High-charge-state limit for the double-to-single ionization ratio of helium in the strong-coupling regime

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We measured the double-to-single-ionization ratios R of helium impacted by the intermediate velocity C^{4+} ion and found that R decreases as $1/v$. This type of velocity dependence is consistent with the data for very highly charged ions ($N^{7+} \sim U^{92+}$). The charge state and velocity dependences of R are interpreted well by the classical over-barrier-ionization (COBI) model. It is found that the ratio R can be written in a manner of $R = 0.28\sqrt{q}/v$ for very high-q projectiles in the strong coupling ($q/v > 1$) regime. This theoretical prediction is in excellent agreement with extensive experimental data.

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I. INTRODUCTION

Ionization of helium by charged-particle impact is the most simple and fundamental many-electron problem in atomic physics. It has been the subject of continuous research for many years, and still attracts considerable attention. Single ionization has been widely studied and a solid understanding has emerged, but nevertheless significant discrepancies with theory persist in measured fully differential cross sections [1]. The investigation of double ionization is aimed at understanding the two-electron transition under different perturbations and the electron-electron correlation [2]. In the past, significant efforts were invested in studies of the double-to-single-ionization ratio R, for it can exhibit the main part of the ionization mechanism [3].

The high-velocity ($q/v \ll 1$) limit of R has been explained by the "shake-off" (SO) model [4]. The SO mechanism states that the first electron is ejected through a binary interaction with the projectile, while the second is ejected due to a sudden change in the effective potential after the removal of the first electron. The limit is expected to be 0.26% and has been established experimentally for fast proton [5], antiproton [6], electron [7], and positron [8] impact.

For intermediate-to-high-velocity ($q/v < 1$) ions, the "two-step 1" (TS-1) mechanism was thought to be dominant [5]. However, more recent studies suggest that "two-step 2" (TS-2) contributions are not entirely negligible either [9]. Furthermore, it was found that a hybrid process between TS-1 and TS-2, labeled TS-1-EL [10] (see below) is the dominant mechanism. In TS-1, the projectile only ejects one target electron directly; the second electron is ejected through electron-electron collision. In TS-2, both electrons are ejected through two independent binary encounters with the projectile, and thus R scales like $(q/v)^2$. For the highly charged ion as Ni^{28+}, the limit of 0.26% cannot be reached even for the velocities close to the speed of light, but the v^{-2} dependence is observed [2]. This indicates that the contribution from TS-2 mechanism is very important for highly charged ions in the $q/v < 1$ regime.

Recently, experiments using novel techniques have focused on other quantities to study the details of double ionization. The fully differential cross sections of helium impacted by swift protons [11] are measured in comparison with the electron data to explore the projectile-charge sign dependent differences. The data clearly show that the TS-1 mechanism becomes more important when the negatively charged ion tends to push one electron toward its parent atom, while it might be much less important when the positively charged ion pulls the electron away from the parent atom. By measuring the probability to find two electrons in the same event and in two independent events, the electron-electron correlation function [12] is found to be sensitive to the momentum difference between the two ejected electrons. Using the method of a four-body Dalitz plot, a new double-ionization mechanism (TS-1-EL) [10] is found in which the fast projectile is elastically scattered by the second ejected electron which is already promoted to the continuum through the TS-1 process. This new mechanism resolved the conflict between the experimental data and the results [9–11] given by TS-1 and TS-2 mechanisms.

In the strong coupling regime ($q/v > 1$), the ratios R for the U^{90+} ion impact [3] deviate drastically from Knudsen's universal curve [13]. The reason may be that the U^{90+} data lie largely outside the perturbation regime [3]. Such deviations are also observed in systematic measurement where fast ($3.6\,\mathrm{MeV/u} \sim 1\,\mathrm{GeV/u}$) and highly charged ($q = 24 \sim 92$) ions are used [14]. As stated by previous authors, helium double ionization for very highly charged ion impact ($q/v > 1$) is still unclear in strong coupling regimes [3–15].

Since the relevant parameter in determining the ionization mechanism is q/v [3], double ionization for very highly charged ions can be understood in some degree by the collision with multicharged ions in the same q/v regime. In the present work, the double-to-single-ionization ratios R of helium by intermediate velocity ($3\tilde{}5v_{\mathrm{Bohr}}$) C^{4+} ion impact are measured and found to be proportional to $1/v$. Such velocity dependence is consistent with the data concerning board q and v regimes [16–22]. The COBI model [23,24] is used to interpret the q and v dependences of R, and the high-q limit of it is found to be $0.28\sqrt{q}/v$. This result is in excellent agreement with the experimental data. The atomic unit (a.u.) is used in the present paper.

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II. EXPERIMENT

The experiment was carried out at the 2×1.7 MV tandem accelerator at Lanzhou University. The details of the apparatus can be found in a previous paper [23]. In this paper, the measurement will be introduced in summary. After the energy and charge-state selection, C^{4+} beams are carefully collimated by slits of 0.2×0.2 mm^2 to ensure the maximum divergence is less than 0.1 mrad, and then they pass through a gas cell filled with helium gas with the pressure of 10^{-4} Torr; the vacuum in the chamber is kept at 10^{-6} Torr.

The scattered projectile is charge-separated by a parallel-plate electric field and deflected to different positions on a multichannel plate (MCP) detector. The recoil helium ions are accelerated by an electric field perpendicular to the beam direction and detected with another MCP detector. The time-of-flight technique is used to measure the charge of the recoil ion. By recording the coincidences between the time of flight of the recoil ion and the position of the scattered projectile, the final charge states of them can be determined. A typical two-dimensional spectrum is shown in Fig. 1 for a 4.8-MeV C^{4+}+ He collision. In Fig. 1 "position" is the position coordinate of the scattered projectile, and "time" is the time coordinate of the recoil ion. All peaks are well resolved from each other and reside well above the background. The major uncertainty in determining R comes from the statistic errors for the small double-ionization cross sections. The overall uncertainty for the double-to-single-ionization ratio R is estimated to be within $\pm 20\%$.

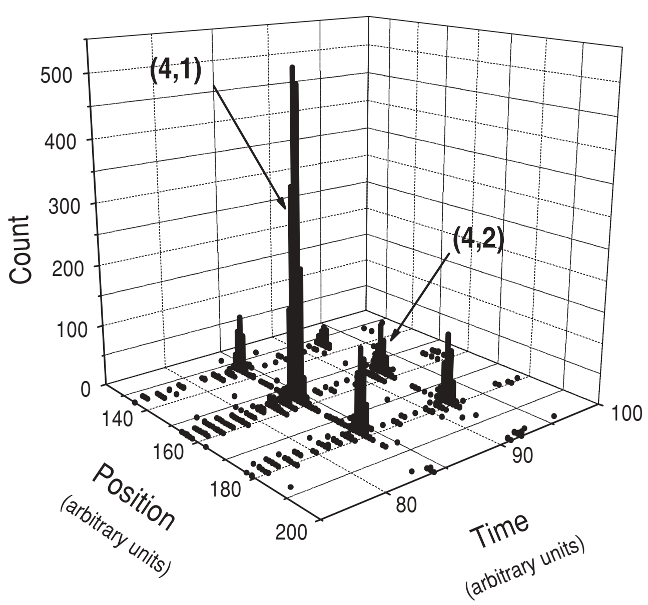

FIG. 1. Two-dimensional spectrum for 4.8-MeV C^{4+}+ He collision. Numbers in brackets represent the final charge states of the projectile and recoil ion. For example, (4,1) corresponds to the single-ionization process of C^{4+}+ He $\rightarrow$ C^{4+}+ He^{+}, and (4,2) corresponds to the double-ionization process of C^{4+}+ He $\rightarrow$ C^{4+}+ He^{2+}.

III. MODEL

Based on Bohr's classical overbarrier model (COBM) [25,26], ionization is introduced in COBI to describe the double-ionization process in the strong coupling regime. According to COBM, electron can be released from the target when the Coulomb potential barrier between the projectile and the target nucleus is lower than the electron's binding energy. The release distance R_r, below which release becomes possible, is given as $R_r = \frac{Z+2\sqrt{qZ}}{I}$, where Z and q are charges of the projectile and the target core, and I is the electron's binding energy. On the other hand, capture takes place only if the release occurs within the capture distance of R_c, in which the electron's potential energy in the ionic field of the projectile is larger than its kinetic energy in the projectile's frame. Hence, the capture distance is $R_c = \frac{2q}{v^2}$.

Now, we concentrate on the electrons which are released outside the capture distance. These electrons will not be ionized until they get enough kinetic energies to escape. When the projectile approached the distance R_I, where the Stark energy transferred to the kinetic energy of the released electron is larger than its binding energy, the ionization will occur. Thus the ionization distance R_I should satisfy the equation $\frac{q}{R_I} \geqslant I + \frac{q}{R_r}$. In brief, the electrons released within the capture distance R_c will be captured; others released outside the capture distance will not be ionized until the ion enters the ionization distance of R_I.

Because the release and capture do not take place instantly but are gradual processes, the corresponding one-electron probabilities P_r, P_c, and P_I for release, capture, and ionization have to be taken into account. The probabilities at any collision parameter are obtained as the ratio of the collision duration, in which the release capture and ionization take place, to the orbital period of the target electron. As a result, one-electron probabilities are

$$P_r = \frac{2\sqrt{R_r^2 - b^2}}{v}\frac{1}{T},$$

$$P_c = \frac{2\sqrt{R_c^2 - b^2}}{v}\frac{1}{T},$$

$$P_I = P_r - P_c = \frac{2}{vT}\sqrt{R_r^2 - b^2} - \sqrt{R_c^2 - b^2},$$

where b is the collision parameter, v is the collision velocity, and T is the orbital period of the target electron. In the independent-electron approximation and for two-electron atoms, the total probabilities are obtained as products of the above one-electron probabilities. The single (σ^+) and double (σ^{2+}) ionization cross sections of helium are

$$\sigma^+ = \int P_{I1}(1 - P_{I2} - P_{C2})2\pi b\, db + \int P_{I2}(1 - P_{I1} - P_{C1})2\pi b\, db,$$

$$\sigma^{2+} = \int P_{I1}P_{I2}2\pi b\, db.$$

Here the subscripts 1 and 2 represent the first and the second released target electrons, i.e., P_{I1} is the ionization probability

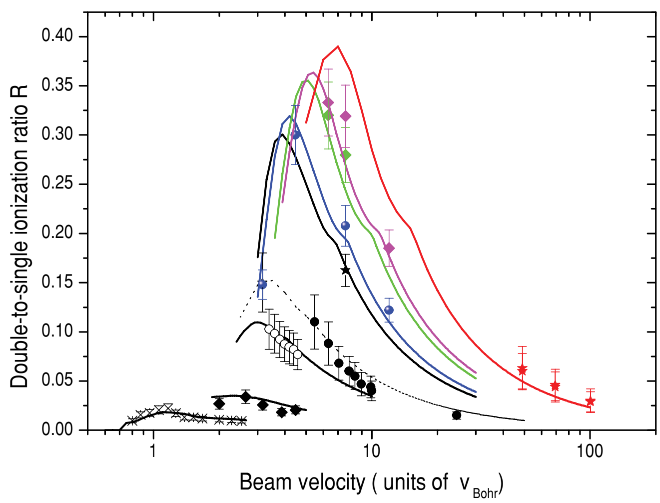

FIG. 2. (Color online) Double-to-single-ionization ratio R of helium impacted by a variety of projectiles ($q = 1\sim92$, $v = 1\sim98 v_{\mathrm{Bohr}}$). The lines are the results of the COBI model. The symbols are experimental data (cross: proton data [16,17]; solid diamond: He^{2+} [18]; open circle: present C^{4+} data; solid circle: O^{7+} [19] and N^{7+} data [20]; solid star: Fe^{15+} [21]; blue solid circle: Fe^{20+} [21] and I^{20+} [15]; green diamond: U^{36+} [14–22]; purple diamond: U^{44+} data [5–22]; red star: $U^{89+}\sim U^{92+}$ data [3–14]).

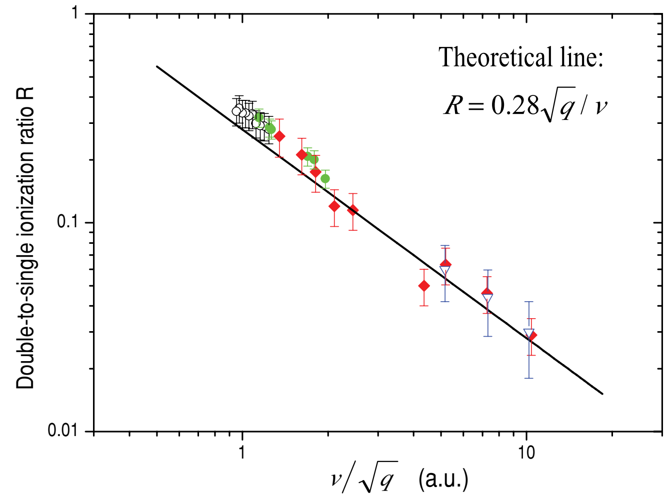

FIG. 3. (Color online) Double-to-single-ionization ratio R of helium impacted by very highly charged fast ions in the strong coupling ($q/v > 1$) regime. The solid line is the calculation result given by COBI model. The symbols are experimental data (open circle: 1 MeV/amu $U^{26+}\sim U^{44+}$ [15]; green circle: 1.4 MeV/amu ions with $q = 15+\sim 44+$ [21]; red diamond: 3.6 ∼ 420 MeV/amu ions with $q = 24+\sim 92+$ [14]; open triangle: 60, 120, and 420 MeV/amu U^{90+} ions [3]).

for the first released electron and P_{c2} is the capture probability for the second released electron.

IV. RESULTS AND DISCUSSION

The calculation results of R for a helium atom impacted by a variety of ions are plotted in Fig. 2. Present and previous data are also plotted in the same figure for comparison. The model correctly predicts the charge state and velocity dependences of R. The percentage of double ionization in total ionization is only 1 ∼ 2% for proton impact, and it rises rapidly to 10 ∼ 15% for multi-charge-state ions as C^{4+} and N^{7+}. For the very highly charged ions as Fe^{20+} and U^{44+}, double ionization becomes more important and contributes nearly half of the total cross sections.

The maxima position of R shifts to higher velocities as the projectile charge q varies to larger numbers. The maxima appear at 1 Bohr velocity for protons and at 2–3 Bohr velocities for He^{2+} ions. For the highly charged ions such as Fe^{20+} and U^{44+}, the maxima appear no more than 5 Bohr velocities and are somehow insensitive to projectile charge q. The model assumes that the electron released outside the capture distance can be ionized by the approaching ion, and therefore the ionization is determined by the total release probability and the uncaptured fraction of it. The total release probability decreases as $\frac{R_r}{v}$, while the uncaptured fraction increases as $[1 - \frac{2q}{R_r}\frac{1}{v^2}]$. The competition between the two gives rise to the appearance of the maxima ionization. When the differential of the ionization probability P_I is set to be zero, the maxima position is obtained as $\sqrt{6q/R_r}$, and it is proportional to $q^{0.25}$ for high-q projectiles. Such weak projectile charge dependence probably leads to the insensitive behavior of the maxima position for very highly charged ions.

Not only for multi-charge-state ions but also for very highly charged ions, the data and model both show that R decreases as $1/v$ when the velocity exceeds the maxima position. In other words, R is equal to β/v and β is a function of projectile charge q. When the projectile charge is sufficiently high, at least larger than 10, the model predicts β has a very simple asymptotic form of $0.28\sqrt{q}$. This gives a conventional way to scale the ratios R for various kinds of projectiles into one universal curve. This provides a scaling rule for very highly charged fast ions in the strong coupling regime. The theoretical curve of $R = 0.28\sqrt{q}/v$ is plotted in Fig. 3 and compared with the data measured by several groups; the overall agreement is very good.

In Fig. 3, all the experimental data fall around the theoretical curve of $R = 0.28\sqrt{q}/v$, and are quite different from the $(q/v)^2$ law valid in perturbation regime. The perturbation law is valid only for the low-q and high-v collisions, where the ionization is treated as binary encounters between the violent projectile and free electrons. This type of ionization is also called "forced impulse" ionization [27], in which the momentum transfer is decided by q/v.

As the projectile charge q increases, the interaction between the projectile and the target electron cannot be regarded as a perturbation. The "free electron" and "forced impulse" approximations break down, but the circulation period of the target electron should be taken into account [28]. With the approach of the projectile, the circulating electron has opportunity to move close to the saddle point of the Coulomb potential barrier, and may pass it if the projectile is within the release distance. Some of the released electrons are ionized when the projectile enters the ionization distance; others will go back to target after collision.

For very highly charged ion impact, the release and ionization distances both reach the same asymptotic form of $R_r = 2\sqrt{qZ}/I$ and $R_I = \frac{1}{I}\frac{2q\sqrt{Z}}{2\sqrt{Z}+\sqrt{q}} \xrightarrow{q\gg Z} \frac{2\sqrt{qZ}}{I} = R_r$. This indicates that nearly all the released electrons can be ionized, for they are pulled away from the parent atom by a sufficiently strong ionic field of the high-q projectile. So, double ionization for very high-q projectile impact might be simple: when the projectile enters the release distance, two helium electrons will be released one after the other; then they will be pulled away strongly and continuously by the passing high-q projectile until they are ejected far away.

The release and the following pulled-away process indicate that the ionization cross section is determined by the release cross section $\sigma_r = \pi R_r^2$ and the release probability P_r. That is, the single-ionization cross section is $\sigma^+ = \frac{1}{2}\pi R_{r1}^2 P_{r1}(0)$, and the double-ionization cross section is $\sigma^{2+} = \frac{1}{2}\pi R_{r2}^2 P_{r1}(0)P_{r2}(0)$. As a result, the double-to-single ionization ratio R is

$$R = \frac{\sigma^{2+}}{\sigma^+} = \frac{\int_0^{R_{r2}} P_{r1}P_{r2}2\pi b\,db}{\int_0^{R_{r1}} P_{r1}2\pi b\,db} = \left(\frac{\pi R_{r2}^2}{\pi R_{r1}^2}\right)\left(\frac{P_{r1}(0)P_{r2}(0)}{P_{r1}(0)}\right) = \left(\frac{R_{r2}}{R_{r1}}\right)^2 P_{r2}(0).$$

We get that $R = (\frac{R_{r2}}{R_{r1}})^2(\frac{2R_{r2}}{vT}) = 2(\frac{I_1}{I_2})^2\frac{Z_2}{Z_1}\frac{1}{T}(\frac{\sqrt{2q}}{v}) = 0.28\sqrt{q}/v$, where the charge Z and the ionization energy I for the first and the second released electron of helium are taken as $Z_1 = 1.3$, $Z_2 = 2$, $I_1 = 24$ eV, $I_2 = 54$ eV $\{Z_i = (2I_i)^{0.5}\}$ and $T = \pi/E_{1s}$. The constant 0.28 indicates that the electron is initially bounded in helium with the parameters of Z, I, and T, and it is expected to be different for other targets (e.g., it is 0.025 for lithium target).

In summary, ionization that occurs in the perturbation regime ($q/v < 1$) can be treated as forced impulse ionization and the energy transfer in binary encounter is determined by $(q/v)^2$. Thus, the cross sections and their ratios agree well with the same curve. In contrast, for very highly charged ions in the strong coupling regime ($q/v > 1$), double ionization is no longer regarded as two independent binary encounters but rather as two sequential release processes occurring at large distances. The dependence of release probability on q and v is proportional to $\sqrt{q}/v$, this leads to the similar dependence of R.

V. CONCLUSIONS

The present work finds that the high-q limit of R for helium is $0.28\sqrt{q}/v$. The release distances and probabilities for two helium electrons are demonstrated to be significant in analyzing the double-ionization mechanism for very high-q projectiles in the strong coupling regime. Though the present work provides some qualitative information, experiments using the multiple and fully differential methods are required to study the details of angular and momentum distribution for the electrons ejected through two release processes.

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