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Empirical Formulae for Direct Double Ionization by Bare Ions: Z= -1 to +92

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Abstract: Experimental cross sections and cross section ratios reported in the literature for direct double ionization of the outer shells of helium, neon and argon atoms resulting from bare ions ranging from protons to uranium and for antiprotons are analyzed in terms of a first-order, second-order interference model originally proposed by McGuire. Empirical formulae for the various contributions to double ionization plus information about the phase difference between the 1st- and 2nd-order mechanisms are extracted from the data. Projectile and target scalings are also extracted. Total cross sections and their ratios determined using these formulae and scalings are shown to be in very good agreement with experimental data for lower Z projectiles and impact velocities larger than 1 a.u. For very high Z projectiles, the amount of double ionization is overestimated, probably due to saturation of probabilities which is not accounted for in scaling formulae.

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Introduction:

Experimental studies have shown that multi-electron transitions (where two or more outer shell electrons are ionized) are major contributors to the total ionization cross section in collisions involving heavy particles. From a dosimetry viewpoint, multiple ionization of the target can be far more important than single ionization. This is because the ionization potentials increase rapidly for multiple electron removal which means the energy deposited when multiple ionization occurs is typically significantly larger than for single electron removal. In addition, multiple ionization of molecules leads to the production of several charged fragments which undergo coulomb explosion to produce energetic ions capable of inducing subsequent chemical or biological processes. In projectile ionization, multiple ionization can be beneficial in that it is exploited for accelerating particles to high energies or it can be detrimental in high energy accelerators or storage rings where it leads to beam losses which cause technical problems such as vacuum degradation, erosion of exposed surfaces and increased radiation levels.

In contrast to excitation or ionization of a single electron by fast bare ions and electrons where perturbation theories such as the first Born approximation can accurately predict total cross sections, modeling multi-electron transitions is much more difficult. Unlike the situation for photo double ionization, for direct multiple ionization of outer shell electrons by charged particle impact the most common approach is to use an independent electron model (IEM) where the projectile interacts with each target electron independently. In the independent electron model, the single and multiple ionization cross sections scale as $(Ze/v)^{2n}$ where Z and v are the projectile charge and velocity, e is the electron charge, and n is the number of ionized electrons; see [1] and [2] and references therein. However, two aspects of the independent electron model that are inconsistent with experimental data are a) it predicts the relative amount of double ionization, i.e., $R_2 = \sigma_2/\sigma_1$ which corresponds to $n = 2$ and 1 in the scaling relationships, to steadily decrease and ultimately

become negligible for very large v, and b) the double to single ionization ratios should be the same for positive and negative values of Z, i.e., for electron and proton, for positron and electron, and for antiproton and proton impact. In contrast, experimental data shows a constant value at high velocities and larger ratios for negative particle impact than for positive particle impact.

Three decades ago, McGuire proposed that for direct double ionization of helium these inconsistencies could be resolved [3] if, in addition to the second-order independent electron mechanism mentioned above (where n=2), first-order mechanisms, such as those used to explain double ionization by photons, are considered. One first-order mechanism is shake-off (SO) where a large energy transfer ejects one of the target electrons with high speed without any subsequent interaction with any other target electron. This causes an abrupt change in potential of the target electronic cloud, which liberates a second electron with low energy. Quantum mechanically, SO is described by an overlap between the initial and final atomic states. Another first-order mechanism, commonly referred to as two-step-one interaction, (TS1), involves a low-energy transfer which ejects one of the target electrons. As this electron exits it knocks out a second electron; this is also known as interchannel coupling. Note, however, that in total cross section measurements there is a fundamental difference between double ionization resulting from photon and charged particle impact, even if only 1st-order processes are considered. This is because in photoionization, excluding Compton scattering, the entire photon energy is deposited whereas for charged particles a distribution of energies, depending on the impact parameter, is transferred. It turns out that the charged-particle impact ratio can be expressed as a weighted integral over the photon ratios at the various energies [4,5]. In fact experimentally, the asymptotic photon ratio is about 1.7% [6], while the charged-particle ratio is 0.26% [7]. The other major difference is that the IEM model does not apply for photoionization because the photon only interacts once; hence the second-order interaction, often referred to as the two-step-two interaction (TS2) mechanism, does not exist.

McGuire pointed out that these first- and second-order mechanisms can interfere with the interference term being proportional to $(Ze/v)^3$. This is illustrated in equation (1) where the double ionization cross section is written as the sum of a 1st- and a 2nd-order cross section plus an interference term,

$$\sigma_2 = \sigma_{1st} + \sigma_{2nd} + \sigma_{Int}. \quad (1)$$

In terms of scaled amplitudes these cross sections are defined as $[(Ze/v)A_{1st}]^2$, $[(Ze/v)^2A_{2nd}]^2$ and $2\cos(\varphi)(Ze/v)^3A_{1st}A_{2nd}$ respectively, where the A's are amplitudes and φ is the phase difference between the matrix elements corresponding to the1st- and 2nd-order mechanisms.

Dividing by the single ionization cross section, a predominantly 1st-order process which scales as $(Ze/v)^2ln(v)$, yields the ratio version of equation (1), namely $R_2 = R_{1st} + R_{2nd} + R_{Int}$. Because of the different projectile charge and velocity dependences, this model predicts that for large Z/v, i.e., at lower velocities for a given Z, the 2nd-order TS2 mechanism dominates and R_2 decreases approximately as $(Z/v)^2$, while for intermediate values of Z/v, i.e., at higher velocities for a given Z, the falloff is slower since the TS2 term decreases faster than the interference term does, and as $Z/v \rightarrow 0$, corresponding to very high velocities, the first-order term dominates and R_2 approaches a constant value. In this model the phases are such that the cosine term is assumed to be positive because then the interference term, R_{Int}, will be positive for negative Z and negative for positive Z, in agreement with the larger ratios that are observed for negative projectiles, i.e., for electrons and antiprotons, with respect to positive particles such as positrons, protons, and positive ions.

The McGuire paper stimulated numerous experimental investigations of double ionization where, to avoid complications resulting from inner shell Auger processes, double ionization of helium was primarily studied and coincidence methods were often used in order to eliminate contributions associated with electron capture or loss by the projectile. A few examples include the work of Haugen et al. [8] and Knudsen et al. [9] who measured double to single ionization ratios for a number of different ions and energies with Z/v ranging from 0.1 to 3, with v in atomic units. Ullrich et al. [7] tested the high velocity limit of R_2 by using relativistic bare neon and nickel ions with velocities ranging from ~40 to 90% the speed of light. In agreement with the findings of Knudsen et al. they found that for larger Z/v the ratios increased as v^{-2} but were velocity independent as $Z/v \rightarrow 0$. Antiproton and proton impact data were compared [10-13] with the observed differences in the double ionization being attributed to a quantum interference, in accordance with the McGuire model. For a synopsis of the antiproton and proton studies, see [11] and references therein.

The McGuire paper plus the advent of antiproton impact data also stimulated numerous theoretical studies and have produced various explanations for the physical mechanisms responsible for the features described above. For example, the observed differences in the double ionization have been attributed to different ionization probabilities, depending on the sign of the projectile charge, at small impact parameters [14,15]; to the Z^3 term, in accordance with the interference model of McGuire [16]; to the two electrons being removed in uncorrelated events at different internuclear distances [17,18] and to electron correlation in the initial and final states [1,19,20]. In general, all of these models yielded reasonable agreement at high energies but overestimated the cross sections at lower energies. So recently, Kirchner et al. [21] addressed the low energy problem and showed that the inclusion of target polarization and binding effects reduced both the single and double ionization cross sections for antiproton impact at low energies, but not enough to match the experimental data. They attributed the remaining discrepancies to the lack of correlation in their model. A different model based up the strong electric field of the projectile leading to ejection or capture of target electrons has also been proposed [22,23] but its range of application should be limited to regions where Z/v is large and, in contrast to the McGuire and several of the other models, has no explicit 1^{st}-order, 2^{nd}-order interference contributions to explain the observed differences in double ionization resulting from positive and negative projectile impact.

Therefore, many different theoretical approaches have been tried and various explanations have been offered with regard to the observed velocity dependences for double ionization and why double ionization is larger at low and intermediate energies for negatively charged projectile impact than it is for positively charged projectiles. However, all of these comparisons have been hampered in several ways. First, in most cases only selected systems have been modeled, e.g., typically only proton and antiproton data or only low Z positive ion data. Second, in certain cases only the 2^{nd}-order mechanism has been modeled with the corresponding 1^{st}-order mechanism and any possible interference contributions lacking. Third, when both 1^{st}- and 2^{nd}-order contributions have been included in the theoretical models, we are aware of only one or two cases where results for the individual contributions have been presented. Almost exclusively only the overall sum has been compared with experiment simply because experimental information about the individual components is lacking, at least at the integral cross section level.

This work addresses these problems within the context of the 1^{st}-order, 2^{nd}-order interference model of McGuire and is a follow-up of an earlier study where, based upon observed velocity dependences, we concluded that 1^{st}-order double ionization processes dominated for Z=1

projectiles [24,25]. Here, we show that this interpretation was incorrect and that the observed velocity dependence is significantly influenced by the large interference contribution for proton and antiproton impact. The present work shows that all existing data for direct double ionization of helium by bare ions are consistent with the 1st-order, 2nd-order interference model of McGuire. More importantly, the present study provides quantitative information and predictions about contributions of the 1st- and 2nd-order double ionization channels as well as for the interference term for a wide range of systems and energies. This is achieved by extracting empirical scalings for the various contributions to double ionization plus information about the phase difference between the 1st- and 2nd-order mechanisms from experimental data. The empirical formulae we obtain can be used either to a) guide theory by showing which process needs to be modeled, b) indicate where interference needs to be included and where it can be ignored, c) provide quantitative predictions for the individual double ionization mechanisms that can be individually compared with various theoretical models, or d) simply used to test the reliability of existing or new data.

Analysis procedure:

Our analysis is based on a large database of published experimental cross sections and double ionization ratios [7-13, 26-38]. To simplify the interpretation, only data for the direct ionization channel, i.e., only data where there are no contributions from the electron capture channel, and fully stripped projectile impact were used. Also, where multiple sets of data are available for a particular ion, we have arbitrarily selected only one which we consider to be "representative and reliable". Figure 1 shows how the measured double to single ionization ratios of helium, R_2, vary as a function of impact velocity, v, for fully stripped ions ranging from protons to bare krypton, U^{90+} and for antiprotons. As illustrated by the proton and neon data, when Z/v is small, i.e. Z is small and/or v is large, the ratios are in near agreement with the theoretical shake limit, (see ref. 7) shown by the arrow on the righthand axis. But as Z/v becomes large the ratios increase approximately as v^{-2}, as shown by the solid line through the He^{2+} data. This increase was attributed to the 2nd-order mechanism becoming dominant. However, for proton and antiproton impact, Fig. 1 shows a dependence more consistent with a v^{-1} behavior, as shown by the dashed line through the proton data. We recently noted this [24, 25] and attempted to interpret the proton and antiproton data solely in terms of the 1st-order SO and TS1 mechanisms which were assumed to interfere. We note that Shao et al. also found a v^{-1} behavior for conditions where Z/v >1. [22,23].

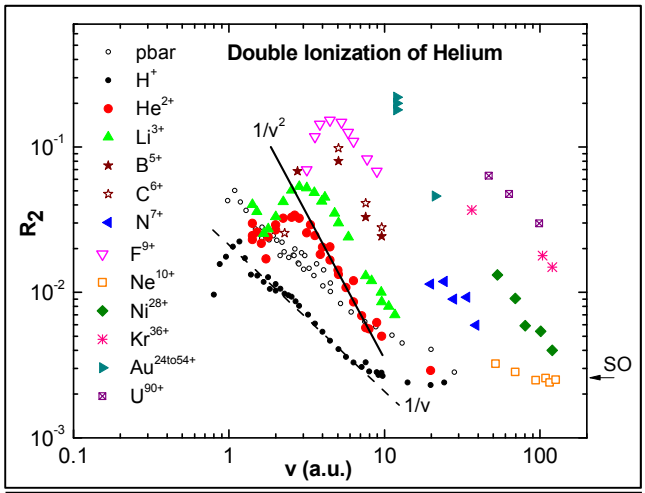

Fig. 1: (color online) Measured ratios for double ionization of helium by bare ion impact as a function of impact velocity in atomic units. Data are from refs. 7-13, 28-40. The lines illustrate v^{-1} and v^{-2} velocity dependences.

To investigate the different velocity dependences for the Z=1 and higher Z projectiles, plus to fully test the 1st-order, 2nd-order interference model of McGuire, the experimental asymptotic high-energy ratio, which is assumed to be entirely first order, $R_{1st}(\infty)$=0.00259 [7], was subtracted

from each experimental R_2 value in Fig. 1. We note parenthetically that this experimental ratio agrees essentially exactly with the theoretical 1st-order i.e., shake, ratio for large v [39]. Here we reiterate that the shake ratio for charged particle impact is considerably smaller than that for photon impact, as mentioned above. This is because the photon deposits its entire energy whereas for charged particles, a distribution of energies which depend on the impact parameter, are transferred. For ion impact, the mean energy transfer is on the order of 20-30 eV and is relatively independent of impact energy [40]. Thus, $R_{1st}(\infty)$ for charged particles should be approximately the same as $R_{1st}(h\upsilon\sim30eV)$ for photon impact, which it is [41]. Also, because the energy transfer is approximately constant and because Fig. 1 shows that with decreasing velocity the measured ratios are considerably larger than their asymptotic limit, subtracting a constant value is reasonable. According to the ratio version of equation (1), this subtraction should isolate the second order plus interference contributions, $R_{2nd} + R_{Int}$, if the McGuire model is applicable.

After subtracting $R_{1st}(\infty)$, it was found that the maxima clearly observed for the Z=2 to 9 data in Fig. 1 could be normalized together by dividing $R_2 - R_{1st}(\infty)$ by $Z^{6/5}$ and v by $Z^{2/5}$. As seen in Fig. 2, after subtracting a constant 1st-order contribution and scaling both the ratios and velocities, the higher velocity dependences of $R_{2nd} + R_{int}$ for Z>1 are consistent with a 2nd-order process being dominant, e.g., either $1/v^2$ or $1/[ln(v)v^2]$. Thus, the scaled velocity dependence, i.e., $(v/Z^{2/5})^{-2}$, combined with the scaled ratio dependence yields the expected IPM cross section scaling, namely $\sigma_n \sim (Z/v)^{2n}$. Further support of a dominant 2nd-order mechanism is the strong similarity between the scaled 2nd-order plus interference contributions and normalized cross sections for direct, predominantly single, ionization of helium by protons [42], shown by the solid black curve in Fig. 2. Finally, the larger and smaller ratios for antiproton and proton impact imply strong interference effects, in accordance with the McGuire model. Also, because all positive ions except protons are seen to scale, the proton and antiproton differences cannot be explained simply by differences in target polarization or distortion due to the sign of projectile charge as was suggested in [20]. The reader should note that some of the proton data around 10 a.u. and a few of the Ne^{10+} data points from Fig. 1 are missing in Fig. 2 since the subtraction yielded negative values which cannot be shown on a logarithmic scale. Also note that scaled asymptotic values, denoted by $R_{1st}(\infty)$, are indicated at the righthand axis in order to illustrate the relative importance of the 1st- and 2nd-order mechanisms as Z increases.

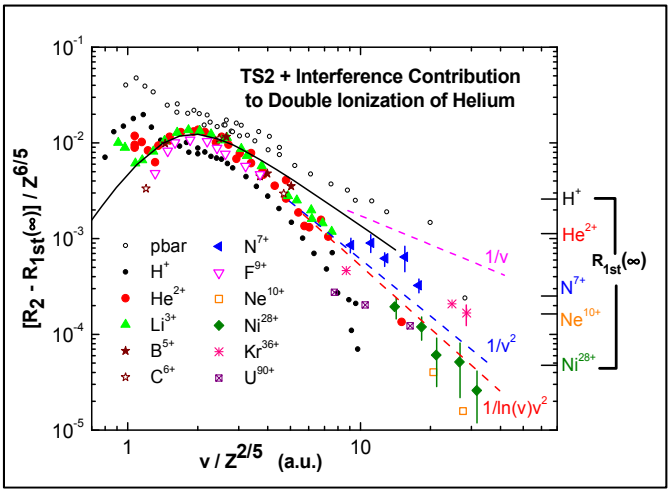

Fig. 2: (color online) Scaled 2nd-order plus interference contributions to the double ionization ratios in Fig. 1. Data sources are the same as in Fig. 1. The dashed lines illustrate v^{-1}, v^{-2}, and $[\ln(v)/v^2]^{-1}$ velocity dependences while the solid line shows normalized single ionization cross sections from ref. 42. Scaled values for $R_{1st}(\infty)$ are shown at the right axis.

Ionization by protons and antiprotons:

To study the dependences seen in Fig. 2 in more detail, experimental cross sections for single and double ionization by antiprotons and protons [9,13-,26,27,36] were investigated. These

cross sections were fit with simple functions in order to provide scaling formulae for the 1st- and 2nd-order and for the interference contributions to double ionization. The scaling formulae were then used to calculate double to single ionization ratios which can be compared to the experimental data in Figs. 1 and 2.

Rather than the more complex procedures and formulae recently presented for determining ionization by any projectile [43], we fit the single ionization data with simple functions of the form $C_{lo}v^m$ below the cross section maxima and $C_{hi}f(v)$ above the maximum. Here C_{lo}, C_{hi} and m are fitting parameters and f(v) is the velocity dependence well above the maxima, e.g., $ln(v)/v^2$. The overall fit was the combination of these functions, e.g., $[\{C_{lo}v^m\}^{-1} + \{C_{hi}f(v)\}^{-1}]^{-1}$. This required extending the "hi" function to scaled velocities less than 1, therefore a constant k was added in the ln term to avoid negative ln values which prohibited achieving a good fit in the low velocity region. Thus, for single ionization, $f(v) = ln(k+v)/v^2$. Using k=0.4, C_{lo}=0.375, m=4, and C_{hi}=4.75, a good fit to the proton data was obtained. Based upon the latest data [13], near the cross section maximum (e.g., ~100 keV) the single ionization cross sections for antiproton impact are approximately 20% smaller than for proton impact but become significantly larger than for proton impact for energies less than 25 keV. A good fit to the antiproton data in the low energy region was achieved for C_{lo}=0.65 and m=1.5. Note that in fitting the proton data, the early single ionization measurements of Afrosimov et al. [36] were not included as they are significantly larger than cross sections measured by Shah, McCallian and Gilbody [27] and the total cross sections of Rudd et al.[42]

The double ionization is more complex since equation 1, the McGuire model, shows that three terms contribute. We obtained information about the individual terms plus about the phase difference, φ, between the 1st- and 2nd-order mechanisms in the following manner. Although the Born approximation implies the 1st and 2nd-order amplitudes are the same for proton and antiproton impact, as stated above experimental data shows small differences, on the order of 20% or less, for velocities between approximately 1 and 3. Therefore, according to the definitions of the terms and equation (1):

$$\sigma_2(\text{proton}) = \sigma_{1\text{st proton}} + \sigma_{2\text{nd}} - \sigma_{\text{Int}} \quad (2a)$$

$$\sigma_2(\text{antiproton}) = \sigma_{1\text{st antiproton}} + \sigma_{2\text{nd}} + \sigma_{\text{Int}} \quad (2b)$$

and

$$\sigma_{\text{Int}} = 2\cos(\varphi)[\sigma_{1\text{st}}\,\sigma_{2\text{nd}}]^{1/2}. \quad (2c)$$

Therefore, adding and subtracting equations (2a) and (2b) isolates $\sigma_{2\text{nd}}$ and σ_{Int}; combining the results will yield the phase information, e.g.,

$$\sigma_{2\text{nd}} = [\sigma_2(\text{antiproton}) + \sigma_2(\text{proton})]/2 - \sigma_{1\text{st ave}} \quad (2d)$$

and

$$\cos(\varphi) = [\sigma_2(\text{antiproton}) - \sigma_2(\text{proton})]/4[\sigma_{1\text{st ave}}\,\sigma_{2\text{nd}}]^{1/2} \quad (2e)$$

where $\sigma_{1\text{st ave}}$ is the average of $\sigma_{1\text{st proton}}$ and $\sigma_{1\text{st antiproton}}$. Note that it is also possible to use double to single ionization ratios, rather than cross sections, in equations (2). For the cross section version, $\sigma_{1\text{st}}$ is obtained from $R_{1\text{st}}(\infty)\sigma_1$. As stated previously, we expect this to be reasonably accurate except perhaps at lower velocities where, in any case, $\sigma_{2\text{nd}} >> \sigma_{1\text{st}}$. $\sigma_{2\text{nd}}$ can also be obtained by multiplying $R_{2\text{nd}}$ by σ_1 with $R_{2\text{nd}}$ being obtained from the ratio version of equation (2d).

Fig. 3 shows $\sigma_{2\text{nd}}$ determined directly by adding the proton and antiproton double ionization cross sections (solid squares) and indirectly by adding their double ionization ratios and multiplying the result by the experimental single ionization cross sections (open squares). Fitting $\sigma_{2\text{nd}}$ with the same functional forms as for single ionization yielded constants for the "lo" region of c_{lo}=0.008 and m=2. For the "hi" region, a fit using $f(v)=v^{-n}$ yielded n between 3 and 3.5 which is significantly slower than the v^{-4} dependence suggested by McGuire for a second order process. However, if a sequential two-step double ionization process is assumed, where the first electron is

ionized via a distant interaction, which scales as $ln(v)/v^2$, followed by a second, closer, interaction, that liberates the second electron and scales as v^{-2}, a slower falloff of $ln(v)/v^4$ is predicted. This is in good agreement with the experimental data, as shown in Fig. 3, and supports the theoretical model used in ref. 17. The solid curve in Fig. 3 has a high velocity dependence of $f(v) = ln(0.4+v)/v^4$ and uses a constant of c_{hi}= 0.5 with the "lo" dependence as defined above. As before, the logarithmic term has been modified in order to extend to velocities less than 1. The dotted curve showing a v^{-4} dependence is provided for comparison purposes.

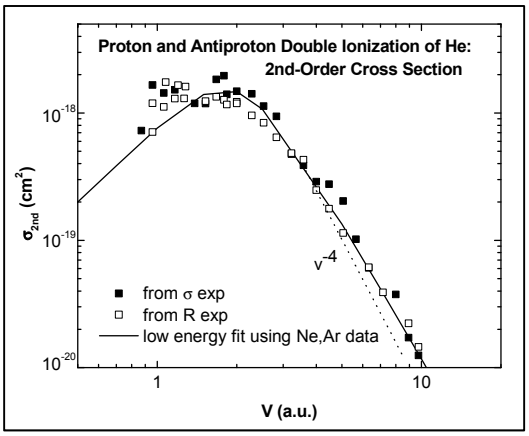

Fig. 3: The 2nd-order cross section, σ_{2nd}, obtained from adding proton and antiproton impact double ionization cross sections from refs. 8-13,26,27,36. The solid line is a fit with a high velocity dependence of $ln(v)v^{-4}$ while the dashed line illustrates a high velocity v^{-4} dependence.

Fig. 4 shows cosines of the phase difference between the 1st- and 2nd-order mechanisms that were extracted from the experimental double ionization ratios combined according to the ratio equivalents of equations (2d) and (2e). Cosines were also calculated using double ionization cross sections but are not shown as the number of energies where both proton and antiproton cross sections are available was smaller, plus where information was available, the scatter in the data was significantly larger. The fitted line to the data shown is given by $\cos(\varphi) = 0.35 + 0.4v^{-2}$.

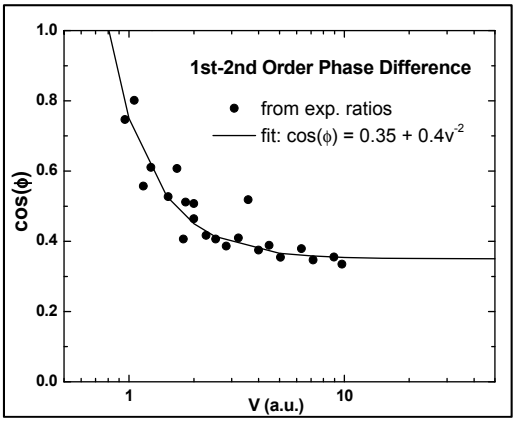

Fig. 4: Cosines of the phase difference between the 1st- and 2nd-order ionization mechanisms obtained from subtracting proton and antiproton impact double ionization cross sections. The data are the same as in Fig. 3. The solid line is a fit.

After rewriting our fitting formulae for proton and antiproton impact ionization of helium, our analysis yields the following models/fits to the data:

Single Ionization: $$\sigma_1 = [1+(C_{hi}/C_{lo})ln(k+v)/v^{2+m}]^{-1} C_{hi} ln(k+v)/v^2 \quad (3a)$$

Double Ionization: $$\sigma_{1st\text{-}order} = R_{1st}(\infty)\sigma_1 = 0.00259\sigma_1 \quad (3b)$$

$$\sigma_{2nd\text{-}order} = [1+(c_{hi}/c_{lo})ln(k+v)/v^{4+m}]^{-1} c_{hi} ln\,(k+v)/v^4 \quad (3c)$$

where $$\sigma_{Int} = 2\cos(\varphi)[\sigma_{1st\text{-}order}\,\sigma_{2nd\text{-}order}]^{1/2} \quad (3d)$$

and $$\cos(\varphi) = 0.35 + 0.4v^{-2} \text{ with the maximum value being 1} \quad (3e).$$

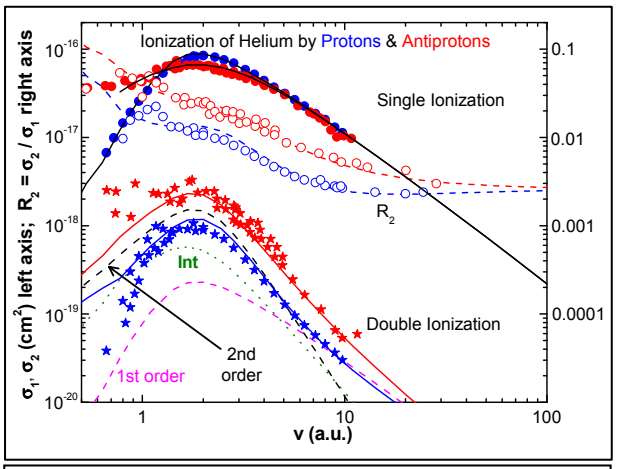

Fig. 5: (color online)Predictions for the various channels leading to single and double ionization of helium by protons and antiprotons, plus double to single ionization ratios, obtained using equations 3 and our fitting parameters compared to experimental data from refs. 8-13,26,27,36. The dotted curves show σ_{Int} and σ_2 determined using the fit to the antiproton single ionization cross sections.

For proton and antiproton impact and velocities in atomic units, in units of 10^{-16} cm^2 equation (3a) yields single ionization cross sections using C_{hi}=4.75, k=0.4, C_{lo}=0.375 (proton), = 0.65 (antiproton) and m=4 (proton) = 1.5 (antiproton), and equations (3b-3e) yields double ionization cross sections using c_{hi}=0.5, c_{lo}=0.008 and m=2. These cross sections for single and double ionization of helium by protons and antiprotons, plus cross section ratios, obtained using these formulae and constants are compared with experimental data in Fig. 5. The solid black curves for single ionization are obtained using equation (3a) and the constants listed above. For double ionization, the solid red (upper) and blue (lower) curves are total cross sections obtained by adding the 1st-order (dashed magenta curve), 2nd-order (dashed black curve), and interference (dot green curve) cross sections shown. Note that here σ_{1st}, σ_{Int} and σ_{2nd} were determined using the fit to the proton single ionization cross sections which for v >1 introduces changes than 10% from values obtained using $\sigma_{1st\ ave}$. Doing so allowed us to determine these quantities in Fig. 5 using only experimental data. Overall, good agreement with measured values is achieved except at the lowest velocities where the early proton impact measurements of Afrosimov et al. [36] are significantly smaller while the antiproton data are larger than our analysis and fits predict. This might imply problems with the low energy double ionization data. Or it could imply that higher order processes need to be included since we have extended our analysis and fits to very low velocities. Another possibility for the discrepancy at low energies is that our method of calculating $\sigma_{1st\text{-}order}$ by multiplying σ_1 by $R_{1st}(\infty)$ is too simplistic. For example, note that if the TS1 mechanism is velocity dependent and decreases with impact velocity, both the 1st-order and the interference terms would be underestimated at low velocities thus leading to an overestimation for proton impact and an underestimation for antiproton impact, as seen in Fig. 5.

The present data and analysis indicate that the 2nd-order mechanism dominates at lower velocity and that interference between the 1st- and 2nd-order double ionization mechanisms accounts for differences between proton and antiproton impact, as predicted by the McGuire model. In the high velocity region, our analysis implies that near v=10 the 1st- and 2nd-order mechanisms are approximately equal in magnitude, but opposite in sign for proton impact, thus cancelling their contributions for proton impact. This is not the case for antiproton impact where the signs of the matrix elements for the 1st- and 2nd-order mechanisms are the same; thus inclusion of the interference term increases the double ionization cross section by a factor of two near v=10 a.u. The effects of this cancellation versus enhancement are clearly seen in Fig. 2. For proton impact velocities greater than 10, σ_{Int} is larger than σ_{2nd} which, because σ_{Int} is negative, means R_2 in this region is smaller than at the asymptotic limit. Overall, reasonable qualitative agreement with measurements is achieved for the cross section ratios, even at low velocities where the proton and antiproton impact ratios increase rather than decrease, as was seen in Fig. 2 for multiply

charged ion impact. The present analysis implies that this increase is due to the different velocity dependences in the threshold region for single and double ionization although we cannot rule out contributions from higher order terms.

Fig. 6 shows a comparison of our empirical curves for the SO and TS2 mechanisms with the ab initio calculations of Gulyás et al.[1] and Nagy et al. [18] As seen, the agreement is typically better than 30% with the time-ordered model of Nagy et al. yielding somewhat better agreement. However, the present work plus the calculations of Gulyás et al. imply that the SO contributions of Nagy et al. are too small at lower velocities. In addition, for these lower velocities the interference contribution that we obtain is larger than that quoted by Nagy et al. which may explain why their calculated values for proton impact are too large plus why their model gave approximately the same cross section for double ionization by protons and antiprotons.

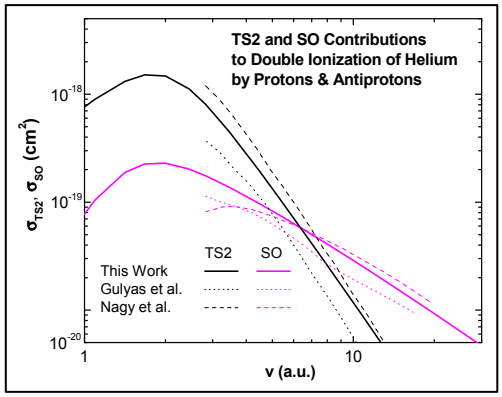

Fig. 6: (color online) Cross sections for the TS2 and SO contributions to double ionization of helium by protons and antiprotons. Thick solid lines, present work; thin solid lines with dots, Gulyás et al. [1]; dashed lines, Nagy et al. [18].

Ionization by bare multiply charged ions:

To extend our analysis to multiply charged ion impact, experimental cross sections for He^{2+}[9,26-28,38], Li^{3+}[26,29], F^{9+}[31] and U^{90+}[34] impact were used. Guided by the scaling found in Fig. 2, the cross sections were divided by $Z^{(6/5)n}$, where n=1,2 for single and double ionization respectively, and the impact velocities were divided by $Z^{2/5}$. To more directly test the scaling of the 2nd-order contribution to double ionization, the 1st-order contribution was subtracted before applying this scaling. Fig.7 shows that these scalings bring the data into near agreement with each other, although the double ionization data seems to indicate that scaling by $Z^{12/5}$ works well for low Z but is too large for higher Z ions. The few outlying points around scaled velocities of unity are the early measurements of Afrosimov et al. [38] which are in qualitative, but not quantitative, agreement with more recent, and presumably more accurate, measurements.

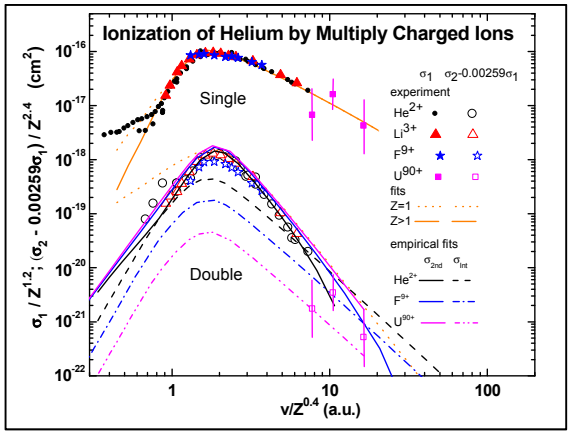

Fig. 7: (color online) Predictions for σ_1 plus σ_{2nd}, σ_{Int} and their sum for ionization of helium by He^{2+}, F^{9+} and U^{90+} obtained by scaling equations (3) and using the fitting parameters for multicharged bare ions, e.g., C_{hi}=4.75; k=0.4; C_{lo}=0.35; m=6; c_{hi}=0.5; c_{lo}=0.0035, m=4. The dotted curves are made using the fitting parameters. The experimental data are for He^{2+}, Li^{3+}, F^{9+} and U^{90+} ions from refs. 9,26-29,31,34, 38.

Applying these scalings to equations (3a) and (3c), i.e., multiplying by $Z^{(6/5)n}$ and replacing v with $v/Z^{2/5}$, yields the orange dotted curves in Fig. 7. These are seen to decrease too slowly in the "lo" region. By increasing the velocity power, m, by 2 for both single and double ionization, e.g., to 6 and 4 respectively, and using C_{lo} =0.35 and c_{lo}=0.0035 with C_{hi} and c_{hi} unchanged, a good match to the data, as shown by the solid orange curves, is obtained. Equation (3b) provided values for σ_{1st}. Then, with the assumption that the phase differences between

1st- and 2nd-order terms in the transition matrix element are the same for all projectiles, σ_{Int} was calculated from equation (3d) and is shown by the dashed and dot-dashed curves. As seen, our predicted cross sections are in relatively good agreement with experimental data although there is a systematic overestimation by our predictions as Z becomes large.

Figure 7 illustrates the decreasing importance of the interference term with increasing Z. Using our fitting formulae and parameters we predict the relative importance of $\sigma_{Int}/\sigma_{2nd\text{-}order}$ in the higher velocity region to be approximately $\frac{1}{6} v_{sc} Z^{-3/5}$, where $v_{sc} = v/Z^{2/5}$. Thus, for Z=1,2,9 between $v_{sc} = 2$ and 6 our analysis predicts the relative importance of σ_{Int} with respect to $\sigma_{2nd\text{-}order}$ to be approximately 0.3 to 1 (Z=1), 0.2 to 0.7 (Z=2), and 0.08 to 0.25 (Z=9). The strong relative importance for protons and antiprotons plus the relative insignificant importance for higher Z ions explains the scaling behavior and different velocity dependences seen in Fig. 2 plus why we originally misinterpreted the mechanisms leading to double ionization [24,25].

Closer looks at the predictions based on our analysis and scalings are shown in Figs. 8 and 9. Fig. 8 shows that our predicted values are in excellent agreement with measurements for He^{2+} impact. Using selected multiply charged ion data, Fig. 9 compares predictions from our fits and scaling with ratios where the 1st-order contributions have been removed. For small Z our predicted values are in excellent agreement and reproduce the dip seen for scaled velocities near 1. For high Z ions, our predictions are in qualitative agreement but are roughly 50% too large. We attribute the overestimation to a breakdown of the Z scaling that was used. Based upon interpolations of the ionization probabilities calculated for proton impact as a function of impact parameter [15], applying a Z^2 scaling yields maximum double ionization probabilities greater than 0.2 for U^{90+} and Ni^{28+} ions having impact velocities smaller than ~20 and 10 a.u. respectively, i.e., for scaled velocities in Fig. 8 less than 4-6 a.u. This breakdown in scaling, either due to a large Z or a small v, means that higher order terms become important, which if additive would be consistent with the increasing overestimation of our calculated ratios observed for F^{9+}, Ni^{28+} and U^{90+} impact. Based upon the good agreement found for antiproton calculations down to $v \sim 0.25$ where only a second-order expansion was used [17] and the good scaling seen in Fig. 9, we don't expect higher order terms to be important for $Z<9$ except perhaps for the lowest scaled velocities.

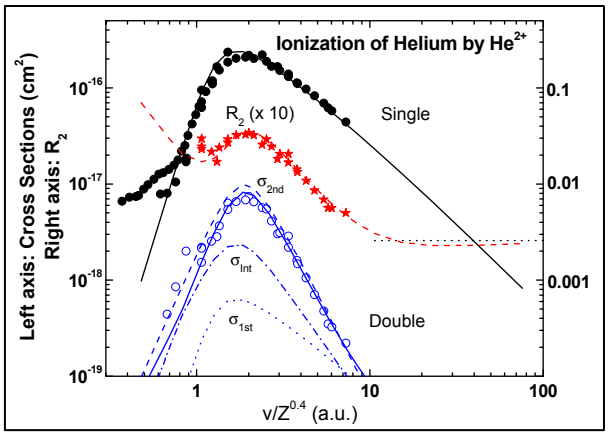

Fig. 8: (color online) Predictions compared to experimental values for single and double ionization of helium by He^{2+} ions. Data are the same as in Fig.7.

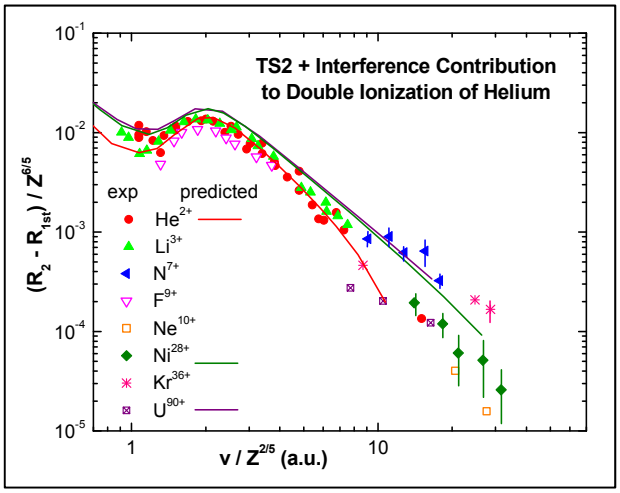

Fig. 9: (color online) Predictions compared to experimental values for the 2nd-order plus interference contributions to double ionization of helium by bare ions. Data are the same as in Fig. 2.

Extension to other targets:

As illustrated above, our analysis provides cross section formulae for the various channels contributing to double ionization of helium that are in reasonable agreement with experimental measurements for fully stripped ions and most impact velocities. To take this analysis one step further, we investigated whether the formulae also apply, or can be modified, for double outer shell ionization of other atoms. For this purpose, single and double ionization cross sections for proton, antiproton, and multiply charged ion impact on helium, neon, and argon were used [He target references are provided above; Ne target data are from refs. 12,28,36 and Ar target data are from refs. 11-13,26,27]. For single ionization, the target scaling formula presented by Montenegro et al. [43,44] combined with our projectile scaling was applied. Thus, σ_1 was divided by $Z^{6/5}\delta_1 N/I_1^2$ and the impact velocity v was divided by $Z^{2/5}I_1^{1/2}$. Here δ_1 is a number on the order of unity used to account for changes in the dipole matrix element for different targets and subshells, N is the number of outer shell electrons (2 for He and 6 was used for Ne and Ar), and I_1 is the first ionization potential in atomic units. For double ionization, based upon our assumption that a distant interaction followed by a closer interaction takes place, we applied the Montenegro et al. scaling twice where for the first ionization, the parameters were identical to those used for single ionization, while for the second ionization it was assumed that the first electron was already far away, meaning a different value for δ_1, one fewer electron, and the second ionization potential, were required. Note that this method is analogous to time ordered model of Nagy et al.[18] Therefore, σ_2 was divided by $(Z^{6/5}\delta_1 N/I_1^2)(Z^{6/5}\delta_2\{N\text{-}1\}/I_2^2)$. This means that for different targets R_{2nd} should scale as $Z^{6/5}\delta_2\{N\text{-}1\}/I_2^2$. For the velocity scaling, it is unclear from our assumptions whether scaling by I_1, I_2, I_1+I_2, or $(I_1+I_2)/2$ should be used. All were tested with I_1 yielding the best overall compression for proton and antiproton impact. For multicharged ion impact, it was found that scaling the helium target velocities by I_1 and the neon and argon velocities by I_2 was required which we assume must have something to do with the fact that Ne and Ar are multi-shell atoms.

Based upon total cross section data, Montenegro et al provided values for δ_1, namely 0.8 for ionization of He, and 0.5 for Ne and Ar [43,44]. We used the recommended value for He and adjusted the values for Ne and Ar slightly to provide a better overall compression of the σ_1 data to a single curve. Assuming both δ_1 and δ_2 are 0.8 for ionization of He, values of δ_2

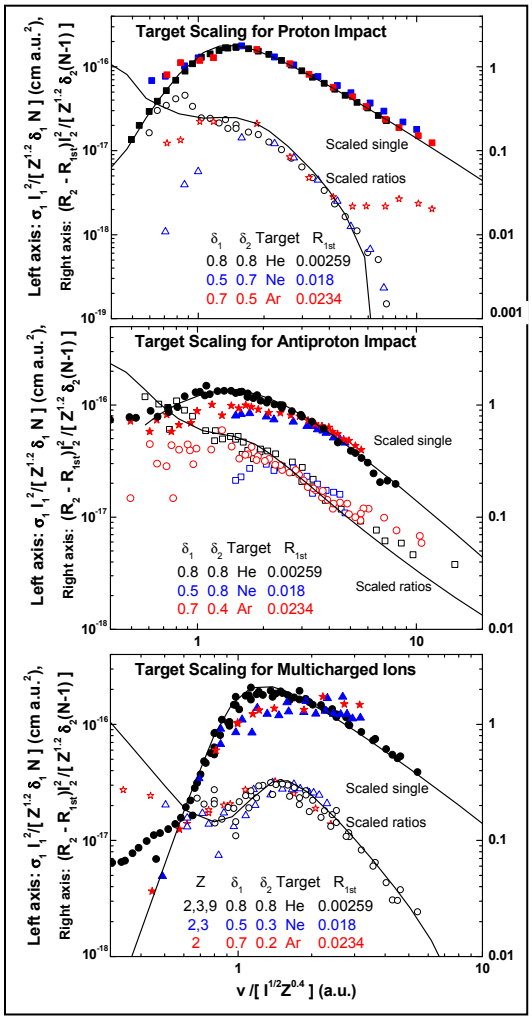

Fig. 10: (color online) Scaled single ionization cross sections and the 2nd-order plus interference contributions to R_2 for proton (top graph), antiproton (middle graph) and He^{2+}, Li^{3+} and F^{9+} (bottom graph) direct ionization of helium, neon, and argon. The curves are determined using equations 3, the parameters listed in the figures, and the target and projectile scaling described in the text.

for Ne and Ar were adjusted to give the best overall compression of the double ionization cross sections as well as f or the cross section ratios. Since our primary goal was to investigate how similar double outer shell ionization of these different targets is, "best values" for δ_2 were determined independently for ionization by protons, by antiprotons, and by multiply charged ions.

Fig. 10 shows the results and the parameters used. The upper curves in each figure are scaled experimental single ionization cross sections compared with our scaled equation (3a). For proton impact, the scaling is quite good. For antiproton impact an increasing deviation is seen at lower velocities. For multicharged ion impact, the scaled data differ by roughly a factor of two near the cross section maxima.

The lower curves are estimates of the second-order plus interference contribution to the double ionization ratios compared to the same values determined from scaled versions of equations (3). In both cases, $R_{1st}(\infty)$ has been subtracted. Since the measured double ionization ratios for neon and argon include contributions from inner shell ionization, the outer shell contribution was estimated by multiplying $R_{1st}(\infty)$ for helium by our target scaling formula for single ionization. This yielded values of 0.0123 for Ne and 0.0234 for Ar as compared to the measured values of 0.0295 and 0.0628 [45] with the difference being attributed to inner shell ionization. For neon, a better agreement with the data was found by using 0.018 rather than 0.0123. Using these values plus values for δ_2 shown in the insets, Fig. 10 shows strong similarities in the second order contributions to double ionization, except in the low scaled velocity regime for proton and antiproton impact. This could indicate problems with the experimental data or it could mean that addition double ionization mechanisms are becoming important. An obvious choice for additional mechanisms are higher order terms. However, the data do not show the strong increase in the ratios that would be expected from the inclusion of higher order terms at low velocities. New measurements and theoretical studies concentrating on impact energies below the cross section maxima are needed to resolve this. As a final comment, the deviations seen for the proton and antiproton impact double ionization ratios for argon at scaled velocities greater than 5 indicate the inner shell ionization channel becoming important. Therefore, with the exception of the low velocity regime Fig. 10 strongly implies that the formulae provided here, when scaled, also apply for double outer shell ionization of more complex atoms.

Summary and conclusions:

Empirical formulae for single and double ionization derived by fitting experimental ionization cross sections and cross section ratios for a wide range of systems and impact energies have been presented. Within the 1^{st}-order, 2^{nd}-order interference model of McGuire, analytical formulae for the various channels leading to single and double ionization as well for the phase difference between the matrix elements for 1^{st}- and 2^{nd}-order ionization mechanisms, were developed. At higher velocities the formulae for the TS2 and SO mechanisms were shown to be in good agreement with ab initio calculations available for proton impact. Multicharged ion data were used to extract projectile Z scaling dependences while proton, antiproton and multicharged ion data for He, Ne, and Ar targets were used to extract target scaling dependences.

The present analysis supports the McGuire model for double ionization and shows that protons do not fit the same scaling found for all other positive ions because the interference term is more important over a broader velocity region for protons, as compared to higher Z positive ions, owing to the different Z-dependences of the 1^{st}- and 2^{nd}-order mechanisms. Thus, the experimental data demonstrate that, in a general sense, the McGuire model is consistent with the differences in proton vs. antiproton results. As the present fits are empirical in nature we cannot

rule out contributions from higher order terms at lower velocities. But, to explain the observed velocity dependences, either the higher order contributions to double ionization must be weak with respect the second order term or they must tend to cancel each other. It is certainly true that correlation and/or polarization effects should engender some changes in the second order (and, thereby, the interference) term in the double-to-single ionization ratio, but the experimental data appears to suggest that differing polarization and/or correlation effects for proton vs. antiproton projectiles are not of crucial importance. Furthermore, it was shown that a combination of these scalings and formulae provide very good to reasonable agreement for double outer shell ionization of any atom by any fully stripped projectile for most impact velocities. Extension to partially stripped ion impact was not attempted as these systems are more complicated in that partial screening of the nuclear charge and elimination of projectile excitation accompanying target ionization must be taken into account and this is difficult to do. Based upon this work, a follow-up paper addressing lepton impact double ionization is planned.

These empirical formulae and scalings presented here can be used to guide theoretical treatments of double ionization by showing what mechanisms are important, or may be ignored, and whether interference effects play major or minor roles. They can also be used to help evaluate the reliability of various experimental data, especially in regimes where the data are difficult to measure. Finally, our analysis indicates the need for experimental and theoretical studies for scaled impact velocities less than unity.

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