5-1-1991

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Recommended Citation

R. S. Schuch and E. L. Justiniano and M. Schulz and S. Datz and P. F. Dittner and J. P. Giese and H. F. Krause and H. Schone and C. R. Vane and S. M. Shafroth, "Population of Highly Excited Intermediate Resonance States by Electron Transfer and Excitation," *Physical Review A*, vol. 43, no. 9, pp. 5180-5183, American Physical Society (APS), May 1991.
The definitive version is available at https://doi.org/10.1103/PhysRevA.43.5180

RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 43, NUMBER 9 1 MAY 1991

Population of highly excited intermediate resonance states by electron transfer and excitation

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(Received 9 May 1990)

Coincidences between two sulfur K x rays were detected from collisions of hydrogenlike S ions with H_2 gas in the projectile energy range between 150 and 225 MeV. These K x rays are emitted in the decay of doubly excited states formed in the collisions via transfer and excitation. The excitation function for two coincident $K\beta$ transitions peaks at about 175 MeV, slightly above the expected *KMM* resonance energy for resonant transfer and excitation (RTE). This demonstrates the occurrence of $\Delta N \geq 2$ transitions (i.e., *KMM* and higher resonances) in the RTE process. The cross sections for the population of the very highly excited states are higher than those predicted by theoretical calculations that use dielectronic recombination rates folded with the Compton profile for the bound electrons.

Resonant transfer and excitation (RTE) occurs in collisions of ions with atoms when a projectile electron is excited in a close collision with a target electron. As a result of this collision, the target electron is captured, and the projectile is left in a doubly excited state.[1-3] This process can be viewed as an inverse Auger decay. From this point of view, a necessary condition for this process to be resonant is that the collision velocity of the target electron must equal the velocity of an Auger electron that would be emitted from the doubly excited state of the projectile. RTE for a hydrogenlike projectile proceeds via the formation of a doubly excited intermediate state of a He-like ion. This state can be described with its main quantum numbers n and N, where for convenience we assume $n \geq N$ (e.g., $2s3p$, with $n=3$, $N=2$). The Auger notation is also often used to describe these transitions where, for example, KLM indicates an electron excited from the K to the L shell and another electron captured into the M shell. The doubly excited intermediate state may decay by either autoionization, causing the ion charge state to return to its initial value as before the collision, or by radiative emission of one, two, or more x rays, in which case the captured electron is stabilized and the charge state decreases by one unit. When radiative stabilization occurs, the process is the ion-atom collision analog of dielectronic recombination (DR) in electron-ion collisions. Because of this relationship, RTE cross sections (σ_{RTE}) may be calculated within the impulse approximation[4] from the DR cross sections (σ_{DR}) by accounting for the Compton profile of the bound target electron.

In previous experiments on RTE with H-like, He-like, and Li-like ions only $\Delta N = 1$ transitions, i.e., *KLn*, were seen as separated groups of resonances.[1-3] There was no clear observation of $\Delta N \geq 2$ transitions (i.e., *KMM* and higher resonances) in the excitation function of RTE cross sections. Recently an indication of these transitions was found indirectly by comparing calculations[5] of the cross section with the measurement[6] for F^{6+} on H_2. Their inclusion lead to a better agreement between theory and experiment at the high-energy part of the resonance. The disagreement with the calculated shape at the high-energy end of the excitation function has been noticed in several publications[6-9] and it has been partly explained by a contribution from the population of very high n states.[5,10,11] But as this cross section was estimated to be very small and as these transitions have not been clearly observed, other mechanisms for enhancing the cross section at high energies such as electron transfer and excitation by a bound target electron (2*e*TE) have been invented[6] to explain the discrepancy (see also below).

The argument for the small RTE cross sections for $\Delta N \geq 2$ relative to $\Delta N = 1$ can be given in the following way. The DR cross section is written as the product of two terms: One term is the rate for populating the intermediate state, which is the autoionization rate $A_a(K,n,N)$ into the K shell. The other term is the branching ratio for its radiative decay, which is given by

$$\frac{\sum_{n,N} A_r(n,N)}{\sum_{n,N} [A_r(n,N)+A_a(n,N)]} ,$$

where here the autoionization rates $A_a(n,N)$ and the radiative rate $A_r(n,N)$ are for all decay channels of the doubly excited state which lead to autoionization and radiative stabilization, respectively. Both the autoionization rate $A_a(K,n,N)$ and the summed radiative rates $A_r(n,N)$ decrease strongly with n and N. For $\sum_{n,N} A_a(n,N)$ the influence of the opening of the *LMM* Auger channel with $\Delta N \geq 2$ transitions is very important. Numerical estimates are not simple in this case, but certainly $A_a(L,M,M) \geq A_a(K,n,N)$. $A_a(L,M,M)$ is probably the largest term in $\sum_{n,N} A_a(n,N)$, as for constant N and high n (Rydberg states) $A_a(n,N)$ decreases strongly with n. From all these factors it can be seen that the DR and therefore also the RTE cross section for $\Delta N \geq 2$ becomes very much smaller than for $\Delta N = 1$ transitions.

K-x-ray–*K*-x-ray coincidences are particularly well suited for detecting the population of such highly doubly excited states, which stabilize by photon emission. The detection of two $K\beta$ x rays or two transitions from higher states in coincidence would be a signature for the population of the highly doubly excited states. Those coincidences had not been seen in previous experiments[3,7] where the energy might have been too low to resonantly populate such states. In the present x-ray–x-ray coincidence measurement we, therefore, significantly extended the ion energy range to search for transitions of this type. We report here clear evidence for a resonant population of highly doubly excited states in ion-atom collisions.

A sulfur beam from the 25-MV Holifield tandem accelerator at Oak Ridge National Laboratory, was post stripped and charge state analyzed for H-like ions. Following collimation the beam entered the gas cell. Since the width of the resonance seen in RTE is determined by the momentum distribution of the target electrons, an H_2 gas target was chosen. The target was about 10 mm long and differentially pumped in multiple stages. Dependences of the true coincidence yield on the target pressure were recorded in order to find the optimum target thickness (~0.5–5 mTorr cm) which allowed the maximum count rate under single collision conditions. Impurities of charge changed ions in the beam before it enters the interaction region are not important in the *K*-x-ray–*K*-x-ray coincidence setup, because ions that capture an electron prior to the gas cell do not contribute to either the true or the random *K*-x-ray–*K*-x-ray coincidences. An exception to this rule would be projectiles that were in the 2*s* metastable state following the stripping process. For those ions even single capture leads to two coincident *K* x rays, but of course these would not show a resonant structure with the beam energy as RTE does. From our results and those in Ref. 3 we see that the fraction of ions in the 2*s* metastable state is negligible. Two Si(Li) x-ray detectors were brought to a distance of 15 mm to the beam. The efficiency of these detectors is close to unity (75% for the S *K* lines) and the respective corrections are straightforward. The x-ray resolution achieved with the Si(Li) detectors was about 160 eV for the S *K* x rays. This was enough to resolve the $K\alpha$ and $K\beta$ x rays, but we were not able to clearly resolve the hypersatellite transitions, which are about 140 eV higher in energy than the satellite transitions.

The intensity of the true coincident *K* x rays was obtained for different ion beam energies. The measurements were performed for small steps in the ion energy covering the RTE resonance, i.e., the ion velocity was varied so as to correspond to the electron-ion relative velocity needed to obtain the associated DR resonances. The data were recorded in the event mode so that random coincidences could be subtracted later. The different runs were normalized with respect to the beam current measured in a Faraday cup. In the results shown only statistical errors from the coincidence counts and the subtraction of the random coincidences are included (see, e.g., also Ref. 3).

In Fig. 1 we show a two-dimensional true coincident x-ray spectrum for 165-MeV energy of the sulfur ions. The energy positions of $K\alpha$, $K\beta$, and $K\gamma$ x rays are indicated. Different event loci can be associated with coincidences between $K\alpha$ and $K\alpha$, $K\alpha$ and $K\beta$, or $K\gamma$, and $K\beta$ or $K\gamma$ with $K\beta$ or $K\gamma$. As these decays occur in a He-like ion, the first of the two emitted *K* x rays must be a hypersatellite transition. Energy shifts due to this can be seen from a closer inspection of the spectra. The visibility of the island from coincidences between $K\beta$ and $K\beta$ or $K\gamma$ x rays is already strong evidence for $\Delta N \geq 2$ transitions in RTE.

Sum energy spectra were formed in the data analysis by adding the energy of the real coincident x rays and in these spectra the three different event types were separated (for details see Refs. 8 and 12), which are $K\alpha$-$K\alpha$ for the time correlated emission of two $K\alpha$ x rays, $K\alpha$-$K\beta$ for the coincident $K\alpha$ and $K\beta$ x rays, $K\alpha$-$K\gamma$ for coincident $K\alpha$ and $K\beta$ or $K\gamma$ x rays, and $K\beta\gamma$-$K\beta\gamma$ for two coincident $K\beta$ and $K\gamma$ and higher x rays. In the excitation function shown in Fig. 2 these cross sections are plotted versus the beam energy (solid diamonds). The open circles in Fig. 2 are taken from Ref. 8 and shown here for completeness. At the lower energy of Ref. 8 the $K\alpha$-$K\beta$ coincidences were separated from $K\alpha$-$K\gamma$ or higher coincident x rays. In the high-energy regime measured here, the small con-

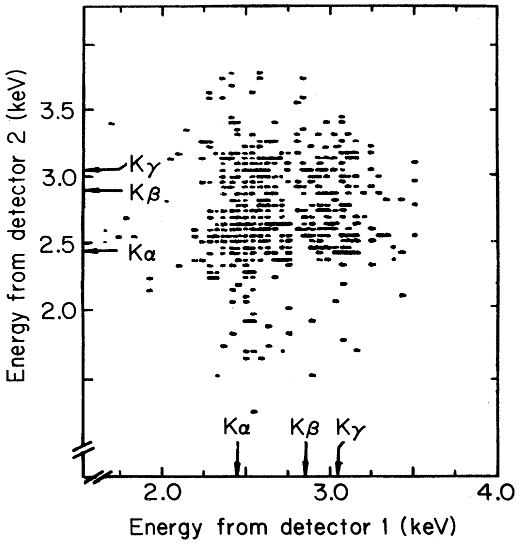

FIG. 1. Two-dimensional true coincident x-ray spectrum for 165 MeV S^{15+}-H_2 collisions as obtained from the two Si(Li) x-ray detectors. The arrows indicate the approximate energies of the $K\alpha$, $K\beta$, and $K\gamma$ satellite transitions.

tribution of *Kα*-*Kβ* coincidences could not be separated from the *Kα*-*Kγ* coincidences. The *Kα*-*Kβ* coincidences are therefore included in the peak labeled *Kα*-*Kγ*, but their inclusion does not alter the cross section much in this region. It should be noted that the cross section scale changes by 1 order of magnitude for the *Kβγ*-*Kβγ* coincidences. The resonance peaks are clearly visible at the expected positions of the resonance energies indicated by arrows at *KLL*, *KLM*, *KLN*, and the *Kβγ*-*Kβγ* coincidences peak at an energy above the expected *KMM* resonance energy. This shows that the maximum of the cross section which leads to these x-ray transitions is for $\Delta N > 2$.

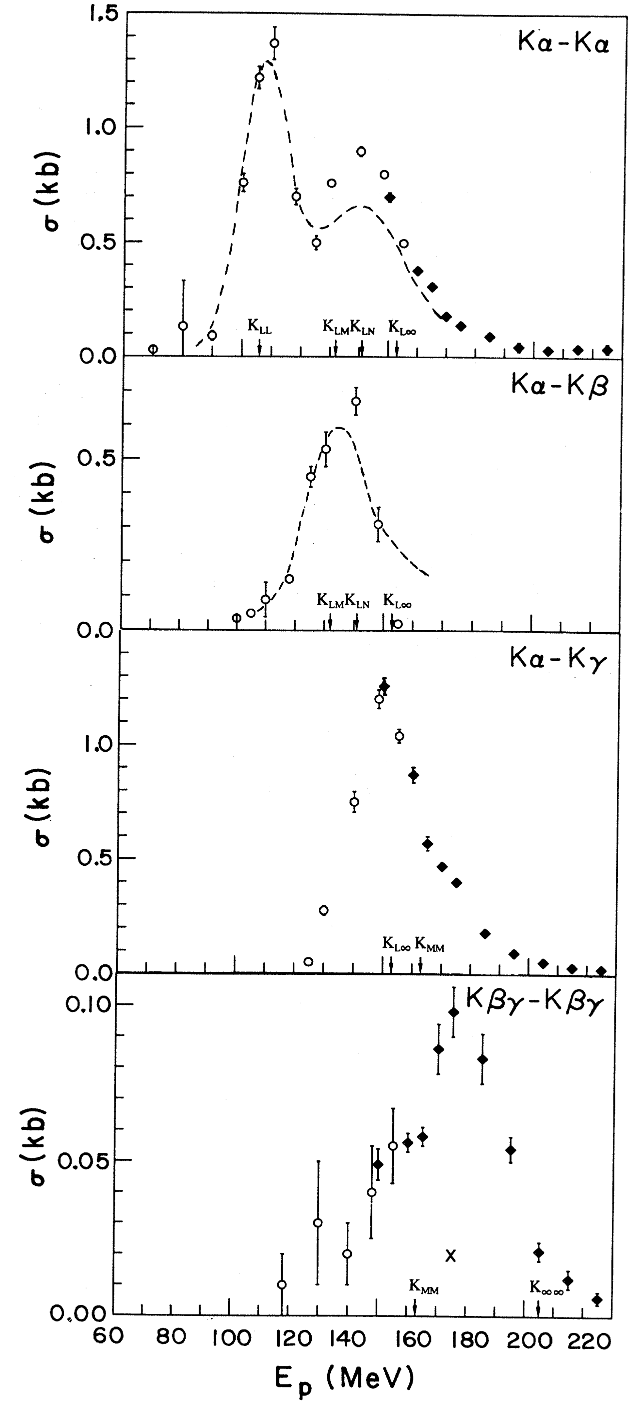

FIG. 2. RTE cross section for S^{15+} on H_2 from coincidences between different *K*-x-ray satellite lines (open circles are those from Ref. 8 for *Kα*-*Kα*, *Kα*-*Kβ*, and *Kα*-*Kγ*). Dashed line and cross represent predictions by McLaughlin and Hahn (Ref. 9).

We want to remark that the cross sections were derived by assuming isotropic and independent emission of the two x rays. It has been predicted[13] that an anisotropy effect could cause an increase of about 25% at 90° for ^1P-^1D-^2S x-ray emission. The effect is even smaller in our case, as we average over many different excited states and have a detector opening angle of about 35°. Additionally an analysis of the x-ray spectra for *K*-x-ray pile-up lines showed no evidence for a correlated emission of the two x rays in one direction.

The dashed lines and a cross in Fig. 2 represent results of calculations by McLaughlin and Hahn,[10] in which the impulse approximation[4] was applied in folding $\sigma_{\rm DR}$ with the Compton profile of H_2 for obtaining the values of $\sigma_{\rm RTE}$. The calculation agrees rather well with the data in the *KLL* maximum of *Kα*-*Kα* and the *KLM* maximum of *Kα*-*Kβ*. Even the second maximum in *Kα*-*Kα* is reproduced when cascading transitions from high *n* states are included.[10] At higher energies (above the *KMM* resonance energy of 163 MeV) coincidences between two *Kβ* and higher x rays also become apparent as one may see in *Kβγ*-*Kβγ* (Fig. 2). This is evidence for $\Delta N \gtrsim 2$ transitions in RTE. The cross section for this process is about 1 order-of-magnitude smaller than that of *Kα*-*Kα*. At 175 MeV McLaughlin and Hahn[10] predict a cross section for *Kβ*-*Kβ* coincidences, that is about a factor of 3 smaller than found experimentally.

There are several possible reasons for the calculated RTE cross section to have underestimated the experimental results. First, we could not experimentally resolve transitions from higher *n* states which are as a result included in the measured RTE cross section. The calculation, however, includes only coincidences between the *Kβ* transitions and does not take into account direct transitions from higher states. Therefore it is possible that this discrepancy is a result of a significant population of very high *n* and *N* states, i.e., transitions with $\Delta n, N \gg 2$. However, because of the arguments stated above, high populations of even more highly doubly excited states are not to be expected.

Another possible reason for this discrepancy is a contribution from a nonresonant transfer and excitation process which competes in ion-atom collisions with RTE. In this process the transfer of the target electron occurs simultaneous with the excitation of the projectile electron, but not necessarily in resonance with an autoionizing intermediate doubly excited state. The nonresonant process can happen when the excitation is caused by either the target nucleus [nonresonant transfer and excitation[2] (NTE)] or for a two-electron target by the "other" target electron (2*e*TE).[14] McLaughlin and Hahn[10] estimate a cross section for NTE which is in the same order of magnitude as the one calculated for RTE at this energy (174 MeV) and for *Kβ*-*Kβ* coincidences. NTE can therefore not be completely ruled out on the basis of the cross section alone as a cause of the discrepancy. The energy dependence of NTE and 2*e*TE is governed by both the excitation and the capture processes, and should have a different energy dependence than RTE. It has been previously observed[2] in a similar collision system that this maximum for NTE is much wider and peaks at a much lower collision energy than for the RTE resonances. In view of this and because the measured cross sections tend very rapidly to zero at collision energies below the resonance energies for all measured x-ray transitions (see Fig. 2), NTE cannot be the process behind the cross sections measured here. As discussed in detail in Ref. 3, small NTE contributions to this data can only be noticed at 70 MeV well below the collision energy range studied in the present work. At 70 MeV we observe if any, only a small contribution of NTE to the $K\alpha$-$K\alpha$ transition,[3] and there, as mentioned above, is where the main contribution of NTE would be expected.

Finally, 2*e*TE transitions occur in the energy range containing the resonance energies of the high *n* states. Using the qualitative arguments described above one can estimate the absolute magnitude of this process to be roughly 1 order-of-magnitude smaller than the RTE cross section being therefore (if at all) only a small contribution to the cross sections measured here and it is not able to account for the observed discrepancy.

In conclusion, we have observed the population of highly doubly excited states with *both* electrons in a high *n* state in heliumlike S and their radiative decay by x-ray emission. The cross section as function of beam energy exhibits a resonance structure with a maximum close to the *KMM* transition energy. The calculations can reasonably well predict the energy position and shape of the resonance but there remain significant discrepancies in the calculated absolute value of the cross section for populating the high *n* states which need further clarification.

This research was sponsored by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Incorporate.

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[1] J. A. Tanis, E. M. Bernstein, W. G. Graham, M. P. Stöckli, M. Clark, R. H. McFarland, T. J. Morgan, K. H. Berkner, A. S. Schlachter, and J. W. Stearns, Phys. Rev. Lett. **53**, 2551 (1984).

[2] J. A. Tanis, Nucl. Instrum. Methods Phys. Res. Sect. A **262**, 52 (1987).

[3] M. Schulz, E. Justiniano, R. Schuch, P. H. Mokler, and S. Reusch, Phys. Rev. Lett. **58**, 1734 (1987).

[4] D. Brandt, Phys. Rev. A **27**, 1314 (1983).

[5] C. P. Bhalla and K. R. Karim, Phys. Rev. A **39**, 6060 (1989).

[6] M. Schulz, R. Schuch, S. Datz, E. L. Justiniano, P. D. Miller, and H. Schöne, Phys. Rev. A **38**, 5454 (1988).

[7] P. H. Mokler, S. Reusch, Th. Stöhlker, R. Schuch, M. Schulz, G. Wintermeyer, Z. Stachura, A. Warczak, A. Müller, Y. Awaya, and T. Kambara, Radia. Eff. Defects Solids **110**, 39 (1989).

[8] E. Justiniano, R. Schuch, M. Schulz, P. H. Mokler, S. Reusch, D. J. McLaughlin, and Y. Hahn, in *Electronic and Atomic Collisions,* edited by H. B. Gilbody, W. R. Newell, F. H. Read, and A. C. Smith (North-Holland, Amsterdam, 1987), p. 477.

[9] J. A. Tanis, E. M. Bernstein, M. Clark, W. G. Graham, R. H. McFarland, T. J. Morgan, J. R. Mowat, D. W. Mueller, A. Mueller, M. P. Stöckli, K. H. Berkner, P. Gohil, A. S. Schlachter, and J. W. Sterns, Phys. Rev. A **53**, 2543 (1986).

[10] D. J. McLaughlin and Y. Hahn, Phys. Rev. A **38**, 531 (1988); and (private communication).

[11] N. R. Badnell, Phys. Rev. A **42**, 209 (1990).

[12] R. Schuch, M. Schulz, E. Justiniano, H. Vogt, S. Reusch, and P. H. Mokler, Nucl. Instrum. Methods Phys. Res. Sect. B **23**, 140 (1987).

[13] C. P. Bhalla, Phys. Rev. Lett. **64**, 1103 (1990).

[14] M. Schulz, J. P. Giese, J. K. Swenson, S. Datz, P. F. Dittner, H. F. Krause, H. Schöne, C. R. Vane, M. Benhemi, and S. M. Shafroth, Phys. Rev. Lett. **62**, 1738 (1989).