Origin of Two-Electron, One-Photon K–X-Ray Transitions*

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Hartree-Fock energy and intensity calculations show that the two-electron, one-photon K^2–L^2–x-ray transitions recently reported are due to $1s^{-2}\rightarrow 2s^{-1}2p^{-1}$ electric dipole transitions. The mechanism which we use to describe the $1s^{-2}\rightarrow 2s^{-1}2p^{-1}$ transition is based on a "shake-down" model.

Recently Wölfli *et al.*[1] have reported evidence of two-electron, one-photon K^2-L^2–x-ray transitions in heavy-ion collisions. An observation of analogous L^2-M^3 Auger electrons has been reported by Afrosimov *et al.*[2] Nothing prevents, in principle, doubly ionized inner-shell hole states of an atom from decaying by radiative or radiationless transitions which result in the simultaneous filling of the two holes. Previous work[3] gives one-electron selection rules for such radiative transitions but estimates of the probability of such transitions in the case of a doubly ionized K shell have to our knowledge not been given. Dow and Franceschetti[4] briefly discuss the simultaneous filling of two L holes by two conduction-band electrons in simple metals. Recently Nagel *et al.*[5] have pointed out that the experimental K^2-L^2 transition energies obtained by Wölfli *et al.* are in disagreement with $1s^{-2}\rightarrow 2p^{-2}$ energies obtained from experimental and Hartree-Fock hypersatellite and satellite transition energies.

The purpose of this Comment is to show that $1s^{-2}\rightarrow 2s^{-1}2p^{-1}$ electric dipole x-ray transitions (defined as $K\alpha\alpha^h$, see Fig. 1) should be observable in heavy-ion collisions where an extensive production of double-K-hole states occurs. We compare our intensity estimates and our results of Hartree-Fock energy calculations with the observations of Wölfli *et al.*[1] and find reasonable agreement. The mechanism for the $1s^{-2}\rightarrow 2s^{-1}2p^{-1}$ transition is described in terms of a "shake-down" model, similar to the model[6] which is used to interpret shifts of autoionizing lines in electron-atom collisions near threshold.

It has been suggested that shake-off and configuration mixing in the final state are responsible for some *low-energy* structures below the characteristic lines in x-ray spectra.[7] The shake-off occurs as a consequence of the change in the average potential acting on the electrons when the vacancy transfers from an inner to an outer shell. This change could also be responsible for a shake-down of a second electron whenever there are two inner-shell holes available initially. The result would be *high-energy* structures considerably above the characteristic line in x-ray spectra. For the case of initial $1s^02s^22p^n$ states, electric dipole transitions to $1s^22s2p^{n-1}$ states would be possible, whereas transitions to final $1s^22s^22p^{n-2}$ states would be parity forbidden. In the following we shall consider the intensity, $I(K\alpha\alpha^h)$, of these transitions with respect to the intensity $I(K\alpha^h)$ of hypersatellite transitions between $1s^02s^22p^n$ and $1s2s2p^{n-1}$ states.

It can be shown that the total hypersatellite x-ray transition rate is given by

$$T(1s^02s^22p^n\rightarrow 1s2s^22p^{n-1}) = \tfrac{4}{9}\alpha^3\omega_1{}^3nD_1(1s2p)^2 \qquad (1)$$

in the frozen-core approximation and in atomic

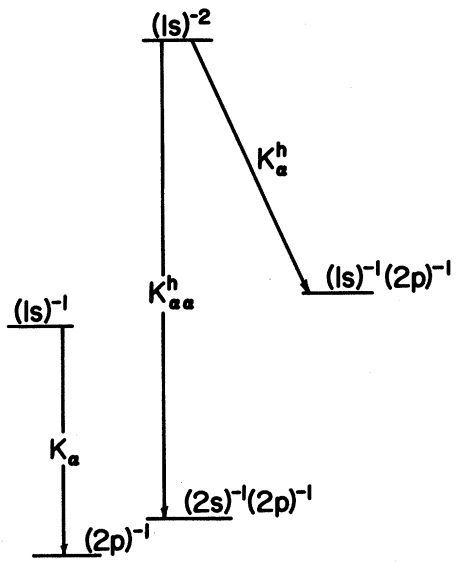

FIG. 1. Energy diagram showing the decay of doubly-ionized K-shell states by the K^2-KL hypersatellite ($K\alpha^h$) transitions and by the competing K^2-L^2 two-electron, one-photon ($K\alpha\alpha^h$) x-ray transitions. The K-L characteristic ($K\alpha$) x-ray transition is also shown to scale.

units. ω_1 is the average $K\alpha^h$ transition energy and $D_1(1s2p)$ is the $2p \rightarrow 1s$ electric dipole radial integral. The rate for $1s^0 2s^2 2p^n \rightarrow 1s^2 2s 2p^{n-1}$ transitions is zero in the frozen-core approximation. However, if we take the relaxation of the electron cloud into account we get in the lowest order

$$T(1s^0 2s^2 2p^n \rightarrow 1s^2 2s 2p^{n-1}) = \tfrac{4}{9}\alpha^3 \omega_2^3 n D_0(1s2s)^2 D_1(1s2p)^2, \quad (2)$$

since the $2s$ orbital of the $1s^0 2s^2 2p^n$ configuration is not orthogonal to the $1s$ orbital of the $1s^2 2s 2p^{n-1}$ configuration. $D_0(1s2s)$ is the $1s2s$ monopole radial overlap integral and ω_2 is the average $K\alpha\alpha^h$ transition energy. Hence, we obtain the branching ratio

$$I(K\alpha\alpha^h)/I(K\alpha^h) = (\omega_2/\omega_1)^3 D_0(1s2s)^2, \quad (3)$$

which attributes the $K\alpha\alpha^h$ intensity to the shake down of the $2s$ electron into a $1s$ hole.

Figure 2 presents nonrelativistic Hartree-Fock (HF) energies[8] for the $1s^0 2s^2 2p^n \rightarrow 1s^2 2s 2p^{n-1}$ electric dipole shake-down transitions as a function of the atomic number Z between Mg ($Z=12$) and Ni ($Z=28$) for the case $n=4$ and 6. The energies are given relative to twice the corresponding HF energies for the $1s^1 2s^2 2p^n \rightarrow 1s^2 2s^2 2p^{n-1}$ transitions. The energies of the parity-forbidden $1s^0 2s^2 2p^n \rightarrow 1s^2 2s^2 2p^{n-2}$ transitions are also shown. The energy spread of the shake-down transitions as a function of n is larger than that of the parity-forbidden transitions. Jundt and Nagel[9] have shown that for Ni-Ni collisions at 60 MeV in a solid Ni target the most probable K-x-ray satellite transitions originate from initial states with two $2p$ holes. According to Fig. 2 the corresponding average HF energy differences are about 30 eV higher than the observed energy differences in Fe and Ni.[1] However, due to the exchange interaction $G^1(2s, 2p)$ the energy difference corresponding to the strongest $K\alpha\alpha^h$ and $K\alpha$ multiplets would be about 50 eV less than the average energy difference in these elements plotted in Fig. 2. As noted by Nagel *et al.*[4] and as shown by Fig. 2 the $1s^0 2s^2 2p^n \rightarrow 1s^2 2s^2 2p^{n-2}$ transition energies are far outside the uncertainty of the experimental values.

Figure 3 displays the calculated relative shake-down rate as a function of Z using Eq. (3) for an initial configuration with $n=4$. This relative rate is decreasing approximately as Z^{-2}, since $D_0(1s2s)^2 \simeq 0.035 Z^{-2}$ according to our HF calculations. The inverse of the most accurate experimental ratios with an error of 75% as reported by Wölfli *et al.*[1] are given for comparison.

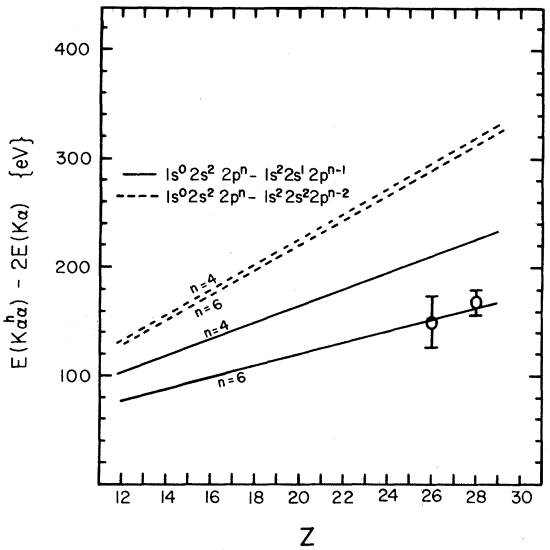

FIG. 2. Relative HF energies of one-photon K^2-L^2-x-ray transitions. Term-average HF energies are used in all the calculations except for the $(1s)^{-2}\,{}^1S_0 \rightarrow (2s)^{-1}(2p)^{-1}\,{}^1P_1$ transition ($n=6$, solid curve). The notation in the figure refers to the subshells involved in the transitions. The measured values based on the data of Wölfli *et al.* (Ref. 1) for Ni-Fe, Ni-Ni, Fe-Fe, and Fe-Ni collisions are given for comparison.

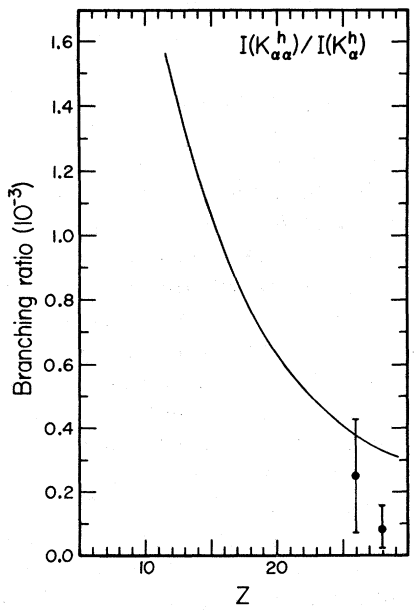

FIG. 3. The branching ratio of K^2-L^2 ($K\alpha\alpha^h$) and K^2-KL ($K\alpha^h$) transitions is given as a function of Z for an initial configuration with $n=4$. The experimental data from Wölfli *et al.* (Ref. 1) are given for comparison.

In order to examine the validity of the shake-down model we relate the monopole interaction element $D_0(1s2s)$ to the change of the average HF potential in the shake-down transition. If the influence of the M orbitals on this potential difference is neglected, then the result is

$$D_0(1s2s) = \frac{-R^0(2s1s1s1s) + R^0(2s2p1s2p) - \frac{1}{6}R^1(2s1s2p2p)}{\epsilon_{2s} - \epsilon_{1s}} . \quad (4)$$

The $1s$ and $2s$ radial wave functions in $D_0(1s2s)$ are assumed to be the nonorthogonal solutions of the term-averaged HF equations for the final and initial configurations, respectively. The corresponding one-electron energies are ϵ_{1s} and ϵ_{2s}. The values of the generalized Slater integrals R^k in Eq. (4) are not appreciably different if the L orbitals are taken from the final or initial state. The integral $R^0(2s1s1s1s)$ which dominates the numerator describes the change of the screening due to the filling of the $1s$ holes. The two other integrals describe the reduction of this screening due to the creation of the final $2p$ hole. Note that Eq. (4) is independent of the number of $2p$ electrons in the initial configuration. Our result indicates also that $D_0(1s2s)$ would not be affected very much by the multiplet splittings.

The intensity of $K\alpha\alpha^h$ is also influenced by final-state configuration interaction primarily because the $1s2s^22p^{n-1}$ and $1s^22s2p^{n-1}$ configurations can mix. Assuming that the orbitals are solutions of the $1s^22s2p^{n-1}$ term-averaged HF equations, it can be shown that the interaction matrix element $\langle 1s2s^22p^{n-1}|H|1s^22s2p^{n-1}\rangle$ is proportional to the exchange integral $R^1(2s1s2p2p)$ which appears in Eq. (4). The proportionality constant for any n and any term is smaller than 1 and can be either positive or negative. Since the absolute value of $R^1(2s1s2p2p)$ is only about 30% of that of $R^0(2s1s1s1s)$ in the cases we have studied this configuration mixing does not change our shake-down results. However, we would like to stress that the relative intensity of $K\alpha\alpha^h$ may be sensitive to the detailed correlation between the two $1s$ electrons in the final state in analogy to the double-photoexcitation cross section of the K shell.[10] Many configurations besides $1s2s^22p^{n-1}$ would be needed to describe this situation properly. Note also that the mixing due to the near degeneracy of the initial $1s^02s^22p^n$ and $1s^02s^02p^{n+2}$ configurations for $n \leq 4$ would suppress the initial-state energy.

In conclusion, we have shown that the shake-down rate of two-electron–one-photon K^2-L^2 transitions relative to K^2-KL hypersatellite transitions is of the order of 10^{-3} for $Z = 12$ and decreases approximately as Z^{-2}. Since there is a significant double-K ionization in many heavy-ion–atom collisions, $1s^{-2} \rightarrow 2s^{-1}2p^{-1}$ transitions should be observable. Our results support the available experimental evidence with regard to both energy and intensity.

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