

HIGH-RESOLUTION AUGER SPECTROSCOPY OF MULTIPLY CHARGED PROJECTILE IONS

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RÉSUMÉ: Recentes études à haute résolution d'électrons Auger émis par des projectiles multichargés sont revus. Mechanismes différents de production d'électrons Auger dans des collisions ion-atome sont discutés pour énergies incidentes de quelques keV/u à quelques MeV/u. Ionization et excitation 'aiguille' sont étudiées pour projectiles de haute vitesse. Pour energies incidentes intermediaire simple capture et transfer-excitation sont considerées. Effets de correlation d'électrons sont discutés pour double capture d'électrons dans des systèmes des collisions lentes et multichargées.

ABSTRACT: Recent high-resolution studies of Auger electrons from multiply charged projectile ions are reviewed. Different mechanisms for Auger electron production in ion-atom collisions are discussed for incident energies from a few keV/u to a few MeV/u. 'Needle' ionization and excitation are studied for fast projectiles. At intermediate incident energies single capture and transfer-ionization are considered. Electron correlation effects are discussed for double electron capture in slow multicharged collision systems.

I. INTRODUCTION

In 1925 Pierre Auger [1] observed remarkable tracks in cloud chamber photographs and attributed them to monoenergetic electrons spontaneously emitted from atomic species. This observation was the discovery of an important decay mode for highly excited atoms competing with the decay mode of photon emission. In the past several decades the Auger effect has received increasing attention in many fields of physics, including atomic, nuclear, solid state, and surface physics.

In recent years the interest has been focused on Auger electrons originating from highly charged ions. This interest has been generated to a large degree in the fields of plasma physics, thermonuclear fusion research, and astrophysics where the properties of highly charged ions play an important role. Multiply charged ions are readily obtained at particle accelerators. Projectiles can be stripped before being directed into the collision region and target atoms can be highly ionized in violent collisions. These different techniques of producing multiply charged ions are involved in the methods of projectile Auger spectroscopy and target Auger spectroscopy [2].

The method of target Auger spectroscopy [3] has been used extensively in ion-atom collision studies. Target atoms may be highly ionized in single collisions when sufficiently heavy projectiles are used in the experiments [4,5]. Also, when projectiles are incident at a high velocity, only small momenta are transferred to the recoil ions so that kinematic broadening effects are negligible. However, target spectroscopy involves serious problems arising from several inner-shell vacancy states randomly created in heavy ion-atom collisions. This is indicated in Fig. 1 which shows the essential features of target and projectile spectroscopy [6].

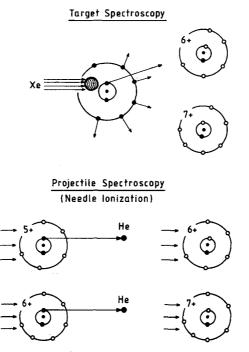


Figure. 1 Schematic diagram showing the main features of target and projectile spectroscopy. The method of ionizing the projectile with light target atoms while preserving the projectile outer shell configuration is called needle ionization. From Ref. [6].

For many years the method of projectile spectroscopy [7] has suffered from the complexity of the Auger spectra composed of broadened lines. The line broadening is due to the strong influence of the Doppler effect for electrons originating from a fast emitter [8,9]. Only recently have the problems of the Doppler broadening been greatly reduced by using the method of zero-degree Auger spectroscopy [10-12]. At an electron observation angle of zero degree the Doppler broadening cancels in first order and, hence, high-resolution measurements are possible for fast projectiles. Itoh et al. [10,13] have exploited the method of zero-degree Auger spectroscopy for projectiles with energies as high as 5 MeV/u. Recent results from this method show that Auger spectra can be recorded with unprecedented high resolution [14].

Projectile Auger spectroscopy has several remarkable features. Once the Doppler broadening is greatly reduced, various kinematic effects may advantageously be applied in experiments, as recently discussed in detail [2]. Also, projectile spectroscopy allows one to limit the number of charge states of the excited ions and, thus, it is possible to gain selective access to specific Auger states. This is achieved by using light target atoms which do not significantly disturb the outer shell of the projectile during the collision (Fig. 1). Hence, the characteristic features of the projectile outer-shell are preserved and they become a signature of the initial state involved in the Auger transition after the collision. The method of using light target atoms in projectile Auger spectroscopy has been called needle ionization [13] as indicated in Fig. 1. The needle ionization is a specific case of the more general concept of **ion surgery** [2,15] where selective excitation or capture is also considered in ion-atom collision processes.

In this article recent measurements using the method of projectile Auger spectroscopy are reviewed. This method can be used as a powerful tool in the analysis of atomic structure and excitation mechanisms in ion-atom collisions. It is shown that detailed information about ionatom collisions may be obtained when methods of high-resolution spectroscopy are combined with methods used to study excitation mechanisms, e.g. the measurements of total yields or angular distributions of Auger electrons. In the following various reaction mechanisms involving one- and two-electron transitions are discussed. Measurements at high, intermediate, and low incident energies are treated separately, since at these energies different reaction mechanisms are favored. It is noted that some of the present topics have also been treated in a recent review by the author [16].

II. SINGLE IONIZATION AND EXCITATION AT HIGH INCIDENT ENERGIES

Before studying the Auger electrons from fast projectiles, a few characteristic features of target spectroscopy shall be recalled. It has been shown previously that the number of states involved in Auger spectra can be greatly reduced when the target atom is stripped to a few-electron system [4,5]. Hence, Auger spectra from, e.g., Li-like ions have been studied. However, it has been found that considerable problems arise from line blending in the Auger spectra when more than three or four electrons are left on the target ion [9].

In particular, line blending problems are encountered when L-Auger spectra from target ions with more than 11 electrons are studied. This is shown in Fig. 2 obtained for the L-Auger spectrum of multiply charged Ar produced by 5.9 MeV/u U⁶⁶⁺ impact by Folkmann et al. [17]. Similar results have been obtained for O^{q+} impact by Schneider et al. [18] and for Ar^{q+} impact by Matsuo et al. [19]. In Fig. 2⁻ the spectrum shows a limited number of well separated lines such as the P_{1/2}-P_{3/2} doublet due to the Na-like configuration $1s^2 2s^2 2p^6 3s^2$ near 102 eV. However, the main bulk of the spectrum consists of overlapping lines which makes it difficult to verify individual structures.

An important method to limit the number of states in target spectra is the observation of the Auger electrons time delayed by nanoseconds after the collision [20]. Thus, electron capture in secondary collisions of the slow recoil ions with target atoms have been studied. Na-like configurations are formed by electron capture into Ne-like Ar^{10+} recoil ions in the metastable state $1s^2 2s^2 2p^5 3s$ °P. This recoil Auger spectroscopy yields detailed information about configurations involving electrons with higher principle quantum number n. Fig. 2 shows that Auger states involving electrons with n=4 to 6 can selectively be produced by adding certain gas components in the target region. It is evident, however, that this method is limited essentially to states based on the Na-like configuration.

Access to other configurations is achieved by using the method of projectile spectroscopy in conjunction with the needle ionization concept. This is shown in Fig. 3 [21] which depicts L-Auger spectra for Arq^+ incident with q=5 to 7 on He at energies of about 100 MeV.

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The prominent lines seen for the incident charge states q=5 and 6 (Fig. 4b and c) are produced by the removal of a projectile 2p (or 2s) electron into the continuum. Hence, the most probable charge state of the emitting ion is one unit higher than the incident charge state. Since needle ionization of the 2p electron leaves the outer shell practically undisturbed, the dominant production of the Na-like configuration $1s^2 2s^2 2p^5 3s^2$ (near 102 eV) is understood for the incident charge state q=6. Accordingly, for q=5 the dominant creation of the Mg-like configuration $1s^2 2s^2 2p^5 3s^2 3p$ (near 112 eV) is plausible.

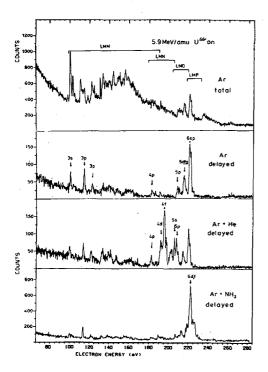
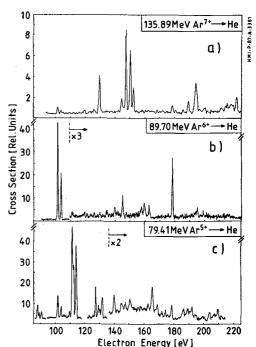


Figure 2. Prompt and delayed Argon-L target Auger spectra produced in collisions of 5.9 MeV/u U⁶⁶⁺ projectiles with Ar admixed with He and CH₄. From Folkmann et al.[17] Figure 3. Argon-L projectile Auger spectra produced in collisions of Ar^{q+} (q=5,6, and 7) with He. The spectra result primarily from excitation and ionization of a 2p electron. From Schneider et al.[21].

Furthermore, there are configurations produced by excitation of the projectile 2p electron to bound states. This process, denoted needle excitation, involves a projectile charge state equal to that of the incident ion. For instance, the Na-like configurations observed for the incident charge state q=7 (Fig. 3a) are predominantly created by needle excitation. It is found that the excitation process favors the creation of the $1s^2 2s^2 2p^5 3s^3 d$ configuration resulting from the dipole transition 2p+3d. In Fig. 3a the corresponding states are seen near 150 eV.

Finally, it can be verified that charge states produced by electron capture are negligible, but that double ionization is found to play a certain role [10]. In this case, when two-electron processes occur, the method of needle ionization reaches its limits.



III. SINGLE CAPTURE AND TRANSFER IONIZATION AT INTERMEDIATE INCIDENT ENERGIES

a. K-AUGER SPECTRA

Two-electron processes gain importance for projectile energies of a few hundred keV/u denoted here as intermediate energies. In this case also single capture may become significant. Before these processes are examined, an application of the needle ionization concept shall be considered. It is emphasized that, although more complex mechanisms occur, the needle ionization method remains important at intermediate incident energies. The different processes are discussed by means of Fig. 4 where carbon-K Auger spectra for C^{2+} , C^{4+} , and C^{5+} incident on He are displayed.

Fig. 4c exhibits Li-like and Be-like configurations which are produced

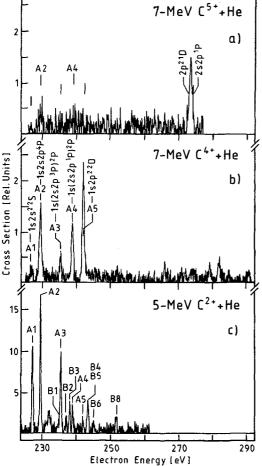


Figure 4. Auger spectra produced in 5- and 7-MeV C^{q+} + He collisions with q=2, 4, and 5. States due to Li-like and Be-like configurations are labeled A and B, respectively. From Ref. [15]

by ionization and excitation of a 1s electron, respectively. An important application of needle ionization is the analysis of incident metastable states. Close inspection of the Auger spectrum for C^{2+} impact (Fig. 4c) shows that the majority of Li-like projectiles are in Auger states which involve outer-shell excitation - see for instance the configuration 1s2s2p. This observation and, in particular, the fact that the guartet state 1s2s2p ⁴P is produced, leads to the conclusion that the C2+ ions are incident in the metastable states 1s² 2s2p ³ Pol2. These states live sufficiently long so that they can survive their travel through the accelerator. Analysis of the line intensities (Fig. 4c) shows that the fraction of incident metastable ions amounts to 75 % [15]. This value appears surprisingly large in view of the relatively high excitation energy of about 8 eV for the metastable ions. It is noted however that the observed fraction of metastable ions is consistent with the assumption of a statistical population of the multiplets involved [2].

For C⁴⁺ and C⁵⁺ impact the states produced by the removal of a single 1s electron are not autoionizing so that processes other than ionization must be responsible for the creation of the corresponding data (Fig. 4a and 4b). The C⁴⁺ spectrum shows lines due only to Li-like configurations such as 1s2s2p which could, in principle, be produced by an electron transfer into the 2s shell accompanied by a 1s-2p transition in the projectile. However as pointed out by Dillingham et al. [22], the Li-like configurations are likely to be produced by single electron transfer to projectiles incident in the metastable state 1s2s 3 S. The most prominent line due to the 1s2p² ²D state is produced by electron transfer to the projectile whose 2s electron is simultaneously excited to the 2p shell. Hence, this two-electron process refers to transfer-excitation into a metastable projectile state.

For the incident one-electron ion C^{5+} states due to Li-like configurations may be created by double electron capture into shells with principle quantum number $n\geq 2$ [22]. These lines, however, are barely seen in Fig. 4b. Instead, at higher electron energies the spectrum exhibits one prominent line which is associated with the He-like states $2p^{2-1}D$ and/or $2s2p^{-1}P$. These states are produced by a transfer-excitation process as discussed by Itoh et al. [23] for He⁺ impact.

The interesting point with the transfer-excitation process is that it may proceed in a resonant manner. If the target electron is regarded as being free, resonant transfer excitation (RTE) is identical with an inverse Auger process which is initiated by electron-electron interaction [24]. The maximum cross section for RTE occurs for a projectile velocity equal to the velocity of the associated Auger electron. This has been verified for Auger-electron emission by Swenson et al. [25] studying O^{5+} impact on He at energies of a few MeV. For the present case it is noted that the velocity of the electrons from the $2p^2$ ¹D/ 2s2p ¹P states matches the velocity of the 7-MeV carbon projectile. Hence, it is rather likely that the dominance of the $2p^2$ ¹D/2s2p ¹P line (Fig. 4a) is due to the importance of the resonant-transfer excitation process.

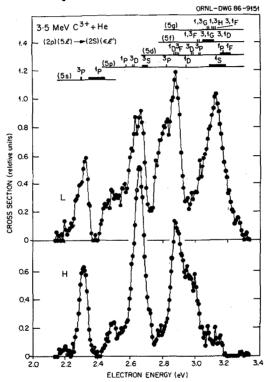
b. COSTER-KRONIG SPECTRA

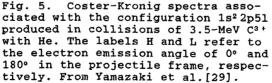
An important class of autoionizing states refers to configurations involving a Rydberg electron. In this connection particular attention has been paid to the Coster-Kronig process where the lower-lying electron undergoes transitions between subshells in the same main shell [2]. Usually, Coster-Kronig transitions are energetically forbidden in multiply charged ions. They become possible, however, when loosely bound Rydberg electrons are present in the ion [26]. Hence, the study of the electrons from Coster-Kronig transitions provides information about the occupation of Rydberg states[27,28].

Coster-Kronig transitions of the type $1s^2 2s\epsilon l-1s^2 2pnl$ have recently been measured with high resolution by Yamazaki et al.[29] in C^{3+} + He collisions. The results for the initial configuration $1s^2 2p5l$ is given in Fig. 5. This configuration is produced by a transfer-excitation process in the 3.5-MeV C^{3+} + He system whose projectile is incident in the configuration $1s^2 2s$. The spectrum transformed into the projectile frame shows the high resolution of 60 meV (FWHM) which was achieved by using kinematic compression effects [6] characteristic of projectile Auger spectroscopy. Comparison with theoretical results (Fig. 5) indicates that the resolution is sufficient to exhibit the term splitting and the quantum defect of the Rydberg electron.

The striking feature of the results in Fig. 5 is the difference in the electron spectra obtained at 0° (labeled H) and 180° (labeled L) in the projectile frame (e.g. see the line group near 3.2 eV). The picture of producing aligned states suggests that, after the collision, the electron density distribution has a forward-backward symmetry along the incident beam axis. Actually, this is not true. As shown by Havener et al.[30] the electron cloud may be asymmetric (polarized)

shortly after the collision. Generally, the electron cloud undergoes rapid oscillations (quantum beats) and the measurement of the ejected electron involves a certain time averaging. If the averaging time is large, one obtains the impression of a forward-backward symmetry. However, if the observation time of the electrons is short, the oscillations of the electron cloud may be observed as in quantum beat experiments [31]. In the present case the observation time corresponds to the lifetime of the autoionizing states. More specifically it is noted that the autoionization width (Fig. 5) is comparable with the energy separation of two or more states differing in parity [29]. Pictorially one may say that the present autoionizing system provides a snapshot over the first instant of the quantum beat event when the electron cloud is still polarized.





Another interesting aspect of the Coster-Kronig spectra is that they yield information about the angular momentum 1 distribution of the associated Rydberg electrons. In Fig. 5 the spectra show four prominent peaks which are composed of various states attributed to certain angular momenta 1. The first peak (at lowest energy) is due to the single term 1s² 2p5s ³P which represents the s state of the Rydberg electron. Similarly, the second peak is composed of lines attributed to the p state of the Rydberg electron. The third peak contains lines due to terms attributed to p,d, and some f states. Estimates show that the p contribution amounts to about 30 % of the intensity of the third peak [32]. The fourth peak is difficult to deconvolute. It contains some contribution from f and g states but its main intensity is due to short lived terms due to p and d states. These latter states are expected to be responsible for the $0^{\circ}-180^{\circ}$ asymmetry seen in Fig. 5.

A preliminary line fitting analysis indicates that in 3.5 MeV C^{3+} + He collisions the angular momenta s, p, d, and f are produced with approximate probabilities of 0.15, 0.5, 0.2 and 0.1, respectively [32]. Hence, the p state is clearly dominant. For lower projectile energies the contribution from the s momentum is found to be enhanced. It is noted that in the present case the method of zero-degree spectroscopy is sensitive only to the magnetic substate M=0. The results given here for the 1 distribution are obtained under the assumption that the magnetic quantum numbers associated with a given term are equally populated. This assumption should be taken with some caution since anisotropic angular distributions of Auger electrons have been observed in various ion-atom collision systems [33]. Future work is needed to study the population of the magnetic quantum numbers involved in Coster-Kronig states.

IV. DOUBLE ELECTRON CAPTURE AT LOW INCIDENT ENERGIES

The process of double electron capture becomes important at incident energies of a few keV/u considered here as low energies. Recently, this process has been studied in multicharged ion-atom collisions[34-37], where the role of correlation effects has received a great deal of attention. Stolterfoht et al. [36] have proposed a direct method to verify correlated double capture in slow, multicharged collision systems. The principle of the method is illustrated in Fig. 6 which shows the orbital energies of the O⁶⁺ + He system. In the incident channel two electrons occupy the He 1s orbital which cross the 31 level of oxygen near 5 a.u. There, uncorrelated double capture may occur by two sequential one-electron transitions and, thus, configurations of equivalent (or near equivalent) electrons are created. As the internuclear distance continues to decrease, resonance conditions are created for the correlated double capture process where one electron is transferred into the 2p state and another electron is excited into a Rydberg level nl. It is interesting to note the similarity of this correlation process with the Auger effect. As pointed out by Winter et al.[38], the 2p and nl levels may also be occupied by one-electron transfer into the 31 orbital followed by a correlated transfer-(de)excitation process. Hence, configurations of the nonequivalent electrons 2pnl are created resulting in Coster-Kronig transitions similar to those considered in Fig. 5.

The states attributed to the equivalent and nonequivalent configurations decay by autoionization producing L-Auger and Coster-Kronig electrons, respectively (Fig. 6). The important point of the present method is that these electrons, which are a signature for uncorrelated and correlated double capture, can be distinguished using high-resolution spectroscopy. Fig. 7 shows the result for the measurements of L-Auger and Coster-Kronig electrons in $60-\text{keV}~0^{6+}$ + He collisions. It is found that the resolution is high enough to separate even Rydberg levels with principle quantum numbers up to n = 12 involved in the configuration 2pnl.

From the line intensities of the Coster-Kronig electrons it was concluded that correlation effects play a significant role for double capture in 60-keV O⁶⁺ + He collisions [36]. Also, this conclusion has been drawn for other slow, multicharged collision systems [37,39]. However, the magnitude of the correlation effects has become a matter of controversy [38,40], since Mack and Niehaus [35] and Bordenave-



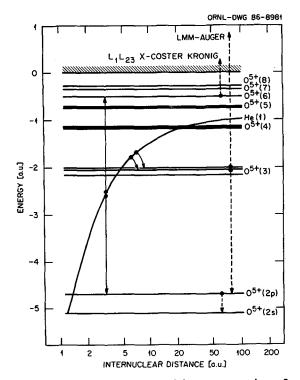


Figure. 6. Diagram of orbital energies for the system O⁶⁺ + He showing that uncorrelated and correlated double capture produce L-Auger and Coster-Kronig electrons, respectively. From Ref. [36].

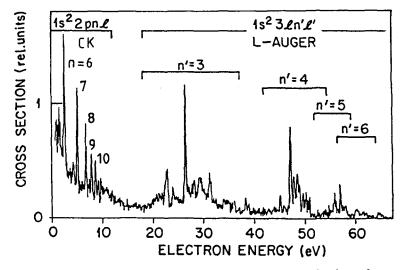


Figure. 7. Spectrum of L_1 -Coster-Kronig (CK) and L-Auger electrons produced in 60-keV O⁶⁺ + He collisions. From Ref. [41].

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Montesquieu et al. [34] have found Coster-Kronig line intensities much smaller (by a factor of about 5) than those observed by Stolterfoht et al. [36] and Mann and Schulte [37]. It was suggested that anisotropic angular distributions may be partially responsible for the observed discrepancies [40], but this supposition could not be substantiated [41]. It appears that future work is needed to clarify the situation. In any case, the method of verifying correlation effects in double capture events by means of the occupation of nonequivalent electron configurations is not questioned.

The Coster-Kronig electrons due to the configuration $1s^2 2pnl$ produced in 0^{6+} + He collisions have recently been measured with improved resolution. The results for n=6 and 7 are shown in Fig. 8 in comparison with theoretical results calculated by Griffin [private commu-

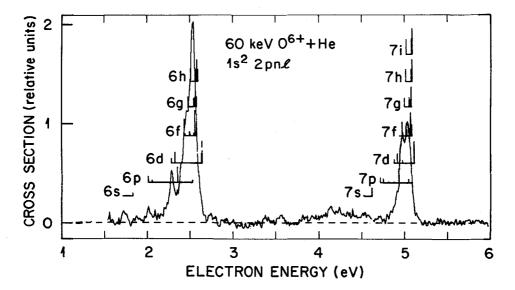


Figure 8. Coster-Kronig electrons attributed to the configuration $1s^2 2pn1$ (with n=6 and 7) produced in 60-keV 0^{6+} + He collisions. From Meyer et al. [41]

nication in [41]) by means of a Dirac-Fock code [42]. The data indicate that the average angular momentum of the Rydberg electrons is 1=3 or 4 which appears to be surprisingly high in comparison with the 1=1 value observed for single electron transfer (Fig. 5).

The high angular momenta observed for the Rydberg electrons are not yet fully understood. It is likely that in the correlated electron capture process, producing the Ryberg electron, not only energy but also angular momentum is exchanged. It is noted that in double electron capture the tranferred electrons receive angular momenta of the same spinning sense. Then, in the correlated transition the electron pushed to the Rydberg state gains angular momentum when the other electron falls to a low-lying (2p) state of relatively small angular momentum. Although this simple picture needs further verification it may be used as a guide to understand the high angular momentum of the Rydberg electron captured in a correlated process.

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