Double x-ray emission following resonant transfer and excitation in collisions of H-like ions with H_2

N. R. Badnell
Department of Physics, Auburn University, Auburn, Alabama 36849-5311
(Received 12 October 1990)

We have solved the capture-cascade equations for double x-ray emission following resonant transfer and excitation in collisions of S^{15+} and Ge^{31+} with H_2. Configuration mixing within the autoionizing complex enhances the cascade-produced $K\alpha$-$K\alpha$ peak and removes the discrepancy between single-configuration *LS*-coupling results and experiment for S^{15+}. Intermediate coupling increases the population of the metastables and so reduces the $K\alpha$-$K\alpha,K\beta,K\gamma$ cross sections by up to 20% over the *LS*-coupling results. The coincidence cross sections for highly excited states are still about a factor of 2 ($K\alpha$-$K\gamma$) or 3 [$(K\beta+K\gamma)$–$(K\beta+K\gamma)$] smaller than experiment for S^{15+}, however. In the case of Ge^{31+}, the total coincidence cross section lies 10–30% below experiment, while the $K\alpha$–$(K\beta+K\gamma)$ cross section is now somewhat larger than experiment. The comprehensiveness of the theoretical calculations suggests that the experimental results may need to be reexamined where significant discrepancies with theory remain.

I. INTRODUCTION

Resonant transfer and excitation in atom-ion collisions and dielectronic recombination in electron-ion collisions have been studied extensively in recent years, both theoretically and experimentally. However, little attention has been paid to the subsequent radiative cascade of the singly excited state that remains after stabilization, even though it may be a useful diagnostic in its own right, as witnessed by theoretical studies [1] of dielectronic recombination at low temperatures in connection with ultraviolet emission in gaseous nebulae. The first laboratory measurement [2] of coincident double x-ray emission following resonant transfer and excitation, for collisions of S^{15+} with H_2, was found to be in broad agreement with theoretical results [3] based on a single-configuration *LS*-coupling approximation, but some significant discrepancies remained. Recently, measurements [4,5] were made for Ge^{31+} on H_2 and these were partially analyzed theoretically [6], and further experiments are in progress.

The sensitivity of the dielectronic recombination of H-like ions to both configuration mixing and intermediate coupling has been noted [7] recently. We now investigate these effects on the double-emission problem. We have already [8] solved the capture-cascade equations in configuration-mixing and intermediate-coupling approximations for dielectronic recombination at low temperatures. In this paper, the Maxwellian source term is replaced by the Compton profile for H_2 and we also need to keep track of the type of K x-ray emission. We outline the required theory in Sec. II and its application to the present calculations in Sec. III. We present our results in Sec. IV and we conclude in Sec. V.

II. THEORY

Consider an ion X^{Z+} whose states are populated by resonant transfer and excitation, from an ion $X^{(Z+1)+}$ in an initial state i, and the subsequent radiative cascade. We may define a cross section $\sigma_x(j)$ for populating a state j and a cross section $\sigma_x(h\rightarrow j)$ for $h\rightarrow j$ photon emission such that, if j is bound,

$$\sigma_x(j)=\sum_{h>j}\sigma_x(h\rightarrow j)\ , \tag{1}$$

where

$$\sigma_x(h\rightarrow j)=\frac{A_r(h\rightarrow j)}{\sum_{k<h}A(h\rightarrow k)}\sigma_x(h) \tag{2}$$

and the sum of transition probabilities in the denominator of Eq. (2) contains both radiative A_r and, if energetically possible, autoionization A_a rates.

If j is autoionizing,

$$\sigma_x(j)=R_c(i;j)\tau_0\sum_l A_a(j\rightarrow i,E_c l)+\sum_{h>j}\sigma_x(h\rightarrow j)\ , \tag{3}$$

where R_c is given by

$$R_c(i;j)=(\pi a_0)^2\frac{\omega(j)}{\omega(i)}J(Q)\left[\frac{I}{E_c}\right]\left[\frac{MI}{E}\right]^{1/2}. \tag{4}$$

$J(Q)$ is the Compton profile of the target gas with Q given by

$$Q=\frac{1}{2I}\left[E_c+E_t-\frac{Em}{M}\right]\left[\frac{MI}{E}\right]^{1/2}, \tag{5}$$

E is the projectile-ion energy in the laboratory frame, E_c is the $j\rightarrow i$ Auger energy, and E_t is the binding energy of the target electron, both in the rest frame of the projectile. M is the ionic mass, m is the electron mass, $\omega(j)$ is the statistical weight of the $(N+1)$-electron doubly excited state, $\omega(i)$ is the statistical weight of the N-electron initial state, I is the ionization potential of the hydrogen atom, and a_0 and τ_0 are the Bohr radius and time with $(\pi a_0)^2\tau_0 = 6.6851\times10^{-33}$ cm^2 sec. The required transition probabilities and energy levels are evaluated in configuration-mixing, *LS*-coupling, and intermediate-coupling approximations using the AUTOSTRUCTURE package [9,10].

The above theory assumes an isotropic source of populating electrons. Allowance for the effect of an anisotropic source on single-photon emission is straightforward, and has been found to be small [11]. In practice, it is nontrivial to apply the same theory to the cascade-produced peak since it would now be necessary to keep track of the magnetic quantum numbers during the cascading. This is beyond the scope of the computer codes used here. However, cascading redistributes the initial population of the magnetic sublevels and so tends to reduce the initial anisotropy, which is small in any case [11].

III. APPLICATION TO H-LIKE IONS

The double *K*-shell vacancy following resonant transfer and excitation in collisions of H-like ions with H_2 gives rise to the possibility of the emission of two coincident *K* x rays. The total cross section for *K*-*K* emission is given by Eq. (1) with $j = 1s^2\,{}^1S_0$. If particular coincidences of emission were observed, e.g., $K\alpha$-$K\alpha$, $K\alpha$-$K\beta$, . . . , it is also necessary to keep track of the type of *K* x ray emitted when solving the capture-cascade equations.

We included all of the following configurations in our calculations:

$$nl + kl_c,\quad n = 1-4,\quad 0 \le l < n,\quad l_c = 0-7$$

$$2snl, 2pnl,\quad n = 2-6,\quad 0 \le l < n$$

$$3snl, 3pnl, 3dnl,\quad n = 3-6,\quad 0 \le l < n$$

$$4pnl,\quad n = 4-6,\quad 0 \le l < n$$

$$5pnl,\quad n = 5-6,\quad 0 \le l < n$$

$$1snl,\quad n = 1-6,\quad 0 \le l < n\ .$$

The contribution from states with $n > 6$ was included by extrapolation of the $n = 6$ data. For the autoionization rates, the 1*s*, 2*s*, and 2*p* orbitals were taken to be hydrogenic and the continuum and $(n > 2)$ Rydberg orbitals were taken to move in a local potential generated by an $n = 1$ or 2 Slater-type orbital. For the radiative rates, all of the orbitals were taken to be hydrogenic. The reasons for these choices are as follows. We are constrained by the angular-momentum algebra of the SUPERSTRUCTURE code [12] to use a unique set of orbitals to represent all of the configurations used to calculate the autoionization rates, and a separate unique set to calculate all of the radiative rates. We found that the configuration mixing within the $2:n:(n > 2)$ complexes was sensitive to the structure used, and so the orbitals were chosen so as to best represent that structure. This may not be a particularly good choice for the $3l'nl$ configurations, but it turns out that they make only a minor contribution to the $K\alpha$-$K\alpha$, $K\beta$,$K\gamma$ coincidences. For the $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ coincidences, we actually took the 3*s*, 3*p*, and 3*d* orbitals to be hydrogenic as well, and the $(n > 3)$ Rydberg orbitals were taken to move in a local potential generated by an $n = 3$ Slater-type orbital. This is not an optimum choice for the $2l'nl$ configurations, but again, they only have an indirect effect on the $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ coincidences. For the radiative rates, we found that the radiative cascades through singly excited states $(1snl \rightarrow 1sn'l')$ were poorly described if orbitals with different principal quantum numbers, but the same angular momentum, were calculated in different potentials, e.g., 2*p* hydrogenic but 3*p* moving in a 1*s* potential, because the configuration mixing between complexes was overestimated. In fact, it makes little difference whether the orbitals are all hydrogenic or whether they all move in a 1*s* potential, say, as long as they all move in the same potential.

In general, we only include electric dipole radiative transitions, since the nonmetastable excited states can always decay this way. The $2\,{}^3S_1$ metastable level can decay through relativistic corrections to the magnetic dipole operator. It depends both on the ion charge and the experimental setup whether the $2\,{}^3S_1$ level decays during the time of flight for which it is capable of being detected. If it does decay, it is generally indistinguishable from other $K\alpha$ radiation observed by the experiments considered here. The back-to-back two-photon emission of the $2\,{}^1S_0$ level gives rise to a characteristic signal [4,5], if indeed it does decay during the lifetime of the experiment. If there were no metastables present, then the cross section for double x-ray emission would be the same as the cross section for single x-ray emission and it would not be necessary to solve the capture-cascade equations to determine the populations of the ground and metastable levels.

IV. RESULTS

A. S^{15+}

We present our results for $K\alpha$-$K\alpha$, $K\alpha$-$K\beta$, $K\alpha$-$K\gamma$ and $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ coincidences in Figs. 1–4 and compare them with experiment [2,11]. We first note that our single-configuration *LS*-coupling results for $K\alpha$-$K\alpha$,$K\beta$,$K\gamma$ coincidences differ by less than 10% from those of similar earlier [3] calculations. It has already been noted [7] that configuration mixing within the autoionizing complex enhances the weak capture cross sections in the dielectronic recombination of H-like ions. This now produces a 60% increase in the cascade-produced $K\alpha$-$K\alpha$ peak over the single-configuration results; the contributing configurations are $2l'nl$, $n > 2$ and all *l*. However, configuration mixing has little effect on the $K\alpha$-$K\beta$ and $K\alpha$-$K\gamma$ coincidence cross sections, since these are dominated by autoionizing configurations of the form 2*pnp*.

The effect of intermediate coupling on double x-ray cross sections is quite different from that on single x-ray cross sections. Again, intermediate coupling enhances the weak capture cross sections and this invariably translates into an increase in the single x-ray cross section, since all of the doubly excited levels can decay

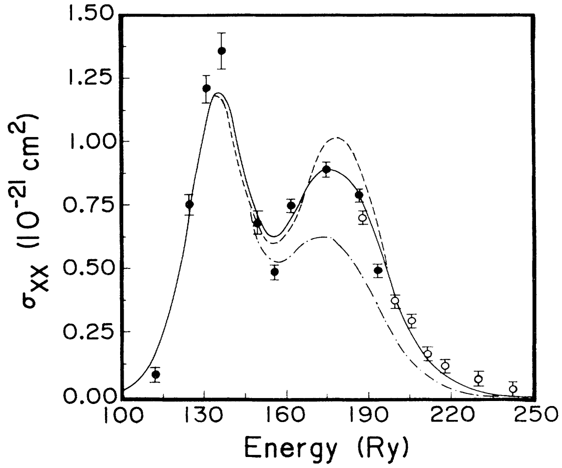

FIG. 1. $K\alpha$-$K\alpha$ coincidence cross sections for $S^{15+}+H_2$ collisions. —·—·—·, single-configuration *LS* coupling; — — —, configuration-mixing *LS* coupling; ——, configuration-mixing intermediate coupling; all this work. ● experimental points from Ref. [2]; ○, experimental points from Ref. [13].

through electric dipole radiation. The case for double x-ray emission is not so straightforward. Intermediate coupling now allows the spin triplets to contribute to double x-ray emission via $1s2p\ ^3P_1 \rightarrow 1s^2\ ^1S_0$ transitions, thus increasing the cross section over the *LS*-coupling result. However, it also reduces the cross sections by allowing spin singlets to end up in the $2\,^3S_1$ metastable, viz., $2pnp\ ^1D_2 \rightarrow 1snp\ ^3P_2 \rightarrow 1s2s\ ^3S_1$. The net result for S^{15+} coincidences is a 10–20 % reduction in the cross section. Overall, there is good agreement between the configuration-mixing intermediate-coupling $K\alpha$-$K\alpha$ cross section and experiment. However, the theoretical and experimental results start to diverge as we consider coincident emission from more highly excited states. In particular, the $K\alpha$-$K\gamma$ cross section is about a factor of 2 smaller than experiment.

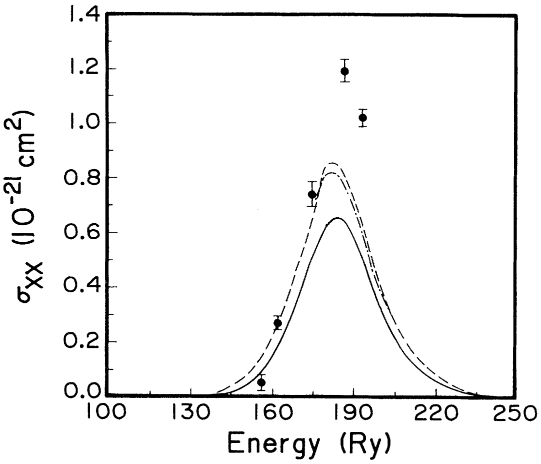

FIG. 3. $K\alpha$-$K\gamma$ coincidences, as in Fig. 1.

The calculated $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ cross section is also much smaller than experiment, by about a factor of 3, and is consistent with an earlier estimate [3]. About 75% of the total $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ cross section is due to the $3l'nl$ configurations, 20% due to the $4pnl$, and 5% due to the $5pnl$, and so we have essentially converged the sum over all higher states γ. The $3snl$ and $3dnl$ configurations do not contribute directly to the $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ cross section, but they do have a significant effect through configuration mixing, this time to reduce the coincidence cross section by one-third. Thus, our results, which include the $4pnl$ and $5pnl$

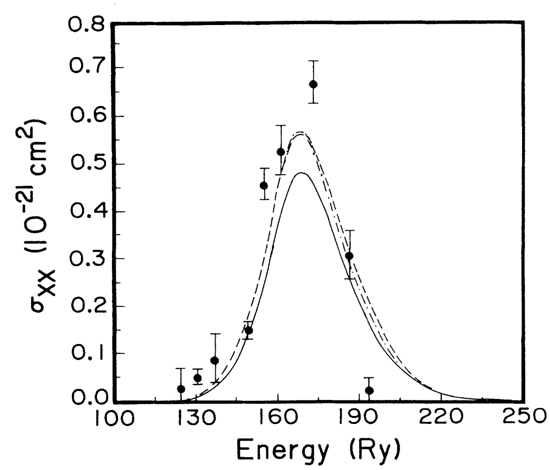

FIG. 2. $K\alpha$-$K\beta$ coincidences, as in Fig. 1.

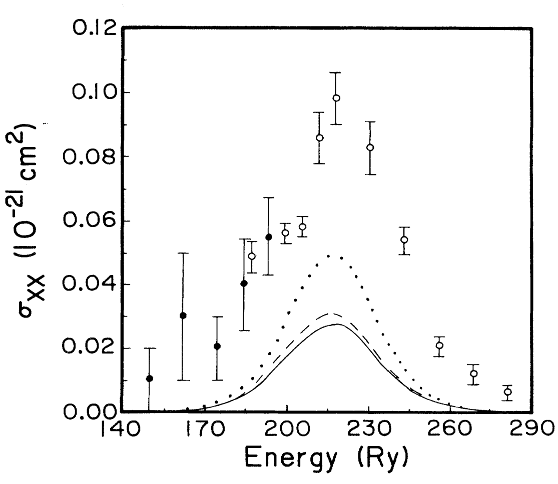

FIG. 4. $(K\beta+K\gamma)$–$(K\beta+K\gamma)$ coincidences, as Fig. 1, plus configuration-mixing *LS*-coupling results omitting the $3snl$ and $3dnl$ configurations (. . . . .).

configurations only, may be somewhat of an overestimate for the 4:n: and 5:n: complexes, but they are not the dominant contribution to the $(K\beta+K\gamma)$-$(K\beta+K\gamma)$ cross section.

The time of flight [2] for which emission will still be detected ($\sim 2.5\times10^{-10}$ s) puts a lower limit of about 4×10^{9} s^{-1} on the radiative rates. Since $A_r(2\,^3S_1\rightarrow1\,^1S_0)=1.4\times10^{6}$ s^{-1}, no error should arise from assuming that the $2\,^3S_1$ level does not radiate during the experiment. The rate from the $2\,^1S_0$ is much larger [$A_r(2\,^1S_0\rightarrow1\,^1S_0)=2\times10^{8}$ s^{-1}], but the two-photon emission would show up on a different part of the experimental coincident spectrum [2,5].

We have assumed that the radiative emission is isotropic; this is only approximately true. We have made a detailed study [11] of anisotropic radiative emission effects on deduced resonant transfer excitation cross sections. From this work [11], we estimate that the allowance for anisotropic radiative emission is unlikely to increase our results by much more than 10%, particularly for the cascade-produced $K\alpha$-$K\alpha$ peak.

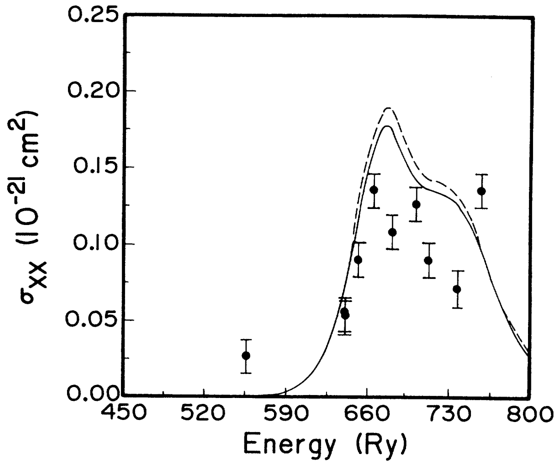

FIG. 6. $K\alpha$-$(K\beta+K\gamma)$ coincidence cross sections, as in Fig. 5.

B. Ge^{31+}

We present our configuration-mixing intermediate-coupling results for the total K-K coincidence cross section for Ge^{31+} in Fig. 5 and compare them with experiment [4]. The time of flight for the experiment [4] puts a lower limit of about 1.4×10^{10} s^{-1} on the radiative rates. We present results both with and without emission from the $2\,^3S_1$ level. Since $A_r(2\,^3S_1\rightarrow1\,^1S_0)=1.7\times10^{9}$ s^{-1} for an isolated ion, the lower cross section should correspond to that being measured, but both sets of results are in broad agreement with experiment. We note that the $2\,^3P_2$ level now decays preferentially through magnetic quadrupole radiation [$A_r(2\,^3P_2\rightarrow1\,^1S_0)=4.4\times10^{10}$ s^{-1}] to the ground level, unlike S^{15+}, where electric dipole radiation to the $2\,^3S_1$ level is preferred. The experiment [4] also measured the contribution from $K\alpha$-$(K\beta+K\gamma)$ coincidences; a comparison of theory and experiment [4] is shown in Fig. 6. We see that the theoretical results are now somewhat larger than experiment, unlike the case of S^{15+}.

The two-photon emission from the $2\,^1S_0$ level can now be seen experimentally [5], since $A_r(2\,^1S_0\rightarrow1\,^1S_0)=1.8\times10^{10}$ s^{-1}. The comparison between theory and experiment [5] is made in Fig. 7. While there is good agreement for the KLL peak, we see that the cross section due to resonant transfer and excitation is much smaller than experiment in the energy region of the KLn ($n>L$) resonances. The experiment [5] only ob-

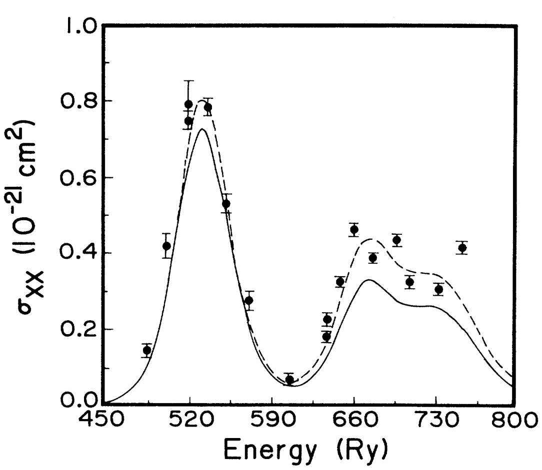

FIG. 5. Total K-K coincidence cross sections for $Ge^{31+}+H_2$ collisions. Configuration-mixing intermediate coupling: ——, with $2\,^3S$ stable; — — —, with $2\,^3S$ radiating. ⧮, experimental points from Ref. [4].

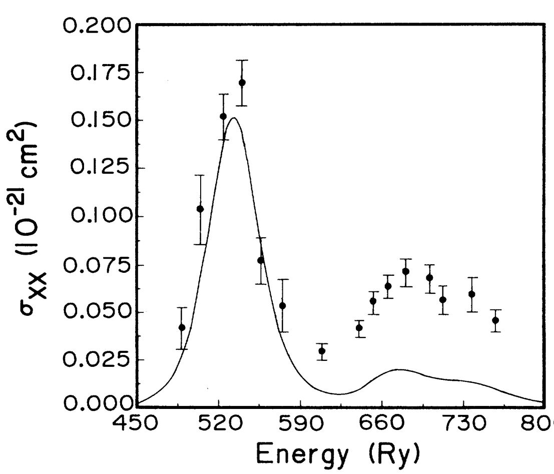

FIG. 7. Two-photon ($2\,^1S_0$) cross section for $Ge^{31+}+H_2$ collisions. ——, contribution from resonant-transfer excitation only. ⧮ experimental points from Ref. [5].

served the coincident two-photon emission from the $2\,^1S_0$ level and did not differentiate between resonant transfer excitation and direct radiative [6] or nonradiative [14] electron capture as the populating process. However, it should be noted, of course, that this nonresonant background cross section should decrease with increasing energy so that, on the face of it, it cannot fully account for the discrepancy between theory and experiment in the KLn ($n > L$) energy region.

Finally, a limited calculation for the Ge^{31+} cross sections presented here has already been carried out [6] based on published [15] rates for the KLL and KLM intermediate states. While we find no disagreement with the individual published [15] rates, their use in Ref. [6] appears to be somewhat in error. In particular, the branching ratio B from $1s3p\,^1P_1$ to $1s2s\,^1S_0$ ($B = 0.625$) is grossly overestimated due to an apparent confusion of units; we obtain $B = 0.061$.

V. CONCLUSION

We have shown the importance of including configuration mixing, and to a lesser extent intermediate coupling, in the calculation of coincidence cross sections. In particular, we have removed the discrepancy between theory and experiment for $K\alpha$-$K\alpha$ coincidences following resonant transfer and excitation in collisions of S^{15+} with H_2, but significant discrepancies remain for the $K\alpha$-$K\gamma$ and $(K\beta+K\gamma)$-$(K\beta+K\gamma)$ coincidences. For Ge^{31+}, there is good agreement between theory and experiment for the total coincidence cross section and, unlike S^{15+}, the theoretical $K\alpha$-$(K\beta+K\gamma)$ cross section is now somewhat larger than experiment. Since we have included all of the important energetically accessible configurations in our calculations with full configuration mixing and intermediate coupling, it suggests that the experimental results may need to be reexamined where significant discrepancies with theory remain.

ACKNOWLEDGMENTS

I would like to thank Dr. M. Schulz and Dr. P. Mokler for some helpful conversations concerning the experiments. This work was supported by the U.S. Department of Energy, Office of Fusion Energy, under Contract No. DE-FG05-86ER53217 with Auburn University.

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