phys. Rev. A* **43** 4861
- [42] Porto J V, Kink I and Gillaspay J D 2000 *Rev. Sci. Instrum.* **71** 3050
- [43] Lopez-Urrutia J R, Beiersdorfer P, Widmann K and Decaux V 2002 *Can. J. Phys.* **80** 1687
- [44] Kim Y S and Pratt R H 1983 *Phys. Rev. A* **27** 2913
- [45] Trzhaskovskaya M B, Nikulin V K and Clark R E H 2009 *At. Data Nucl. Data Tables* **95** 987

- [46] Lotz W 1967 *Z. Phys.* **206** 205
- [47] Kramida A, Ralchenko Y, Reader J and NIST ASD Team 2013 *NIST Atomic Spectra Database* version 5.1 (<http://physics.nist.gov/asd>)
- [48] Van regemorter H 1962 *Astrophys. J.* **136** 906
- [49] Sampson D H and Zhang H L 1992 *Phys. Rev. A* **45** 1556
- [50] Janev R K 1992 *Atomic and Plasma material Interaction Data for Fusion* vol 3 (Vienna: IAEA) p 71
- [51] Adler H, Meyer E S, Serpa F G, Takacs E, Gillaspay J D, Brown C M and Feldman U 1995 *Nucl. Instrum. Methods Phys. Res. B* **98** 581
- [52] Beiersdorfer P, Decaux V, Elliott S R, Widmann K and Wong K 1995 *Rev. Sci. Instrum.* **66** 303
- [53] Liang G Y *et al* 2009 *Astrophys. J.* **702** 838
- [54] Matranga M *et al* 2003 *Nucl. Instrum. Methods Phys. Res. B* **205** 244