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Resonant Behavior in the Projectile X-Ray Yield Associated with Electron Capture in S + Ar Collisions

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Experimental evidence is presented for a new resonant process in ion-atom collisions which is analogous to dielectronic recombination in free-electron-ion collisions. Resonant behavior observed in the yield of projectile $K \ge K$ rays in coincidence with single-electron capture in 70-160-MeV S+Ar collisions is attributed to simultaneous electron capture and K-shell excitation. The data indicate that this resonant process is an important mechanism in inner-shell vacancy production in the energy range studied.

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It was recently suggested that projectile Kshell excitation may occur simultaneously with electron capture in ion-atom collisions.¹ Such a process, which is due to the Coulomb interaction of the projectile with the target electrons, is qualitatively analogous to an inverse Auger transition and is expected to be resonant for projectile velocities corresponding to the energy of an exiting electron in the Auger process. Since the captured electron is initially bound in the target, the width of the resonance should be reflective of the distribution of electron momenta in the target.

In the case of free-electron recombination, this process is called dielectronic recombination which occurs when a highly stripped ion captures a continuum electron and simultaneously excites an electron from the ground-state configuration of the ion. Since radiation can be emitted following the formation of this excited state, dielectronic recombination is believed to be an important energy-loss mechanism in high-temperature fusion plasmas.² Dielectronic recombination has been identified in plasmas but cross sections for this process have never been successfully measured in laboratory experiments.

In an ion-atom collision, radiation may also be emitted following formation of the intermediate excited state, i.e., $Z_1^{q^+} + Z_2 \rightarrow (Z_1^{(q-1)+})^* \rightarrow Z_1^{(q-1)+}$ $+h\nu$. In this Letter we report the first observation in an ion-atom collision of a *resonant* behavior in the cross section for projectile x rays associated with ions having captured a single electron. The existence of resonant behavior in this yield is expected for simultaneous electron capture and excitation. The present results indicate that this mechanism is significant in producing inner-shell vacancies in highly stripped ions. The *simultaneous* electron-capture and excitation process has recently been referred to as resonant transfer and excitation (RTE).³

In the present work, sulfur K x rays coincident with single-electron capture and loss have been measured for 70–160-MeV S^{13^+} ions incident on argon under single-collision conditions. Li-like S is used since it has no long-lived excited states. This work was done at Brookhaven National Laboratory with the MP tandem Van de Graaff accelerator facility. The beam of S^{13+} ions was collimated by two 0.5-mm square apertures separated by about 2.8 m prior to entering a differentially pumped gas cell. X rays from the interaction region were detected with a 200-mm² Si(Li) detector located about 1 cm from the beam line and positioned at 90° to the incident beam direction. Ions emerging from the gas cell passed between a pair of electrostatic deflection plates which separated the various charge-state components of the beam. Ions undergoing single-electron capture (q-1) and loss (q+1) were detected in surface-barrier detectors. The non-charge-changed component of the emerging beam (q = 13 +) was collected in a Faraday cup located about 40 cm behind the surface-barrier detectors. A capacitance manometer was used to measure target gas pressures which were less than 7 mTorr for all measurements. All cross sections of interest were found to be independent of pressure in the range studied, indicating single-collision conditions.

Coincidences between S K x rays and singleelectron capture and loss events were recorded with a time-to-amplitude converter (TAC). The singles x-ray spectrum and the coincidence spectra from the q-1 and q+1 TAC's were stored in a two-dimensional x-ray energy versus time array. This allowed energy "cuts" to be made in the TAC spectra to give the coincident events corresponding to the projectile x rays. Figure 1 shows the various projectile cross sections obtained from our measurements. Relative errors for a given cross section are about 5%. Absolute errors in the values shown are estimated to be about 25%.

From the figure it is seen that the cross section for sulfur K x-ray emission associated with single-electron capture σ_x^{q-1} exhibits a maximum in the region of about 120 MeV. We attribute this peak to resonant transfer and excitation. RTE is expected to occur as a result of the formation of intermediate resonant states such as $1s^22s$ $\rightarrow 1s2s^22p$, $1s2s2p^2$, $1s2s^23p$, 1s2s2p3d, etc. Qualitatively, a maximum in the cross section σ_x^{q-1} would occur when the energy of the captured electron in the projectile rest frame matches the Auger electron energy for a given transition. Since the velocity component of the target electrons along the beam axis contributes to the rela-



FIG. 1. Projectile cross sections for 70–160–MeV $S^{13+} + Ar$ collisions. σ_x^{q-1} is the cross section for projectile *K* x-ray emission associated with single-electron capture; σ_x^{q+1} is the cross section for projectile *K* x rays associated with single-electron loss; $\sigma_{K\alpha}$ is the cross section for sulfur singles $K\alpha$ x-ray emission; and $\sigma_{q,q-1}$ is the total electron-capture cross section. The curve labeled σ_{RTE} is a theoretical calculation (Ref. 4) of the *resonant* part of σ_x^{q-1} . The solid curve for σ_x^{q-1} is a normalization of σ_{RTE} to the data with the background indicated by the dashed line. All other curves are drawn through the data to guide the eye.

tive velocity, the shape of σ_x^{q-1} should reflect the target electron momenta distribution, i.e., their Compton profile.

The experimental results indicate that there is a nonresonant contribution under the σ_x^{q-1} peak. This background may be due to *uncorrelated* excitation and single-capture events taking place during the same collision with a single target atom. Since the single-capture cross section is large (~ 10⁻¹⁷ cm²) it is possible that an ion which undergoes *K*-shell excitation due to the Coulomb interaction *with the target nucleus* will also capture during the same collision.

Also shown in Fig. 1 is the cross section for sulfur K x-ray emission associated with singleelectron loss σ_x^{q+1} , the cross section for singles $K\alpha$ x-ray emission $\sigma_{K\alpha}$, and the total single-capture cross section $\sigma_{q,q-1}$. In both σ_x^{q+1} and $\sigma_{K\alpha}$ evidence for anomalous variation is noted in the region of the "resonance." A possible explanation for the origin of these anomalies will be given below.

Strong evidence that the observed maximum in σ_x^{q-1} is due to a real resonance and, in fact, is produced by RTE is provided by a comparison of the data with theoretical calculations of the RTE process. A calculation of the resonant part (RTE) of σ_x^{q-1} for S^{13^+} + Ar has been carried out by Brandt⁴ using the dielectronic recombination cross sections of McLaughlin and Hahn⁵ and the Compton profile of the target electrons.⁶ It should be noted that this calculation requires the projectile velocity to be much greater than the bound electron velocity. Hence, the two 1s electrons which do not satisfy this condition were not included. At any rate the 1s electrons would not contribute significantly to the RTE cross section. Furthermore, all intermediate states which can result in $K\alpha$ emission were included in Brandt's computations since the x-ray detector used in the present experiment could not resolve $K\alpha$ transitions corresponding to the various intermediate states.

The theoretical curve based on this calculation is labeled σ_{RTE} in Fig. 1. The relatively large width of the RTE peak is partly due to the distribution of target electron momenta. An additional contribution to the width as well as a slight asymmetry in σ_{RTE} occurs because this curve is not due to a single state but is the envelope of the contributions from the various intermediate states.

The solid curve for σ_x^{q-1} in Fig. 1 was obtained by normalizing the magnitude of the theoretical σ_{RTE} curve to the peak in σ_x^{q-1} and adding this to a slowly varying background. The background used is indicated by the dashed line in Fig. 1. Clearly, the position and width of the peak in σ_x^{q-1} are in good agreement with the theoretical calculations.

The absolute magnitude of the maximum in the data is about a factor of 2 larger than the theory. Reasons for this discrepancy could be twofold: (1) the calculated dielectronic recombination cross sections⁵ are too small and/or (2) important effects arise from the initial binding energies of the target electrons which are not accounted for in the calculations. These points need further study.

As previously noted the data show evidence for anomalies in $\sigma_{K\alpha}$ and σ_{X}^{q+1} . Since $\sigma_{K\alpha}$ is the total x-ray yield, a true resonance in σ_x^{a-1} would produce an equal strength resonance in $\sigma_{K\alpha}$. If it is assumed that both $\sigma_{K\alpha}$ and σ_{X}^{q+1} have nonresonant contributions which increase *linearly* with energy, it is found that $\sigma_{K\alpha}$ shows a small peak relative to its nonresonant part and σ_x^{q+1} displays a small asymmetry (like an interference shape) centered near 120 MeV relative to its nonresonant part. An estimate of the magnitude of the intensity of the maximum in $\sigma_{K\alpha}$ gives a value about a factor of 2 larger than that for σ_x^{q-1} . This implies that other charge states in addition to q-1 must be contributing to the "peak" in $\sigma_{K\alpha}$. Since all other outgoing charge states (q - 2,q+2, etc.) except q-1, q, and q+1 have negligible intensities and the asymmetry in σ_x^{q+1} is small, the *additional* intensity in the $\sigma_{K\alpha}$ "peak" is attributed almost entirely to charge state q. The origin of the anomalies in these cross sections is not obvious and requires further consideration.

A possible explanation for the appearance of anomalies in the q and q + 1 channels is the existence of coupling between the three intense charge states (q-1, q, and q+1). Such coupling between these various charge states is reasonable in view of the high probability for charge exchange in the relatively close collisions expected for RTE. Since charge state q is related to q - 1 by a *sin*gle electron exchange, a resonance in q - 1 could have a large effect in the charge-state-q channel. Also, a smaller resonant effect for σ_x^{q+1} is consistent with the less probable two-electron exchange involved in the coupling of the resonant q-1 channel to the q+1 channel. Channel-coupling effects such as these are well known in nuclear reactions. We emphasize that the channel-coupling interpretation is speculative and further experimental and theoretical work is needed in order to confirm or to reject this possibility.

The existence of anomalous effects in the q and q + 1 channels would provide additional support for the RTE interpretation of the peak in σ_x^{q-1} . In particular, an explanation of the observed peak in σ_x^{q-1} as due to an accidental consequence of an increasing excitation cross section and a decreasing probability for capture in an uncorrelated multistep process could not *also* explain effects in the q and q + 1 channels. Also, the possibility that *unrelated* anomalies occur in the same energy region in these different channels is unlikely.

If the discussion above concerning channel coupling is assumed to be valid, the calculation of Brandt for σ_{RTE} should be compared with the "peak" strength in $\sigma_{K\alpha}$ instead of σ_{X}^{q-1} . In this case the theoretical intensity is about a factor of 4 lower than the experimental results.

In conclusion, strong experimental evidence is presented for a new resonant process (RTE) in high-energy ion-atom collisions. Support for the interpretation of this resonant process as the analog of dielectronic recombination is provided by comparison of the results with theoretical calculations. The data indicate that RTE is very important in the production of inner-shell vacancies in the velocity regime investigated. Furthermore, if the RTE interaction occurs primarily with the weakly bound target electrons for which the projectile velocity is much greater than the electron velocity then RTE may, in fact, approximate dielectronic recombination. If such is the case, RTE could be used as a benchmark for theoretical calculations of dielectronic recombination cross sections. We emphasize, however,

that such a close association between RTE and dielectronic recombination remains to be established.

Additional theoretical calculations as well as experimental investigations should be made in order to confirm the interpretations presented here. We plan to extend the measurements to other target atoms, ionic species, and charge states. A particularly interesting target to investigate is He because of its simple electronic structure.

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