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TESTING DOUBLE IONIZATION MECHANISMS WITH HIGHLY-CHARGED IONS

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ABSTRACT

Double ionization of He target atoms by intermediate- to high-velocity charged ions is considered in terms of two mechanisms: (1) a two-step (TS) process in which both target electrons are removed in separate direct interactions with the projectile, and (2) a one-step mechanism in which one electron is liberated in a single projectile-target interaction followed by subsequent "shakeoff" (SO) of the second electron. Measured double-to-single ionization ratios are used to exhibit and test the underlying mechanisms of double ionization. It is found that these ratios exhibit a high degree of universality as a function of v/q , where v is the velocity of the ion and q is its charge. The results strongly suggest that the important factor in determining the regions of validity for TS and SO is the ionic charge divided by the impact velocity and not just the impact velocity itself.

INTRODUCTION

Double ionization of helium by charged particles has been the subject of numerous experimental and theoretical investigations for nearly three decades.¹⁻¹³ Much of the interest in double ionization stems from its fundamental nature^{1,2,4} as well as its expected relationship to double ionization by photons.^{3,13-15} The single ionization of helium by fast, fully-stripped ions has been studied extensively, both theoretically¹⁶ and experimentally,¹⁷ and is well understood. For double ionization, however, additional physical mechanisms come into play. Thus, the study of the double-ionization process provides an opportunity to obtain a better understanding of two-electron transitions.

McGuire has proposed⁴ that double ionization of He by a fast highly-charged projectile can result from either a single or a double interaction of the projectile with the target electrons. In a single projectile-target interaction, generally referred to as the one-step mechanism or shakeoff (SO), the projectile interacts with only one of the target electrons, transferring it to the continuum. This electron must be ejected rapidly so that the impulse approximation is valid. Then, subsequent rearrangement from the two-electron wavefunction of the undisturbed helium system to the single-electron wavefunction of the remaining ion can lead to the ejection of the second electron. This type of double-ionization mechanism is expected to dominate for high-velocity collisions where the collision time is small, and thus there is a small probability for two separate interactions.

At lower velocities, a two-step process referred to as TS-2,¹⁸ in which the projectile interacts with each of the target electrons separately, dominates the double-ionization process. In yet another

type of process involving a single projectile interaction with a target electron, Knudsen et al.¹⁹ have suggested that the projectile may interact with one of the target electrons, and this electron in turn may subsequently interact with the second electron in a binary collision, thereby ejecting it. This latter two-step process is labelled TS-1. It is important to note, however, that it may not be possible experimentally to distinguish TS-1 from shakeoff^{18,20} and, in fact, in the limit of high ejected electron velocities these mechanisms may be identical in magnitude but opposite in sign.²⁰ This being the case, both TS-1 and SO will not be differentiated here, and will be referred to collectively as SO. Furthermore, the TS-2 mechanism will be referred to simply as TS.

For intermediate- to high-velocity collisions (i.e., $q/v < 1$, where q is the projectile charge and v is the projectile velocity in atomic units), perturbative methods⁵ can be used to treat double ionization. Since the one-step mechanisms (shakeoff and TS-1) are first-order (in q/v) processes in the projectile-target interaction, a sophisticated calculation involving the electron-electron interaction is required to treat double ionization theoretically. The two-step mechanism TS-2 (second order in q/v), on the other hand, is much easier to handle theoretically since it can be treated in the independent-particle model. Because the theoretical approaches required for the one- and two-step mechanisms are drastically different, several studies^{4,5,18,21} have focussed on establishing the velocity regimes where the different mechanisms dominate.

In this work, the ionization of helium by intermediate- to high-velocity projectiles is reviewed. Of particular interest are those results obtained for high charge states at high projectile velocities. Regimes of validity are delineated for each of the mechanisms, and ratios of double-to-single ionization are presented and compared with theoretical predictions.

THEORETICAL FORMULATION

Ionization (single and double) of neutral targets by charged ions depends on the charge q and the collision velocity v . It is, in fact, well known that the important parameter governing ionization is q/v . From various studies,^{4,5,18,21} it can be inferred that the TS mechanism should dominate for projectile charges q and velocities v (in atomic units) such that $q/v > 0.2$, while shakeoff (SO) should dominate for $q/v < 0.05$. Both TS and SO are likely to be important in the intermediate range $0.2 \geq q/v \geq 0.05$ where interference effects may be important. The regions of validity for the different double ionization mechanisms are displayed graphically in Fig. 1. The dashed line corresponding to $q/v=1$ indicates the approximate upper limit where perturbation methods may be used to treat the TS mechanism. We have chosen the lower and upper limits for the two-step and shakeoff regimes as $q/v=0.2$ and 0.05 , respectively, by noting that at these points the competing process of shakeoff or two-step, respectively, is negligible.^{4,5,21} These do not represent absolute boundaries; they serve, however, to indicate where one process ceases to dominate and the other becomes non-negligible. Of course, between these two limits, both the two-step and shakeoff terms

Fig. 1. Plot showing the q and v (in atomic units) regimes where the two-step and one-step (shakeoff) mechanisms of double ionization are expected to be dominant, as well as the intermediate region where both mechanisms are expected to be important. The values along the right-hand axis, namely, 1.0, 0.2, and 0.05, represent the approximate maximum q/v values corresponding to the perturbative TS, Intermediate (interference), and SO regimes, respectively. For $q/v > 1$ the TS mechanism is also dominant, but perturbation methods cannot be used. q and v values for the following ions are indicated: $\blacklozenge$, O^{7+} ; $\blacksquare$, N^{7+} ; $\times$, S^{13+} ; $\bullet$, U^{90+} ; and $\times$, $Nb^{23,41+}$.

are important and interference between these two amplitudes can play a significant role.

Since we are interested here mainly in the mechanisms leading to the double ionization of helium, we will focus on the ratio R of double-to-single ionization which, as shown below, can be written in a manner which explicitly exhibits the double-ionization mechanisms. Single ionization by fast, highly-charged ions striking light targets is well understood theoretically,¹⁶ and, in the range of energies considered here, the cross section σ_1 can be written as

$$\sigma_1 = (\text{const}) \times (q/v)^2 \ln v \quad (1)$$

where q is the charge of the ion and v is the velocity. (The $\ln v$ term

comes from integration over the tail of the long-range Coulomb potential of the projectile and is important only at very high velocities.) The one-step and two-step double-ionization cross sections are then given by

$$\begin{aligned}\sigma_{SO} &\propto \sigma_1 & (2a) \\ \sigma_{TS} &\propto \sigma_1^2 & (2b)\end{aligned}$$

where the notation SO refers to "shakeoff," and TS refers to "two-step." Since the TS and SO mechanisms can interfere, the total double-ionization cross section σ_2 cannot be written simply as the sum of these cross sections, but must be written as a coherent sum of amplitudes, i.e.,

$$\begin{aligned}\sigma_2 &= |a_{SO}(q/v) + a_{TS}(q/v)|^2 & (3a) \\ &= \sigma_{SO} + \sigma_{int} + \sigma_{TS} & (3b)\end{aligned}$$

where a_{SO} and a_{TS} are the corresponding amplitudes for the one-step (shakeoff) and two-step processes, respectively. Since double ionization must occur at considerably smaller impact parameters than single ionization, the $\ln v$ term doesn't occur in σ_2 .⁴ Then, using Eqs. 1, 2, and 3 the ratio $R = \sigma_2/\sigma_1$ is given by

$$R = C_{SO} + C_{int} \frac{(q/v)}{\ln v} + C_{TS} \frac{(q/v)^2}{\ln v} \quad (4)$$

In Eqs. 3b and 4, the first term is due to the SO mechanism, the last term results from the TS mechanism, and the middle term comes from the interference between the SO and TS mechanisms.

Knudsen et al.⁵ have found empirically, for charges and velocities in the perturbative two-step regime, i.e., $1 \geq q/v \geq 0.2$, that R scales as

$$R = 4.55 \times 10^{-3} q^2/E \ln(13.12vE) \quad (5a)$$

where E is measured in MeV/u. Hence, R is expected to be a linear function of $q^2/[E \ln(13.12vE)]$ in this particular two-step regime. Additionally, these authors found that in the asymptotic limit, i.e., $q/v \ll 1$,

$$R = 2.20 \times 10^{-3} \quad (5b)$$

Equations 5a and 5b were obtained using data for projectiles with atomic numbers ranging from 1-8 and energies from 0.13 to 15 MeV/u incident on helium targets. More recently, Reading and Ford⁸ have presented ab initio calculations which are consistent with Eqs. 5a and 5b.

RESULTS AND DISCUSSION

Extensive measurements of double-to-single ionization ratios in the intermediate- to high-velocity regimes are available only for H^+ (Refs. 5, 18, and 22) and He^{2+} (Refs. 5, 12, and 18). Additionally, there are published data for N^{7+} (Ref. 12), O^{7+} (Ref. 23), S^{13+} (Ref. 24), U^{90+} (Ref. 25), and several other highly-charged (not fully-stripped) ions (Ref. 26); new data for Ne^{10+} , Kr^{36+} , and $Bi^{67,83+}$ ions obtained by Ullrich et al.²⁷ were presented at this conference. As mentioned above,

the ratio of double-to-single ionization is the parameter used most frequently to exhibit and test double ionization mechanisms. From Eq. 4, R is mainly (except for the $\ln v$ dependence) a function of q/v (or v/q),⁴ and thus a high degree of universality should be exhibited in a plot of R vs. v/q . Furthermore, in the perturbative two-step regime, i.e., $v/q = 1-5$ ($q/v = 1.0-0.2$), R is expected to vary nearly as $(q/v)^2$ or $(v/q)^{-2}$ (see Eq. 4), and deviations from this dependence may be attributable to the interference term in Eq. 4.

Following Ref. 28, values of R for a diverse set of projectile ions with different velocities and charges are shown in Fig. 2, plotted as a function of v/q . Most of the available data are seen to fall along a common curve with the "crossover" from the TS to the SO regime being clearly evident. Furthermore, in the TS regime, i.e., $v/q = 1-5$, the data agree quite well with the predicted $(v/q)^{-2}$ behavior (solid line in the figure); for $v/q > 15$, R approaches a constant value in agreement with predictions of the SO mechanism.⁵

Fig. 2. Double-to-single ionization ratios R of helium by several ions as a function of v/q (in atomic units). Data are as follows: $\circ$ - H^+ (Refs. 5, 18, and 22); $\square$ - He^{2+} (Refs. 5, 12, and 18); $\blacksquare$ - N^{7+} (Ref. 12); $\blacklozenge$ - O^{7+} (Ref. 23); $\times$ - S^{13+} (Ref. 24); $\bullet$ - U^{90+} (Ref. 25). The solid line indicates a $(v/q)^{-2}$ dependence (see text). The dashed line indicates the shakeoff (SO) limit ($= 0.0022$) determined by Knudsen et al.⁵ The two-step (TS), shakeoff (SO), and intermediate (Int.) regions are also indicated.

Available data for O^{7+} (Ref. 23) and S^{13+} (Ref. 24) lie entirely in the TS regime as seen in Fig. 1. In fact, the S^{13+} data are out of the perturbative two-step regime. In Fig. 2, the O^{7+} data are seen to mainly follow the $(v/q)^{-2}$ dependence while the S^{13+} data deviate strongly from this behavior as might be expected. There is a continuity, however, from the O^{7+} data to the S^{13+} data as the value of v/q decreases.

The data obtained by Heber et al.¹² for N^{7+} in the 10-40 MeV/u energy range (solid squares in Fig. 2) appeared to yield a fairly constant (and thus presumably asymptotic) double-to-single ionization ratio. A constant value for this ratio with increasing energy is generally interpreted^{4,5,18} to indicate that the one-step mechanism is principally responsible for double ionization at these velocities. However, the ratio was considerably larger than that previously measured for fast proton impact^{5,18}. On the other hand, Heber et al.¹² also measured the double-to-single ionization ratio for 20 MeV/u He^{2+} impact, and this ratio was in good agreement with the values obtained for fast protons.^{5,18} Consequently, these authors suggested that a new double-ionization mechanism may exist for heavy-ion impact. They did not take into account, however, differences in projectile charge states which drastically affect the onset of the high-velocity SO regime as can be seen in Fig. 1. It has been previously shown that the projectile charge is fully as important as the projectile velocity in single ionization.²⁹

In a Comment by Andersen et al.³⁰ it was argued that the data of Heber et al.¹² could be explained within the framework of the one-step shakeoff mechanism and the two-step TS-1 and TS-2 mechanisms and that no new double-ionization mechanism was required. More recently, it has been shown²⁸ that there is no physical basis to interpret these data in terms of the one-step mechanism, and, furthermore, that the data of Heber et al.¹² are consistent with existing formulations. From Fig. 1 it is seen that the 10-40 MeV/u N^{7+} data fall well within the two-step perturbative regime, just bordering on the region where interference effects may start to come into play. Furthermore, although there is scatter in the measured ratios, the trend of these data tend to decrease with increasing v/q as seen in Fig. 2, thus indicating that the asymptotic region has not been reached.

In other work²⁵ for very high charge states and very high velocities, the ratios of double-to-single ionization of helium by 60-, 120-, and 420-MeV/u U^{90+} projectiles were investigated. The charge-state and velocity regime for these 60-420 MeV/u U^{90+} data is indicated by the solid circles in Fig. 1. It is seen that the U^{90+} data are fully within the two-step regime, and, furthermore, lie in the region where the perturbative treatment is expected to break down. Thus, both the N^{7+} data and the U^{90+} data are at velocities far removed from the one-step regime in which the asymptotic limit can be tested.

However, it is noted that the U^{90+} data fall farthest from the common curve and below the $(v/q)^{-2}$ dependence in Fig. 2. The deviation of the U^{90+} data from universality may be, in part, due to: (1) the fact that these data lie largely outside the perturbative regime (see Fig. 1), (2) the high charge state of these ions, or (3) relativistic effects. In order to differentiate between these two possibilities, or, perhaps, to find an alternative explanation, more theoretical and

experimental work will be required for ions with very high velocities and charge states.

Finally, in Fig. 1, two points are indicated for v (a.u.) > 100 and charges of $23+$ and $41+$, respectively. The points indicate the regimes for planned measurements³¹ to be conducted at the Lawrence Berkeley Laboratory Bevalac Facility. These measurements will be conducted for $770 \text{ MeV/u Nb}^{23+}$ and $1300 \text{ MeV/u Nb}^{41+}$. The measured ratios of double-to-single ionization for these ions may shed light on the large discrepancy observed for U^{90+} in Fig. 2.

In Table 1, a summary of existing (and planned) high energy measurements is given. This table shows that, to date, only for H^+ and He^{2+} projectiles are there double ionization data in the asymptotic regime, i.e., $q/v < 0.05$. Also shown are the expected regimes for the planned measurements with $770 \text{ MeV/u Nb}^{23+}$ and $1300 \text{ MeV/u Nb}^{41+}$ ions. It should be noted, however, that according to Fig. 1, if the theoretical formulations are correct, the asymptotic regime can never be reached for projectiles with charge > 7 . This latter result still needs to be verified experimentally, however.

Table 1. Measured values R of double-to-single ionization at high energies, where q is the incident charge of the projectile, T_{max} is the maximum kinetic energy for which data are available for the given charge state, v is the (relativistic) velocity in atomic units, and R is the ratio of double-to-single ionization. The ratio for high-energy photons ($\geq 200 \text{ keV}$) is also listed (Refs. 13 and 15).

q	$T_{\text{max}}(\text{MeV/u})$	$v(\text{a.u.})$	q/v	$R(\%)$
1	15	24	0.04	0.2
2	20	28	0.07	0.2
7	40	39	0.18	1.0
90	420	99	0.91	3.0
23	770	115	0.20	planned
41	1300	124	0.33	"
photon	$\geq 200 \text{ eV}$	--	----	1.5-3.5

CONCLUSIONS

Double ionization of helium has been investigated for very high-velocity (up to $v/c = 0.72$) and very high charge-state (up to $q=90+$) projectiles. Even for these extremely high-velocity projectiles, it is found that the asymptotic limit, where the ratio of double-to-single ionization is expected to approach a constant value, is not yet reached. These results tend to confirm that the charge of the projectile ion is fully as important as the projectile velocity in determining the regime

where double ionization due to the one-step shakeoff mechanism dominates. In fact, by comparison with other data, it is apparent that the asymptotic limit has only been reached to date for incident electrons, protons, and alpha particles.

By plotting ratios of double-to-single ionization for several ions, velocities, and charge states it was shown that this ratio exhibits a high degree of universality as a function of v/q (or q/v) in agreement with the expectations of the perturbative treatment of double ionization. Recent data of Heber et al.¹² for 10-40 MeV/u N^{7+} ions tend to lie on this universal curve, and are consistent with previous results. On the other hand, the double-to-single ionization ratios for U^{90+} deviate from the universal curve and the reason for this deviation is unclear.

In summary, the parameter of significance in analyzing double ionization mechanisms is demonstrated to be q/v as expected. Future studies of the asymptotic limit for highly-charged ions must take this fact fully into account in interpreting double ionization results for high-velocity projectiles, and additional measurements are needed to determine that range of validity of the theoretical formulations.

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