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Resonant transfer and excitation in Li-like F colliding with H_2

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We have measured coincidences between x rays and projectiles that have captured one electron in $F^{6+}+H_2$ collisions at projectile energies between 15 and 33 MeV. The cross sections for capture and simultaneous x-ray emission as a function of projectile energy show clear structures. Indications of an unexpectedly high population of high-n states predominantly formed by resonant transfer and excitation (RTE) were found. Above the KLn ($n>1$) RTE resonance energies another maximum was observed.

Resonant transfer and excitation (RTE) (Ref. 1) has been studied intensively the past few years.[2-6] In this process, a weakly bound (quasifree) target electron excites a projectile electron and is itself captured to a bound state of the projectile. Capture and excitation proceed via an interaction between the two active electrons and are therefore correlated. In the approximation that the target electron is truly free, RTE followed by radiative decay to a singly excited state (RTEX) is equivalent to dielectronic recombination (DR) (Refs. 7-9) which can be considered as the time reverse of the Auger process. Both RTE and DR are resonant processes.

The fact that in RTE the target electron is not exactly free leads to a broadening of the resonances in the RTE cross sections by the momentum distribution (Compton profile) of the target electron. Therefore, in most RTEX experiments performed so far where coincidences between projectile x rays and the projectiles which have captured electrons or coincidences between two x rays were measured,[2-6] targets have been used which do not contain strongly bound electrons with a large momentum distribution, i.e., He and H_2.[2-6] But, even for these targets, the broadening is so strong that the resonances for most intermediate states overlap to yield in many cases just one broad maximum in the observed energy dependence of the cross sections. The energy separation between the intermediate states scales approximately as Z^2, whereas the width of the resonances only scales with Z. Therefore, the resolution of the different resonances should become better with increasing Z of the projectile. This is one reason why most experiments done so far were carried out with ions having the highest possible Z. Indeed two groups of resonances could be resolved for Li-like Ca (Ref. 3) and Ti,[5] and three groups were resolved in Li-like Ge.[6] It was also shown that for H-like S ions much better resolution can be obtained than for Li-like S ions.[4]

In the low-Z regime ($Z<14$), however, very little experimental work has been done yet on RTEX. The reason for this is that the detection of x rays is more difficult for x-ray energies below about 1 keV than for higher energies. With Si(Li) detectors, the efficiency for detecting x rays with energies <1 keV, as well as the energy resolution, is very poor. The only study on RTEX for $Z<10$ was performed by Pepmiller *et al.*[10] The x rays were measured there in high resolution with a crystal spectrometer, using a gas proportional detector. However, in that experiment, RTE resonances could not be conclusively observed. This was explained by large contributions from a competing process to RTE, which is called nonresonant transfer and excitation (NTE).

RTE, followed by Auger electron emission (RTEA) in collisions of $O^{5+}+He$ has been studied by Swenson *et al.*[11] In that work, the population of doubly excited states was investigated by using high-resolution electron spectroscopy to observe Auger electrons emitted from nonradiative decays of the doubly excited states. Clear evidence for resonances from the $[1s2s2p^2(^1D$ and $^3D)]^{**}$ states was found. However, the x-ray decay channel should look significantly different from the Auger decay channel because the Auger yield, being very close to unity for low-Z ions, does not vary very much for different doubly excited states. Thus, the Auger emission cross sections should reflect quite well the cross sections for initially populating the doubly excited states. For the x-ray decay channel, in contrast, the measured cross section and, in particular, the n-state dependence might be influenced to a large extent by large relative differences in the small fluorescence yields.

In this report, we present evidence for RTEX in the regime of $Z < 10$. This was possible by detecting the x rays with gas proportional counters which were optimized for good time resolution.

At the EN-tandem of the Oak Ridge National Laboratory, we obtained Li-like F beams at energies between 15 and 33 MeV. After collimating down to 2×2 mm^2, the beam passed through a differentially pumped, windowless H_2 gas target. Target pressures of up to 100 mTorr were used. The length of the gas cell was 70 mm. The emergent beam was then charge-state analyzed by an electrostatic analyzer. Projectiles that had captured one electron were detected by a ceratron; the beam fraction that did not undergo charge exchange was collected in a Faraday cup and used for normalization.

The x rays were measured by two gas proportional counters. A mixture of 90% Xe and 10% CO_2 at a pressure of about 2 atm was used as counter gas. The gas volume was separated from the vacuum by a 3.5-μm-thick Mylar foil. A plate with a backgammon structure served as the cathode. About 2 mm above the cathode tungsten wires were stretched separated from each other by 4 mm across the active area. The detectors measured 70 mm in the beam direction and 30 mm perpendicular to the beam direction. A potential of 1.3 kV was applied to the wires.

The fast signals of the x-ray detectors and the ceratron particle detector were measured in timing coincidence (XP coincidence). The data consisting of the XP coincidence time spectra, the total count rate of the ceratron, and the integrated beam current were stored on a VAX 11/750.

In order to determine the detection efficiencies ϵ_x of the x-ray detectors, data were also taken for bare F projectiles colliding with H_2. Except for capture to the K shell or to metastable states each projectile with a charge reduced by one unit should lead to an x ray. Therefore, neglecting capture to the K shell and to metastable states, which is estimated to be not more than 10% of the total capture,[12] ϵ_x is given by the ratio of the true XP coincidence count rate to the count rate of the projectiles that have captured one electron. For each detector, we find a value of $\epsilon_x = 7\times10^{-5}$.

In Fig. 1, a time spectrum of the XP coincidences is shown for 25 MeV $F^{6+}+H_2$. A clear peak representing the true coincidences can be seen at about 110 nsec on top of a flat background from random coincidences. The width of the time peak indicates a time resolution of the x-ray detectors of better than 20 nsec. It is this good resolution which made it possible to observe a time peak with a true to random ratio of up to 4:1 (depending on beam energy).

The true XP coincidences are a signature of capture and simultaneous emission of an x ray following an excitation process. The measured cross sections σ^x_{q-1} for this process are given by

$$\sigma^x_{q-1} = N_{XP}/N_0 \Delta x\, \epsilon_x \epsilon_p\ . \qquad (1)$$

Here N_0 is the initial beam and Δx the target thickness. ϵ_p is the efficiency of the particle detector. N_{XP} is the number of true coincidences which is obtained by integrating the time peak of the coincidence spectra and subtracting the contributions from random coincidences in the region of the time peak.

In Fig. 2, σ^x_{q-1} is plotted versus projectile energy. The error bars shown are only statistical errors. The systematic errors, mainly due to the uncertainty in ϵ_x and Δx, we estimate to be 30%. In the data, clear structures can be

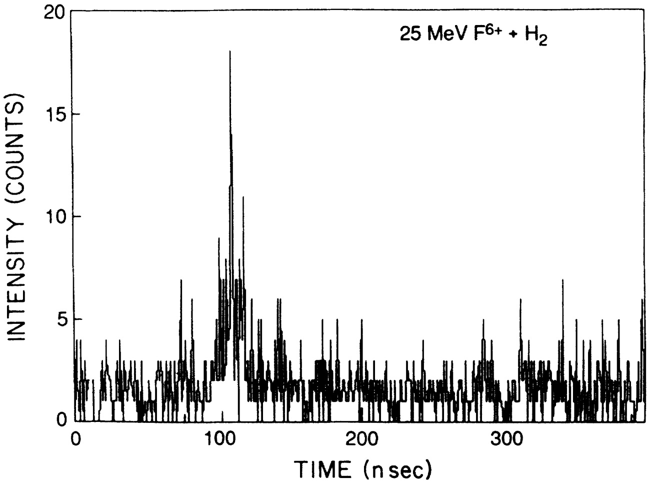

FIG. 1. Time spectrum of the x-ray particle (XP) coincidences for 25-MeV $F^{6+}+H_2$.

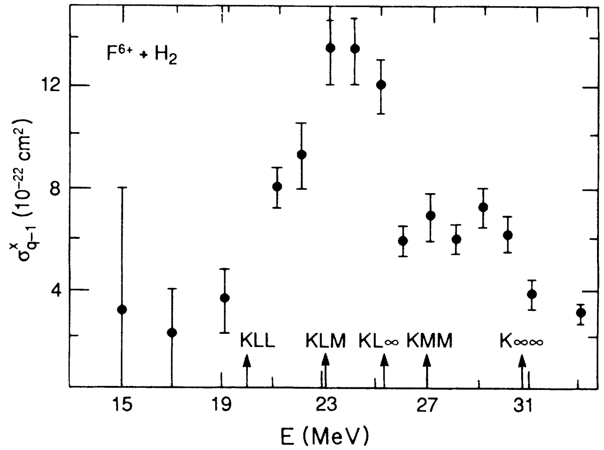

FIG. 2. Measured cross sections for capture and simultaneous emission of an x ray vs projectile energy. The arrows indicate the resonance energies for some intermediate states.

seen. There is a pronounced maximum at about 24 MeV and a shoulder around 28 MeV. The width of the first maximum is about 6 MeV full width at half maximum (FWHM). The arrows in this figure indicate projectile energies for which RTE resonances are expected. As it is commonly used, we also employ Auger notation for the intermediate states populated; e.g., *KLL* means a *K* electron is excited to the *L* shell and an electron is also captured to the *L* shell. The first maximum in the data falls in a projectile energy range where RTE resonances for the *KLn* ($n > 1$) series are expected.

However, in addition to RTEX, the competing process NTE (Ref. 13) followed by x-ray emission (NTEX) can also contribute to σ_{q-1}^{x}. In this process, as in RTE, a target electron is captured to the projectile and simultaneously a projectile electron is excited. However, here the excitation is due to an interaction with the target nucleus and is thus not correlated with the capture process. Estimates for the NTE cross sections, following the method of Brandt,[14] show that its projectile energy dependence should look quite different from the one for RTE cross sections.[10] For NTE, a maximum is expected at energies much lower than the RTE resonance energies; for the collision system studied here, the NTEX maximum should be at about 8 MeV.[10] At the low-energy side of the NTEX maximum, the cross section decreases steeply. The high-energy side, however, displays a much flatter decline giving rise to a strongly asymmetric and much broader (> 10 MeV FWHM) maximum than the RTE resonances. Such a projectile energy dependence was indeed observed for the collision system S^{13+}+He.[15]

The projectile energy dependence of our data clearly look much more like that expected for RTEX than that for NTEX. The first maximum matches in its position the resonance energies for populating *KLn*($n > 1$) states by RTE, it has a shape that is not strikingly asymmetric, and its width agrees quite well with the calculated Compton width of a single RTE resonance (5.1 MeV FWHM using the Compton profile for H_2 from Eisenberger).[16] Furthermore, according to a calculation using the method of Brandt,[14] the contribution from NTEX, in our case, should be only < 5% in the region of the RTE maximum.

In the work of Pepmiller *et al.*[10] where the collision system F^{8+}+He was studied, a dominance of NTEX over RTEX was found. It should be noted, however, that the NTEX cross sections decrease strongly with the target *Z*. Therefore, it is expected that with an H_2 target used in our experiment the NTEX cross sections are smaller than for He by about a factor of 8.

One interesting result is that apparently the *KLL* resonances do not give the major contributions to the measured cross sections. At the resonance energies for these states the data just barely start rising, having a cross section of at most $\frac{1}{3}$ of the value of the maximum. Judging from the position of the maximum, the main contributions seem to come from the *KLM* states. Also, given the energy separation between the *KLL* and *KLM* resonances of 3 MeV, the observed width of 6 MeV FWHM can be taken as an indication that the *KLL* resonances are not very strong. With a Compton width of a single resonance of 5.1 MeV, the maximum should have a width of at least 8 MeV if the *KLL* resonances were comparable to the *KLM* resonances. The maximum is even close to the resonance energies for populating $KL\infty$ states, where ∞ denotes a high *n* state near the Rydberg limit. This shows that even higher *n* states than *KLM* might have considerable contributions to the measured cross sections. The importance of high *n* states in RTEX has been demonstrated in previous experiments[3-6] and is also expected from theoretical calculations.[17,18] However, it has never been observed in such a pronounced form as in the present data, in particular the dominance of the higher *n* states over the *KLL* states.

In principle, one could think of two reasons for the small contributions from the *KLL* resonances. Either these states are much less populated than higher *n* states or the fluorescence yields are much smaller than for higher *n* states. The population cross section should be proportional to the Auger rate λ_A, which in turn scales approximately like n^{-3}.[19] Therefore, the population of *KLL* states should be larger compared to higher *n* states. The fluorescence yield, ω, is given by

$$\omega = \lambda_x / (\lambda_A + \lambda_x) ,$$

where λ_x is the radiative rate. A *KLn* state can decay radiatively by a *L* to *K* or an *n* to *K* transition. Since the rate of the *L* to *K* transition is dominant over the *n* to *K* transition rate,[20] one should mainly observe $K\alpha$ x rays independent of *n* of the *KLn* states. The relevant λ_x in ω is then obviously independent of *n*. Therefore, for $\lambda_x \ll \lambda_A$ which is the case for low *Z* ions, ω scales approximately as n^3. For a single resonance, the RTEX cross section is then approximately independent of *n*. Since with increasing *n* more states are contributing in a given energy range, pile up of these states would indeed lead to a dominance of higher *n* states over the *KLL* states. One should note that this argument no longer holds for very high *n* states where the condition $\lambda_A \gg \lambda_x$ is not fulfilled.

This latter condition also loses validity with increasing *Z*. In the other extreme case, $\lambda_x \gg \lambda_A$, the RTEX cross section should scale like λ_A, i.e., like n^{-3}. Therefore, the relative cross section for the *KLL* resonances compared to the higher *n* state resonances should systematically increase with increasing *Z*. Such systematics can indeed be seen in those experiments, where the *KLL* resonances could be resolved from the higher *n* state resonances.[3-6] Theoretical calculations also predict that the ratio of the *KLL* resonances to the sum of all higher *KLn* resonances decreases with decreasing *Z*.[21] Extrapolating the values for $Z = 14$–26 given in Ref. 21 down to $Z = 9$ yields a value for the *KLL* to *KLn* ratio of $\simeq 0.4$–0.5, which is not inconsistent with our data.

Another surprising result in the present data is the shoulder at about 28 MeV, which is above the *KLn* series limit. The energy range for these contributions coincides with the resonance energies for populating *Knm* ($n,m > 2$) states by RTE. Nevertheless, it is not evident that the shoulder can be entirely attributed to these resonances. It is commonly held that the cross sections for the *Knm* ($m \geq n$) RTE resonances should drop off relatively fast with increasing *n*. Even for the sum of all resonances with $n > 2$, half the contribution of the *KLm* series as seen in our data was not necessarily expected. Also in all experimental studies performed so far, resonaces above the *KLm* series limit could never be identified (e.g., Refs. 2–6).

Hahn and McLaughlin[22] have proposed that there is another competing process to RTE, which we term two electron transfer and excitation (2*e*TE), which might yield considerable contributions to σ^x_{q-1} near the resonance energies for the *Knm* $(n,m > 2)$ states. Again, in this process, a target electron is captured by the projectile and simultaneously a projectile electron is excited. Here, however, the excitation is not due to an interaction with the captured electron, as in RTE, or with the target nucleus, as in NTE, but with a second target electron. Thus, in 2*e*TE excitation and capture are not correlated and no resonant behavior is expected. If the binding energy of the target electrons is neglected, then 2*e*TE should have a threshold at the *K* to *L* excitation energy (25.5-MeV projectile energy in the electrons rest frame).

There are no accurate numerical calculations available yet for 2*e*TE cross sections. Rough estimates of 2*e*TE cross sections (using calculated cross sections for excitation by free electrons[23] and Oppenheimer-Brinkman-Kramers capture probabilities) show that the magnitude near the threshold (2×10^{-22} cm^2) is consistent with the magnitude of the shoulder observed in the data. On the other hand, the present data do not provide conclusive evidence for either 2*e*TE or high cross sections for the *Knm* $(n,m > 2)$ RTE resonances. This question needs further investigation.

In summary, we have found evidence for RTE in $F^{6+} + H_2$ collisions. We observed an unexpectedly strong contribution from capture to high *n* states. The lowest lying states (*KLL*) in contrast, display surprisingly small contributions to the measured sections. Above the *KLn* series contributions were observed which might be due to *Knm* $(n,m > 2)$ RTE resonance or to a process called 2*e*TE which has not been observed before.

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