

Recombination in $\text{Kr}^{34+} + \text{H}_2$ collisions

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Recombination of Kr^{34+} ions in collisions with H_2 has been investigated. The principal motivation for this work was to search for resonant electron-transfer double excitation (RT2E), a three-electron correlation mechanism that is expected to occur in an ion-atom collision when electron transfer is accompanied by the simultaneous (resonant) excitation of two electrons to form an intermediate excited state. As a benchmark for RT2E, measurements of resonant transfer excitation (RTE) were made first. In this work, Kr (mostly K -shell) x-ray emission associated with single-electron-capture events was detected. Events associated with RTE were readily observed, but no events associated with RT2E were observed to a sensitivity of about 10^{-25} cm^2 . However, x rays associated with capture did occur at about twice the Kr K x-ray energy. The change in the x-ray energy of these latter events with beam energy indicates that radiative electron capture (REC) (inverse photoelectric effect) is the mechanism responsible. The observed cross sections for both the RTE and REC mechanisms are in reasonable agreement with theoretical calculations.

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INTRODUCTION

Recombination of atomic ions is a subject of intense interest and importance [1]. In general, recombination can occur resonantly via dielectronic recombination (DR) or non-resonantly via radiative recombination. In a collision between a highly charged ion and a neutral target atom, the interaction of a target electron with the projectile may result in the process of resonant electron-transfer excitation [2] (RTE) in which the excess energy from charge transfer (electron capture) results in projectile excitation. RTE, illustrated schematically in Fig. 1(a), proceeds via the inverse of an Auger transition and is resonant for projectile velocities equal to outgoing Auger electron velocities. The resulting intermediate excited state may deexcite by emitting a photon or by Auger electron emission. In the case of x-ray emission, recombination occurs and RTE may be investigated experimentally by detecting the emitted photon in coincidence with a projectile ion that has captured an electron. RTE, which is a correlated two-electron mechanism, is completely analogous to the ion-electron recombination process of dielectronic recombination [3] and both processes lead to the same intermediate excited states. The theoretical formulation of RTE has been based on the impulse approximation [4].

DR is of considerable interest in astrophysical studies [5], accelerator development (storage rings), and high-temperature plasmas for fusion [6]. During the past decade, RTE, as the ion-atom analog of DR, has been studied extensively and today the close relationship between RTE and DR is clearly established. Therefore, in addition to its fundamental importance as a correlated two-electron process in ion-atom collisions, RTE has been of considerable interest since it provides a means of studying DR.

The mechanism investigated in this work, namely, resonant transfer double excitation (RT2E), is similar to RTE except that two inner-shell electrons are excited during the

capture process as illustrated in Fig. 1(b). RT2E is expected to occur resonantly via a correlated three-electron interaction and the intermediate excited state that is formed can subsequently decay by photon or Auger emission. As for RTE resulting in x-ray emission, a signature for RT2E is the de-

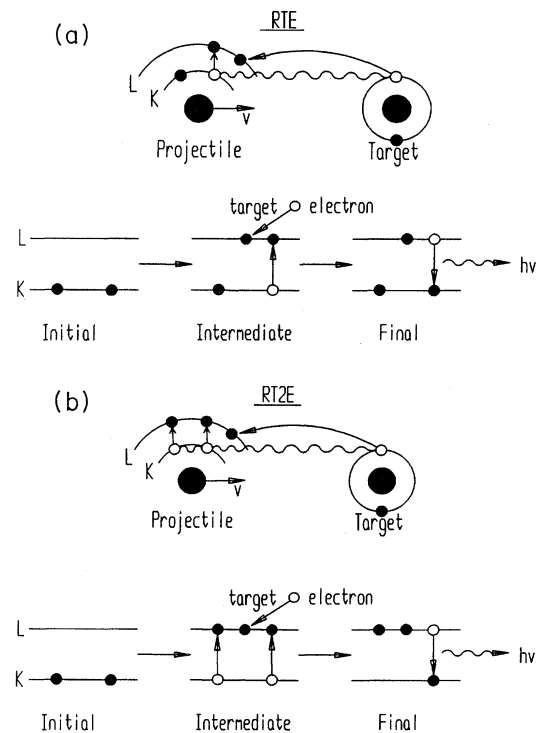


FIG. 1. (a) Schematic of RTE for a heliumlike ion. (b) Schematic of RT2E for a heliumlike ion.

tection of a photon in coincidence with a projectile that has captured an electron. RT2E can be distinguished from RTE, however, since the resonant energies for RT2E are slightly more than twice those for RTE. A process analogous to RT2E has been observed [7] in collisions of free electrons with Li^+ ions in which the intermediate triply excited state formed as a result of the single-capture and double-excitation process was observed from decay occurring by the emission of two correlated electrons. The inverse of the RT2E process, involving double inner-shell deexcitation associated with the emission of a single electron, was reported [8] several years ago. Competing with this latter process is double decay accompanied by single-photon emission [9]. The study of such correlated multielectron transitions dates back to the early days of the quantum theory of atoms [10].

Another process, namely, nonresonant transfer excitation (NTE) [11], could also contribute to the observation of x rays coincident with capture in the energy region where RT2E is expected to occur but should not exhibit the resonant behavior expected for RT2E. For the collision system used in this work, the expected magnitude of these nonresonant processes is not presently known, but based on previous calculations of NTE [12] is likely to be negligible in the region of interest.

In this work, recombination in Kr^{34+} ions is investigated. RTE and radiative electron capture (REC) (ion-atom analog of radiative recombination) are isolated and identified and cross sections obtained. No evidence for RT2E is found within the sensitivity of the present measurements. Preliminary results of this work have already been published elsewhere [13,14].

EXPERIMENTAL PROCEDURE

The present measurements were made at the National Superconducting Cyclotron Laboratory (NSCL) at Michigan State University for Kr^{34+} ions colliding with H_2 . The choice of H_2 as a target was dictated by the desire to keep the "background" from nonresonant processes [11] small and to use a target with only two electrons, as has been the case for

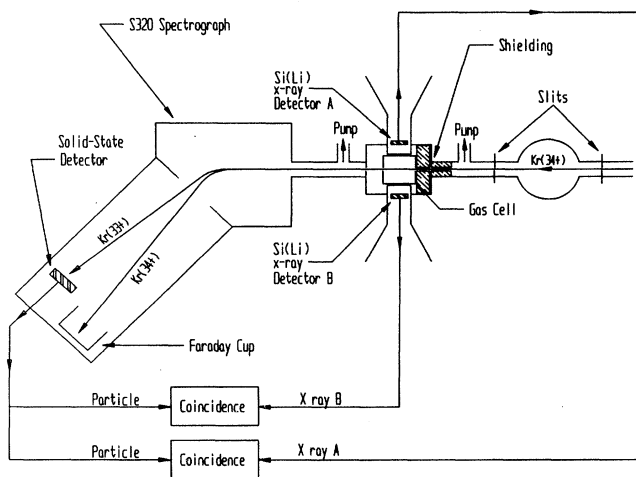


FIG. 2. Schematic of the experimental apparatus.

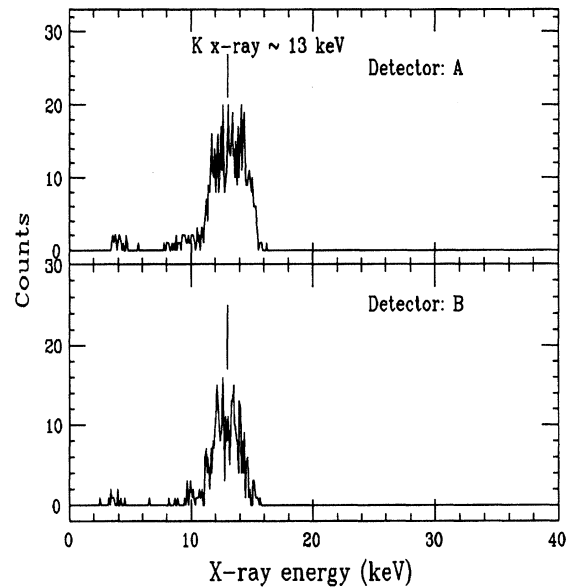


FIG. 3. Spectra of x rays coincident with single-electron capture for detectors A and B (see Fig. 2) for 16.5-MeV/u $\text{Kr}^{34+} + \text{H}_2$ collisions at 50 mTorr gas cell pressure.

most studies of RTE. The charge state was selected and the desired beam energy obtained by using the energy degrader and the A1200 beam analysis device at NSCL [15]. A schematic of the experimental apparatus is shown in Fig. 2. The arrangement and the techniques used are very similar to those used previously [2] for the investigation of RTE.

A projectile beam composed of Kr^{34+} ions was accelerated and directed through a gas cell containing the H_2 target molecules. As the projectile passes through the target cell, it can interact with a target molecule to form intermediate ex-

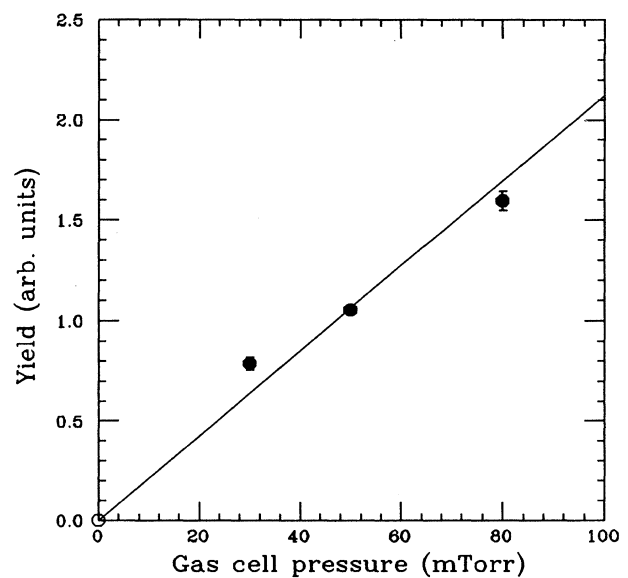


FIG. 4. Plot of the yield for x rays coincident with capture versus the gas cell pressure for 16.5-MeV/u $\text{Kr}^{34+} + \text{H}_2$ collisions.

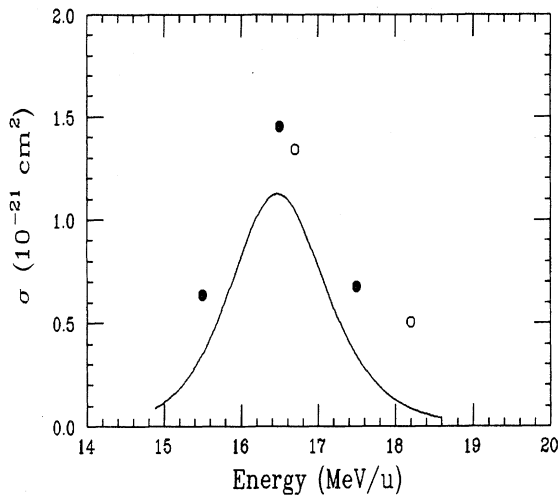


FIG. 5. RTE cross sections for $\text{Kr}^{34+} + \text{H}_2$: \bullet , present data; \circ , Ref. [13]. The solid curve is the calculated RTE cross section. The statistical errors are about the size of the symbols; the overall systematic uncertainty is estimated to be less than about 30%.

cited states characteristic of RTE or RT2E (or NTE). This excited state can then decay by emitting x rays. Since the formation of the intermediate state of interest here requires the capture of a target electron by the ion, the projectile ion becomes less positive (i.e., $q \rightarrow q - 1$, where q is the projectile charge) and can be detected by magnetic separation of the charge-changed components of the projectile beam emerging from the gas cell. By collecting the charge-changed components and the non-charge-changed component, the fraction of projectile ions that have undergone capture can be determined. However, in order to determine that an intermediate excited state characteristic of RTE or RT2E has been formed, an x ray that results from the relaxation of the excited state must also be detected. In this work, detection of a K -shell x ray implies that excitation of the projectile K shell took place in the collision interaction. Since a He-like ion has two electrons initially in the K shell (the lifetime of metastable states in Kr^{34+} is too short [16] to be of importance here), the only way K -shell x-ray emission can occur is for at least one of the two K -shell electrons to be promoted to a higher shell.

For x-ray detection, two Si(Li) detectors, labeled A and B, were mounted at 90° with respect to the beam axis as shown in Fig. 2. Two sets of slits were used to collimate the incident beam. The beam components emerging from the collision were separated by the S320 spectrograph that was

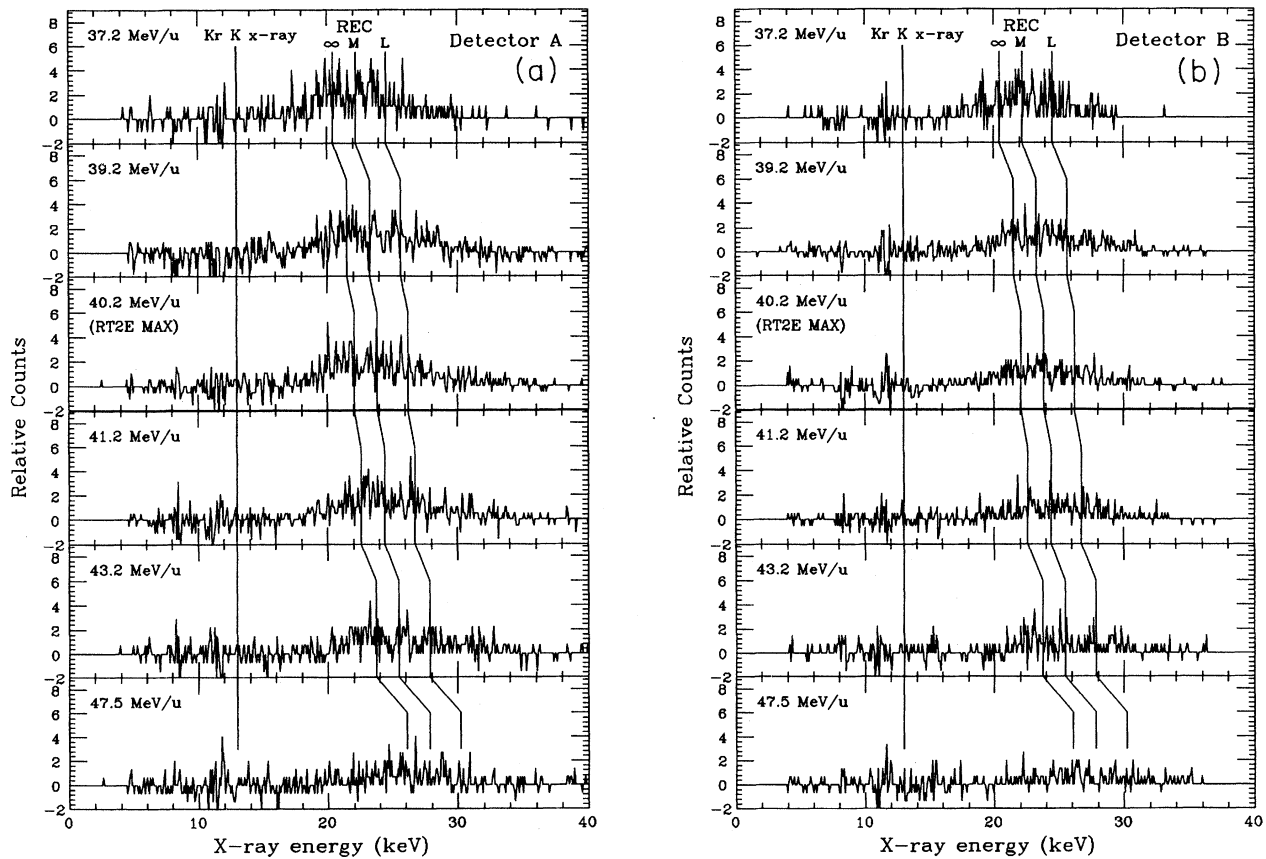


FIG. 6. Spectra of x rays coincident with single-electron capture for detectors A and B (see Fig. 2) in the projectile energy region 37.2–47.5 MeV/u. The expected position of Kr K -shell x rays is indicated as well as the expected centroid energies for REC to the L shell, M shell, and series limit (∞), respectively (see the text).

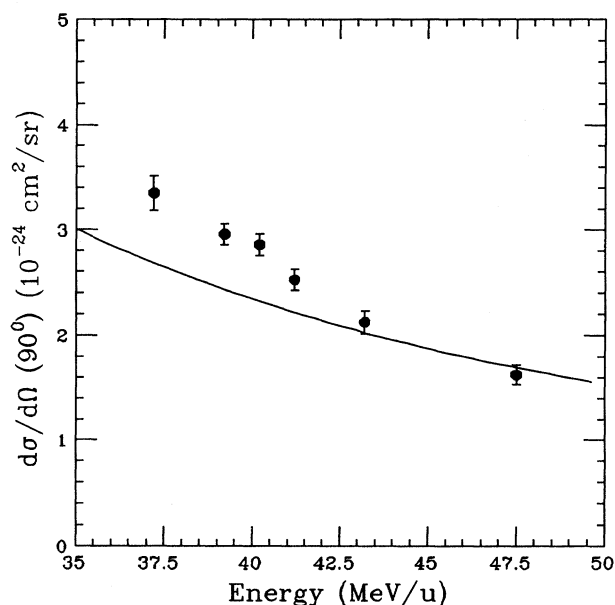


FIG. 7. Measured differential cross sections at 90° for x rays coincident with capture for x-ray energies greater than 20 keV. Error bars are relative uncertainties based on the total number of observed events for both detectors. The overall systematic uncertainty is estimated to be about 30%. The solid curve is the calculated REC cross section.

used as a charge-state analyzing device following the target region. Finally, a solid-state detector was used to detect the charge-changed (single capture) beam component while a Faraday cup was used to collect the main beam component.

Data were taken for two projectile energy regions: 15.5–17.5 MeV/u (the RTE maximum occurs near 16.5 MeV/u) and 37.2–47.5 MeV/u (the RT2E maximum is expected to occur near 40.5 MeV/u). RTE was measured first to test the experimental setup and to provide a benchmark for the RT2E measurements.

Typical spectra for x-ray emission coincident with capture are shown for both detectors in Fig. 3. Data were collected for a specific target cell gas pressure and several different pressure runs were conducted at each projectile energy in order to verify the linear dependence of the measured x-ray and particle yields with respect to the gas cell pressure. This is necessary to ensure that single-collision conditions prevail. At least three runs for each projectile energy (typically 0, 50, and 80 mTorr) were conducted in the RTE energy region, i.e., 15.5–17.5 MeV/u. The total coincidence counts were obtained by integrating the region of interest (the peak region in Fig. 3) of each spectrum. By plotting the yield (i.e., the number of counts per number of incident ions) of coincidence counts versus the gas cell pressure, one can verify the linear dependence of the measured yields with respect to the gas cell pressure as shown in Fig. 4. For the 37.2–47.5 MeV/u energy region, however, total x-ray and coincidence yields were measured only at 0 and 80 mTorr target gas pressure because of very low counting rates.

RESULTS AND DISCUSSION

As discussed above, Fig. 3 shows typical spectra of x rays coincident with single-electron capture for both detectors A

and B for 16.5-MeV/u $\text{Kr}^{34+} + \text{H}_2$ collisions, which is near the energy where the RTE *KLL* (excitation to the *L* shell is accompanied by capture to the *L* shell) maximum should occur. In these spectra, Kr *K*-shell x rays of energy about 13 keV are clearly seen.

The cross sections derived from these spectra, along with theoretical calculations of RTE, are shown in Fig. 5, where a pronounced maximum is observed to occur near 16.5 MeV/u beam energy as expected. It is seen that the measured cross sections are systematically larger than the theoretical values, but the discrepancy is within the systematic error of about 30%. Measurements of RTE for Kr^{35+} ions have previously been reported by Stöhlker [17].

Spectra for x rays coincident with capture in the projectile energy region 37.2–47.5 MeV/u (the RT2E *KKLLL* maximum is expected to occur near 40.5 MeV/u) are shown in Fig. 6 for detectors A and B. (The notation *KKLLL* means that double excitation to the *L* shell is accompanied by capture to the *L* shell.) All of the spectra were normalized to the one associated with the 37.2 MeV/u beam energy so that the relative number of events can be compared for each spectrum. It is seen that there are essentially no real coincidences associated with Kr *K*-shell x rays (near 13 keV), in contrast to the RTE results shown in Fig. 3. An explanation for this observation would be that the triply excited intermediate state formed in RT2E decays principally by first emitting an x ray and then by ejecting an Auger electron (or vice versa), thus returning the projectile ion to its original charge state. In this case, RT2E events would not be observed by detecting x-ray events coincident with electron capture. A theoretical estimate of the RT2E cross section for *KKLLL* transitions involving radiative stabilization by two sequential photons gives about 10^{-27} cm^2 , while the RT2E cross section for one photon followed by Auger emission (or vice versa) is about 10^{-28} cm^2 . The sensitivity of the present measurements is about 10^{-25} cm^2 . So, for the present experimental sensitivity it would not be possible to observe RT2E unless the theoretical results are a severe underestimate.

Coincidences do exist, however, for higher x-ray energies (greater than or approximately equal to 20 keV), as seen in Fig. 6. (The electronic setup allowed for the observation of coincidence events for x-ray energies as high as 40 keV.) From the figure it is seen that there is an apparent shift in the centroid energy of these high-energy x-ray events with beam energy, a feature that is characteristic of the REC mechanism [18] (inverse photoelectric effect). The positions labeled REC-*L*, REC-*M*, and REC- ∞ are the expected centroid x-ray energies for REC associated with electron capture into the *L*, *M*, and $n = \infty$ (zero binding energy) shells, respectively.

The measured differential cross sections, observed at 90° with respect to the beam axis, associated with electron-capture coincidences for x-ray energies greater than 20 keV are shown in Fig. 7 along with the calculated values for REC. There is reasonable overall agreement between experiment and theory and the measured cross sections decrease monotonically with beam energy as does the REC theoretical prediction. However, there seems to be a systematic discrepancy at the lower beam energies. The reason for this discrepancy is unknown.

CONCLUSIONS

In summary, recombination has been investigated in high-energy $\text{Kr}^{34+} + \text{H}_2$ collisions. Three recombination mechanisms are considered, namely, resonant transfer excitation, resonant transfer double excitation, and radiative electron capture. Kr *K*-shell x rays associated with RTE involving *KLL* transitions are readily observed and the measured cross sections are in good agreement with theory. Higher-energy x rays associated with single-electron capture are attributed to REC. Here, there is reasonable overall agreement with theory, but there seems to be a systematic discrepancy at the lower beam energies.

No evidence for the RT2E mechanism is observed, however, in the present work. Although this mechanism probably occurs, its estimated cross section (less than or approximately equal to 10^{-27} cm^2) is too small to be measured within the sensitivity (approximately 10^{-25} cm^2) of the present measurements.

ACKNOWLEDGMENT

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- [1] For a review, see *Recombination of Atomic Ions*, edited by W. G. Graham, W. Fritsch, Y. Hahn, and J. A. Tanis (Plenum, New York, 1992).
- [2] See J. A. Tanis, in *Recombination of Atomic Ions* (Ref. [1]), pp. 241–257.
- [3] M. J. Seaton and P. J. Storey, in *Atomic Processes and Applications*, edited by P. G. Burke and B. L. Moiseiwitsch (North-Holland, New York, 1976), pp. 133–197.
- [4] D. Brandt, *Phys. Rev. A* **27**, 1314 (1983).
- [5] A. Burgess, *Astrophys. J.* **139**, 776 (1964).
- [6] A. L. Merts, R. D. Cowan, and N. H. Magee, Jr., Los Alamos Scientific Laboratory Informal Report No. LA-6220-MS, 1976 (unpublished).
- [7] A. Müller, G. Hofmann, B. Weissbecker, M. Stenke, K. Tinschert, M. Wagner, and E. Salzborn, *Phys. Rev. Lett.* **63**, 758 (1989).
- [8] V. V. Afrosimov, Yu. S. Gordeev, A. N. Zinov'ev, D. Kh. Rasulov, and A. P. Shergin, *Pis'ma Zh. Éksp. Teor. Fiz.* **21**, 535 (1975) [*JETP Lett.* **21**, 249 (1975)].
- [9] W. Wölfli, Ch. Stoller, G. Bonani, M. Suter, and M. Stockli, *Phys. Rev. Lett.* **35**, 656 (1975).
- [10] W. Heisenberg, *Z. Phys.* **32**, 841 (1925).
- [11] P. L. Pepmiller, P. Richard, J. Newcomb, J. Hall, and T. R. Dillingham, *Phys. Rev. A* **31**, 734 (1985).
- [12] T. M. Reeves, J.M. Feagin, and E. Merzbacher, in *Abstracts of Contributed Papers, XIVth International Conference on the Physics of Electronic and Atomic Collisions, Palo Alto, 1985*, edited by M. J. Coggiola, D. L. Huestis, and R. P. Saxon (North-Holland, Amsterdam, 1986), p. 392.
- [13] K. E. Zaharakis, R. R. Haar, J. A. Tanis, M. W. Clark, and V. L. Plano, in *VIth International Conference on the Physics of Highly Charged Ions*, edited by P. Richard, M. Stöckli, C. L. Cocke, and C. D. Lin, AIP Conf. Proc. No. 274 (AIP, New York, 1993), pp. 303–306.
- [14] K. E. Zaharakis, R. R. Haar, O. Voitke, M. Zhu, J. A. Tanis, and N. R. Badnell, *Nucl. Instrum. Methods Phys. Res. Sect. B* **98**, 300 (1995).
- [15] For a discussion of the energy degrader and the A1200 analysis device, see, for example, NSCL Newsletter No. NSCL-N12, April 1991, Michigan State University.
- [16] G. W. F. Drake, *Phys. Rev. A* **3**, 908 (1971).
- [17] Th. Stöhlker, Ph.D. thesis, University of Giessen, Germany, 1991 (unpublished).
- [18] H. W. Schnopper, H. D. Betz, J. P. Delvaille, K. Kalata, and A. R. Sohval, *Phys. Rev. Lett.* **29**, 898 (1972).