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Double ionization of helium by high-velocity U^{90+} ions

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Double ionization of helium is investigated for 60-, 120-, and 420-MeV/u ($v/c = 0.34$–0.72)U^{90+}-ion impact. The measured double-to-single ionization ratios indicate that, even for these very high velocities, double ionization of the He target results predominantly from independent interactions of the projectile with both target electrons. It is concluded that the asymptotic high-velocity regime for one-step double ionization (i.e., "shakeoff") has not yet been reached even for U^{90+} projectiles at 420 MeV/u, and, in fact, cannot be reached for projectiles with $q \gtrsim 7$, thereby verifying that the ionic charge is fully as important as the velocity in determining the importance of a given ionization mechanism.

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I. 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 theoretically [16] and experimentally [17], and is well understood. Additional physical mechanisms come into play in double ionization. Thus, the study of the double-ionization process provides an opportunity to obtain a better understanding of two-electron transitions.

McGuire has proposed [4] 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, the projectile interacts with only one of the target electrons, transferring it to the continuum. Subsequent rearrangement from the two-electron wave function of the undisturbed helium system to the single-electron wave function 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, in which the projectile interacts with each of the target electrons separately, dominates the double-ionization process. In 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 labeled 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].

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 methods [5] 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 focused on establishing the velocity regimes where the different mechanisms dominate.

Heber *et al.* [12] have recently published measurements of the ionization of helium by fast, fully stripped nitrogen-ion impact. The purpose of this work was to investigate the high-velocity double-to-single ionization ratio and test whether the asymptotic value established using fast proton [5] and electron [22] impact applies for highly charged ion impact. Their measurements [12] for N^{7+} in the (10–40)-MeV/u energy range appeared to yield a 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 effect the onset of the high-velocity regime. It has been previously shown that the projectile charge is fully as important as the projectile velocity in single ionization [23].

In a Comment by Andersen *et al.* [24], 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 shown [25] that there is no physical basis for interpreting these data in terms of the one-step mechanism, and, furthermore, that the data of Heber *et al.* [12] are consistent with existing formulations.

In the present work, we investigate the ionization of helium by a projectile at considerably higher energies (and higher velocities) and for a much higher charge state than in the work of Heber *et al.* [12], specifically 60-, 120-, and 420-MeV/u U^{90+}. Double-to-single ionization ratios and absolute cross sections for single and double ionization of the target are presented. For the present work, the measured double-to-single ionization ratios are even larger (>0.03) than those observed by Heber *et al.* [12]; however, even for these extremely high velocities, the results indicate that double ionization is mainly due to the TS-2 mechanism, in which the projectile interacts separately with both target electrons. Thus, this finding provides experimental evidence that the important factor in determining the one-step and two-step regimes is the *ionic charge divided by the impact velocity* (q/v), and not just the impact velocity itself.

II. EXPERIMENTAL TECHNIQUE

The measurements were carried out at the Lawrence Berkeley Laboratory BEVALAC facility using techniques similar to those previously described by Berg *et al.* [26]. A schematic diagram of the apparatus is shown in Fig. 1. Beams of 60-, 120-, and 420-MeV/u U^{90+} ions were passed through a gas cell filled with helium at pressures of approximately 10^{-4} Torr. Emerging projectiles were detected with a plastic scintillator, while helium recoil ions produced in the encounter were accelerated by a transverse (to the beam) electric field and detected with a microchannel plate. Provision was also made for detection of electrons from the target, but these events were not used in the analysis. By recording coincidences between the recoil ions and the projectile ions, the charge state of the recoils could be determined from the difference in the time-of-flight for singly and doubly charged helium ions. A typical recoil-ion time-of-flight spectrum is shown in Fig. 2.

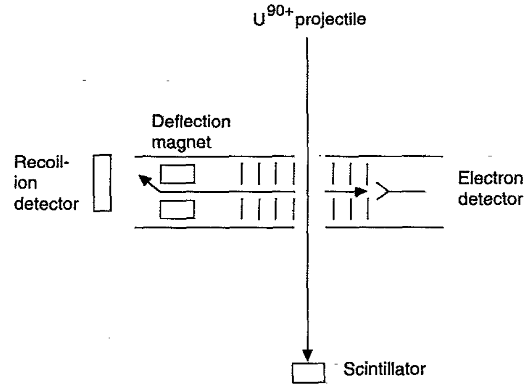

FIG. 1. Schematic diagram of the experimental apparatus (not to scale).

In order to reduce background events that give rise to accidental coincidences, two halo-veto detectors were used, one just in front of the gas cell and the other just after the gas cell. Each of these detectors had apertures slightly smaller than the gas cell apertures. Then, by recording coincidence events only for those projectiles that emerged from the gas cell without hitting either of the veto counters, the background from secondary particles created by uranium hitting various surfaces was reduced considerably.

Double-to-single ionization ratios were determined directly from the measured intensities of He^{2+} to He^{+}, as shown in Fig. 2, while absolute cross sections were deter-

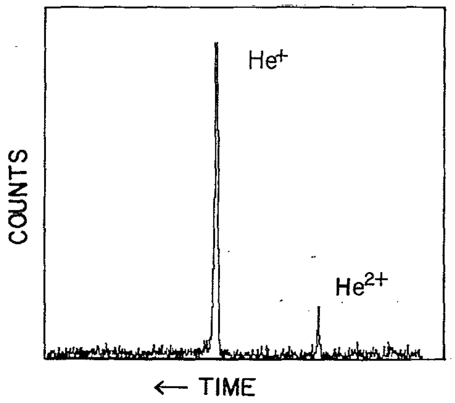

FIG. 2. Typical time-of-flight spectrum for 60-MeV/u U^{90+} colliding with He at a pressure of 0.15 mTorr. Peaks due to He^{+} and He^{2+} recoil ions are indicated. Cross sections were determined from the fractional yield of each of these peaks, while the double-to-single ionization ratio was determined directly from the relative intensities of the peaks.

mined by measuring the pressure dependence of the coincidence yields for single and double ionization, respectively. Due to experimental limitations in determining the target gas pressure precisely, there were large uncertainties in the target thickness. Consequently, the absolute ionization cross sections were determined only to factors of 2–3. However, as discussed below [see Eq. (4)], it is the ratio of the double-to-single ionization cross sections that is the essential parameter in establishing which double ionization mechanism is important, and these ratios were determined much more accurately.

The double-to-single ionization ratios are subject to systematic uncertainties such as (1) unequal detection efficiencies of He^+ and He^{2+} recoil ions, (2) charge-state impurities in the beam that could lead to charge-transferring collisions with the target gas, and (3) impurities in the target gas leading to H_2^+ recoil ions that are indistinguishable from He^{2+} ions using time-of-flight analysis. The overall uncertainty resulting from these processes is small since (1) the fields and geometry of the recoil-ion detector assured total collection of the recoil ions, and the relative detection efficiencies for He^+ and He^{2+} recoil ions impacting on the channelplate detector were measured in ancillary experiments; (2) using information from Ref. [23], combined with beam-line pressures, the charge-state impurity components of the beam were found to be negligibly small; and (3) no contamination from H_2^+ was expected since contamination of the high purity target gas by H_2 could be inferred from the presence or absence of an H^+ dissociation fragment, which was, in fact, not observed. Dissociation of any water vapor contamination would also lead to H^+ production but not to H_2^+ production. The major uncertainty in determining the ratios was then due to limited statistics because of the small double-ionization cross sections. The overall uncertainty for the double-to-single ionization ratios is estimated to be ±25%.

III. RESULTS AND DISCUSSION

The absolute cross sections obtained in the present work are shown in Fig. 3(a), and the dashed line shows the predicted single-ionization cross section from the empirical scaling of McKenzie and Olson [27]. Within the large experimental uncertainties, the magnitudes of the predicted cross sections are of the same order as the measured ones. The double-to-single ionization ratios are shown in Fig. 3(b). The ratios obtained here for U^{90+} ions are considerably larger than the value of 0.0022 found by Knudsen *et al.* [5] for fast protons. This result is similar to previous findings with heavy projectiles [12]. The solid line in Fig. 3(b) is the predicted double-to-single ionization ratio from the empirical curve of Knudsen *et al.* [5], which will be discussed in more detail below.

We note that ionization of the He target could also occur via electron capture by the incident U^{90+} ions, and such an occurrence would complicate the interpretation of the results. However, for the energies considered here, ionization of the target by electron capture [28] is expected to be orders of magnitude smaller than direct ionization [27].

We are interested here mainly in the mechanisms leading to the double ionization of helium, so we will focus on the ratio R of double-to-single ionization, which can be written in a manner that 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

$$\sigma_1 = (\mathrm{const}) \times (q/v)^2 \ln v \;, \tag{1}$$

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 given by

$$\sigma_{SO} \propto \sigma_1 \;, \tag{2a}$$

$$\sigma_{TS} \propto \sigma_1^2 \;, \tag{2b}$$

where the notation SO refers to "shakeoff," and TS refers to "two-step."

Since the TS and SO mechanisms can interfere, the to-

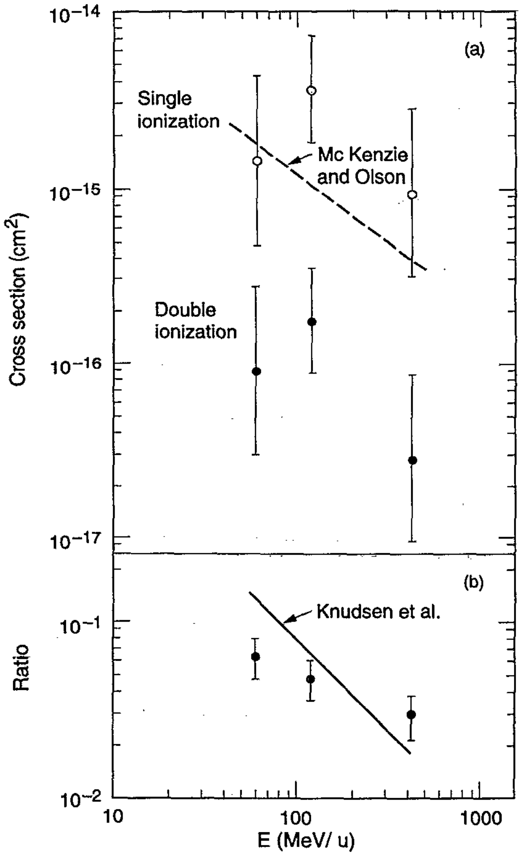

FIG. 3. Results of the present work for 60-, 120-, and 420-MeV/u U^{90+} ions incident on He. (a) Absolute cross sections for single and double ionization. The dashed curve is the calculated single-ionization cross section from the predictions of McKenzie and Olson (Ref. [27]). (b) Double-to-single ionization ratios. The solid line is the predicted ratio from the empirical scaling of Knudsen *et al.* [Eq. (5a) and Ref. [5]]. (See text.)

tal 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.,

$$\sigma_2=|a_{\rm SO}(q/v)+a_{\rm TS}(q/v)^2|^2 \quad (3a)$$

$$=\sigma_{\rm SO}+\sigma_{\rm int}+\sigma_{\rm TS}\ , \quad (3b)$$

where $a_{\rm SO}$ and $a_{\rm TS}$ 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 does not occur in σ_2 [4]. Then, using Eqs. (1)–(3), the ratio $R=\sigma_2/\sigma_1$ is given by

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

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

$$R=4.55\times10^{-3}q^2/E\ln(13.12\sqrt{E}\,)\ , \quad (5a)$$

where E is measured in MeV/u. Hence, R is expected to be a linear function of $q^2/[E\ln(13.12\sqrt{E}\,)]$ in this particular two-step regime. Additionally, these authors found that in the asymptotic limit, i.e., $q/v\ll1$,

$$R=2.20\times10^{-3}\ . \quad (5b)$$

Equations (5a) and (5b) were obtained using data for projectiles with atomic numbers ranging from 1–8 and energies from 0.13–15 MeV/u incident on helium targets. More recently, Reading and Ford [8] have presented *ab initio* calculations that are consistent with Eqs. (5a) and (5b).

The regions of validity for the different double-ionization mechanisms depend strongly on the velocity and charge of the incoming ion, and can be displayed graphically [25], as shown in Fig. 4. 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 are important and interference between these two amplitudes can play a significant role. The charge-state and velocity regime for the present (60–420)-MeV/u $\rm U^{90+}$ data is indicated by the solid circles in Fig. 4, and, for comparison, the (10–40)-MeV/u $\rm N^{7+}$ data of Heber *et al.* [12] are indicated by the solid squares. The $\rm U^{90+}$ data are fully within the two-step regime, and, furthermore, lie in the region where the perturbative treatment is expected to break down. Similarly, the $\rm N^{7+}$ data fall well within the two-step perturbative regime, just bordering on the region where interference effects may start to come into play. Thus both sets of data are at velocities far removed from the one-step regime in which the asymptotic limit can be tested. For this reason, the double-to-single ionization ratios obtained in the present work for $\rm U^{90+}$ are compared with the empirical predictions of Knudsen *et al.* [5] in Fig. 3(b), in which the solid line was calculated from Eq. (5a). Although the predicted energy dependence is somewhat steeper than the data, the overall agreement is reasonable.

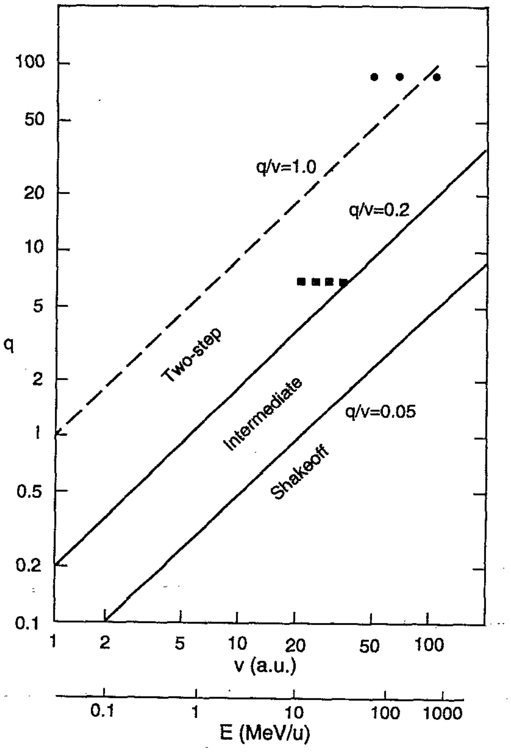

FIG. 4. Plot showing the q and v (in atomic units) regimes where the two-step and one-step (shakeoff) mechanisms of double ionization are expected to be dominant, as well as the intermediate region where both mechanisms are expected to be important. The q and v values for the present $\rm U^{90+}$ data are indicated by the solid circles, while the q and v values for the $\rm N^{7+}$ data of Ref. [12] are indicated by the solid squares.

Following Ref. [25], the q and v dependence of the double-to-single ionization ratio is displayed explicitly in Fig. 5, where R is plotted as a function of v/q. In addition to the present data (solid circles) for $\rm U^{90+}$ and the data (solid squares) of Heber *et al.* [12] for $\rm N^{7+}$, ratios of double-to-single ionization are plotted for several other incident ions of varying velocity and charge. From Eq. (4), the ratio R is mainly (except for the $\ln v$ dependence) a function only of q/v (or v/q) and, thus, a high degree of universality is exhibited in Fig. 5, as already noted by Tanis *et al.* [25]. Furthermore, in the perturbative two-step regime, i.e., $v/q=1$–5 ($q/v=1.0$–0.2), R is expected to vary very nearly as $(q/v)^2$ or $(v/q)^{-2}$ [see Eq. (4)]. This latter dependence is indicated by the solid line in Fig. 5. Deviations from this line can possibly be attributed to the interference term in Eq. (4).

It is noted that the present $\rm U^{90+}$ data fall farthest from

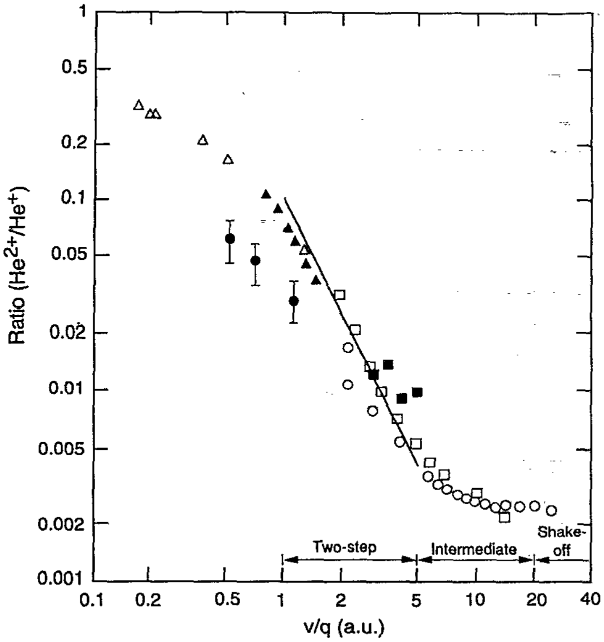

FIG. 5. Ratios R of double-to-single ionization of helium by several ions as a function of v/q (in atomic units). Data are as follows: ○, H^+ (Refs. [5], [18], and [29]); □, He^{2+} (Refs. [5], [12], and [18]); ■, N^{7+} (Ref. [12]); ▲, O^{7+} (Ref. [30]); △, various (Ref. [31]); ●, U^{90+} (present). The solid line indicates a $(v/q)^{-2}$ dependence (see text).

the common curve and below the $(v/q)^{-2}$ dependence, while the N^{7+} data are generally consistent with the trend of all the other data, although perhaps slightly above. 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. 4), or (2) the high charge state of these ions. 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.

IV. CONCLUSIONS

Double ionization of helium has been investigated for very high-velocity ($v/c = 0.34-0.72$) and very high-charge-state ($q = 90+$) 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 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 double-to-single ionization ratios for several ions, velocities, and charge states, it can be 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. The recent data of Heber *et al.* [12] for (10–40)-MeV/u N^{7+} ions tend to lie on this universal curve and thus are consistent with previous results. On the other hand, the double-to-single ionization ratios for the present U^{90+} data deviate from the universal curve; 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.

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