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# Cascade theory for double *K* x-ray emission in transfer and excitation collisions

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We propose a cascade model to interpret a recent, novel experiment of $S^{15+}+H_2$ collisions with simultaneous excitation capture in which two *K* x rays were detected. Cascade effects leading to double *K* x-ray emission are carefully examined to obtain $K\alpha$-$K\alpha$, $K\alpha$-$K\beta$, and $K\alpha$-$K\gamma$ (and higher states) coincidence cross sections. The overall spectral shapes and absolute magnitudes are consistent with the predictions of the model.

Resonant electron transfer and excitation (RTE) processes in ion-atom and ion-molecule collisions have been extensively investigated experimentally in recent years with coincidence techniques[1-7] where the stabilizing x rays and charge-changed ions were detected as the signature of RTE. The mechanism for this correlated excitation-capture process can be viewed as an electron-electron interaction between a projectile electron and a *bound* electron in the target, with simultaneous capture of the same target electron, resulting in a doubly excited, autoionizing intermediate resonance state which subsequently decays by either photon emission (RTEX) or by electron emission (RTEA). This is described schematically by

$$\begin{aligned} A^{Z+}+B &\Rightarrow (A^{(Z-1)+})^{**}+B^{+} \quad \text{(RTE)} \\ &\hookrightarrow (A^{(Z-1)+})^{*}+\gamma \quad \text{(RTEX)} \\ &\hookrightarrow (A^{Z+})^{*}+e' \quad \text{(RTEA)}\,. \end{aligned} \tag{1}$$

RTE is, effectively, the inverse of Auger ionization in the analogous electron-ion collision and is thus resonant for ion velocities corresponding to those of the ejected Auger electrons.[8] The resonant capture of a *free* electron in electron-ion collisions results in a doubly excited intermediate state, which similarly decays either radiatively by photon emission [dielectronic recombination (DR)][9,10] or by electron emission [resonance excitation (RE)] as described by

$$\begin{aligned} e+A^{Z+} &\Rightarrow (A^{(Z-1)+})^{**} \\ &\hookrightarrow (A^{(Z-1)+})^{*}+\gamma \ \text{(DR)} \\ &\hookrightarrow (A^{Z+})^{*}+e' \ \text{(RE)}\,. \end{aligned} \tag{2}$$

Direct observation of DR has proven difficult because of the small cross section. Only the field-enhanced[11] cases with intrashell excitations ($\Delta n_t=0$) have been observed,[12-15] and one preliminary result was reported in which excitations to different shells ($\Delta n_t\neq 0$) were involved.[16] On the other hand, RTEX has been uniquely successful[17] in indirectly measuring the DR cross sections for the $\Delta n_t\neq 0$ case. As noted above, the stabilizing radiation and charge-changed ions ($A^{(Z-1)+}$) are measured in coincidence.[5] Agreement between theory and experiment has been excellent.[18] In addition, preliminary data[19] on RTEA were also analyzed[20] in terms of the corresponding resonance excitation (RE) process, as indicated in (1) and (2).

An ingenious, new measurement of RTE was reported[21] in which two *K* x rays were detected in coincidence as the signature of RTE, rather than the *K*-x-ray–charge-changed-ion coincidence measurement. This process is denoted as RTEXX. Here we propose a cascade model for the analysis and interpretation of this experiment and confirm its potential utility as a state selective study of both RTE and DR. Considering the fact that both the experimental approach described in Ref. 21 and the theoretical model are new, the agreement we obtain here is sufficient enough to show that the model is essentially a correct representation of the experiment. The cascade theory described here has greatly helped to clarify the physics involved and some of the difficulties in the experimental procedure encountered at the early stage. Both our calculation and the final data quoted here are the result of many refinements in theory and experiment. The process of interest is

 

$$
\begin{array}{l}
S^{15+}(1s)+H_2 \Longrightarrow [S^{14+}(2pnl)]^{**}+H_2{}^{+} \\
\qquad\qquad\qquad \lfloor\!\longrightarrow S^{14+}(1s^2)+\gamma_K'+\gamma_K'' \ \text{(RTEXX)} \\
\qquad\qquad \lfloor\!\Longrightarrow S^{15+}+e'+\gamma_K \ \text{(RTEAX)}\,.
\end{array}
\tag{3}
$$

In order to produce two $K\alpha$ photons, the recombined ion $(S^{14+})^{**}$ has to go through several cascade decays. Generally, there are many cascade channels available for each of the intermediate states. Therefore, although the initial state of resonant excitation capture is identical to that in RTEX (and/or DR), RTEXX is treated quite differently in so far as the decaying aspect is concerned. In general, $\sigma^{\text{RTEXX}}$ is expected to be smaller than $\sigma^{\text{RTEX}}$, as will be discussed in more detail below.

The RTEXX cross section is obtained from the corresponding electron-ion cross sections by folding the DRX cross section over the Compton profile of the target. Thus, we have the DRX process

$$
e^- + S^{15+}(1s) \rightarrow [S^{14+}(2pnl)]^{**} \rightarrow \cdots \rightarrow S^{14+}(1s^2)+\gamma_K'+\gamma_K''\,,
\tag{4}
$$

and the cross section is given, in the isolated resonance approximation,[9] by

$$
\sigma^{\text{DRX}} = \sum_d \left[ \frac{4\pi^2 a_0^2}{(p_c a_0)^2} \right] V_a(i\rightarrow d)\tilde{\delta}(E-E_d,\Gamma) \sum_{d_1,\ldots,d_m} \omega(d\rightarrow d_1)\omega(d_1\rightarrow d_2)\cdots\omega(d_m\rightarrow f) \equiv \sum_d \sigma_d^{\text{DRX}}\,,
\tag{5}
$$

where $\omega(d_j\rightarrow d_k)$ is the partial x-ray yield given by $\omega_{jk}=A_r(d_j\rightarrow d_k)/[\Gamma_a(d_j)+\Gamma_r(d_j)]$ with the radiative and Auger transition probabilities $A_r$ and $A_a$, and $\Gamma_r=\Sigma A_r$ and $\Gamma_a=\Sigma A_a$. In (5), $V_a$ is the probability for simultaneous excitation-capture, and $\delta$ is the normalized Lorentzian profile,[9] $p_c$ is the momentum of the continuum electron, and $e_c=(p_c[\text{Ry}])^2$. The RTEXX cross section is then given by[8]

$$
\sigma^{\text{RTEXX}} \approx \sum_d W(q_{dz})\bar{\sigma}_d^{\text{DRX}}\Delta e_c[\text{a.u.}] \left[ \frac{M_{\text{ion}}[\text{a.u.}]}{2E_{\text{ion}}[\text{a.u.}]} \right]^{1/2}
\tag{6}
$$

where $W$ is the Compton profile of the target $B$, $\bar{\sigma}_d^{\text{DRX}}$ is the energy averaged DRX cross section $\sigma_d^{\text{DRX}}\equiv(1/\Delta e_c)\times\int\sigma_d^{\text{DRX}}de_c'$, $E_{\text{ion}}$ is the ion kinetic energy in the lab frame, and

$$
q_{dz}=(e_c-E_{\text{ion}}m_e/M_{\text{ion}}) \left[ \frac{M_{\text{ion}}[\text{a.u.}]}{2E_{\text{ion}}[\text{a.u.}]} \right]^{1/2}\,.
$$

Specifically, the cascade structures for the $K\alpha - K\alpha$ process, for example, are

$$
\begin{array}{l}
1s+k_cl_c \overset{V_a}{\Longrightarrow} 2p^2 \xrightarrow{\omega_{12}} 1s\,2p \xrightarrow{\omega_{23}\approx 1} 1s^2 \ \text{(dominant)} \\
\quad \lfloor\!\overset{V_a}{\Longrightarrow} 2p\,3s \xrightarrow{\omega_{45}} 1s\,3s \xrightarrow{\sim 1} 1s\,2p \xrightarrow{\sim 1} 1s^2 \ \text{(dominant)} \\
\qquad\quad \lfloor\!\xrightarrow{\omega_{46}} 2p^2 \xrightarrow{\omega_{67}} 1s\,2p \xrightarrow{\sim 1} 1s^2,\ \text{etc.}\,,
\end{array}
\tag{7}
$$

and so on for the $2p3d$, etc. The $2p3p$ contribution is small, but not $2pnl$, $n\geq 4$, and $l\geq 0$. Thus we have, for example,

$$
\begin{array}{l}
1s+k_cl_c \overset{V_a}{\Longrightarrow} 2p4s \xrightarrow{\omega} 1s4s \not\longrightarrow 1s\,2p \longrightarrow 1s^2 \ \text{(dominant)} \\
\qquad\qquad\qquad\qquad\quad \lfloor\!\not\longrightarrow 1s\,3p \longrightarrow 1s\,3s \longrightarrow 1s\,2p \longrightarrow 1s^2 \\
\qquad\qquad\quad \lfloor\!\xrightarrow{\omega} 2p\,3p \longrightarrow 1s\,3p \not\longrightarrow 1s\,3s \longrightarrow 1s\,2p \longrightarrow 1s^2 \\
\qquad\qquad\qquad\quad \lfloor\!\longrightarrow 2p\,3s \longrightarrow 2p^2 \longrightarrow 1s\,2p \not\longrightarrow 1s^2 \\
\qquad\qquad \lfloor\!\xrightarrow{\omega} 2p^2 \longrightarrow 1s\,2p \not\longrightarrow 1s^2,\ \text{etc.}
\end{array}
\tag{8}
$$

It turns out that, in all cases, the decaying modes corresponding to the decay branches where the initial $2p\rightarrow 1s$ occurs at the earliest possible stage, dominates the process. By contrast, the DR cross section for the $2p4s$ state, for example, is simply given by the truncated cascade series in (8), as marked by the slashes. This difference is very important from a theoretical point of view, because RTEXX is a totally new way of sampling the structure of the cascade process, thus providing a different handle in the study of the TE process.

The $K\alpha$-$K\beta$ processes are given, for example, as

$$
\begin{array}{l}
1s+k_cl_c \overset{V_a}{\Longrightarrow} 2p\,3p \xrightarrow{\omega_{12}} 1s\,3p \xrightarrow{\omega_{23}\approx 1} 1s^2 \\
\qquad\qquad\qquad \lfloor\!\longrightarrow 1s\,2p \xrightarrow{\sim 1} 1s^2\,,
\end{array}
\tag{9}
$$

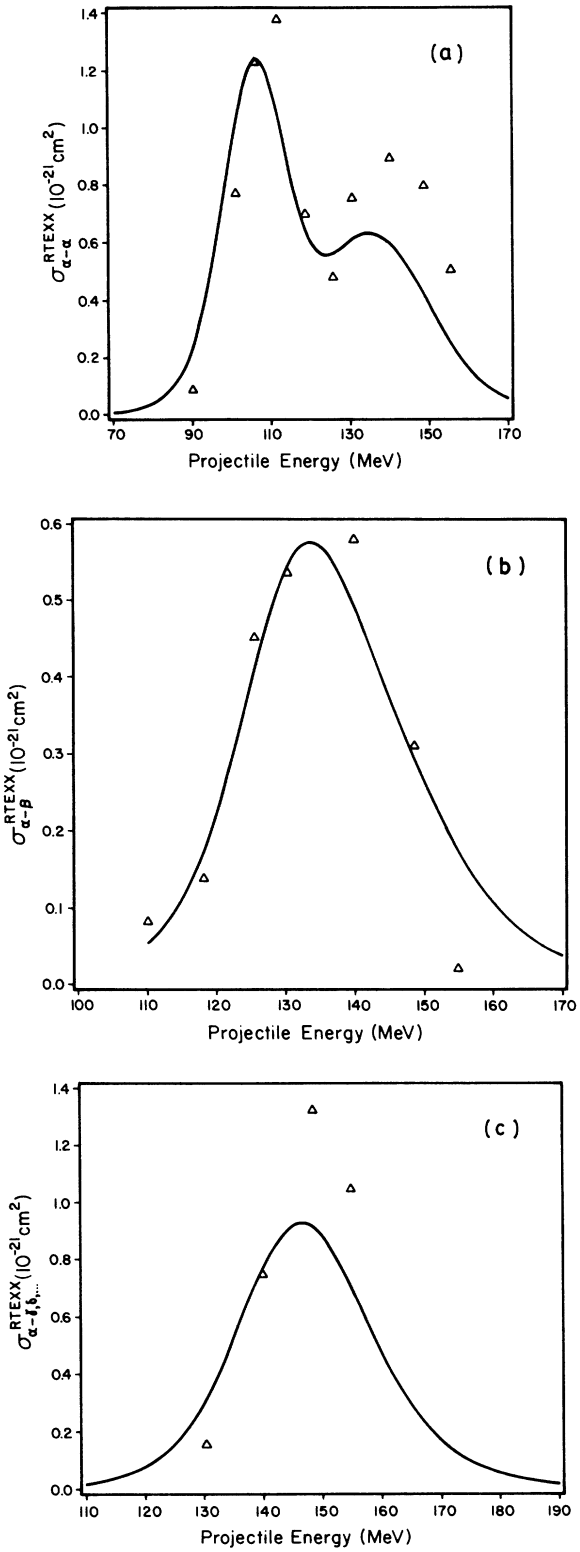


FIG. 1. (a) RTEXX cross sections for capture into doubly excited intermediate states, which cascade to produce $K\alpha$-$K\alpha$ coincidences, are presented as a function of projectile energy (MeV). The first peak at 106 MeV corresponds to *KLL* resonances. The second peak centered around 135 MeV corresponds to KLM, KLN, and higher resonances that cascade to $K\alpha$-$K\alpha$ emission. The triangles represent coincident technique measurements. (b) Cross sections for $K\alpha$-$K\beta$ cascade decays. (c) Cross sections for $K\alpha$-$K\gamma$, and higher, cascade decays.

and so on, and for $K\beta$-$K\beta$. The intermediate states are of the types $3p^2$ and $3pnl$ with $n \geq 3$. The calculation showed that the cross section for $K\beta$-$K\beta$ emission is very small, which is in agreement with experimental data. Many cascade processes similar to (7)–(9) were also considered for $K\alpha$-$K\gamma$ and higher states, and again found to be quite different from the DR and RTEX modes.

The calculation of $\sigma^{\mathrm{DRX}}$ was carried out[10] in a single-configuration, nonrelativistic, Hartree-Fock approximation in LS coupling, and the Auger rates were evaluated by a distorted-wave method. All the cascade contributions were explicitly included for the initial intermediate states $(2pnl)$ with $n \leq 6$, and the higher $n \geq 7$ contributions were estimated by using the $n$-scaling behavior.[9] The role of metastable states, such as $1s2s$ produced by $2s2p \rightarrow 1s2s + K\alpha$ followed by $1s2s \rightarrow 1s^2 + 2\gamma$ is negligible and is excluded because the probability of one of these photons having the $K\alpha$ energy is small. Similarly, other metastables such as $2p^3(^3P)$ were also considered and found to give marginal contributions for the experimental time window of 250 ps. The metastable $2s$ component in the $S^{15+}$ beam makes only nonresonant contributions to the background and not to RTEXX. Figure 1(a) contains the energy averaged cross section $\bar{\sigma}^{\mathrm{DRX}}$ for the $K\alpha$-$K\alpha$ measurement, with $\Delta e_c = 1$ Ry obtained by folding the cross section with the $H_2$ Compton profile[22] in accordance with (6). For comparison, experimental data of Schultz *et al.*[22] are also included. The positions of both the first and second peaks, as well as the overall profile and the relative peak heights, were very well reproduced. Figure 1(b) contains the theoretical and experimental cross sections for the $K\alpha$-$K\beta$ emission, and Fig. 1(c) for the $K\alpha$-$K\gamma$ (and higher states) emission. Agreements here are even better than the $K\alpha$-$K\alpha$ case. However, some discrepancy remains in spite of our extensive collaborative efforts with experimentalists to understand the problem. The reasons for this discrepancy are not clear at this stage, particularly in the case of the second peak in the $K\alpha$-$K\alpha$ data. Further refinements in both experiment and theory may clarify this, but as Figs. 1(a)-1(c) clearly show, we are confident that the basic assumptions of the model are correct, as reflected both in absolute magnitude and in the locations of the peaks.

The new experimental technique introduced in Ref. 21 to measure the RTEXX cross section by detecting the two $K$ x rays in coincidence is shown to be a viable approach to RTE and an important alternative to the x-ray–charge state coincidence measurements of the earlier RTEX studies. Although the two groups of excitation modes involving the intermediate states $(2p2l)$ and $(2pnl)$ with $n \geq 3$ are resolved, the fine structures were not separated due to the wide Compton profile. However, the present experiment samples the DR process in a *different* form, providing an additional handle on the choice of intermediate states to be examined. Furthermore, the cascade process plays an all-important role here, which provides a stringent test of the theory employed. Considering the fact that this is the first experiment of this kind, the approach is very promising and, with some further refinements, can provide valuable information on RTE and DR. Details of the calculation will be given elsewhere.

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