

Multi-ionization of helium by slow highly charged ions

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The double to single ionization ratios R_{21} of helium bombarded by slow ($v_{\text{Bohr}} < v < 4v_{\text{Bohr}}$) C^{2+} and C^{3+} ions are measured by the time-of-flight and coincidence technique. When the velocity is about $3v_{\text{Bohr}}$, the values of R_{21} by C^{q+} reach a maximum and are found to be 2 times the values for bare ions such as He^{2+} and Li^{3+} . A simple model is presented to estimate the single and double ionization cross sections of helium by various ions, and to present the scenario of multi-ionization in the low to intermediate energy range. The calculation results by the model are in excellent agreement with the experimental data.

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I. INTRODUCTION

For ion-atom collisions in general, single-electron processes are in many cases well understood quantitatively [1–4], while multielectron processes are less well understood and are often difficult to describe even qualitatively. Much attention had been given to the process of multiple ionization of helium [5–14] for it is the simplest multielectron collision system and allows direct comparison with theory.

Besides the direct measurement of the single ionization (SI) and double ionization (DI) cross sections, the cross-section ratios R_{21} between DI and SI are also important to reveal the multi-ionization mechanisms [5,7–9,12,15–23]. In the intermediate to high energy range ($v > 10v_{\text{Bohr}}$), McGuire had shown that the double ionization can be understood in terms of two mechanisms [15,17,18]: (1) Two-step process (TS), in which both target electrons are removed in separate direct interactions with the projectiles, and the value of R_{21} increases with the projectile charge q and decreases with the collision velocity v as $q^2/v^2 \ln v$; (2) shake-off process (SO), in which the first electron is removed directly while the second is ejected when the resulting ion “relaxes” to a continuum state, so the ratio R_{21} is expected to be independent of q and v . In the very low energy range ($v < v_{\text{Bohr}}$), capture is the dominant process and the direct ionization can be neglected. The well-known classical over-barrier model (COBM) of Bohr and Lindhard [24] gives a reasonable description of the capture mechanism in terms of transitions over the Coulomb barrier between the nuclei. The capture cross sections given by COBM are independent of the collision velocity v ; as a result, the cross-section ratios R_{21} are also expected to be constant.

In the low to intermediate energy range ($v \sim v_{\text{Bohr}}$), experiments focused on the absolute values of cross sections for SI and DI and also on the trends of R_{21} varying with v and q of the projectile [5,6,9,19–23]. No matter what the projectiles are, all experiments indicated that cross sections for SI and DI increase very rapidly with the collision velocity v and reach their maximum values when v is about 2 or $3v_{\text{Bohr}}$, then decrease slowly for higher v . Even the values of

R_{21} have the similar trend [5,6,9,19–23]. For such multielectron systems in such a strongly perturbative energy range, the results of SI given by first-principles quantum calculations [continuum distorted wave–eikonal initial state (CDW-EIS); multichannel close-coupling (MCC)] are in good agreement with experiments [1–4], but the understanding of multiple ionization is still inadequate.

In the present work, the cross-section ratios R_{21} of helium bombarded by slow ($v_{\text{Bohr}} < v < 4v_{\text{Bohr}}$) C^{2+} and C^{3+} ions are measured by the time-of-flight and coincidence technique. When the velocity is about $3v_{\text{Bohr}}$, the values of R_{21} by C^{q+} reach a maximum and are found to be roughly double those by bare ions such as He^{2+} [6] and Li^{3+} [9]. Meanwhile, a simple model is presented to estimate the values of SI and DI of helium by various ions, and to present the scenario of multi-ionization in the low to intermediate energy range. How the ratios R_{21} vary with the collision velocity v and the projectile charge state q is discussed, as well as the differences between the ratios R_{21} for bare and partially stripped ions. The experimental method is presented in Sec. II and the model in Sec. III.

II. EXPERIMENTAL METHOD

The experimental apparatus and procedure used to measure double to single ionization ratios R_{21} for C^{2+} and C^{3+} impact are identical to those used for F and Cl ions in Ref. [25], to which the reader is referred for additional information. Summarized briefly, the ion beam is energy and is charge selected and passes through a gas cell. The product target ions are extracted with an electric field and counted in coincidence with post-collision charge state analyzed projectile ions. A typical two-dimensional spectrum is shown in Fig. 1 for 2.3 MeV C^{2+} -He collision. In Fig. 1 “position” is the position coordinate of the scattered projectile which determines the charge state of the scattered projectile which determines its charge state, and “time” is the time coordinate of the recoil ion which determines the charge state of the ionized target atom. In Fig. 1 all peaks are well resolved from each other, and reside well above the background. The double to single ionization ratios R_{21} are measured for helium by 0.3–6 MeV C^{q+} ions ($q=2,3$) in the present work.

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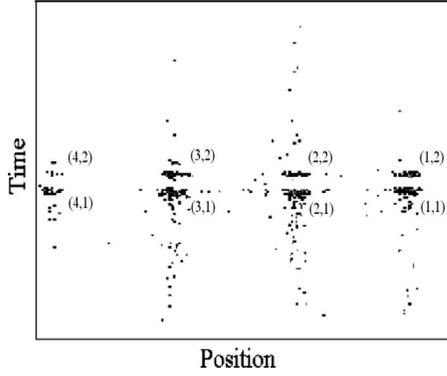


FIG. 1. The two-dimensional spectrum of 2.3 MeV C^{2+} -He. Numbers in the brackets of the spectrum are the final charge states of the projectile and of the target ion. For example, (2,1) corresponds to the SI process $C^{2+} + He \rightarrow C^{2+} + He^+$ and (2,2) corresponds to the DI process of $C^{2+} + He \rightarrow C^{2+} + He^{2+}$.

III. SIMPLE THEORETICAL ESTIMATION

The model we proposed to estimate the single and double ionization cross sections of helium is based on Bohr-Lindhard's COBM [24]; therefore, it is called a classical-over-barrier-ionization model (COBI).

Two important ion-atom interaction distances are introduced in COBM. First is the release distance R_r of the electron from the target nucleus, which satisfies

$$R_r = \frac{Z + 2\sqrt{qZ}}{I}, \quad (1)$$

where I is the ionization energy of the electron, q and Z are the charge of the projectile and of the target seen by the electron. Equation (1) indicates that the electron can be released when the Coulomb-potential barrier between the projectile and the target is lower than the energy of the target electron.

Second, the capture distance R_c is introduced. R_c is the ion-atom distance in which the electron can be captured by the projectile. R_c satisfies the following equation:

$$\frac{q}{R_c} = \frac{1}{2}v^2, \quad (2)$$

where v is the velocity of the ion. Equations (1) and (2) indicate that the electron can be captured only if the release occurs within the capture distance R_c , otherwise the release is temporary.

In the low energy range, $R_c < R_r$, the released electrons are all captured; the capture cross section is given by

$$\sigma_c = \pi R_r^2. \quad (3)$$

For higher energy, $R_c < R_r$. Only the electrons released within the distance of R_c can be captured and as a result, the capture cross section

$$\sigma_c = \pi R_c^2 f. \quad (4)$$

f is the probability of the release process occurring within the distance of R_c , where $f = \left(\frac{R_c v_e}{v a_e}\right)$ means the ratio of the duration of the collision to the orbital period of the bound electron. a_e and v_e are the orbital radius and the orbital velocity of this electron.

In Bohr's theory of COBM [24], the electrons which are released but not captured will go back to the target atom after collision. The energy level of the released electron had been estimated by Bohr [24], Barany [26], and Niehaus [27]; it is $I_n + \frac{q}{R_r}$, the ionization energy of the target electron increased by the Stark energy by the ion.

We consider that the released but not captured electrons will be accelerated by the approaching ion. At some distance R_f , the electrons will get enough kinetic energy from the ion to escape from the target atom; meanwhile, these electrons will not be captured; they will be ionized. The ionization distance R_f satisfies

$$\frac{q}{R_f} \geq I + \frac{q}{R_r}. \quad (5)$$

It means when the Stark energy transferred to the kinetic energy of the electron is larger than the ionization energy of the quasimolecular states, the electron will escape from the target atom. If it is not captured, it will be ionized. That is, in our model, the released electrons which have sufficient energy to escape from the target atom but cannot form a stable bound state of the ion are ionized. This means that the electrons which are released within the distance R_c will be captured; those released between R_r and R_c will not be ionized until the ion approaches the distance R_f .

Before we calculate the cross sections for a two-electron system, it is necessary to calculate the ionization and capture probabilities for a single electron. Then, those cross sections for two electrons may be calculated easily using the independent-electron model (IEM).

Several approximations are made:

(1) Projectiles move along linear trajectories with the collision parameter b and the velocity v .

(2) The orbital period of the electron is $T = 2\pi \frac{a_e}{v_e}$, neglecting the influence of the ions which is important in very slow collisions as v is much less than v_e .

For the given collision parameter b and velocity v , the probability that an electron will be released is $P_r(b) = \frac{2\sqrt{R_r^2 - b^2}}{v} \frac{1}{T}$, the ratio of the duration of collision to the period of the bound electron, for $b \leq R_r$.

The capture probability is $P_c(b) = \frac{2\sqrt{R_c^2 - b^2}}{v} \frac{1}{T}$ for $b \leq R_c$. The ionization probability is $P_f(b) = P_r(b) - P_c(b)$ for $b \leq R_f$.

When we construct the cross sections of the two-electron system with these single-electron probabilities, it is important to distinguish these two electrons with subscripts 1 and 2, e.g., R_{r1} , R_{c1} , R_{f1} , the distances for the first electron; R_{r2} , R_{c2} , R_{f2} , the distances for the second electron. They satisfy the following equations:

$$R_{rn} = \frac{Z_n + 2\sqrt{q_n Z_n}}{I_n},$$

$$\frac{q_n}{R_{cn}} = \frac{1}{2}v^2,$$

$$\frac{q_n}{R_{ln}} \geq I_n + \frac{q_n}{R_{rn}} \quad (n = 1, 2), \quad (6)$$

where q_n and Z_n are the charges of the projectile and of the target seen by the n th target electron. During the collision, the target electrons will cross over the potential barrier one after the other. To the second released electron, the first released electron will lose its ability to screen the target nucleus while it will screen the projectile nucleus. The method to determine the values of q_n and Z_n of the n th released electron is $q_{n+1} = q_n - 1$ and $Z_{n+1} = Z_n + 1$, the same as Barany [26] does in his work to deal with the multicapture processes. It should be noted that the value of q_2 for H^+ ions is estimated from the $1s$ binding energy of the negative hydrogen ion to be 0.3.

The values of $P_{c1}(b)$, $P_{l1}(b)$, and $P_{c2}(b)$, $P_{l2}(b)$ are calculated from R_{r1} , R_{c1} , R_{l1} and R_{r2} , R_{c2} , R_{l2} . After integrating $P_{c1}(b)$, $P_{l1}(b)$, and $P_{c2}(b)$, $P_{l2}(b)$ for all collision parameter b , we get the cross sections of the two-electron system.

The single ionization cross section (SI) is

$$\sigma_{SI} = 2\pi \int P_{l1}(b)[1 - P_{l2}(b) - P_{c2}(b)]bdb$$

$$+ 2\pi \int P_{l2}(b)[1 - P_{l1}(b) - P_{c1}(b)]bdb.$$

The double ionization cross section (DI) is

$$\sigma_{DI} = 2\pi \int P_{l1}(b)P_{l2}(b)bdb.$$

The ratio of double to single ionization is $R_{21} = \frac{\sigma_{DI}}{\sigma_{SI}}$. The calculation results are plotted in Figs. 2–14 together with the results of our experiment and other experiments in the same velocity range.

IV. DISCUSSION

As seen in Figs. 4–11, both single (SI) and double ionization (DI) cross sections of helium are described well by the COBI model. The experimental data show that SI and DI have a sharp increase when the collision velocity is less than 2 and reach the maximum values as v is about 3, then will decrease slowly. To understand this conveniently, the ionization probability when the impact parameter $b=0$ will be discussed, because the velocity dependences of SI and DI are similar to that of the probability $P_l(0)$,

$$P_l(0) = \frac{2}{Tv}(R_r - R_c) = \left(\frac{2R_r}{v}\right)\left(\frac{1}{T}\right)\left(\frac{R_r - R_c}{R_r}\right).$$

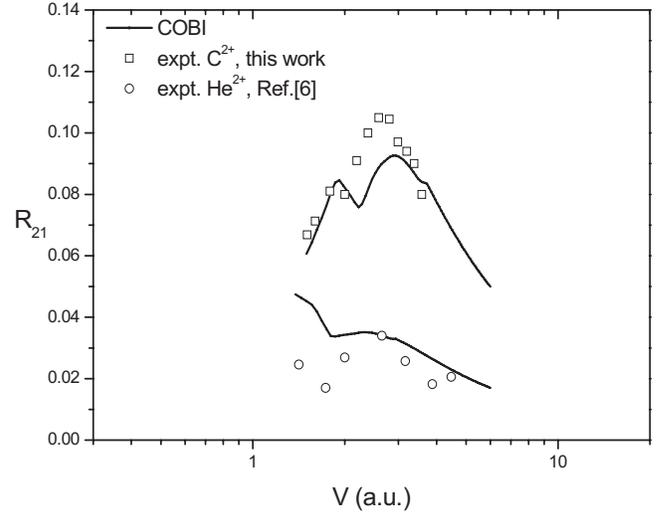


FIG. 2. Double to single ionization ratios R_{21} of helium impact by He^{2+} and C^{2+} .

Let $h = \frac{2R_r}{v} \frac{1}{T}$; $g = \frac{R_r - R_c}{R_r}$. h is the ratio of the collision duration $\frac{2R_r}{v}$ to the period of the bound electrons T . Obviously, h is the probability of the release process occurring within the collision duration $\frac{2R_r}{v}$. It is equal to 1 in the low velocity range and decreases with the collision velocity v as $1/v$ for higher velocities. g is the uncaptured fraction of the released electrons, $g = 1 - \frac{2q}{R_r v^2}$, which increases strongly with the collision velocity v and is equal to 1 for higher v .

We conclude that (a) the uncaptured fraction of the released electrons g increases as v^2 , which is the main reason for the sharp increase of SI and DI in the low velocity range. (b) The interaction time h decreases, causing the cross sections for SI and DI also to decrease for higher v . (c) In the velocity range from 2 to $4v_{Bohr}$, the uncaptured fraction of the released electrons g increases while the interaction time h decreases; such competitions between h and g will lead to the appearance of the maximum value of SI and DI when the velocity v is about 2 or $3v_{Bohr}$.

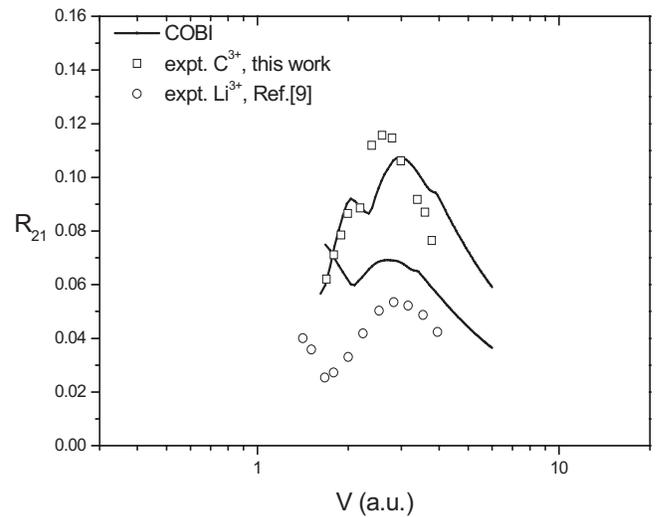


FIG. 3. Double to single ionization ratios R_{21} of helium impact by Li^{3+} and C^{3+} .

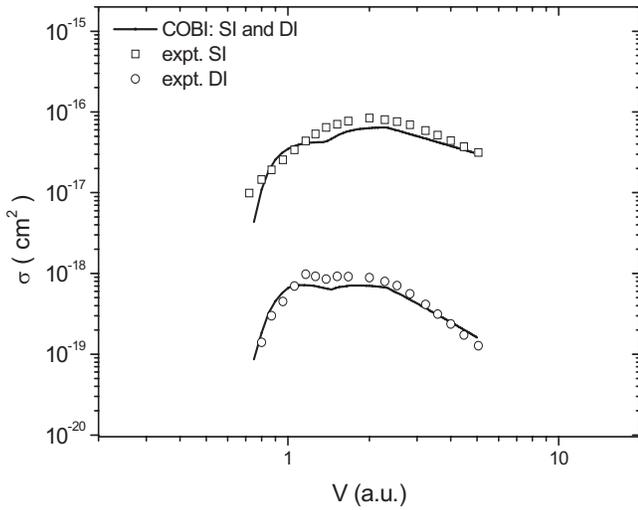


FIG. 4. SI and DI cross sections of helium by H^+ impact. Solid lines: COBI; experiment [5,28].

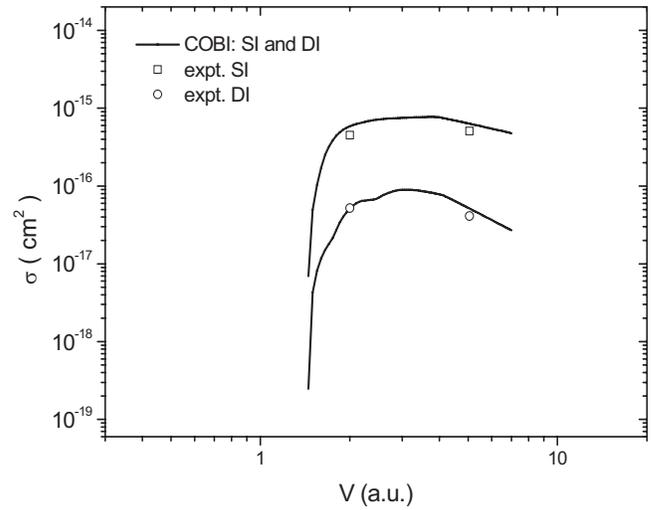


FIG. 7. SI and DI of helium by I^{5+} and B^{5+} impact. Solid lines: COBI; experiment [7,14].

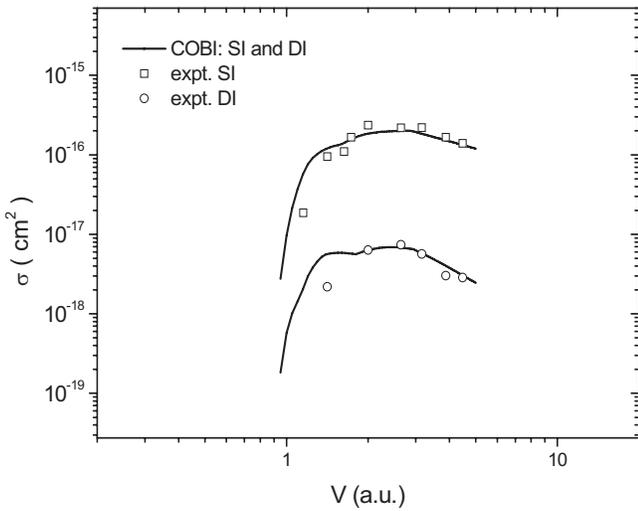


FIG. 5. SI and DI cross sections of helium by He^{2+} impact. Solid lines: COBI; experiment [6].

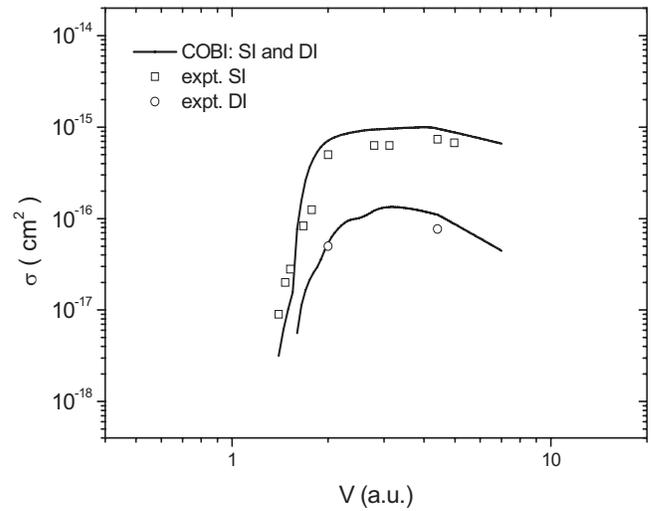


FIG. 8. SI and DI of helium by C^{6+} and Au^{6+} impact. Solid lines: COBI; experiment [4,13].

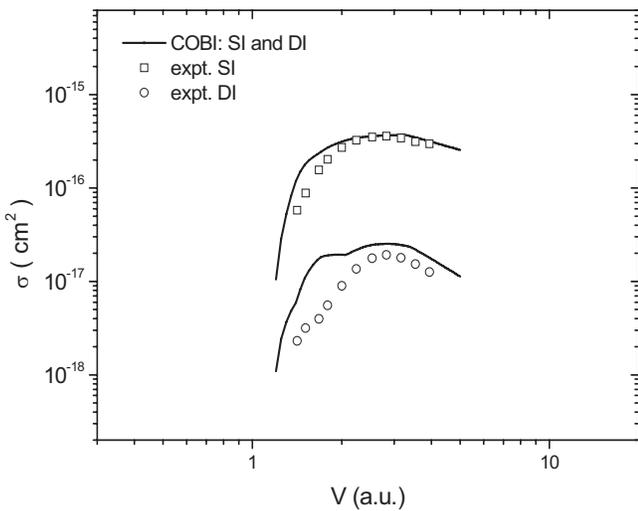


FIG. 6. SI and DI cross sections of helium by Li^{3+} impact. Solid lines: COBI; experiment [9].

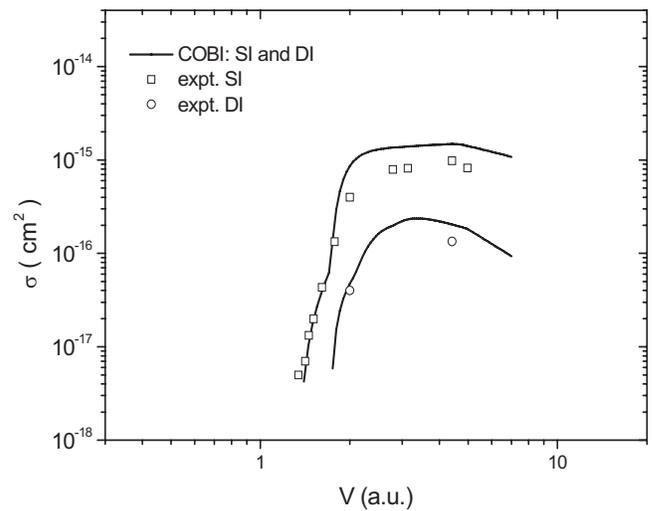


FIG. 9. SI and DI of helium by O^{8+} and Au^{8+} impact. Solid lines: COBI; experiment [4,13].

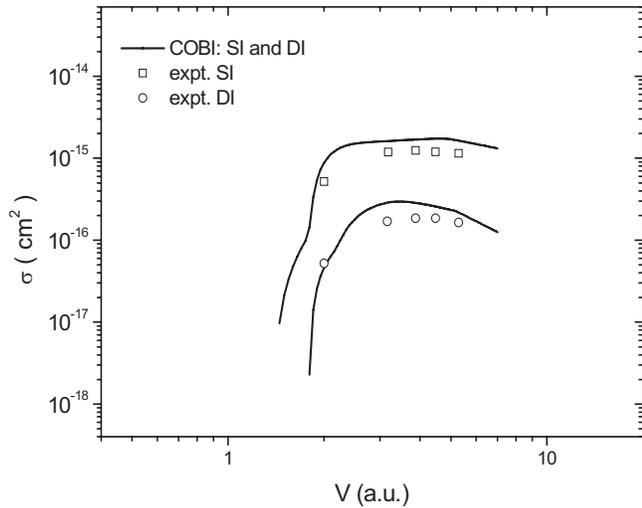


FIG. 10. SI and DI of helium by F^{9+} and I^{9+} impact. Solid lines: COBI; experiment [12,14].

The maximum values of SI and DI are plotted in Fig. 13 as a function of the charge states of the projectile. From Fig. 13, the maximum value of DI increases with q faster than SI does; this will cause the cross-section ratios R_{21} to also increase with projectile charge state q , as seen in Fig. 14. The cross-section ratios R_{21} by various charge-state projectiles are plotted in Fig. 12 as a function of the collision velocity v . In Fig. 12, the trends of R_{21} varied with q and v are described well in the COBI model: (1) R_{21} increases with charge state q ; (2) R_{21} , SI, and DI have their maxima near $2-3v_{\text{Bohr}}$; (3) in addition to maxima, R_{21} for H^+ , He^{2+} , and Li^{3+} ions have a shoulder at about $3/2v_{\text{Bohr}}$ because SI and DI do not reach their maxima at the same v for these projectiles.

In the present work, the cross-section ratios R_{21} by slow C^{2+} and C^{3+} ions are measured. It is found that the ratios by these partially stripped carbon ions are roughly double those by He^{2+} and Li^{3+} , which are plotted in Figs. 2 and 3. DuBois [29] had already investigated the SI by those low charge state

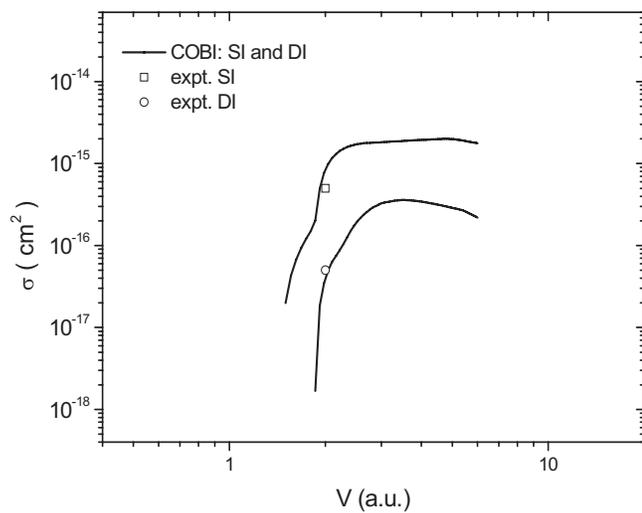


FIG. 11. SI and DI of helium by I^{10+} impact. Solid lines: COBI; experiment [14].

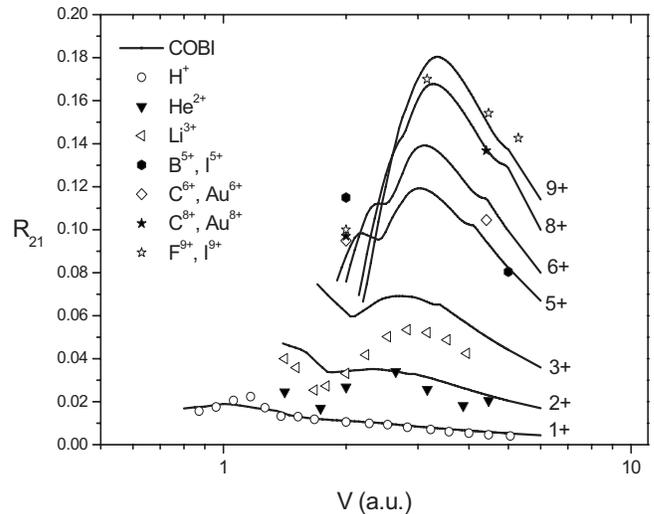


FIG. 12. Double to single ionization ratios R_{21} of helium by various charged projectiles. Solid lines are the results of COBI, the charge states q range from 1 to 9. The points are the experimental data: H^+ [5,28], He^{2+} [6], Li^{3+} [9], B^{5+} [7], I^{5+} and I^{9+} [14], Au^{6+} and Au^{8+} [13], C^{6+} and O^{8+} [4], F^{9+} [12].

partially stripped ions; it is found that the values of SI by C^{1+} , C^{2+} ions are larger than that by H^+ and He^{2+} . The size of these low charge state partially stripped ions is of the order of several a.u.² which is comparable with the ionization cross sections ($\sim 10^{-16} \text{ cm}^2$) by these ions, so the effective charge should be taken into consideration. Using the formula $q_{\text{eff}} \approx n\sqrt{2I}$, the effective charges of C^{2+} and C^{3+} are 3.7 and 4.5, respectively; the number 3.7 for C^{2+} is in accord with the estimation by DuBois [29]. The cross-section ratios calculated by using these effective charges are plotted in Figs. 2 and 3. These effective charges are larger than the charge states q of the projectiles. Since the cross-section ratios R_{21} increase with increasing charge state q as seen in Fig. 12, they are larger than those for bare ions of the same charge state q .

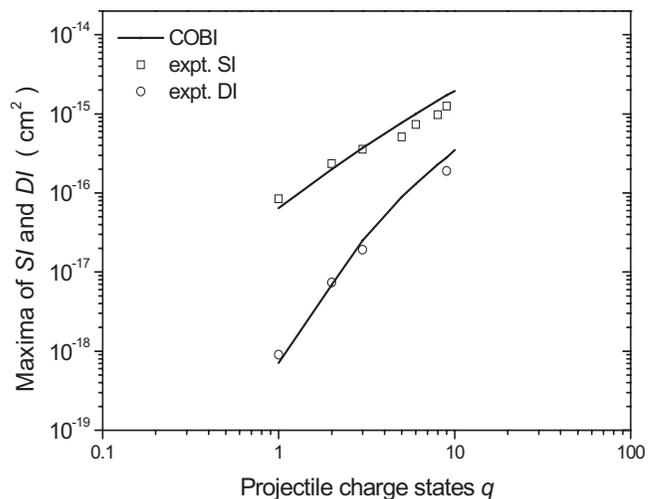


FIG. 13. Maximum values of SI and DI of helium by various projectiles.

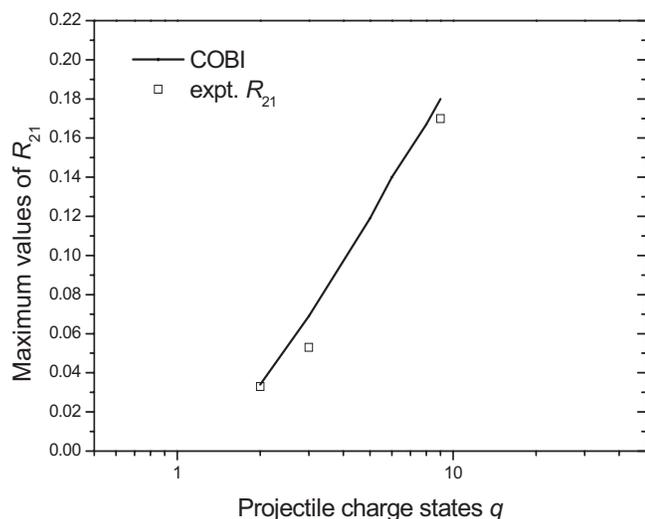


FIG. 14. Maximum values of R_{21} of helium by various projectiles.

V. CONCLUSION

In the low to intermediate velocity range, the cross sections for SI and DI of helium by various ions and their ratios

R_{21} are described well in the COBI model. The scenario of ionization in such a strongly perturbative velocity range is the following: The electron released outside the capture distance R_c will be ionized until the ion approaches the ionization distance R_I where sufficient Stark energy has been transferred to the electron to escape from the target. The experimental data show that SI and DI have a sharp increase when the collision velocity is less than $2v_{\text{Bohr}}$ and reach the maximum values as v is about $3v_{\text{Bohr}}$, then will decrease slowly for higher v ; this is the result of the competition between the uncaptured fraction of the released electrons and the interaction time. The experiments also show some differences between the ratios R_{21} for bare and partially stripped ions. After taking the effective charge of these partially stripped ions into consideration, the differences between the values of R_{21} may be interpreted within the framework of COBI.

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