PHYSICAL REVIEW A VOLUME 55, NUMBER 6 JUNE 1997

Doubly differential electron emission for transfer ionization in 100-keV H^+ on Ar

G. Bernardi, P. Focke, and W. Meckbach
Centro Atómico Bariloche and Instituto Balseiro, Comisión Nacional de Energia Atómica, 8400 Bariloche, Argentina

(Received 3 March 1997)

We have measured doubly differential electron emission for transfer ionization produced by 100-keV H^+ impact on an Ar target. Data were obtained by measuring coincidences between electrons of selected energy and angle of emission with outgoing H^0 projectiles. The measurements covered emission angles in the range of $\theta=0°$ to 160° and energies starting from 10 eV. We present a detailed discussion on the experimental conditions used and give a tentative interpretation of the main features observed. [S1050-2947(97)51006-7]

PACS number(s): 34.50.Fa, 34.70.+e, 34.90.+q

The ionization of atoms by ion impact has been the subject of extensive investigations for a long time. Although total cross sections give valuable information for theoretical discussions and a variety of applications, more detailed knowledge of the collision dynamics is obtained from measurements of the differential cross sections for electron emission.

The simplest process, and in many cases the main contribution to ionization, is the emission of one target electron. Accordingly, a great amount of experimental and theoretical work has been dedicated to collisions with one active electron. Experimental data, as well as theoretical discussions, are scarcer in the case of multielectron processes, such as direct double ionization and transfer ionization, for which mainly total cross sections are available. A main goal of the study of these processes is to elucidate the role of the electron correlation, or to what extent these processes can be described by an independent electron model. Much interest was devoted to the study of direct (without charge exchange) double ionization of He by charged projectiles and photon impact, recently reviewed by McGuire *et al.* [1]. Two primary mechanisms leading to double ionization have been proposed for charged projectiles. At high collision velocities, a one-step or shake-off mechanism dominates: The projectile ionizes the target; then a second electron is emitted following rearrangement in the remaining target ion. At lower collision velocities, double ionization is produced predominantly by a two-step mechanism, in which the projectile interacts with each of the target electrons.

We concentrate here on a simple case of transfer ionization (TI), produced by 100-keV H^+ impact on Ar, for which the final state essentially comprises a neutral H^0 atom and a free electron. This process predominantly involves two M-shell target electrons [2], contributions from inner shells being negligible. Although the transfer ionization process has not been as fully discussed as direct double ionization [1] for the charge ($Z=1$) and impact velocity ($v_p=2$ a.u.) used in the present measurements, a two-step mechanism could be expected to dominate. A reasonable agreement with total cross sections for transfer ionization at intermediate collision energies was obtained in independent-electron-model calculations by Chatterjee, Prasad, and Roy [3] and Shingal and Lin [4]. The present measurement of the doubly differential electron emission for transfer ionization gives additional and more detailed information that is useful in discussing this subject.

Besides the available data, concerned with total cross sections, experimental work has been done to study the contribution of transfer ionization to the differential electron emission at velocities close to that of the ionic projectile, the so-called ‘‘cusp electrons’’ [5–11]. Other measurements were performed at emission angles $\theta\cong90°$, looking for the contribution of the Thomas double-scattering mechanism in the transfer ionization process [12–14].

Our equipment for the measurements of doubly differential electron emission has been described in detailed elsewhere [15]. In the present measurements, the 100-keV proton beam was collimated to 0.25×0.3 mm^2 before entering the collision chamber. The target consisted of an effusive Ar source, localized at the object focus of a rotatable cylindrical mirror spectrometer. A half-angle of the acceptance cone of 2° and an energy resolution of 6% was used. An electrostatic-charge-state analyzer separated the neutral and charged beam components exiting the collision chamber. Neutral atoms (H^0) were detected with a projectile-secondary-electron converter [16], provided with a high-count-rate channeltron detector. Standard electronics were used to obtain coincidences between electrons of selected energy and angle of emission and the neutralized H^0 projectiles. Total electron counts (start pulses) and total H^0 counts (stop pulses) were also registered.

As pointed out by Sarkadi *et al.* [9], Závodszky *et al.* [10], and Víkor *et al.* [11], a main source of error in the measurement of the relatively small transfer ionization cross section is due to double collision events. We analyze this problem as follows. The H^+ beam that arrives at the target contains a small contamination of H^0, produced by electron capture in the beam transport line and in the first part of the collision chamber, before the target. Furthermore, the projectiles (H^+,H^0) emerging from the target can have a charge-exchange collision in the path up to the charge-state analyzer. A true transfer ionization event is produced when an incident H^+ captures a target electron, an additional electron being emitted in the same single collision. Contamination is due to other collision events that lead to the same final products. (a) A H^+ can produce an electron by ionization and, in a subsequent collision (in the target or afterward), be neutralized. (b) A capture process may occur, followed in a second collision by an ionization of an Ar atom, both in the target. (c) In the case of an incident H^0, a first collision in the target can produce a free electron by loss of the bound electron;

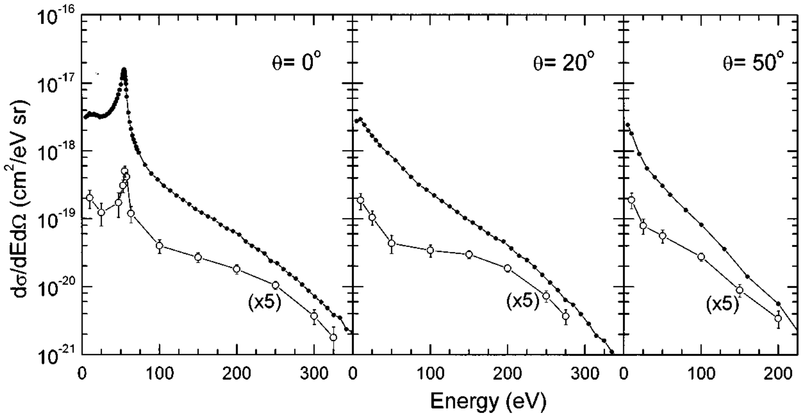

FIG. 1. Doubly differential cross sections. Open circles: transfer ionization (coincidence spectra). Dots: electron emission (singles spectra); $\theta=0°$ and 20°, present data; $\theta=50°$, data from Ref. [18]. Lines are guides for the eye.

then a second collision (in the target or afterward) regenerates the H^0 by capture. (d) In a single collision, a H^0 projectile can produce an ''untrue transfer ionization event'' by ionization of an Ar atom in which the H^0 does not lose its bound electron.

We note that double collision processes can be important due to the large cross sections involved, as opposed to a transfer ionization cross section. In our case of 100-keV H^+ on Ar, the cross sections for ionization of one and two electrons (without charge transfer) are $\sigma_1^{11}=4\times10^{-16}$ cm^2 and $\sigma_2^{11}=6\times10^{-17}$ cm^2, respectively. The total capture cross section is $\sigma^{10}=1.2\times10^{-16}$ cm^2, whereas for transfer ionization with two active electrons it is $\sigma_{TI}\equiv\sigma_2^{10}=3\times10^{-17}$ cm^2. These data and the notation are from Ref. [2]. Cross sections for three-electron processes are considerably smaller. For an incident H^0 projectile, the loss cross section is $\sigma^{01}\cong5\times10^{-16}$ cm^2 [17].

Contamination from double collision events leads to electron count rates with a quadratic dependence on target thickness. The transfer ionization process that we want to measure has, up to a certain maximum target thickness, a linear dependence. Then, in principle, by reducing the target thickness, this contamination could be reduced to a negligible value. In the case of contamination due to single-collision ionization of an Ar atom by a H^0 projectile, special care must be taken. Even for small Ar pressure in the collision chamber, there is a fraction F_0 of H^0 in the beam, produced in the beam transport line before the collision chamber. Due to the fact that the contamination event is produced in a single collision, the effect on the measured coincidences depends on the value of $(\sigma^{00}/\sigma_{TI})F_0$, with σ^{00} the cross section for ionization of Ar by H^0. Therefore, this contamination cannot be reduced by further decrease of the Ar pressure in the collision chamber.

Now, we summarize the experimental conditions used in order to show how the contamination effects discussed above were maintained within acceptable values. Our measurements were performed with a proton-beam current of the order of 15 pA. We observed a linear behavior for the electron count rate in noncoincidence measurements, as a function of Ar pressure in the collision chamber (i.e., target thickness) in the range up to 10^{-5} Torr. In the present measurements, an Ar pressure of 3.5×10^{-7} Torr was used. Under this condition, a rate of 1.2×10^5 counts/s of incident H^0 is obtained. Background pressure in the collision chamber was below 10^{-7} Torr, and in this case the count rate for H^0 decreased to 0.8×10^5 counts/s. We estimate the fraction of H^0 projectiles in the beam, at the position of the target and for the Ar pressure of 3.5×10^{-7} Torr used, to be of the order of 0.1%.

In order to investigate the amount of contamination from double-collision events, we measured the rate of coincidences between electrons and outgoing H^0 projectiles as a function of chamber pressure (Ar target), for electrons emitted at $\theta=0°$ at the maximum of the cusp ($E_c=55.3$ eV). The expected behavior is of a linear pressure term, for true transfer ionization coincidences, and a quadratic one, for double collisions. Then the ratio between coincidence counts and the pressure is expected to follow a linear behavior, with an extrapolated value to zero pressure that is proportional to the transfer ionization cross section [11]. Effectively, we observed for this ratio a linear dependence up to 7×10^{-6} Torr. A fitting of the experimental data allows one to estimate, for the Ar target pressure of 3.5×10^{-7} Torr used, that double collisions account for 15% of the measured coincidences. For electrons emitted at $\theta=50°$ and $E=100$ eV, where the contribution of TI to the total electron emission is not as small as at the cusp maximum (as shown in Fig. 1), we observe that double-collision contamination is strongly reduced.

As we mentioned above, the contamination due to Ar ionization in a single collision by H^0 impact depends on the value of $(\sigma^{00}/\sigma_{TI})F_0$. With a roughly estimated $\sigma^{00}\cong10^{-16}$ cm^2 [17] and a neutral fraction of 0.1%, the contamination results in 0.3% of the total TI cross section. Additionally, for some selected electron emission angles and energies, we determined the amount of this contamination by a direct measurement of the Ar ionization by H^0 projectiles. We used a 100-keV pure H^0 beam and measured the coincidences between electrons and outgoing H^0 projectiles. Then, with the estimated neutral fraction (0.1%) in the beam used in the TI measurements, we obtained the percentage of coincidences due to this effect. The results were as follows: $\theta=0°$ at the cusp maximum, $E_c=55.3$ eV, 15%; $\theta=0°$, $E=10$ eV, 0.7%; $\theta=20°$, $E=25$ eV, 1%. At the energy of the binary encounter peak (target ionization), $E=4\,E_c\cos^2\theta$, the results were $\theta=0°$, 0.6%; $\theta=20°$, 0.2%; $\theta=50°$, 0.1%.

Absolute cross-section values for the present transfer ionization data were obtained by normalizing, at each electron angle and energy of emission, our total electron counts to the doubly differential cross section for electron emission in

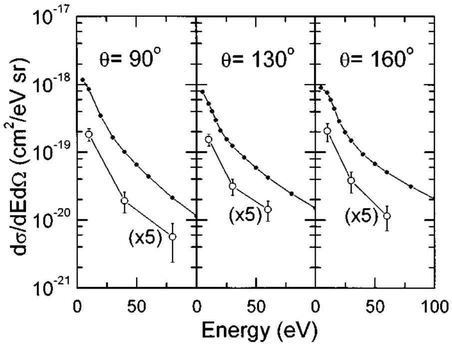

FIG. 2. Same as Fig. 1. Electron emission data (singles spectra) are from Ref. [18].

100-keV H^+ on Ar measured by Rudd, Toburen, and Stolterfoht [18]. As we measured the singles (noncoincidence) spectra for $\theta=0°$ and 20°, not contained in the data mentioned, our spectrum for $\theta=20°$ was normalized, within an uncertainty of 15%, to an interpolation of the $\theta=10°$ and 30° spectra of Rudd, Toburen, and Stolterfoht [18]. This normalization procedure includes a correction factor due to the efficiency of the H^0 projectile detector, estimated as 85% [16].

In Figs. 1 and 2 we show the doubly differential cross section for transfer ionization, together with that corresponding to the noncoincidence measurements of electron emission. At first glance, the doubly differential cross section for TI shows a general trend like that of the electron emission. For the latter, in accordance with tabulated total cross sections [2], the ionization of one and two electrons (without charge transfer) contributes 71.2% and 21.4%, respectively; 5.3% corresponds to TI and the remaining is due to processes involving three or more electrons.

It calls attention to the fact that, for $\theta=0°$, 20°, and 50°, the contribution from binary electrons, at an energy $E=4E_c\cos^2\theta$, is more clearly seen in TI than in the total emission. This may be attributed to a predominantly small-impact-parameter contribution to the electron emission in TI, as was suggested by McGuire *et al.* [1]. In fact, calculations of total cross sections at intermediate energies by Shingal and Lin [4], within an independent electron model, show that TI is produced at smaller impact parameters than single ionization.

At $\theta=0°$, a cusp is clearly seen in the TI data, as already observed by Víkor *et al.* [11] for 50-keV H^+ on Ar. Here, the cusp height in TI accounts for 0.6% of the cusp electron production, in good agreement with the tendency of measurements at lower impact energies [11]. In the case of single ionization by H^+, the cusp has been attributed to electron capture into the continuum (ECC) by the projectile Coulomb field. For TI, a detailed discussion of the cusp production mechanisms was done by Víkor *et al.* [11]. Here we limit ourselves to pointing out that, due to the evidence of cusp electron production in neutral impact and outgoing projectile collisions [9,16], an independent electron model can explain the observed cusp in TI. Nevertheless, only a detailed comparison of the cusp shape obtained for TI and one obtained for incident (and outgoing) H^0 can confirm the validity of this argument. Otherwise, a specific mechanism, such as a proposed correlated double capture [7], would be necessary. For emission at large angles, $\theta=90°$, 130°, and 160°, the three data points for TI roughly follow the behavior of the total electron production.

Another interesting feature is observed at low electron emission energy. At the minimum energy measured ($E=10$ eV) the TI cross section is, within uncertainties, almost independent of the emission angle. On the contrary, the total electron emission shows a decrease with increasing angles. For electron emission in the forward direction by H^+ impact, it is known that a broad ridge is observed [19]. This was attributed to a typical two-center effect [20,21], where the emitted electron is subjected to the Coulomb fields of both the projectile and the residual target ion. This ridge joins the low-energy side of the strongly asymmetric ECC peak, with the also-asymmetric low-energy peak [22]. The behavior of the low-energy TI data could be attributed to the fact that in the final state we have a neutral H^0 outgoing projectile, which is unable to influence the emitted electron, mainly subjected to the field of the residual target ion.

In summary, we have measured the doubly differential electron emission in a TI process for 100-keV H^+ on Ar giving detailed information, including an ample range in energy and angle, with which to discuss this two-electron process. A tentative interpretation of the main features observed has been given, but it remains to be determined whether the experimental results can be reproduced by a two-step model, where ionization and capture take place in direct projectile-electron interactions, or if a mechanism including electron correlation would be necessary. We note that, even in an independent two-step model, the ionization must take into account an initial state with a charged H^+ projectile and a final state with a neutralized H^0 projectile. Additionally, a specific mechanism for the cusp observed in TI would be required.

We would like to thank A. González and S. Suárez for collaboration during the measurements. We thank the Consejo Nacional de Investigaciones Científicas y Técnicas (Argentina) for financial support.

[1] J. H. McGuire *et al.*, J. Phys. B **28**, 913 (1995).
[2] R. D. DuBois and S. T. Manson, Phys. Rev. A **35**, 2007 (1987).
[3] S. N. Chatterjee, S. Prasad, and B. N. Roy, J. Phys. B **21**, 1209 (1988).
[4] R. Shingal and C. D. Lin, J. Phys. B **24**, 251 (1991).
[5] C. R. Vane *et al.*, Phys. Rev. Lett. **43**, 1388 (1979).
[6] L. H. Andersen, M. Frost, P. Hvelplund, and H. Knudsen, Phys. Rev. Lett. **52**, 518 (1984).
[7] S. Datz, C. Bottcher, L. H. Andersen, P. Hvelplund, and H. Knudsen, Nucl. Instrum. Methods Phys. Res. B **10/11**, 116 (1985).
[8] A. Kövér *et al.*, J. Phys. B **22**, 1595 (1989).
[9] L. Sarkadi, J. Pálinkás, A. Kövér, D. Berényi, and T. Vajnai,

Phys. Rev. Lett. **62**, 527 (1989).

[10] P. A. Závodszky *et al.*, Nucl. Instrum. Methods Phys. Res. B **79**, 67 (1993).

[11] L. Víkor *et al.*, J. Phys. B **28**, 3915 (1995).

[12] J. Pálinkás, R. Schuch, H. Cederquist, and O. Gustafsson, Phys. Rev. Lett. **63**, 2464 (1989).

[13] R. Hippler, G. Schiwietz, and J. Bossler, Phys. Rev. A **35**, 485 (1987).

[14] A. D. González, D. Fregenal, and S. Suárez, Nucl. Instrum. Methods Phys. Res. B (to be published).

[15] G. Bernardi, S. Suárez, D. Fregenal, P. Focke, and W. Meckbach, Rev. Sci. Instrum. **67**, 1761 (1996).

[16] P. Focke, G. Bernardi, D. Fregenal, and W. Meckbach (unpublished).

[17] L. J. Puckett, G. O. Taylor, and D. W. Martin, Phys. Rev. **178**, 271 (1969).

[18] M. E. Rudd, L. H. Toburen, and N. Stolterfoht, At. Data Nucl. Data Tables **23**, 405 (1979).

[19] G. Bernardi, S. Suárez, P. D. Fainstein, C. R. Garibotti, W. Meckbach, and P. Focke, Phys. Rev. A **40**, 6863 (1989).

[20] P. D. Fainstein, V. H. Ponce, and R. D. Rivarola, J. Phys. B **24**, 3091 (1991).

[21] N. Stolterfoht, R. D. DuBois, and R. D. Rivarola (unpublished).

[22] S. Suárez, C. R. Garibotti, G. Bernardi, P. Focke, and W. Meckbach, Phys. Rev. A **48**, 4339 (1993).