Experimental evidence for dielectronic excitation producing Ne *K* vacancies in 35-keV $N^{7+}+Ne$ collisions

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The method of Auger electron spectroscopy was utilized to measure cross sections for the production of *K*-Auger electrons in $N^{7+}+Ne$ and $N^{6+}+Ne$ collisions at an impact energy of 35 keV. In addition to the *K*-Auger electrons from the nitrogen projectile following multiple electron capture, *K*-Auger electrons of the Ne target are observed for the system $N^{7+}+Ne$. This observation is attributed to dielectronic excitation, produced by electron-electron interaction, where a *K*- and an *L*-shell electron of Ne are transferred into the *K* shell of N^{7+}. [S1050-2947(96)50112-5]

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In the past few years, considerable work has been devoted to multiple electron capture in slow collisions between a multicharged ion and a neutral target. Processes responsible for electron capture have been extensively studied experimentally [1–3] and theoretically [4,5]. Particular attention has been devoted to dynamic electron-correlation effects that occur in ion-atom collisions [6,7]. These effects are produced by mutual interactions of two electrons whose description leads beyond the independent particle model. A characteristic dielectronic process is *autoexcitation* [8], where one electron is transferred to a deeper level, while another electron is excited to a high Rydberg state. This process can occur in separated atoms as well as in collisional transient molecules. Examples for autoexcitation processes occurring during the collision are the processes of correlated double capture [2,6,7] and correlated transfer excitation [9].

In this paper, we focus our attention on the inverse autoexcitation process, denoted *dielectronic excitation* [10]. Due to electron-electron interaction, an electron from a higher-lying level is deexcited transferring its excess energy to another electron which, in turn, is removed from a deeper-lying level. Hence, dielectronic excitation involves a mechanism producing vacancies in a rather deep inner shell. It is noted that this process can also take place in the transient molecule that is formed during the collision. The most important feature of dielectronic excitation is its dominance at low collision energies at which other mechanisms (excitation, ionization) fail to produce inner-shell vacancies. It should be emphasized that dielectronic excitation and autoexcitation are rather similar, as both processes are produced by dynamic electron correlation.

The first indication for dielectronic excitation of a *K* shell has been provided by Afrosimov *et al.* [11], studying the singly charged system $N^{+}+Ar$. Similar measurements of vacancy creation in the *L* shell of the heavier collision partner in the collision $Ar^{+}+Si$ have been performed by Dubois, Stolterfoht, and Schneider [12]. In the latter example, a promotion of two *L*-shell electrons of Si occurs, so that resonance conditions are created for the inverse autoexcitation process. The two vacancies are simultaneously filled by transitions of an electron from a higher-lying orbital and an electron from the $2p$ orbital of Ar. Recently, this process was confirmed by model calculations by Stolterfoht [13] using first-order perturbation theory in conjunction with the semiclassical approximation.

The study of the $Ar^{+}+Si$ system has been motivated by related work about ion-solid collisions involving Ar^{+} projectiles on crystalline Si. Recently, new interest in the field was created since much attention has been devoted to the interaction of slow, highly charged ions with solid surfaces where ''hollow'' atoms are produced [14,15]. These studies have clearly exhibited the need for conclusive evidence regarding the process of dielectronic excitation.

In the previous study [13] the existence of dielectronic excitation was made evident by comparison with a model calculation. As this model is relatively simple, a direct experimental proof would be desirable. For the process of dielectronic excitation, at least two vacancies are needed in an inner shell. In singly charged ion-atom collisions, these vacancies are produced during the collision. In highly charged ion-atom collisions, these vacancies can be prepared prior to the collision. Hence, for a projectile with two vacancies in the *K* shell, the dielectronic excitation process is rather likely and the related cross sections are expected to be relatively large.

In this work, we provide direct experimental evidence for dielectronic excitation in the case of the multiply charged system $N^{7+}+Ne$. This is done by comparison with the results of the $N^{6+}+Ne$ system. It is noted that for both systems the capture of a $1s$-Ne electron by means of an electron single transition is very unlikely at low projectile energy. In the case of $N^{6+}+Ne$ collisions, only one *K* vacancy exists in the N^{6+} projectile and, consequently, the process of dielectronic excitation is expected to fail. On the contrary, in the $N^{7+}+Ne$ system, two *K* vacancies exist in the N^{7+} projectile before the collision. A simulta-

neous capture of a $1s$ and a $2l$ target electron may occur, due to dielectronic excitation, populating the $1s$ orbital of nitrogen.

After dielectronic excitation in the system $N^{7+}+Ne$, the target is in a highly excited state with a vacancy in the Ne K shell, which may decay via the emission of Auger electrons. Hence, dielectronic excitation in the system N^7+Ne can be studied by means of target Auger spectroscopy. In this work, Auger-electron spectra from both projectile and target were compared for the systems $N^{6+}+Ne$ and N^7+Ne. The mechanism of dielectronic excitation is interpreted within the framework of a molecular orbital diagram. Absolute cross sections for the production of Ne K-shell vacancies are deduced from target Auger-electron spectra, showing that the mechanism for dielectronic excitation is important.

The measurements were carried out at the 14-GHz electron cyclotron resonance (ECR) ion sources at the Grand Accélérateur National d'Ions Lourds (GANIL) in Caen and at the Ionenstrahl-Labor of the Hahn-Meitner Institut (HMI) in Berlin, using the electron-spectroscopy apparatus [16] from HMI. Ions of N^{6+} and N^{7+} were extracted from the ECR source and collimated to a diameter of ~ 2 mm, with typical currents of about 30 nA. In the collision chamber, the beam was colliding with an effusive Ne-gas jet. During the acquisition, the pressure of $\sim 3\times 10^{-5}$ Torr was maintained in the chamber. This pressure was sufficiently low to avoid multiple charge-exchange collisions for the incident ions.

Auger electrons produced in $N^{q+}+Ne$ collisions were detected at several angles up to 150°, with respect to the incident beam direction, using a single-stage spectrometer which consists of a parallel-plate analyzer [16]. The resolution of the analyzer was 5% full width at half maximum (FWHM). The length l_0 of the ion beam, seen by the spectrometer at 90° with respect to the incident beam, was ~ 4 mm. This length increases according to $l=l_0/\sin\theta$ as the observation angle decreases.

To evaluate absolute cross sections, we used methods described in detail previously [16]. Absolute values for the production of Auger electrons were obtained using a uniform gas pressure in the scattering region. This was achieved by moving the jet upwards far away from the ion beam and flooding the scattering chamber uniformly with the Ne target gas. Auger spectra were measured with low resolution both with the gas jet and in the uniform-gas mode.

Figure 1 shows typical Auger-electron spectra for the systems 35-keV N^{6+} and $N^{7+}+Ne$ at an observation angle of 90°. Two groups of lines are clearly separated. In the range 0–550 eV, the spectra show peaks attributed to electrons emitted by the projectile, whereas the group of lines in the range 600–900 eV follows the deexcitation of the target. The origin of the Auger electrons from projectile and target was verified by analyzing the Doppler effect, due to the velocity of the projectile. The Doppler effect leads to a shift of the peaks associated with the projectile when changing the observation angle. On the contrary, the group of peaks associated with the target is insensitive to the Doppler effect, because of the low velocity of the target after the collision.

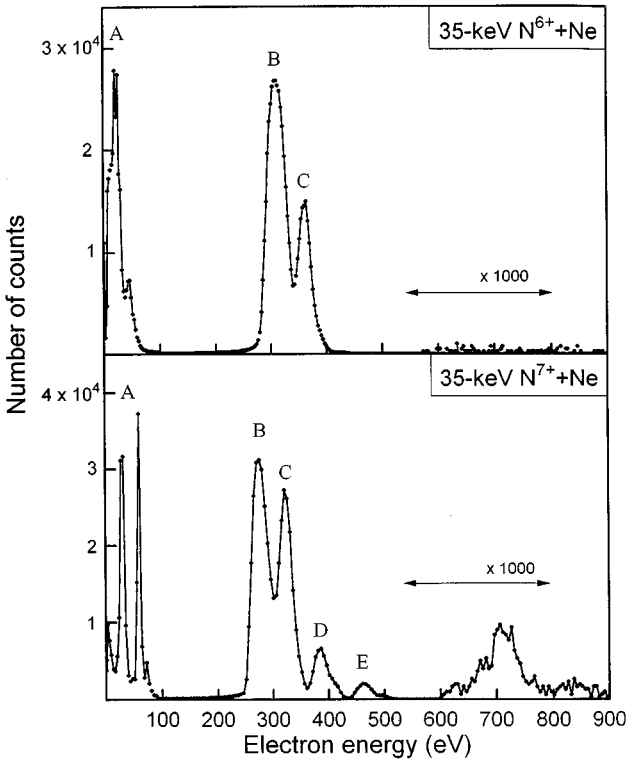

FIG. 1. Spectra of Auger electrons produced in 35-keV $N^{6+}+Ne$ and $N^{7+}+Ne$ at an observation angle of 90°. The peaks in the range 0–550 eV correspond to the decay of projectile states associated with configurations $2lnl'$ ($n\geq 2$) and $3lnl'$ ($n\geq 3$). The lines in the range 280–550 eV are essentially due to capture into triple excited states $2ln_1l_1n_2l_2$ ($n_2\geq n_1\geq 2$) and $3ln_1l_1n_2l_2$ ($n_2\geq n_1\geq 3$). The peaks centered at ~ 700 eV in the collision $N^{7+}+Ne$ are produced by dielectronic excitation, which simultaneously creates a vacancy in a target K shell and a transfer of a $2l$ target electron. It is seen that the collision $N^{6+}+Ne$ does not create a K vacancy in the target.

We first examine the peaks associated with the projectile (denoted A to E in Fig. 1). The emission of these electrons follows the capture of two or more $2l$ electrons from the target, at relatively large internuclear distances. Since our task is not the detailed analysis of these peaks, a brief comparison is made of the present systems $N^{q+}+Ne$ with the system $N^{7+}+Ar$ studied previously [17]. First, it is likely that double capture populates configurations of quasiequivalent electrons $3lnl'$ and $4lnl'$ ($n\geq 4$), which give rise to L-Auger electrons in the range 0–100 eV (line A in Fig. 1). The lines in the range 280–550 eV are essentially due to capture into triple excited states $2ln_1l_1n_2l_2$ ($n_2\geq n_1\geq 2$) and $3ln_1l_1n_2l_2$ ($n_2\geq n_1\geq 3$), which give rise to K-Auger electrons. Hence, for both spectra in Fig. 1, we expect that the lines B and C (D and E) correspond to Auger electrons following the capture of three $2l$ electrons into configurations $2ln_1l_1n_2l_2$ ($3ln_1l_1n_2l_2$). It is noted that the Coulomb interaction between the projectile nucleus and target electrons is mainly responsible for the charge-exchange process producing these configurations.

We now focus on electrons emitted in the range 600–900 eV (Fig. 1). First, a remark about the width of the target-Auger peaks is in order. There are several broadening effects present in $N^{q+}+Ne$ collisions. It has been shown previously

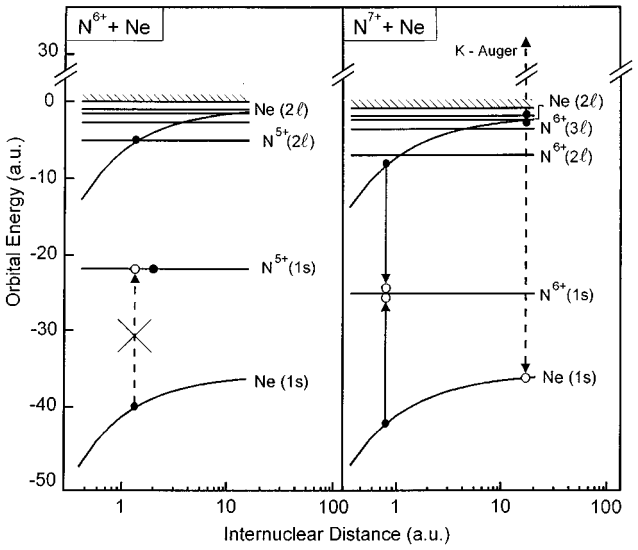

FIG. 2. Diagram of orbital energies for the systems $N^{6+}+Ne$ and $N^{7+}+Ne$. In $N^{6+}+Ne$ (left diagram), Ne K-shell excitation is unexpected, since the probability for single-electron transfer can be neglected in slow collisions. In $N^{7+}+Ne$ collisions, resonant energy conditions lead to a simultaneous transfer of a $1s$ and a $2l$ target electron by means of dielectronic excitation (left diagram). This process produces target configurations decaying by K-Auger-electron emission (dashed lines).

[18] that, at low collision energies, line broadening of a few 10 eV is expected for each peak. Consequently, since the group of lines associated with the target is composed of a large number of individual lines, it is impossible to perform high-resolution studies of the Auger electrons. Therefore, the present study is performed with low resolution.

A group of lines due to the deexcitation of the target is observed for the system $N^{7+}+Ne$, whereas the lines which are in the same range are missing for the system $N^{6+}+Ne$. This is attributed to the occurrence of dielectronic excitation in the $N^{7+}+Ne$ system. The mechanism of dielectronic excitation leading to the capture of a $1s$ target electron is illustrated in Fig. 2, which shows approximate orbital electron energies for the collisional systems $N^{6+}+Ne$ and $N^{7+}+Ne$. In the incident channel, two electrons occupy the Ne-$1s$ orbital and eight electrons fill the Ne-$2l$ orbital. In both systems, the Ne-$1s$ orbital does not cross the orbitals of the projectile. Thus, the capture of a $1s$ target electron by means of a single-electron transition is expected to be unlikely in both collisional systems.

In the case of the system $N^{7+}+Ne$ (right side of Fig. 2), it is seen that at internuclear distances of ~ 1 a.u. resonance conditions are created for dielectronic excitation process in which one $2l$ target electron is transferred into the $1s$ projectile orbital, giving its excess energy to a $1s$ electron of neon. This electron, in turn, is excited and transferred into the $1s$ shell of the projectile. It is important to note that the presence of two $1s$ vacancies initially in the projectile is essential for the dielectronic excitation process. It is clear that the dielectronic excitation process is impossible for the collision system $N^{6+}+Ne$ (left side of Fig. 2) because the $1s$ projectile orbital is already occupied by one electron. Indeed, the $N^{6+}+Ne$ system does not indicate measurable intensity between 600 and 900 eV that can be associated with the Ne K-Auger emission (Fig. 1).

TABLE I. Auger emission cross sections in the systems $N^{7+}+Ne$ and $N^{6+}+Ne$. The quantities σ_p (σ_t) refer to the deexcitation of the projectile (target) after the collision.

Collision system	Auger emission cross section σ_p (cm^2)	σ_t (cm^2)
$N^{7+}+Ne$	$(2.3\pm0.4)\times10^{-14}$	$(3\pm1.5)\times10^{-18}$
$N^{6+}+Ne$	$(1.5\pm0.3)\times10^{-14}$	$<10^{-20}$

The measured Auger spectra were used to evaluate total Auger emission cross sections. Cross sections for Auger emission attributed to projectile configurations is denoted σ_p, whereas σ_t refers to Auger emission cross sections associated with the creation of a K-shell target vacancy due to dielectronic excitation. Differential cross sections $d\sigma_p/d\Omega$ and $d\sigma_t/d\Omega$ for Auger electron emission were obtained by integration of the spectra with respect to electron energy. The emission of Auger electrons was found to be isotropic, within the experimental uncertainties. Thus, total Auger-electron emission cross sections σ_p and σ_t were derived by multiplying the corresponding differential cross sections by 4π.

The results are given in Table I. The capture of $2l$ target electrons due to nucleus-electron interaction leads to a value of about 10^{-14} cm^2 for both systems N^{6+}, $N^{7+}+Ne$. This value is consistent with cross sections estimated for the system $N^{7+}+Ar$ [17]. For the peaks associated with the deexcitation of the Ne target in the system $N^{7+}+Ne$, the Auger emission cross section was found to be 3×10^{-18} cm^2. The same order of magnitude was found in collisions in which electron correlation plays an important role. For example, cross sections for the production of configurations $2lnl'$ ($n\geqslant3$) in the collision $C^{6+}+He$ [7] are of the order of 10^{-17} cm^2. Hence, the dielectronic excitation process is significant in the system $N^{7+}+Ne$. For the system $N^{6+}+Ne$, we estimated an upper limit of $\sim10^{-20}$ cm^2, which corresponds to the electron background. Hence, the K-vacancy production is very small in $N^{6+}+Ne$ collisions.

In conclusion, the dielectronic excitation process involving the capture of K-shell electrons has been clearly identified in the present experiment, by comparison of the collisional systems $N^{7+}+Ne$ and $N^{6+}+Ne$. This process provides a mechanism producing a vacancy in a deep inner shell in slow collisions. The corresponding cross section was found to be large. Further experiments are suggested to study the projectile energy dependence of dielectronic excitation processes. It has been previously shown that the importance of dielectronic processes increases with decreasing projectile velocity [19]. Furthermore, the dielectronic excitation process is of great interest for collisions of multiply charged ions with solids, which have often been studied at very low energies [20]. For these reasons, we plan to study the dielectronic excitation in collisions at energies smaller than the one used in this work.

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