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Citation: AIP Conference Proceedings **680**, 24 (2003); doi: 10.1063/1.1619656
View online: http://dx.doi.org/10.1063/1.1619656
View Table of Contents: http://scitation.aip.org/content/aip/proceeding/aipcp/680?ver=pdfcov
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HTML ABSTRACT + LINKS

DOUBLY-EXCITED KLL STATES FORMED IN TRIPLE ELECTRON CAPTURE

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Abstract. Doubly excited KLL states of C^{3+} were populated by triple electron capture in collisions of fast bare C^{6+} ions with Ar atoms. Measurements of the Auger electron emission at zero degree with respect to the beam direction were used to determine the absolute single differential cross sections for the triple electron capture to all autoionizing KLL states. In addition, independent particle model was utilized to predict the differential cross section for the population of all doubly excited states of carbon. Theoretical cross sections were critically compared with experimental results.

INTRODUCTION

The study of multiple electron capture in collisions of highly charged ions with multielectron atoms or molecules has become a very active area of atomic physics research in the last decade. Transfer processes with more than two active electrons represent a fundamental problem of a many–body dynamic system, thus providing tests for most advanced atomic models. The present understanding of multiple electron transfer is primarily reached in terms of the classical quasimolecular description of the process. To date, however, a unified treatment that accounts for a wide range of energies, has not yet come forth. In particular, the role of electron–electron correlation effects in multielectron capture still remains unclear, raising the demand for a more comprehensive model.

The experimental study of multiple electron capture has been instigated by the introduction of more advanced ion sources during the 1980s, as attested by a number of publications [1, 2, 3, 4, 5, 6]. Projectile charge–change, recoil–ion production and total charge–transfer [1, 2, 3, 4, 7, 8] cross sections were obtained for collisions of Ar^{q+}, I^{q+} and Xe^{q+} projectiles with various targets. The Auger electron spectroscopy was initially implemented in coincidence experiments with charge–state analyzed recoil ions [5, 6] and then subsequently employed in time–of–flight (TOF) triple–coincidence with scattered projectile and target ions to study collisions of O^{7+} and $^{15}N^{7+}$ with Ar gas targets [7, 8]. Although the investigation of multielectron capture has yielded a significant number of articles, experimental studies were primarily concentrated on low velocity collisions ($v << 1$ a.u.). On the other hand, multielectron transfer in collisions of fast ions with atomic targets received much less attention and poses tremendous challenges.

Experimental investigation of multiple electron transfer resulting from fast, highly charged ion–atom collisions is hampered by the rapid fall off of capture probabilities with increasing velocity of the ion. To the best of our knowledge, the study of true triple electron capture in fast ($v > 1$ a.u.) collisions has not yet been attempted experimentally. Meanwhile, the problem of multiple electron transfer at high impact velocities becomes more appealing, instigated by the dominant role of the process in populating lower–lying multiexcited states, recently a very dynamic area of atomic research.

In this work, high resolution zero–degree Auger electron projectile spectroscopy has been used to study triple electron capture to doubly excited KLL states of carbon in collisions of fast (v = 4.5–6.6 a.u.) C^{6+} ions with Ar gas targets. Measured absolute single differential cross sections (SDCS) were used to test the predictions of the independent particle model (IPM), in which the simultaneous capture of all three target electrons is assumed. Single electron capture probabilities, employed by the model, were calculated using the two–center semiclassical close–coupling method [9], based on an atomic orbital expansion [10]. Model calculations were found to be in good agreement with the experimental data.

EXPERIMENTAL PROCEDURE

The experiments were performed in the J. R. Macdonald Laboratory at Kansas State University, using the 7 MV

CP680, *Application of Accelerators in Research and Industry: 17th Int'l. Conference*, edited by J. L. Duggan and I. L. Morgan
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EN tandem Van de Graaff accelerator. The C^{6+} beam was magnetically selected after colliding the primary Li–like C^{3+} beam with a 5 $\mu g/cm^2$ carbon foil, and then focused into a 5 cm long differentially pumped gas cell. In collisions with Ar atoms, doubly excited KLL states of C^{3+} ions were populated. These states decay dominantly by emitting Auger electrons at energies corresponding to the $1s^2\,{}^1S$ ground state transition. The Auger electrons, emitted in the forward direction were detected with a zero–degree hemispherical spectrograph [11], capable of analyzing electrons in an energy bandwidth of about 20%, which significantly facilitates experimental measurements of low intensity processes, such as triple electron capture.

The Ar gas target was maintained at constant pressure, utilizing a feedback–controlled baratron gauge. The target pressure was set at 20 mTorr that corresponds to the middle of the linear interval of the yield–pressure curve. A potential source of error is the possible beam contamination with C^{5+} and C^{4+} ions. In this case, the KLL states of C^{3+} may be populated by single or double electron capture from the residual gas. Therefore, the effect of the vacuum pressure on the KLL Auger electron emission was studied. The bare C^{6+} ions travel a distance of about 2 m after the analyzing magnet at a residual gas pressure of 1×10^{-6} Torr before entering the gas cell. To evaluate the contribution of a possible contamination, the vacuum pressure between the analyzing magnet and the gas cell was increased up to 1×10^{-4} Torr, and the measurement of the Auger electron emission from KLL lines was repeated. For both vacuum pressure settings the integrated KLL Auger electron emission was found to be the same. Therefore, the formation of C^{5+} or C^{4+} ions between the analyzing magnet and the gas cell that contribute to the population of KLL states is negligible. Consequently, the above study clearly showed that the observed doubly excited states of C^{3+} are formed in a single collision of C^{6+} ions with Ar.

THE INDEPENDENT PARTICLE MODEL

In the case of high impact velocities, if several electrons are transferred during the collision, the projectile's energy levels will not fully readjust to reflect the dynamics of changes due to the insufficient interaction time. Therefore, the projectile charge will be only partly screened during the collision by captured electrons. In order for the interaction time to be considered small, the impact velocity has to exceed the projectile electron orbital velocity of a state to which the target electron is transferred. In the investigated system, 6–13 MeV C^{6+}+ Ar, impact velocities (4.5 – 6.6 a.u.) are greater than K (4.2 a.u.)

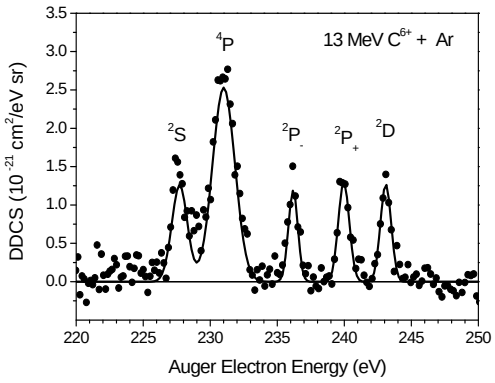

FIGURE 1. zero–degree Auger electron DDCS spectra for the 13 MeV C^{6+} + Ar collision system. KLL states of C^{3+}, populated by triple electron capture are shown

and L (2.1 a.u.) shell velocities of the projectile. The extended classical overbarrier model cannot be employed in this case since the collision time is not sufficient for target electrons to be molecularized. Instead, captured electrons should rather be treated as independent particles, captured by an unscreened potential. The total triple electron capture probability, P, is then a statistical product of single electron capture probabilities. The detailed calculation of P from single electron capture probabilities is presented at the end of this section.

Since the triple electron capture process is expected to have a very low cross section at high collision energies, the collision system should be optimized to increase the electron transfer probabilities. This can be achieved by matching the electron orbital velocities of the target and the projectile. For the investigated collision system the binding energy of the Ar L shell is located between K and L shell energy levels of C^{5+}. Thus, the electron capture probability from the Ar L shell to the K and L shells of C^{5+} is enhanced. At the same time, the contribution from K and M shells to the total capture is significantly reduced.

The L shell of Ar consists of three energetically resolved sublevels (nlm), recognized in the non–relativistic close coupling code as: 2s0, 2p0 and 2p$\pm$1, filled with 2,2, and 4 electrons each. Out of these 8 electrons, one is captured to the K shell (1s0) of C^{5+} and two others are captured to the L shell (2s0, 2p0, 2p$\pm$1). If C^j is the number of possible ways for any three electrons from the L shell of Ar in a certain electron configuration j to be captured to the KLL state in carbon, then the total triple electron capture probability can be expressed as

$$P(b) = \sum_s C^s \prod_{i=1}^{3} P_i^{kj}(b) \prod_{i=4}^{8} Q_i^j(b), \qquad (1)$$

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where s is the sum over all possible statistical configurations. P_i^{kj} is a probability for the i^{th} electron of Ar in the j level (2s0, 2p0, 2p±1) to be transferred to the k level (K or L shells) of C^{5+}. Q_i^j is a probability for the i^{th} electron of Ar in the j level not to be transferred to either the K or L shell of C^{5+}.

RESULTS AND DISCUSSION

Fig. 1 shows the Auger electron spectrum measured in collisions of 13 MeV C^{6+} ions with Ar gas targets. The formation of the $C^{3+}(1s2l2l')$ doubly excited states, namely $1s2s^2\,^2S$, $1s2s2p\,^4P$, $[1s(2s2p)^3P]\,^2P_-$, $[1s(2s2p)^1P]\,^2P_+$ and $1s2p^2\,^2P\,^2D$, which Auger decay to the $C^{4+}(1s^2)$ ground state, is prominent. All the above states were populated by triple capture, which allows for the population of all possible Li–like doubly excited states. Therefore the ^2S, ^2P and ^4P states corresponding to the $1s2p^2$ configuration are also populated. However, the ^2P and ^4P states are not allowed to Auger decay to the $C^{4+}(1s^2)$ ground state due to parity conservation considerations, while the ^2S state has a very low Auger decay rate to be observed.

First, in order to correct the experimental cross sections for the undetectable in this study x–ray decay channel, the Auger yields, Y, for the observed transitions were evaluated from theoretical autoionization and fluorescence rates [13, 14, 15]. Also, the decay path of the metastable $1s2s2p\,^4P$ state was taken into account. It is a metastable state, with lifetimes of 25 ns for J=5/2, 9.1 ns for J=3/2 and 3.0 ns for J=1/2 [16, 14], which are in general comparable to the ion's time of flight (TOF) in the gas cell (e.g. in the case of 13 MeV C^{6+}, TOF = 3.5 ns). However, due to the spectrograph geometry, electrons emitted in the area between the gas cell and the analyser entrance will still be recorded, thus enhancing the electron yield of the state and also decreasing the energy resolution of the line, as it is seen in Fig. 1. The correct electron yield for the $1s2s2p\,^4P$ state was obtained after biasing the gas cell by a voltage of -30 V, which energetically separates the yield fractions originating from inside and outside the gas cell area. The total electron yield was then calculated from the available ion's time of flight and the exponential decay rate of the state [17]. The above corrections were included in the same factor, R, for each collision energy. Finally, the four doubly excited states ^2D, ^2P, ^4P and ^2S corresponding to the same electron configuration, $1s2p^2$, were populated by triple capture according to their statistical weight. However, since only the ^2D state is observed as discussed earlier, the production of other states was accounted for as shown by the inverse term in Eq. (2).

Under the above considerations the single differential cross section for the triple electron capture to the KLL states of C^{3+} is given by:

$$\left(\frac{d\sigma}{d\Omega}\right) = \sum_i \left(\frac{d\sigma}{d\Omega}\right)^i = \sum_i Z^i_{exp}\,\frac{1}{Y^i_A}\,\frac{1}{R^i}\left[\frac{2J+1}{\sum_J (2J+1)}\right]^{-1} \qquad (2)$$

where i is the sum over the observed KLL doubly excited states. In Table I, the discussed correction factors are presented for the 13 MeV C^{6+} + Ar collision system.

The triple electron capture probability P(b) was obtained within the IPM as given by Eq. (1). The total triple electron capture cross section, σ, was then determined after integrating the probability, $P(b)$, over the impact parameter.

In Fig. 2 the experimental zero degree triple electron capture cross sections to KLL states measured for 6, 9, and 13 MeV C^{6+} + Ar collisions are compared with the independent particle model calculations for the corresponding collision energies. The experimental error bars are calculated by taking the quadrature sum of the statistical and absolute uncertainties. The latter incorporates mainly the electron detection efficiency of the spectrograph.

It is seen clearly from Fig. 2 that the independent particle model calculations systematically overestimate the experimental triple capture SDCS by a factor of about 1.5. The predicted collision energy dependence of the process is in good agreement with the experimental one. Considering the fact that theoretical single electron capture amplitudes contain certain error not shown in Fig. 2,

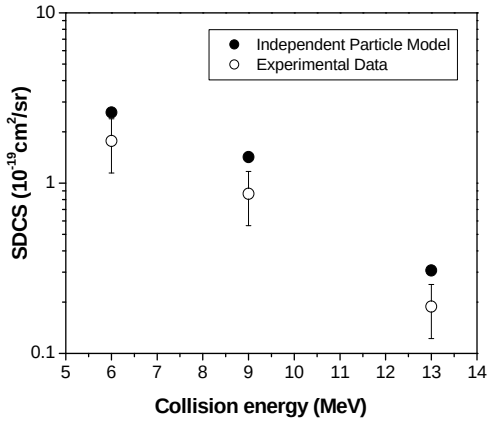

FIGURE 2. Absolute SDCS measurements of triple electron capture to the C^{3+} KLL states, populated in fast collisions of bare carbon ions with Ar targets (open circles). IPM calculations (solid circles) are seen to be in fairly good agreement with the data.

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TABLE 1. All data are referred in the case of the 13 MeV C^{6+} + Ar collision system. Both experimental and calculated SDCS are in units of $10^{-21} cm^2/sr$. Transition energies are given relative to the $C^{4+}(1s^2)$ ground state in units of eV.

Intermediate State	Transition Energy	Integrated Peak Area	Y	R	Statistical Weight	$SDCS_{exp}$	$SDCS_{calc}$
$1s2s^2\,^2S$	227.5[1]	1.8±0.6	≃1[1]	1.00	1	1.8±0.6	
$1s2s2p\,^4P$	229.7[1]	5.3±1.6	≃1[1]	0.51	1	10.5±3.2	
$1s2s2p\,^2P_-$	235.9[1]	1.1±0.5	0.92[1]	1.00	1	1.1±0.3	
$1s2s2p\,^2P_+$	239.3[1]	1.3±0.4	≃1[1]	1.00	1	1.3±0.4	
$1s2p^2\,^2D$	242.3[1]	1.3±0.4	≃1	1.00	3	3.8±1.1	
					SDCS(sum)	18.6±0.6	30.6

* From Ref. [18, 14, 13]

the comparison gives enough evidence to conclude that the independent particle model provides an adequate description of the triple electron capture in fast ion–atom collisions. In view of that, the screening dynamics of the incident projectile ion by captured target electrons can be understood through simultaneous electron transfer, in which the energy levels of the projectile do not completely readjust during the course of the collision due to the insufficient interaction time. Therefore, if the impact velocity exceeds the projectile electron orbital velocities, the ion nucleus remains essentially unscreened during the collision. In addition, since the independent particle model does not account for the electron–electron correlation explicitly, the role of the electron–electron correlation in triple electron capture by fast ions is believed to be insignificant.

CONCLUSIONS

Zero–degree Auger electron spectroscopy has been used to determine the absolute SDCSs for triple electron capture to doubly excited KLL states of carbon in collisions of fast (v = 4.5–6.6 a.u.) C^{6+} ions with Ar atoms. Experimental data were used to test the predictions of the independent particle model that utilizes single electron capture probabilities calculated using the two–center semiclassical close–coupling method. Model calculations were found to be in fairly good agreement with the experiment, signifying that the IPM provides an adequate description of the triple electron capture in fast ion–atom collisions. In particular, it was concluded that target electrons are captured simultaneously by an essentially unscreened potential of the projectile, reflecting the fact that the collision time is too small for the ion energy levels to readjust. Thus, the effects of electron–electron correlation do not appear to be significant in multiple electron transfer at high collision velocities.

ACKNOWLEDGMENTS

This work was supported by the Division of Chemical Sciences, Geosciences and Biosciences, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

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