Electron-capture processes of low-energy Si³⁺, Si⁴⁺, and Si⁵⁺ ions in collisions with helium atoms

H. Tawara,¹ K. Okuno,² C. W. Fehrenbach,¹ C. Verzani,¹ M. P. Stockli,¹ B. D. Depaola,¹ P. Richard,¹ and P. C. Stancil³

¹J. R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506-2601

²Department of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-03, Japan

³Department of Physics and Astronomy, The University of Georgia, Athens, Georgia 30602-2451

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Single- and double-electron-capture cross sections for Si^{q^+} (q=3, 4, and 5) ions in collisions with He atoms have been measured at collision energies of a few hundred to a few thousand eV. The observed cross sections for single-electron capture are found to be of the order of 10^{-15} cm², relatively independent of the collision energy, and are generally in agreement with recent quantal calculations for Si³⁺ and Si⁴⁺ ions and with the present Landau-Zener calculations for Si⁵⁺ ions. It is found that the measured cross sections for double-electron-capture processes, for which calculations are available only for Si⁴⁺ ions, are roughly one order of magnitude smaller than those for single-electron capture and tend to increase as the collision energy increases.

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I. INTRODUCTION

Electron-capture processes involving low-energy, highly charged ions in collisions with light neutral atoms are understood to play a key role in many fields such as laboratory (~0.1 eV/amu-10 keV/amu) and astrophysical (~1 meV/ amu-10 eV/amu) plasmas. In such electron-capture processes, an electron is generally captured into an excited state of the ion, followed by line emission. The observed ratios of line intensities provide important information on the electron temperature as well as density and spatial and temporal distributions in the emitting region of the plasma. In some cases, the electron is captured into the ground state of the ion without emitting radiation, in which case the ion can easily be ionized back to the original charge state (reionization) via inverse electron-capture processes between two charged ions. Therefore, precise knowledge of such electron-capture processes is critical in determining the abundances of ions having a particular charge in the plasma.

Among various ions in space, silicon ions, one of the most abundant ions in astrophysical plasmas, are particularly interesting and important. The lines emitted from silicon ions are often used to model various plasmas in space. It was pointed out sometime ago that inclusion of electron-capture processes on plasma modeling significantly changes the predicted abundance and ionization balance of silicon ions in coronal plasmas [1]. Immediately upon this realization, some calculations using the Landau-Zener approximation were performed [2].

There are only a few detailed experimental and theoretical investigations of the electron-capture processes involving low-energy (lower than 1 keV/amu) silicon ions in various charge states colliding with neutral (light) atoms. In particular, their collisions with atomic hydrogen are expected to play an important role in some applications such as nuclear-fusion plasma research [3]. A summary and compilation of the electron-capture data for Si ions up to 1995 has been given [4].

Recently, some accurate quantum-mechanical, closecoupling calculations have been reported for low-energy (~ 1 meV/amu-10 keV/amu) Si²⁺, Si³⁺, and Si⁴⁺ ions colliding with helium atoms, mostly from the viewpoint of astrophysical interest and applications to material development [5–11]. On the other hand, so far there is almost no experimental confirmation of these calculations, except for measurements of rate coefficients of Si^{3+} and Si^{4+} at ~4000 K [12,13]. While the Si^{3+} measurement agrees with theory, that for Si^{4+} is two orders of magnitude smaller than the theoretical prediction [13].

In the present work we report the measured cross sections of single-and double-electron capture for Si³⁺, Si⁴⁺, and Si⁵⁺ ions colliding with helium atoms over a range of ion collision energies from a few hundred to a few thousand eV; we also compare these measured values with recent quantum-mechanical calculations and the present multichannel Landau-Zener calculations. To date, no work, either experimental or theoretical, has been reported on the doubleelectron-capture processes of these silicon ions at low energies, to our knowledge, except for Si⁴⁺ ions [11]. Here we report on two electron processes in low-energy Si^{q+} + He (q = 3, 4, and 5) collisions.

II. EXPERIMENT

We have used an octopole ion beam guide (OPIG) system in order to avoid significant divergence of the decelerated and product ions. The OPIG used here is only slightly modified from the original one [14] which has already been described in detail. In Fig. 1 the present experimental setup is schematically shown for investigations of the collision processes with the OPIG system. In the present studies, the OPIG, which is floated on a deceleration potential, is followed by an ion retardation-detector system consisting of four meshed electrodes, an ion detection system, and a beamprofile-monitoring viewer system. Inside the OPIG, the octopole electric field of length over 15.0 cm (along the primary-ion direction), which is driven by a 15-MHz radiofrequency (rf) signal with a maximum amplitude of about 300 V, confines the ions throughout the entire collision region. The ion retardation-detection system is used to separate the ions of the primary-ion beam from those that have captured electrons. All ions that pass through the retardation



FIG. 1. Schematic of the present system for measuring electron-capture cross sections, which is combined with the OPIG, focusing, retardation, and fluorescent viewer system.

field are counted on a detector consisting of a pair of microchannel plates (MCP's) in a chevron arrangement followed by a phosphor screen. Ions hitting the MCP's are counted using standard particle-counting techniques. In addition, the fluorescence from the screen is monitored with a chargecoupled device camera, allowing us to verify that all of the ions, both the primary and the product ions, are collected by the MCP's. In order to facilitate control of the ion beam, simple electrostatic lenses are placed immediately before and after the OPIG. The intensity ratios of the product ions to the primary ions are measured as a function of the helium gas pressure, ensuring that single-collision conditions prevail during data taking. The absolute pressure of helium gas is monitored with a calibrated spinning-ball gauge. Other general features relevant to the OPIG have been published previously [14].

The projectile Si^{q+} (q=3, 4, and 5) ions are produced in the KSU EBIS ion source [15] by leaking a small amount of SiH₄ gas into the ion source. The ions are extracted from the trap portion of the source at a potential of 3 kV and masscharge selected with a 90°-deflection analyzing magnet. They are then decelerated down to the final energy of 1 $\times q$ keV. After passing through a switching magnet, the ions are sent through an aperture of 0.85 mm in diameter into a gas collision chamber. Here they are further decelerated down as low as $10 \times q$ eV. The first entrance electrode in the OPIG is used to monitor the intensity of the primary-ion beam and to integrate the current during the measurements. As the electron-capture cross sections at low energies are known to strongly depend on the internal electronic state of the primary ions [9], it is also important to note that the EBIS ion source produces predominantly ground-state ions because the ionizing electron energy is high and because the neutral gas pressure in the ion source is so low that the electron-capture probabilities of the ions trapped inside the ion source are very small, thus avoiding the formation of excited-state ions. Indeed, as seen later, the observed cross sections seem to support this feature of the ions from EBIS. Thus, the following discussion is limited to ground-state silicon ions colliding with ground-state neutral He.

III. RESULTS AND DISCUSSION

Before reporting the observed cross sections for the electron-capture processes, a short description of some features of the present system is in order. The whole system is relatively simple and has been found to work nicely but it is noted that, in the present system, there are some serious limitations in going to very low collision energies and higher charge states.

The first of these is the energy spread of the primary-ion energy from the EBIS. By monitoring the transmitted-ion intensities of the primary-ion beams through the OPIG as a function of its deceleration voltage, operated through the "cutoff" region, we measured what is, in effect, the integral of the primary-ion beam energy. That is, the derivative of this curve represents the energy spread of ions from the EBIS. For example, the median energy of the primary Si⁴⁺ ions accelerated at 0.815 kV was measured to be 3.106 keV, suggesting that the space-charge potential formed in the ionizing electron beam operating at 3 keV and 25 mA dc current but with pulsed ion extraction is 154 eV, and the energy spread was approximately 5 eV [full width at half maximum (FWHM)]. This energy spread was observed to be a strong function of the current and energy of the ionizing electrons in the ion source. This observed energy spread seems to be slightly larger than that in another EBIS in dc mode operation (electron beam as well as ion expulsion), which was reported to be $0.8 \times q$ eV at 2.5 keV and 10 mA [16]. This difference could be largely due to the intense electron-beam current used in the present work.

The second limitation is the inherent limitation of the energy resolution of the present retardation system. Although simple, the present four-meshed retardation system has serious limitations in separating the product ions (in particular, single-electron-captured products) from the primary ions at large OPIG deceleration voltage V_c (and, therefore, the corresponding low collision energy). When the single-electron-capture processes occur at a deceleration voltage of V_c in the OPIG for primary ions with charge q at an acceleration voltage of V_{ac} in the EBIS ion source (neglecting the negative

space potential caused by the ionizing electron beam mentioned above), the voltage difference between the primary ion and the single-electron-captured ion in the retardation system is given by

$$[qV_{\rm ac} - V_c]/(q-1) - V_{\rm ac} = [V_{\rm ac} - V_c]/(q-1).$$
(1)

Here, the first term on the left-hand side corresponds to the retardation voltage required to stop the single-electroncaptured ions and the second term to the primary ions. Thus it is clear that, with large deceleration resulting in a small collision (energy) voltage $[V_{ac}-V_c]$, this difference becomes increasingly small, making it difficult to separate these two kinds of particle. For example, when the collision voltage for the primary Si⁴⁺ ions where some reasonable fraction of the primary ions are still found to pass through the OPIG is 10 V, this difference is only 3.3 V, which makes it almost impossible to separate the two ions. It is also clear that, when the primary-ion charge increases, the difference becomes small and the ions are difficult to separate. In Figs. 2(a) and 2(b) are shown typical features of the present retardation system where the ion intensities are plotted as a function of the retardation voltage V_r . In Fig. 2(a), the strongest ion intensity corresponding to the flat part on the extreme left-hand side represents the primary ions plus the singleand double-electron-captured product ions and background, the second most intense part to the single- and doubleelectron-captured ions plus background, the third to the double-electron-captured ions plus background, and finally the fourth part at the far right-hand side to background, which is believed to be mostly due to neutral particles produced in collisions with the surfaces of the four retardation meshes. Clear changes of the ion intensities between two different ions have been observed, making it easy to measure the ion intensities in each charge state. On the other hand, in Fig. 2(b) where a large deceleration voltage V_c is applied at the OPIG, the ion intensities decreased gradually and no clear change of the ion intensities between the primary ions and the single-electron-captured ions can be seen, although there are still slight differences between the single- and double-electron-captured ions. It is believed that the main reason for this limitation is the resolution of the present retardation system, which is strongly influenced by the uniformity of the retardation electric field among the four meshes, each having more than 90% transmission. In fact the observed resolutions are measured by differentiating the retardation curve to be about 22.5 V (FWHM) for ions accelerated at 814.9 V, corresponding to 2.8%, which can still be improved through better mechanical construction of the retardation system. This limitation prevents us from studying very low collision energies.

The third limitation inherent in this experimental configuration is related to the accuracy of the cross-section measurements. This is influenced mainly through the following parameters.

(1) Although integration of the primary-ion current was performed to reduce the effect of fluctuations during the stepping of the retardation voltage, nevertheless relatively large variations were observed. It is possible that slight



FIG. 2. Ion intensities as a function of the retardation voltage V_r for 2.3295-keV Si³⁺ ions colliding with helium atoms. The deceleration voltage V_c in the OPIG is zero in (a) where three steps can be seen (see text for details). On the other hand, the deceleration voltage in (b) is $V_c = 737.7$ V, resulting in the final collision energy of 116.4 eV where the first step is no longer seen and the second and third steps are barely seen. The arrows on the left and right show the expected cutoff positions for single- and double-electron-captured product ions, respectively. Note that there are some offset voltages in V_r .

movement of the primary-ion position on the first plate of the OPIG might have changed the ion current in the collision region of the OPIG by roughly 10%.

(2) The limited resolution of the present retardation system tends to strongly influence the cross-section measurements for single-electron capture, in particular at the lowest collision energies, by roughly 20-30 %.

(3) Because the background rates, due mainly to neutral particles, are relatively large compared with the count rates of double-electron-captured product ions, the uncertainty is estimated to be 50% for the double-electron-capture cross sections.

(4) At large count rates, the pulse-height distributions of the output signals from the MCP's become strongly dependent on the count rates. They were therefore carefully monitored during all measurements to avoid changes, and count rates were kept below 5000 counts per second in order to minimize this source of error, which is then estimated to be approximately 20%.

Adding quadratically to these sources of error the uncertainty in gas target thickness (uncertainty in effective target length and pressure, a few percent), the final uncertainty in the observed cross sections is estimated to be 35-40% for single-electron capture and 65% for double-electron capture.

The measured cross sections of single-electron capture for Si^{3+} , Si^{4+} , and Si^{5+} ions in collisions with helium atoms are shown as a function of the ion collision energy in Figs. 3(a), 3(b), and 3(c), respectively, together with theoretical calculations where available.

A. Single-electron-capture processes

As seen in Fig. 3(a), for Si^{3+} ions, the cross sections for single-electron capture in the collision-energy range investigated here are of the order of 10^{-15} cm², and relatively independent of the collision energy. This is in general agreement with the calculations, although the theories predict a very broad peak ranging from 10 to 5000 eV, with the maximum cross section at around 150 eV, and a sharp decrease on both the high-and low-energy sides. In the electron-capture processes of Si³⁺ ions in collisions with helium atoms, the following channels are expected to be important [see Fig. 4(a) for the energy correlation diagram]:

$$\operatorname{Si}^{2+}(3s^{2}S) + \operatorname{He}^{+}(1s), \quad (2)$$

$$\operatorname{Si}^{3+}(3s^2S) + \operatorname{He}(1s^2) \rightarrow \begin{cases} \operatorname{Si}^{2+}(3s^3p^3P) + \operatorname{He}^+(1s), & (3) \end{cases}$$

$$\int \operatorname{Si}^{2+}(3s3p P) + \operatorname{He}^{+}(1s).$$
 (4)

Over the present collision-energy region, calculations [6,7] predict the dominance of electron capture into the ground state, process (2), which occurs mainly through an avoided crossing at an internuclear distance of about 6 a.u. Capture into an excited state, process (3), becomes noticeable only above about 100 eV. This is expected to occur at an avoided crossing at smaller internuclear distances of around 3.5 a.u. Process (4) is generally small as the incident ions have to go to much smaller internuclear distances to find proper level crossings and the process is endoergic by 0.4 eV/amu. These arguments can be easily understood from the energy correlation diagram shown in Fig. 4(a).

Cross sections for Si^{4+} ions colliding with helium atoms are shown in Fig. 3(b). The general trend in the observed results seems to be similar to that for Si^{3+} ions, although the theoretical predictions show weaker collision-energy dependence over the present energy range. In the electron-capture processes for Si^{4+} ions, the following two channels are noticeable [see Fig. 4(b)]:

$$Si^{4+}(2p^{6}{}^{2}S) + He(1s^{2}) \rightarrow \begin{cases} Si^{3+}(2p^{6}3s^{2}S) + He^{+}(1s), & (5) \\ Si^{3+}(2p^{6}3p^{2}P) + He^{+}(1s). & (6) \end{cases}$$



FIG. 3. The measured cross sections for single-electron capture for (a) Si^{3+} , (b) Si^{4+} , and (c) Si^{5+} ions colliding with helium atoms as a function of the collision energy. In (a), the calculated cross sections for single-electron capture for Si^{3+} ions are by Stancil *et al.* [7] (long-dashed line) and by Honvault *et al.* [6] (dotted line). In (b), those for Si^{4+} ions are by Stancil *et al.* [9] (solid line), by Bacchus-Montabonel and Ceyzeriat [10] (dotted line), by Opradolce *et al.* [8] (long-dashed line), and by Suzuki *et al.* [11] (dot-dashed line). In (c), the present results for Si^{5+} ions based upon multichannel Landau-Zener calculations are shown (the solid and the dotted lines represent those using the Olson-Salop-Taulbjerg and Butler-Dalgarno coupling schemes, respectively). The error bars represent those mentioned in text.



FIG. 4. Diabatic energy correlation diagrams involving singleelectron-capture processes for (a) Si^{3+} +He, (b) Si^{4+} +He, and (c) Si⁵⁺+He collisions. Note that in the first two processes an electron is captured into the ground state or a very low excited state but in the third into higher excited states, which are common in highly charged ion collisions. In (c), the ranges of the 3p and 3d manifolds are indicated.

Looking at the energy correlation diagram shown in Fig. 4(b), one can see that the dominant contribution comes from the electron capture into the excited (3p) state, process (6), which occurs at an internuclear distance of \sim 7 a.u. Capture into the ground (3s) state becomes comparable to process (6) only at collision energies above 1 keV. The latter process is expected to occur only at small internuclear distances (~ 4 a.u.). This feature is clearly seen in the correlation diagram shown in Fig. 4(b).

For Si⁵⁺ ions, the single-electron-capture cross sections are roughly constant over the present collision energy, as shown in Fig. 3(c). Other features are similar to the other charged ions, although, for technical reasons mentioned earlier, we were unable to measure the cross sections at energies as low as for the lower-charge-state ions. As no detailed theoretical analysis of the Si⁵⁺+He collision system has been reported so far to our knowledge, we have performed multichannel Landau-Zener calculations for single-electroncapture cross sections using the two different model rotational coupling schemes of Butler-Dalgarno [2] and Olson-Salop-Taulbjerg [17,18]. The energy correlation diagram is shown in Fig. 4(c) and indicates that, among the processes

$$\int \operatorname{Si}^{4+}(2p^6) + \operatorname{He}^+(1s)$$
 (7)

$$\operatorname{Si}^{5+}(2p^{5\,1}S) + \operatorname{He}(1s^{2}) \rightarrow \begin{cases} \operatorname{Si}^{4+}(2p^{5}3l) + \operatorname{He}^{+}(1s) & (8) \\ \operatorname{Si}^{4+}(2p^{5}4l) + \operatorname{He}^{+}(1s), & (9) \end{cases}$$

$$\operatorname{Si}^{4+}(2p^{5}4l) + \operatorname{He}^{+}(1s), \quad (9)$$

process (8), in particular capture into the excited 3d manifold states, is the most dominant channel for single-electron capture in the present collision-energy range. Indeed, these states cross the incident channel in the so-called "capture window" near an internuclear distance of 7 a.u. [17]. Capture into the ground state, process (7), and that into the 3sstate are expected to become important only at higher collision energies as the avoided crossing occurs only at small internuclear distances. Other channels such as capture to the 4d and 4f states in process (9) are endoergic and thus become important only at higher collision energies. The 4s and 4p states cross the incident channel at larger internuclear distances and thus are diabatic. In contrast to Si³⁺ and Si⁴⁺ ions colliding with helium atoms, this feature of capture into highly excited states of Si⁵⁺ ions is common in electroncapture processes involving highly charged ions [19].

It is noted that, in the present collision-energy region, theoretical cross sections calculated based upon both coupling schemes show general agreement with the measured results, as shown in Fig. 3(c), although a significant difference between these two coupling schemes is seen at much lower energies. Indeed there is an obvious difference in the contributions of the 3d manifold and in their collisionenergy dependence. In the present collision-energy region, the Butler-Dalgarno scheme predicts that $3d^{1}F^{0}$ is the most dominant, but $3d^{3}P^{0}$ becomes dominant in the Olson-Salop-Taulbjerg scheme. This observation may suggest that the Butler-Dalgarno scheme can work for relatively low-charged ions $(q \leq 5)$, while the Olson-Salop-Taulbjerg scheme is more appropriate for higher-charge ions.

B. Double-electron-capture processes

Double-electron-capture processes in Si^{q+} ions colliding with He atoms,

$$\operatorname{Si}^{q^+} + \operatorname{He}(1s^2) \to \operatorname{Si}^{(q-2)+**}(nl,n'l') + \operatorname{He}^{2+}, \quad (10)$$

are interesting but complicated, compared with those for single-electron capture (** indicates that both of the two electrons may be captured into doubly excited states) [20]. The observed cross sections for double-electron capture are shown in Fig. 5. They are mostly of the order of 10^{-16} cm² or less, that is, one order of magnitude smaller than those for single-electron capture. In addition, they tend to increase as the collision energy increases. So far, we know of no detailed calculations of the cross sections for processes (10), except for Si^{4+} +He collisions [11].

In double-electron-capture processes, there are two possible mechanisms [21-23]: in the first case two electrons are captured into the same or similar n states (symmetric capture, $n \approx n'$) and in the second the electrons go into very different *n* states (asymmetric capture, $n \neq n'$). After capturing two electrons, thus forming the doubly excited states, the highly charged projectile ions may be stabilized through radiative decay (in asymmetric capture) or autoionizing decay (in symmetric capture). Through coincidence experiments between a charge-changed projectile ion and a chargeselected target recoil ion [21,24], it is known that, for highly



FIG. 5. The observed cross sections for double-electron capture for Si^{3+} (circles), Si^{4+} (squares), and Si^{5+} (crosses) ions colliding with helium atoms as a function of the collision energy. The threshold energy for Si^{3+} ions is shown with the arrow.

charged ions, a significant fraction of these doubly excited states decay via autoionizing transitions.

For Si³⁺ ions, where one of the electrons may be captured into the ground state, the states formed in double-electron capture decay only via radiative transitions. The threshold for double-electron capture forming the Si⁺ $(3s^23p^2P^0)$ ion state is about 8.44 eV, which correspond to a projectile-ion energy of 237 eV. Nonzero cross sections are observed, although they are small ($\sim 10^{-17} \text{ cm}^2$) and have large uncertainties. One of the reasons for this might be the two successive single-electron-capture collisions, which are estimated to be about 50% of the total at the lowest collision energies. The situation for the Si⁴⁺ ions seems to be similar to that for the Si³⁺ ions, although the autoionizing decay rates are likely to be slightly enhanced. The calculation of double-electroncapture cross sections for Si⁴⁺+He collisions by Suzuki et al. [11] including capture only to the ground state $Si^{2+}(3s^2)$ gives values about three orders of magnitude smaller than the present measurements. The calculation may have underestimated the total double-electron-capture cross sections since it neglected capture to excited states. However, while the threshold for capture to the $Si^{2+}(3s^2)$ ground state is only 0.1 eV/amu, the threshold for capture to $\text{Si}^{2+}(3s3p\,^1P^0)$, the first excited state for which a transition is allowed, is about 3 eV/amu. It thus seems unlikely for double-electron capture into excited states to contribute significantly in the measured energy range.

On the other hand, Si^{5+} ions are expected to have relatively high probabilities of autoionization as both electrons are primarily captured into excited states [24], since capture to the entire spectrum of Si^{3+} ion energies is exoergic. However, as crossings of the double-electron-capture channels with the single-capture and initial channels are expected to occur at very short internuclear distances $R \leq 2$ a.u., one could expect an effective threshold of 1–10 eV/amu. Coincidence experiments would provide more information on the decay mechanism of the doubly excited states formed in double-electron-capture processes of relatively low-charge-state ions.

IV. CONCLUDING REMARKS

In the present work, we have reported the measured total cross sections for single- and double-electron-capture processes for Si^{q+} (q=3, 4, and 5) ions colliding with helium atoms for collision energies ranging from a few hundred to a few thousand eV. The observed single-electron-capture cross sections for all these ions have been found to show very weak dependence on the collision energy in the present energy range. They also tend to increase slightly as the incident-ion charge increases from q=3 to 5. Furthermore, the experimental data for Si^{3+} and Si^{4+} ions are found to be in general agreement with recent fully quantal calculations and those for Si⁵⁺ ions are in reasonable agreement with the present multichannel Landau-Zener calculations. The only theoretical calculation known to be available for doubleelectron capture is for the Si⁴⁺ ion and is found to disagree with the present measurements.

To further rigorously test these theories and also to use them for astrophysical and other applications, it is necessary to determine not only total cross sections at much lower energies, but also partial cross sections, namely, the (n,l)distributions, for different capture channels for these ions over a wide range of collision energies.

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