

Multi-electron transitions in F^{q+} -Ne and Na^{q+} -Ne collisions^{*}

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Abstract. The ratios of the multiple ionization cross-section to those of the single ionization of neon are measured for 2.0-8.0 MeV F^{q+} and Na^{q+} ($q = 2-5$) ions bombardment. By means of the coincidence between the charge state-selected scattered projectiles and recoil ions the contribution of the electron capture is separated from the total multiple ionization. A theoretical method is proposed to exclude the Auger transitions from the considered multiple ionization processes in the present work. The (q/v) dependence of the obtained ratios of the “pure” direct multiple ionization to those of the single ionization is discussed. The electronic structure dependence of the electron transition occurring in ion-neon collisions is studied and discussed for the symmetrical collision systems F^{q+} -Ne and Na^{q+} -Ne.

PACS. 34.50.Fa Electronic excitation and ionization of atoms (including beam-foil excitation and ionization)

1 Introduction

Single and double ionization of one- electron and two-electron targets by charged particle bombardment have been intensively studied in past two decades [1–3]. For multi-electron target atoms the multiple ionization presents a problem with much more increased complexity owing to the contribution of the Auger transitions of the subsequent rearrangement of the initial ionized states [4]. Most of the current investigations to the multi-electron target atoms concentrate on the measurements of the total [3, 5,6] and differential [7] cross-sections of the production of the recoil ions (including the contributions of the direct multiple ionization and Auger transitions), electron capture and electron loss processes [8], δ -electron emission [9] and the recoil ion momentum spectroscopy [10]. However, in order to evaluate which multiple ionization process is important it is necessary to separate the multiple ionization contributions resulting from outer-shell ionization and inner-shell ionization followed by an Auger relaxation [4]. Experiments where only the initial and final charge states of the collision partners are measured cannot distinguish direct multiple outer-shell ionization from the inner-shell ionization followed by an Auger relaxation, and spectroscopic information is required [11].

It is known that the most dominant process of the ionization of the target atom by light ion is the direct ejection of a single electron, *i.e.* single ionization. Here, it is generally accepted that higher states of ionization (multi-

ple ionization) occur via inner-shell ionization followed by Auger decay, and this process is sometimes accompanied by additional outer-shell ionization [12]. In this opinion the direct ejection of more than one outer-shell electron in light ion and multi-electron atom collisions is regarded as a second-order process which can be neglected. Manson *et al.* [12] studied the double ionization of neon by proton impact and Cai *et al.* [13] studied the multiple ionization of argon by carbon and oxygen ions, respectively. They found that the direct multiple ionization of the outer-shell electrons of the target atom is the dominant mechanism leading to the double ionization of neon induced by proton impact, and the double and triple ionization of argon induced by carbon and oxygen ion impact and not the Auger channel. At present time the investigation of the multiple ionization of neon by F^{q+} and Na^{q+} ion is still unavailable in the literature.

In the present work the ratios of the multiple ionization cross-sections to those of the single ionization of neon are measured for 2.0-8.0 MeV F^{q+} and Na^{q+} ($q = 2-5$) ion impact. By means of the coincidence between the charge state-selected scattered projectile and recoil ion the contribution of the electron capture is separated from the pure multiple ionization channel. A theoretical method is proposed to exclude the Auger transitions from the considered multiple ionization processes. The (q/v) dependence of the obtained ratios of the direct multiple ionization to those of the single ionization of neon is studied and discussed. The electronic structure dependence of the electron transition occurring in ion-neon collisions is studied by measuring the relative yields of recoil ion Ne^{1+} for the

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scattered projectiles in different charge states in 5.0 MeV F^{q+} and Na^{q+} -Ne collisions.

2 Experimental

The ion beams were provided by a 2×1.7 MV tandem accelerator [14]. After energy- and charge state-selected by a magnet, the beams were carefully collimated with two two-dimensional apertures of 0.2×0.2 mm² to ensure the maximum divergence of 0.1 mrad, and then crossed a thermal beam of target gas emerging from a small hollow needle in gas cell. The pressure in gas cell should be so low that the projectile undergoes only a single collision with one target atom in the gas cell, in which the pressure has a magnitude of 10^{-4} Torr [15]. Several test experiments were carried out to determine the value of the gas pressure, which is kept to be 2×10^{-4} Torr during each measurement. A differential pumping system was used to maintain the vacuum outside the gas cell to be better than 2×10^{-6} Torr. The scattered projectile was charge-separated by a parallel-plate static electric field next to the gas chamber to deflect the scattered projectiles with different charge states to different positions x . A one-dimensional position-sensitive detector (Microchannel plate with a resistance anode) was used to measure the charge state of the scattered projectile by determining its position x . The recoil ions produced in the collision of the projectiles with the target atoms were accelerated out of the gas cell by an electric field oriented transverse to the beam axis into a flight tube, which consists of an acceleration region and a field-free drift tube. Both the acceleration field and the drift tube were equipped with grids of stainless steel meshes, with a transparency of about 90%. Upon reaching the end of the flight tube, the recoil ions were further accelerated into a channel-electron-multiplier (CEM), where they generated the start signals for a time-to-amplitude converter (TAC). The time-of-flight technique was employed to measure the charge state of the recoil ions, with the signal of the recoil ions as the start signal and the delayed scattered projectile signal as the stop signal. A CAMAC data acquisition system was used to record the events of the scattered projectile and the recoil ion with the definite charge states produced in the same collision.

A typical measured two-dimensional spectrum of ion-neon collision is shown in Figure 1. In Figure 1 the abscissa is the position coordinate of the scattered projectile which determines the charge state of the scattered projectile, and the ordinate is the time coordinate of the recoil ion which determines the charge state of the ionized target atom (recoil ion). In partially stripped ion and atom collisions both the projectile and the target atom may lose electrons. In direct ionization (DI) the projectile does not change its charge state during the collision, and the neon atom may lose one electron via the so-called direct single ionization (DSI) or lose multiple electrons via the direct multiple ionization (DDI for direct double ionization, DTI for direct triple ionization and so on). On the other hand, the projectiles with charge state q may change the charge state to $q+i$ via i -fold stripping process or to $q-i$ via i -fold

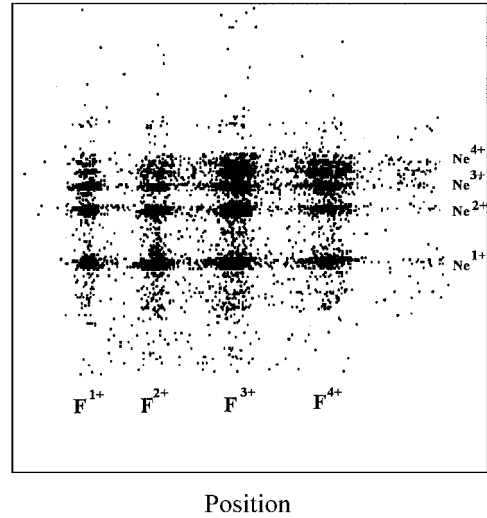


Fig. 1. The two-dimensional charge-state spectrum of 5.0 MeV F^{3+} -Ne.

electron capture. The process in which both the projectile and the target atoms lose one electron simultaneously may be referred to as SS + SI (single loss + single ionization). The SS + DI represents the process in which the projectile loses one electron and target atom loses two electrons simultaneously. The process in which the projectile captures one electron from the target atom is the so-called single capture (SC), and the process is referred to the transfer ionization (TI) when such a process is accompanied by one electron loss of the projectile. The projectile may directly capture two or more electrons from the target atom via the double capture (DC) or multiple capture (MC). Since neon has two principal shells one initial vacancy in its K -shell (produced by the direct single ionization or single capture) may decay via the KLL Auger transition and may lead to the ionization of a second electron of neon. In the same way, the triple ionization of neon may result from the Auger decay of the initial two-vacancy state. The measured Ne^{2+} , Ne^{3+} and Ne^{i+} peaks in Figure 1 include the contributions of the Auger transitions.

To eliminate most of the experimental uncertainties the ratios of the double-to-single ionization and triple-to-single ionization were measured and studied for neon by 2-8 MeV F^{q+} and Na^{q+} ions ($q = 2-5$) in the present work. A theoretical method is proposed in this work to subtract the contributions of Auger transitions from the total multiple ionizations in order to investigate the mechanism of the multiple ionization of neon.

3 Results and discussions

Since neon has two electron shells the initial vacancy in its inner-shell (K -shell) may decay in a variety of ways which lead to different charge states. Thus, after the initial production of inner-shell vacancies, there are branching ratios for the various subsequent processes. Where the branching ratio is quite close to unity, it has been set equal to

Table 1. Various channels leading to single, double and triple ionization of neon: $q' = q$ represents the direct ionization process in which the projectile does not change its charge state; $q' = q - 1$ represents the single electron capture process; $q' = q - 2$ represents the double electron capture process; σ_i^T ($i = 1, 2, 3$) is the total i -fold ionization cross-section of Ne; σ_i^{qq} ($i = 1, 2, 3$) is the direct i -fold ionization cross-section of Ne; σ_i^{qq-1} ($i = 1, 2, 3$) is the contribution of the single electron capture to the i -fold ionization cross-section of Ne, and σ_i^{qq-2} ($i = 1, 2, 3$) is the contribution of the double electron capture to the i -fold ionization cross-sections of Ne.

Ne^{i+}	q'	Initial vacancy	Subsequent Processes	σ_i^T	σ_i^{qq}	σ_i^{qq-1}	σ_i^{qq-2}
$i = 1$	q	L		1	1		
	$q - 1$					1	
$i = 2$	q	L^2		1	1		
	$q - 1$					1	
	$q - 2$					1	1
	q	K	KLL Auger	1	1		
$q - 1$					1	1	
$i = 3$	q	L^3		1	1		
	$q - 1$					1	
	$q - 2$					1	
	q	KL	KLL Auger	1	1		
	$q - 1$					1	1
	$q - 2$					1	1
	q	K	$KLLL$ Auger	a	a		
	$q - 1$					a	a

one in Table 1. This is shown in the case of the KLL Auger decay following an initial K -shell or KL vacancy. In the case of triple ionization we get from Table 1 that $\sigma_3^{qq} = \sigma_{L^3}^{qq} + \sigma_{KL}^{qq} + a\sigma_K^{qq}$, where σ_3^{qq} is the cross-section of the triple ionization, $\sigma_{L^3}^{qq}$ is the cross-section of the direct production of three L vacancies, σ_{KL}^{qq} is the cross-section of the production of one L vacancy and one K vacancy accompanied by KLL Auger process, a is the fraction of K vacancies undergoing $KLLL$ Auger decay, and σ_K^{qq} is the cross-section of the production of one K vacancy.

Table 1 lists the various channels leading to single, double and triple ionization of neon. Table 1 shows how single ($i = 1$), double ($i = 2$) and triple ($i = 3$) ionization of neon are related to the fundamental (initial vacancy) cross-sections ($\sigma_i^{qq'}$) and to those of the measurable final target ionization charge states $q' = q, q - 1, q - 2$: σ_i^{qq} , σ_i^{qq-1} and σ_i^{qq-2} .

Single ionization of neon occurs only via L -shell vacancy production. This can be a result of direct L -shell ionization (the cross-section of the direct L -shell single ionization: σ_L^{qq}) or via L -shell capture (the cross-section of the single capture from L -shell: σ_L^{qq-1}). Table 1 shows that these are related to the measurable quantities by

$\sigma_L^{qq} = \sigma_1^{qq}$ and $\sigma_L^{qq-1} = \sigma_1^{qq-1}$. On the other hand, double ionization can result from the production of two L -shell vacancies or from an initial K -shell vacancy followed by a KLL Auger relaxation. Double L -shell vacancies result from direct double ionization ($\sigma_{L^2}^{qq}$), single capture plus one additional ionization ($\sigma_{L^2}^{qq-1}$), or double capture events ($\sigma_{L^2}^{qq-2}$); single K vacancies result from direct ionization (σ_K^{qq}) or capture (σ_K^{qq-1}) of a K -shell electron.

Triple ionization occurs via ionization and capture of three L -shell electrons or via K -shell ionization followed by Auger relaxation. The K -shell ionization can be accompanied by L -shell ionization, either in the initial or in the relaxation stage, via shake-off plus KLL Auger emission or via double Auger emission ($KLLL$ Auger).

As an example, we discuss here the double ionization of neon induced by Auger transition. As one K -electron of neon ionizes, the subsequent Auger process KLL occurs, and forms the double ionization state of neon. The K -Auger yield of neon is 0.982 [11]. The total double ionization cross-section would be

$$\sigma_2^{qq} = \sigma_2^{qq}(\text{DDI}) + \sigma_K^{\text{Auger}} = \sigma_2^{qq}(\text{DDI}) + 0.982 \sigma_K^{qq}, \quad (1)$$

where $\sigma_2^{qq}(\text{DDI})$ is the cross-section of the direct double ionization, σ_K^{Auger} is the contribution of the KLL Auger process.

The measured double-to-single cross-section ratios, $R_{21}^{qq}(\text{exp})$, are the ratios of the total double ionization cross-sections, σ_2^{qq} , with those of the single ionization, $\sigma_1^{qq}(\text{DSI})$, where $\sigma_1^{qq}(\text{DSI}) = \sigma_K^{qq} + \sigma_L^{qq} - 0.982 \sigma_K^{qq}$, *i.e.*

$$R_{21}^{qq}(\text{exp}) = \frac{\sigma_2^{qq}}{\sigma_1^{qq}}. \quad (2)$$

Using equations (1) and (2) the cross-section ratio of the double ionization to that of the single ionization (DI-to-SI), which excludes the Auger contribution, can be obtained from the following formula:

$$R_{21}^{qq} = \frac{\sigma_2^{qq}(\text{DDI})}{\sigma_1^{qq}(\text{DSI})} = R_{21}^{qq}(\text{exp}) - \frac{0.982 \sigma_K^{qq} / \sigma_L^{qq}}{1 + 0.018 \sigma_K^{qq} / \sigma_L^{qq}} = R_{21}^{qq}(\text{exp}) - R_{21}^{qq}(\text{Auger}), \quad (3)$$

where $R_{21}^{qq}(\text{exp})$ is the measured DI-to-SI ratios which include the contribution of Auger transitions, σ_K^{qq} and σ_L^{qq} are the single ionization cross-sections of K -shell and L -shell, respectively. $R_{21}^{qq}(\text{exp})$ includes the contribution of Auger cascading. Since the dual parameter data acquisition is used in our measurement, the contribution of the electron capture can be experimentally separated from the total double ionization. Equation (3) does not include the contribution of the electron capture. The cross-section ratio $\sigma_K^{qq} / \sigma_L^{qq}$ may be calculated by using Bohr theory [16]:

$$\frac{\sigma_K^{qq}}{\sigma_L^{qq}} = \left(\frac{1}{I_K} - \frac{1}{2mv^2} \right) / \left(\frac{1}{I_L} - \frac{1}{2mv^2} \right), \quad (4)$$

where I_K and I_L are the binding energies of the K -shell and L -shell [17]. m is the mass of the electron and v is the

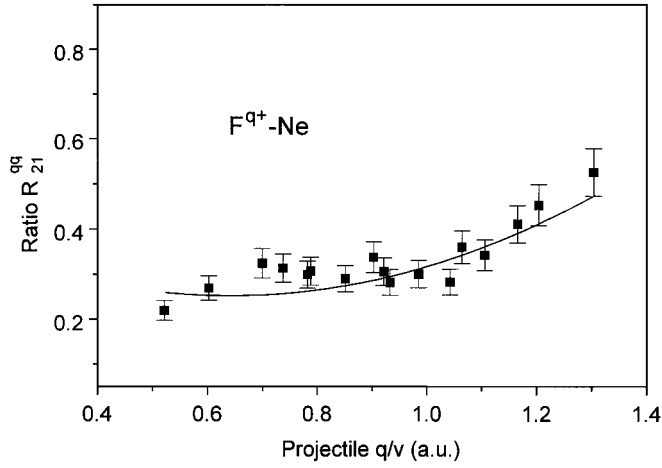


Fig. 2. The corrected double-to-single ionization cross-section ratio for F^{q+} -Ne: ■ present measured data; solid line: fitted result using $(q/v)^2$ scaling.

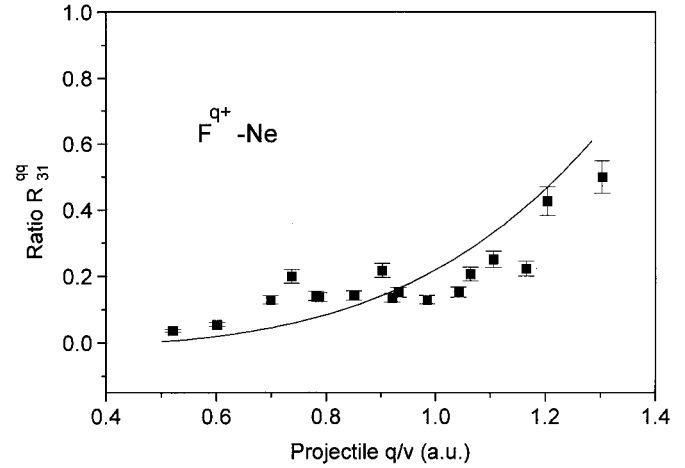


Fig. 3. The corrected triple-to-single ionization cross-section ratio for F^{q+} -Ne: ■ present measured data; solid line: fitted result using $(q/v)^4$ scaling.

projectile velocity. In fact it would be more reasonable if the predictions of the well-working theoretical models or the published experimental cross-section ratios could be used here instead of the calculated results of equation (4). The calculated cross-section ratios of equation (4) are used in the present work for the following two reasons: (a) The present paper studies the collision processes in the intermediate energy range, and there is no successful theoretical model to describe the ionization processes induced by projectiles with $Z > 2$ in this energy range. (b) For the K -shell ionization there are many experimental cross-section data available in the literatures, but there are only very few published L -subshell ionization cross-section data, especially for a projectile which is heavier than proton and helium, and there is no published inner-shell ionization cross-section for F^{q+} -Ne and Na^{q+} -Ne collisions available in the literatures. Using the calculated single ionization cross-section ratios and the present measured cross-section ratios $R_{21}^{qq}(\text{exp})$, the ratios R_{21}^{qq} , which exclude the contribution of Auger process, can be deduced from equation (3).

For the triple ionization of neon a similar method can be used to deduce the cross-section ratio of the triple ionization to that of the single ionization (TRI-to-SI), R_{31}^{qq} . Neglecting the terms of higher orders we get

$$R_{31}^{qq} = R_{32}^{qq} \cdot R_{21}^{qq} \approx R_{32}^{qq}(\text{exp}) \cdot R_{21}^{qq}, \quad (5)$$

where $R_{32}^{qq}(\text{exp})$ is the measured cross-section ratio of triple ionization to double ionization (TRI-to-DI) which is approximately equal to R_{32}^{qq} , excluding the Auger transition. The measured $R_{21}^{qq}(\text{exp})$ and $R_{31}^{qq}(\text{exp})$ in the present work are listed in Table 2 for F-Ne and Na-Ne collisions with projectile energy of 2.0 to 8.0 MeV and charge state of +2 to +5. The deduced ratios R_{21}^{qq} and R_{31}^{qq} are shown in Figures 2 to 5. The solid line in Figures 2 and 4 are the fitted lines using $(q/v)^2$ scaling, and the solid lines in Figures 3 and 5 are the fitted lines using $(q/v)^4$ scaling. The experimental error bars in Figures 2 to 5 are 10%.

Table 2. The measured ratios $R_{21}^{qq}(\text{exp})$ and $R_{31}^{qq}(\text{exp})$ for F-Ne and Na-Ne collisions.

q	E (MeV)	F^{q+} -Ne		Na^{q+} -Ne	
		$R_{21}^{qq}(\text{exp})$	$R_{31}^{qq}(\text{exp})$	$R_{21}^{qq}(\text{exp})$	$R_{31}^{qq}(\text{exp})$
2	2	0.3140	0.1995	0.2726	0.0925
	3	0.2696	0.0542	0.1744	0.0361
	4	0.2198	0.0357	0.1564	0.0062
3	2	0.3427	0.2503	0.3386	0.3366
	3	0.3385	0.2171	0.2987	0.1071
	4	0.2997	0.1406	0.2228	0.0766
4	5	0.3260	0.1302	0.2032	0.1394
	3	0.4536	0.4259	0.5411	0.3084
	4	0.2834	0.1522	0.2955	0.1371
	5	0.2837	0.1520	0.2704	0.1154
5	6	0.2932	0.1440	0.3260	0.1557
	7	0.3115	0.1393	0.3240	0.1345
	4	0.5261	0.4986		
	5	0.4122	0.2235	0.4533	0.3529
	6	0.3632	0.2078	0.2695	0.1125
7	7	0.3044	0.1309	0.3251	0.1460
	8	0.3102	0.1372	0.3001	0.1252

It is found that Auger process is less important when the projectile energy is low, and it becomes more and more important as the projectile energy increases. When the impact energy is high enough, the Auger transition may become one of the most dominant processes of the multiple ionization of atom. Since the ionization cross-section ratio of the K -shell to the L -shell is in the order of 10^{-3} in the present energy range, the Auger process plays only a minor role in the ionization of target atom for the collision systems studied in this work.

The deduced DI-to-SI cross-section ratios corrected by Auger transition in Figures 2 to 5 show agreement with the $(q/v)^2$ scaling, which is predicted by the two-step

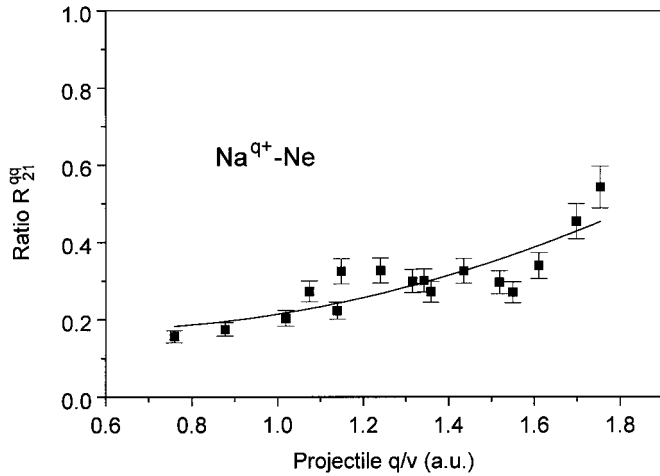


Fig. 4. The corrected double-to-single ionization cross-section ratio for Na^{q+} -Ne: ■ present measured data; solid line: fitted result using $(q/v)^2$ scaling.

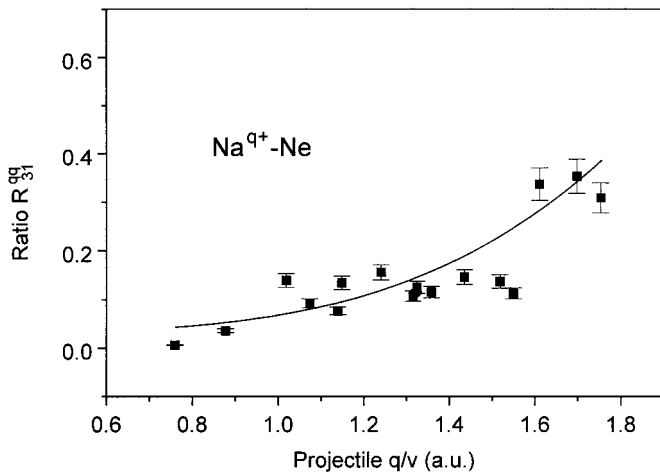


Fig. 5. The corrected triple-to-single ionization cross-section ratio for Na^{q+} -Ne: ■ present measured data; solid line: fitted result using $(q/v)^4$ scaling.

mechanism, and the TRI-to-SI cross-section ratios agree with the $(q/v)^4$ scaling. It may be suggested that the multi-step (two-step and three-step) mechanism may dominate the direct double and triple ionization of neon by F^{q+} and Na^{q+} ions. This is in agreement with the results of Manson *et al.* [12] and Cai *et al.* [13].

The electronic structure dependence of the electron transition occurring in ion-neon collisions is studied in the present work. Figure 6 shows the relative yields of recoil ion Ne^{1+} for different charge states q' of the scattered projectile in 5.0 MeV F^{5+} and Na^{5+} -Ne collisions. The error bars of Figure 6 are 12%. In Figure 6 $q' = 5$ means the direct single ionization ($q' = q = 5$), $q' = 6$ means the single stripping (projectile)+single ionization of target atom, $q' = 4$ means the single capture and so on. Since fluorine, neon and sodium atoms have similar atomic numbers the study based on the collision systems of these three atoms may provide effective and clear infor-

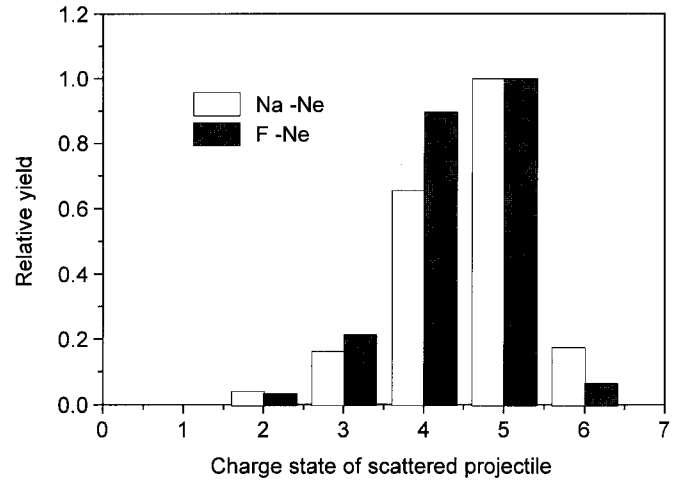


Fig. 6. Relative yields of Ne^{1+} for 5.0 MeV F^{5+} -Ne and Na^{5+} -Ne collisions.

mation about the electronic structure dependence of the electron transitions during ion-atom collisions.

It is apparent from Figure 6 that the fivefold fluorine ions more easily capture electrons from the target atom during their collisions with the target, and the fivefold sodium ions lose their electrons more easily than the fluorine ions. We may consider the different electronic structure of these two ions to understand the phenomenon. The fivefold fluorine ion has the electron configuration of $1s^2 2s^2$, and the fivefold sodium ion has the configuration of $1s^2 2s^2 2p^2$. The binding energies of the outer-most shell electron of F^{5+} and Na^{5+} could be calculated using [18]:

$$E = -\frac{Rhc(Z-s)^2}{n^2} - \frac{Rhc\alpha^2(Z-s)^4}{n^3} \left(\frac{1}{l+1} - \frac{3}{4n} \right) \quad (6)$$

here R is the Rydberg constant, h is the Planck constant, α is the fine structure constant, c is the velocity of light, n is the principal quantum number, l is the angular momentum quantum number, and $(Z-s)$ is the effective nuclear charge considering the screening effect. For the electron of the outer-most shell of F^{5+} ion, $n = 2$, $l = 0$, $Z - s \approx 7$, and for that of Na^{5+} ion, $n = 2$, $l = 1$, $Z - s \approx 7$. The binding energy of the fivefold fluorine ions is nearly four times larger than that of the Na^{5+} based on the calculated results of equation (6). Thus F^{5+} ions capture electrons more easily than Na^{5+} ions, while Na^{5+} ions lose electrons more easily than F^{5+} ions when meeting other atoms. It may be concluded that the electronic structure (chemical property) of the projectile may considerably affect the probability and the mechanism of the electron transitions in the collisions with other atoms.

Summary

The cross-section ratios of the double ionization (DI) and triple ionization (TRI) to those of the single ionization (SI), R_{21}^{qq} and R_{31}^{qq} , have been measured using time-of-flight and dual parameter data acquisition methods for

2.0-8.0 MeV F^{q+} -Ne and Na^{q+} -Ne collisions. A practical method is proposed in this work to subtract the contribution of the Auger transition from the total multiple ionization in order to investigate the multiple ionization mechanism of neon. It is found that the Auger process plays only a minor role in the ionization of target atom for the collision systems studied in this work.

The obtained cross-section ratios R_{21}^{qq} and R_{31}^{qq} , which exclude the Auger channels, are in agreement with the $(q/v)^2$ and $(q/v)^4$ scalings for F^{q+} -Ne and Na^{q+} -Ne collisions. It may suggest that the multi-step process may dominate the multiple ionization of neon by 2.0-8.0 MeV F^{q+} and Na^{q+} ions bombardment.

The electronic structure dependence of the electron transition occurring in ion-neon collisions has been studied for F^{q+} -Ne and Na^{q+} -Ne collisions. It may be concluded that the electronic structure (chemical property) of the projectile may considerably affect the probability and the mechanism of the electron transitions in ion-atom collisions.

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