Multielectron Processes in Heavy Ion–Atom Collisions at Intermediate Velocity

D. Vernhet,[1] L. Adoui,[2] J. P. Rozet,[1] K. Wohrer,[1] A. Chetioui,[1] A. Cassimi,[2] J. P. Grandin,[2] J. M. Ramillon,[2]
M. Cornille,[3] and C. Stephan[4]
[1]*GPS (CNRS UMR 75-88), Universités Paris 7 et 6, 2 Place Jussieu, 75251 Paris Cedex 05, France*
[2]*CIRIL (UMR N°11 CEA/CNRS), Rue Claude Bloch, B.P. 5133, 14070 Caen Cedex 5, France*
[3]*DARC, Observatoire de Paris Meudon, Place Jansen, 92195 Meudon Cedex, France*
[4]*NIM, IPN, Université Paris 11, B.P. N°1, 91405 Orsay Cedex, France*

(Received 3 February 1997)

Using high resolution x-ray spectroscopy, we have measured projectile electron single and multiple cross sections when a two-electron Ar^{16+} ion collides with neutral target atoms. For a fixed impact velocity ($v_p = 23$ a.u.), but using various targets from He to Xe, a range from the perturbative regime to the strong interaction regime has been investigated. Double excitation cross sections are found to be well reproduced by an independent electron model. First measurements of capture-ionization cross sections are also reported and show the importance of this often-neglected process. [S0031-9007(97)04505-5]

PACS numbers: 34.50.Fa, 34.10.+x, 34.90.+q

In ion-matter interactions where highly charged ions collide with neutral targets, the intermediate velocity regime is of particular interest. In this regime, the primary processes, namely, excitation, ionization, and capture, have cross sections of the same order of magnitude; coupling between these channels is important. Thus, complete calculations leading to a unified treatment of those different processes, including an explicit representation of the continuum, are very difficult to handle [1]. Nevertheless, in this velocity regime, the stopping power is nearly maximum; for applied purposes, the understanding of mechanisms accounting for radiation damage in materials is important. For example, this is the first step needed to achieve a good knowledge of the parameters governing lethal effects of ions colliding with living cells [2], or track diameters in crystals [3].

Furthermore, as will be shown in this Letter, cross sections of multiple processes may be as high as single ones when the perturbative partner (the target) is heavier than the perturbed one (the projectile). Until today, very few measurements have been made and then just for light atoms at high velocities [4–8]. Theoretical interpretation remains controversial in terms of correlation effects needed to correct the independent electron model (IEM). A quantitative evaluation of electron correlation during the collision (the so-called intermediate state correlation) requires a complete treatment. Theoretical work has been limited mainly to the multiple ionization and excitation of He [9–14]. Furthermore, no one has investigated heliumlike systems for double excitation, but for ionization correlation has been shown to fall off as $1/Z_p$ [15].

In this Letter, we have measured cross sections of multiple processes for a two-electron ion in collision with neutral targets at intermediate velocity: Ar^{16+} on He, N_2, Ne, Ar, Kr, and Xe at $v_p = 23$ a.u. Thus we investigate a range of interactions spanning the perturbative regime to the strong interaction regime. More precisely, in this Letter we report measurements of capture-ionization (CI), capture-excitation (CE), capture-excitation-ionization (CEI), double excitation (DE) and excitation-ionization (EI) cross sections as well as a single excitation (SE) one. All of these processes are presented in Table I. Note that the CI process is competitive to the SE one; it populates the same final configurations. We note, however, that SE will give rise to just singlet states, while CI will populate triplet states as well. The same argument holds for DE versus CEI. A spin-selective experiment allows us to differentiate between these various multiple-electron processes. We use the fact that each populated state of the projectile, as it decays, emits x rays with slightly different energies. In the case of an Ar^{16+} ion, it is necessary to achieve a resolution of about 10 eV to distinguish between all of these different ~3 keV transitions. To fulfill these conditions, we have, in this Letter, used very high resolution and high transmission crystal spectroscopy.

The experiment has been performed at the Sortie Moyenne Energie facility at Grand Accélérateur National d'Ions Lourds in Caen. A high intensity beam ($1\ \mu\mathrm{A}_e$) of Ar^{16+} at 13.6 MeV/u was directed at various atomic targets confined in an open gaseous cell. A metastable fraction was present in the incoming Ar^{16+} beam [$1s2s\,^3S_1$ state]; this has to be taken into account in the data analysis. The fraction was found to be $(3.2 \pm 0.5) \times 10^{-3}$ at the entrance of the collision area [16]. The specially designed spectrometer used was composed of a flat mosaic graphite crystal and a localization chamber. The spectrometer was placed at 30° with respect to the beam axis and used in a vertical geometry. This allows a first order compensation of Doppler broadening and reduces the sensitivity to any polarization effect in the x-ray emission

0031-9007/97/79(19)/3625(4)$10.00

TABLE I. Review of all the processes studied here: single excitation (SE), double excitation (DE), excitation-ionization (EI), capture-ionization (CI), capture-excitation-ionization (CEI), capture-excitation (CE). The quoted final states are those observed experimentally.

	Processes involving just projectile electrons		Processes also involving target electrons	
Initial state	Processes	Final states	Processes	Final states
$Ar^{16+}(1s^2\,^1S_0) + X$	SE	$Ar^{16+}(1s2p, 1s3p\,^1P) + X$	CI	$Ar^{16+}(1s2p\,^1P$ and $^3P) + X^+$
	DE	$Ar^{16+}(2l2l'\,^1L) + X$	CEI	$Ar^{16+}(2l2l'\,^1L$ and $^3L) + X^+$
	EI	$Ar^{17+}(2p, 3p) + X$	CE	$Ar^{15+}(1s2l2l'\,^2L$ and $^4L) + X^+$

pattern. The global efficiency was accurately determined by comparison of hydrogenlike and heliumlike line intensities, in the case of a He target, with a well calibrated Si(Li) detector. It amounted to 1.3×10^{-7} ($\pm 15\%$) and was nearly constant in a range of 260 eV around 3.7 keV. The resolving power was 1.4×10^{-3}.

Spectra we have obtained for all of the excited states studied (up to $n = 5$) are presented in Fig. 1 in the case of N_2 and Xe targets. One can see from a direct analysis of these spectra as a function of target atomic number that (i) the $1snp\,^1P_1 \rightarrow 1s^2\,^1S_0$ transitions, mainly due to a SE process, keep the same relative intensity; (ii) on the other hand, the intensities of all of the lines due to multiple processes (filled in black) increase very rapidly with Z_t when compared to the SE process. We have extracted cross sections for each of these processes and the results are listed in Table II. We give here a brief survey of the procedure used to deduce the cross sections [17]. We use atomic structure calculations, done by one of us (M. C.), for branching ratios and configuration mixing coefficients. In the data analysis, single excitation from the metastable fraction of the beam has been taken into account. Cross sections of CI and CEI processes are deduced from the observed triplet state populations (respectively, $1s2p$ and $2l2l'$). Their contribution to the population of singlet states is then extracted, assuming a $2J + 1$ statistical population of $^{2S+1}L_J$ states. SE and DE cross sections are finally determined from the (remaining) populations in singlet states. Cross sections of CE and EI processes are derived from the observed $1s2l2l'$ and

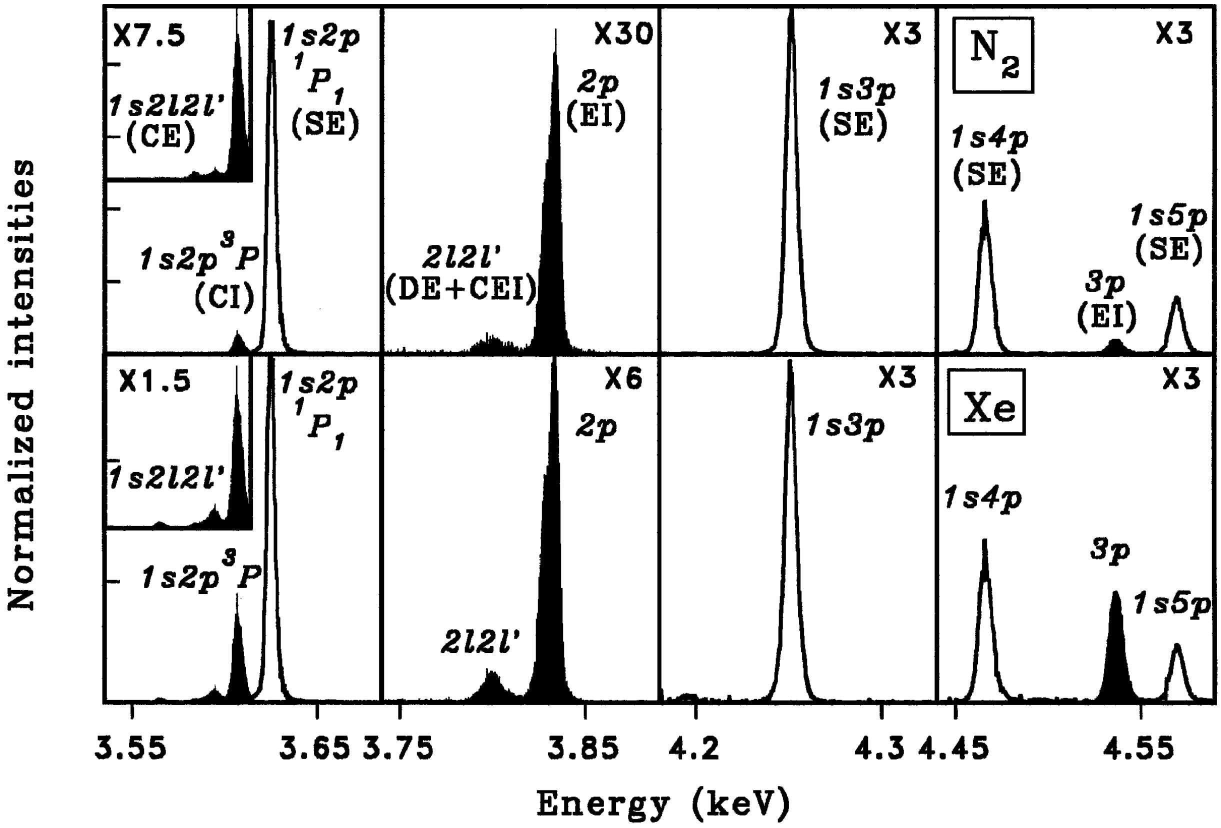

FIG. 1. X-ray spectra recorded with the crystal spectrometer in the case of Ar^{16+} colliding with N_2 and Xe neutral targets at $v_p = 23$ a.u. For each target, intensity of the transitions have been normalized to $1s2p\,^1P_1 \rightarrow 1s^2\,^1S_0$—the spectra recorded for the N_2 and Xe targets have independent normalization. The indicated processes are those responsible mainly for the emission of the corresponding transitions. The transitions filled in black are the lines due mainly to multiple processes.

TABLE II. Experimental cross sections ($\times 10^{-21}$ cm^2) for SE, DE, EI, CI, CEI, and CE processes (as quoted in Table I) in $\mathrm{Ar}^{16+} \rightarrow Z_t$ neutral targets at $v_p = 23$ a.u. Reported values include cascade contributions. In the case of CE, cross sections are extracted within a factor of 2. For the He target, multiple processes are found to be negligible.

Cross sections (10^{-21} cm^2)	Targets					
	He	N_2	Ne	Ar	Kr	Xe
$\sigma_{\mathrm{SE}}(1s^2 \rightarrow 1s2p)$	11.2	102.5	178	293	488	484
	(±15%)	(±20%)	(±21%)	(±24%)	(±33%)	(±29%)
$\sigma_{\mathrm{SE}}(1s^2 \rightarrow 1s3p)$	⋯	29.14	⋯	86.64	⋯	159.77
		(±22%)	⋯	(±22%)	⋯	(±23%)
$\sigma_{\mathrm{DE}}(1s^2 \rightarrow 2s2p + 2p^2)$	⋯	0.7	2.2	6	15	14
		(±30%)	(±30%)	(±28%)	(±30%)	(±38%)
$\sigma_{\mathrm{EI}}(1s^2 \rightarrow 2p)$	⋯	4	10	37	127	119.5
		(±24%)	(±27%)	(±31%)	(±29%)	(±33%)
$\sigma_{\mathrm{EI}}(1s^2 \rightarrow 3p)$	⋯	1.2	⋯	12.5	⋯	46.5
		(±25%)	⋯	(±31%)	⋯	(±33%)
$\sigma_{\mathrm{CI}}(1s^2 \rightarrow 1s2p)$	⋯	4	15	59	216	260
		(±100%)	(±45%)	(±27%)	(±27%)	(±29%)
$\sigma_{\mathrm{CEI}}(1s^2 \rightarrow 2s2p + 2p^2)$	⋯	0.15	0.5	3.6	17	19
		(±51%)	(±37%)	(±26%)	(±25%)	(±28%)
$\sigma_{\mathrm{CE}}(1s^2 \rightarrow 1s2l2l')$	⋯	1	4	19	51	71

$2p, 3p$ line intensities. The quoted error bars (Table II) include uncertainties of all experimental parameters; there are important contributions due to spectrometer efficiency, statistics, and spectra deconvolution. In the following, we will restrict our analysis to the $n = 2$ level, except for the EI process which has been measured as well for $n = 3$.

As the target atomic number increases, all of the processes including both target electrons—via capture channels—and projectile ones increase more rapidly than those involving just projectile electrons. For example, the CI cross section, negligible for the He target, reaches 54% of the SE cross section for Xe. Furthermore, even in the case of the EI process, involving only projectile electrons, the cross section is found to be as large as 25% of the SE for the heavier target (for $n = 2$ as well as for $n = 3$). These measurements, made for a heavy ion, demonstrate the importance of these often-neglected multiple processes at intermediate velocity.

For such systems, calculations taking into account the coupling between all of the possible collisional channels are not yet available. However, in the case of DE, a tentative analysis can be made in the frame of the independent electron model since, in the case of heavy ions, electron correlation in the initial state can be neglected. It is well known that the first Born approximation—which scales as Z_t^2—is not valid when the perturbation strength becomes too large (typically for $Z_t \geq 8$ in the present case). From our single excitation measurements, it is possible to extract a coefficient depending on the target atomic number, $C_E(Z_t)_{\mathrm{expt}}$, and defined as the ratio between our data and a Z_t^2-scaling law. One can also derive a coefficient $C_E(Z_t)_J$ from a similar comparison between the recent Janev calculations [18] and the Z_t^2-scaling law. This approach, based upon the dipolar law (for the perturbative regime) or the adiabaticity of the collision (for the strong interaction regime), provides an analytic fit of the experimental data in the case of single excitation of light ions. The comparison between $C_E(Z_t)_{\mathrm{expt}}$ and $C_E(Z_t)_J$, reported in Table III, shows that the Janev calculations reproduce very well the relative evolution of the single excitation cross section, even for a heavy ion, over all of the perturbation ranges studied (details will be given in a forthcoming paper).

For the double excitation process, we assume that the impact parameter dependent probability for a given Z_t, $b \times P(Z_t, b)$ can be deduced from $b \times P\ (Z_t = 1, b)$ by using the expression $P(Z_t, b) \propto Z_t^2 \times C_E(Z_t) \times P\ (Z_t = 1, b')$ taken from the recent $b \times P(b)$ MO's calculations for single excitation [19]. Then, in the IEM, one can expect DE cross sections to scale as $\sigma_{\mathrm{DE}} \propto C_E(Z_t) \times Z_t^2 \times \sigma_{\mathrm{SE}}$ for a given projectile Z_p, at least up to a symmetrical collision. We have reported in Fig. 2 the experimental evolution of DE over SE cross sections ratio

TABLE III. Experimental $C_E(Z_t)_{\mathrm{expt}}$ and theoretical $C_E(Z_t)_J$ single excitation coefficients (see text). $C_E(Z_t)_J$ coefficients have been normalized to 1 for the He target.

	Targets					
C_E	He	N_2	Ne	Ar	Kr	Xe
$C_E(Z_t)_{\mathrm{expt}}$	0.97	0.73	0.62	0.31	0.13	0.06
	(±15%)	(±20%)	(±21%)	(±24%)	(±33%)	(±29%)
$C_E(Z_t)_J$	1.00	0.63	0.51	0.32	0.12	0.05

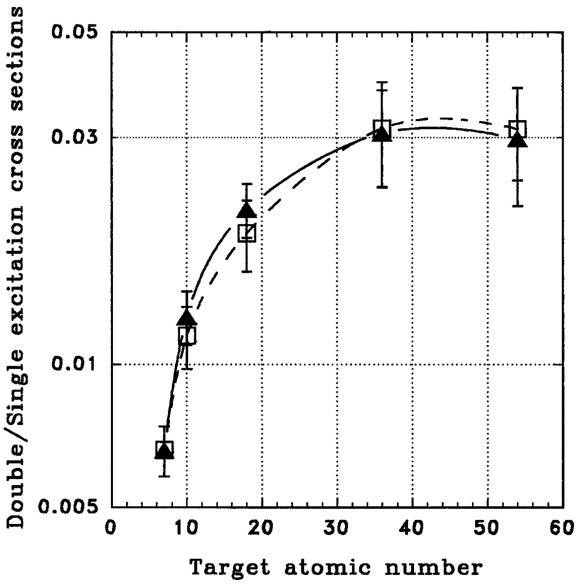

FIG. 2. Experimental ratio of double to single excitation cross sections (full triangles and full line) and comparison with the IEM scaling law $C_E(Z_t)_{\mathrm{expt}} \times Z_t^2$ (open squares and dashed line).

and the $C_E(Z_t)_{\mathrm{expt}} \times Z_t^2$ scaling law (normalized to N_2). The very good agreement obtained shows the validity of a simple independent electron approximation for the treatment of double excitation for such a heavy ion (Ar) at intermediate velocity.

Our measurements of EI cross sections in $2p$ and $3p$ final states are presented in Fig. 3. The cross sections change as a function of the target atomic number in a similar way for these two final states. The single excitation cross sections in $n = 2$ and 3 (see Table II) have similar behaviors, thus the result for EI is again consistent with an independent electron model.

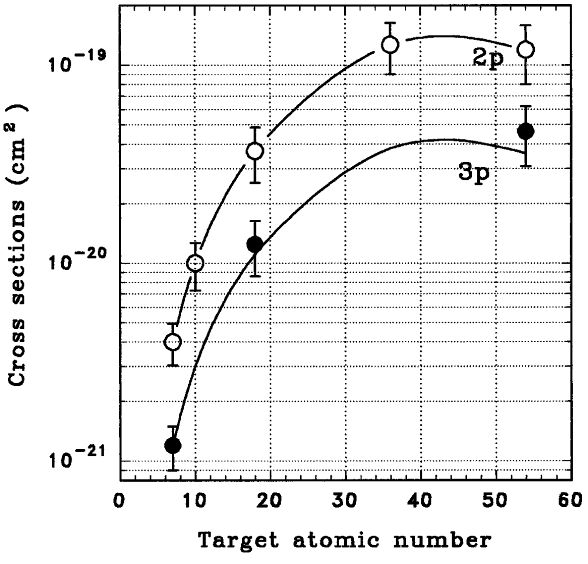

FIG. 3. Excitation-ionization (EI) cross sections as a function of Z_t for $2p$ and $3p$ final states of Ar^{17+}. The fit made for the $2p$ final state is scaled for the $3p$ final state case, with a normalization to the N_2 target.

In this Letter, we have measured cross sections of single excitation and multiple processes from the perturbative regime to the strong interaction regime, varying the asymmetry of the collision system Z_p/Z_t from 9 to 0.33. The very first measurements of the multiple processes for a heavy ion are presented, and show their large importance in this velocity range. The theoretical understanding requires a complete treatment including the n bodies involved in these collision systems. In particular, all of the multiple processes involving capture warrant a more thoroughly developed theoretical treatment; here, competing processes (e.g., CI and SE or CEI and DE) can be separated experimentally via the spin signature of the observed transitions. In the present case, as electron correlations in the initial state can be neglected, a simple independent electron treatment, based upon experimental single excitation cross sections, appears to be successful for predicting the observed DE cross sections. This result is also consistent with our measurements of EI cross sections. The experimental conditions will be described in detail in a forthcoming paper.

[1] H. J. Lüdde *et al.*, J. Phys. B **28**, 4101 (1995).
[2] A. Chetioui *et al.*, Adv. Space Res. **18**, 15 (1996).
[3] A. Meftah *et al.*, Phys. Rev. B **48**, 920 (1993).
[4] J. O. P. Pedersen and P. Hvelplund, Phys. Rev. Lett. **62**, 2373 (1989).
[5] J. O. P. Pedersen and F. Folkmann, J. Phys. B **23**, 441 (1990).
[6] N. Stolterfoht *et al.*, Phys. Rev. A **51**, 350 (1995).
[7] B. Skogvall *et al.*, Phys. Rev. A **51**, R4321 (1995).
[8] A. Bordenave-Montesquieu *et al.*, J. Phys. B **28**, 653 (1995).
[9] J. F. Reading and A. L. Ford, Phys. Rev. Lett. **58**, 543 (1987); J. Phys. B **20**, 3747 (1987).
[10] F. Martin and A. Salin, J. Phys. B **28**, 639 (1995).
[11] F. Martin and A. Salin, Phys. Rev. Lett. **76**, 1437 (1996).
[12] F. Martin and A. Salin, Phys. Rev. A **54**, 3990 (1996).
[13] F. Martin and A. Salin, Phys. Rev. A **55**, 2004 (1997).
[14] J. F. Reading, T. Bronk, and A. L. Ford, J. Phys. B **29**, 6075 (1996).
[15] A. L. Ford and J. F. Reading, J. Phys. B **23**, 3131 (1990).
[16] L. Adoui *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **98**, 312 (1995).
[17] L. Adoui, Thèse de Doctorat, Université Pierre et Marie Curie, Paris, 1995 (unpublished).
[18] R. K. Janev, Phys. Rev. A **53**, 219 (1996).
[19] A. Macias *et al.*, Phys. Rev. A **53**, 2869 (1996).