X-ray transitions between correlated many-electron quantum states*

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The projectile energy dependence of apparent two-electron, one-photon x-ray transitions is determined from x-ray spectra resulting from collisions of nickel ions with thin nickel targets. Hartree-Fock energy calculations show that the observed projectile energy dependence can be attributed to the production of L-shell vacancies. It is also demonstrated that these transitions are fundamentally many-electron processes which cannot be described in terms of independent-particle models alone. Theoretical transition probabilities based on the non-closed-shell many-electron theory of Sinanoğlu yield a $K\alpha^h$ ($K\alpha$ hypersatellite) to $K\alpha\alpha$ branching ratio for nickel of 6700.

INTRODUCTION

X-ray transitions at energies approximating twice the characteristic K x-ray energies have recently been reported by Wölfli *et al.*[1] in Ni+Ni and Fe+Fe collisions. The original identification of these x-rays with two-electron (one-photon) transitions has received further support recently from additional investigations.[2-6] The Wölfli *et al.* observations, however, involved only a single value of the projectile energy. In this work, we report new measurements which show that these x-ray transition frequencies vary systematically with projectile energy, and we present results of Hartree-Fock calculations which indicate that this frequency variation reflects the increase in the number of L-shell vacancies with increasing projectile energy. Moreover, we demonstrate that the intensities of these transitions cannot be explained in terms of independent particle approximations.

DEPENDENCE OF TRANSITION ENERGY ON PROJECTILE ENERGY

An example of the x-ray spectra obtained in this work is shown in Fig. 1. Data were accumulated with a 5-mm-thick intrinsic Ge spectrometer possessing a resolution of approximately 250 eV at a photon energy of 6 keV. Labeled in the figure are the $K\alpha\alpha$ and $K\alpha\beta$ peaks previously[1-6] attributed to two-electron one-photon transitions. Measurements were carried out both at 90° and 0° to the beam direction. The observation of the Doppler shifted projectile $K\alpha\alpha$ line at 0° (under the 90° $K\alpha\beta$ line), as illustrated in the 91.5-MeV spectrum, precludes the possibility that the lines are due to target contaminants.

The peak energies are given in Table I. The errors quoted for the $K\alpha$ and $K\beta$ frequencies principally reflect uncertainties in the energy calibration and errors arising from the line-shape fitting procedure used in determining the peak centroid of the radiation standards and $K\alpha$, $K\beta$ peaks. The larger errors in the $K\alpha\alpha$ and $K\alpha\beta$ energies originate from additional statistical uncertainties in the measured $K\alpha\alpha$ and $K\alpha\beta$ peak centroids. While $K\alpha\alpha$ and $K\alpha\beta$ measurements required the use of a 0.034-in. Al absorber to prevent pulse pileup of the $K\alpha$ and $K\beta$ x rays, characteristic K x-ray data were taken with no absorber in order to prevent distortion of the measured energies due to differential absorption of the α_1, α_2, β_1, and β_3 characteristic K lines. Further experimental details for obtaining and analyzing the data are reported elsewhere.[6]

Theoretical energies for $K\alpha$, $K\alpha^h$ ($K\alpha$ hypersatellite), $K\beta$, $K\alpha\alpha$, and two types of $K\alpha\beta$ transitions are given in Table II. These values were obtained from restricted Hartree-Fock (RHF) energies with corrections for relativistic mass and

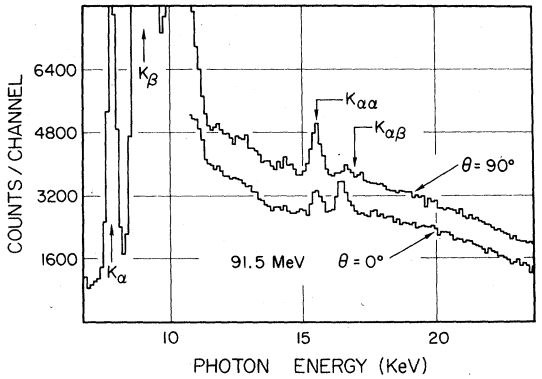

FIG. 1. Sample of x-ray spectra emitted in $^{58}Ni+^{58}Ni$ collisions. Beam energies quoted here and elsewhere are average projectile energies in the thin (~200 μg/cm^2 oriented at 45° to the beam direction) targets.

TABLE I. Experimental nickel atom x-ray energies obtained in this work and by Wölfli *et al.* Uncertainties in each measurement are given in eV in parentheses.

Projectile energy (MeV)	x-ray energy (keV) $K\alpha$	$K\beta$	$K\alpha\alpha$	$K\alpha\beta$
17.9	7.525(6)	8.446(8)	15.194(37)	...
37.1	7.549(6)	8.522(8)	15.278(17)	16.331(49)
64.8	7.576(6)	8.607(8)	15.312(10)	16.457(26)
91.5	7.593(6)	8.660(8)	15.359(15)	16.551(48)
(40)[a]	7.530[a]	...	15.228[a]	16.174[a]

[a]See Ref. 1. (Projectile energy quoted in this case is actual beam energy and not mean projectile energy in target.)

spin-orbit effects. The RHF energies were calculated using the expansion SCF (self-consistent-field) method of Roothaan.[7] Separate SCF calculations were performed on both states in each transition in order to account for the charge redistribution which accompanies the transition. The basis sets used in these calculations consisted of seven to eight Slater-type orbitals (STO's) of s-symmetry, five to six p-symmetry STO's and five d-symmetry STO's. These calculations were carried out using the computer program of Roothaan and Bagus,[7] which has been modified to allow inner shell vacancies.

Because of the inner-shell vacancies, most of the electronic states involved in these calculations do not have the lowest energy for their symmetry (i.e., L, S, and parity). Consequently, RHF (single configurational) wave functions are not variationally bound by the states of interest.[8] Therefore, in order to avoid the possibility of variational collapse, the STO orbital exponents used in these calculations were not optimized, but were fixed at values determined by SCF calculations[9] on various states of Ni, Cu, and Zn ions expected to have nearly the same effective nuclear charges (subshell by subshell) as the states of interest. Even this procedure does not totally guarantee the prevention of variational collapse. However, examination of the wave functions obtained in these calculations indicates that each has converged to the state of interest.

Contributions from the relativistic mass effect and spin-orbit interaction to the energy of each state were calculated using the hydrogenic formula,

$$\Delta E_{\mathrm{REL}} = (\alpha^2 Z_{\mathrm{eff}}^4/8n^4)\,[3 - 8n/(2j+1)]\ , \qquad (1)$$

for each $1s$, $2s$, and $2p$ electron. Appropriate values of Z_{eff} were estimated separately for each subshell of each state. Relativistic effects were completely neglected for M-shell electrons. Transitions involving a $2p$ electron ($K\alpha$, $K\alpha^h$, $K\alpha\alpha$, and $K\alpha\beta$) were calculated for a $2p_{1/2}$ (L_{II}) jumping electron. Transitions involving $2p_{3/2}$ (L_{III}) electrons will occur at energies approximately 18 eV larger. The RHF transition energies are increased by about 100 to 150 eV by these effects. These corrections are expected to be accurate to approximately 10 to 15 eV.

The effects of electron correlation on transition energies were neglected in these calculations. These effects, however, as shown below are extremely important in the calculation of the two-electron transition probabilities. Electron correlation is expected to influence the $K\alpha$, $K\alpha^h$, $K\beta$, and $K\alpha\alpha$ transition energies by only about 3 to 5 eV. The $K\alpha\beta$ transition energies, however, could be affected by as much as 30 to 50 eV due

TABLE II. Theoretical nickel atom x-ray energies given by restricted Hartree-Fock calculations with relativistic corrections. For each type of transition, the orbitals in parentheses indicate holes in the subshell occupation levels given at the left.

Subshell occupation						Transition energy (keV) $K\alpha$	$K\alpha^h$	$K\beta$	$K\alpha\alpha$	$K\alpha\beta$	$K'\alpha\beta$
$1s$	$2s$	$2p$	$3s$	$3p$	$3d$	$(1s)\rightarrow(2p)$	$(1s^2)\rightarrow(1s2p)$	$(1s)\rightarrow(3p)$	$(1s^2)\rightarrow(2s2p)$	$(1s^2)\rightarrow(2s3p)$	$(1s^2)\rightarrow(3s2p)$
2	2	6	2	6	10	7.463	7.739	8.271	15.093	15.957	16.055
2	2	6	2	6	0	7.458	7.734	8.323	15.083	16.014	16.107
2	2	6	2	0	0	7.490	7.768	...	15.157	...	16.264
2	2	3	2	6	0	7.560	7.854	...	15.347	...	16.498

to interactions between the final state configurations.

The results in Table II indicate that individual M-shell electrons, especially $3d$ electrons, have relatively little influence on the energies of the transitions. The $K\alpha$ energies are increased by an average of 34 eV for each $2p$ vacancy, in agreement with the results of previous Hartree-Fock-Slater calculations.[10] The $K\alpha\alpha$ and $K\alpha\beta'$ energies, however, are much more sensitive, being increased by 88 and 130 eV per $2p$ vacancy, respectively.

The effects of $2s$ vacancies were estimated by subtracting the corresponding $2s$ SCF orbital energies from the total energies of the initial and final states in each transition. These calculations indicate that a $2s$ vacancy increases the energy of a $K\alpha$ transition by approximately 29 eV, and the energy of a $K\alpha\alpha$ transition by roughly 85 to 103 eV, depending on the number of $2p$ vacancies. These results, based on Koopmans's theorem,[11] are less accurate than the estimates of the effects of $2p$ vacancies because these calculations do not permit the remaining electrons to adjust to the presence of the $2s$ vacancy.

Comparison of Tables I and II strongly suggests that the dependence of the $K\alpha\alpha$ x-ray energies on projectile energy is primarily due to the production of increasing numbers of L-shell (mainly $2p$) vacancies with increasing projectile energy. From a detailed examination of the tables, it is estimated that there are an average of one to two L-shell vacancies at 17.9 MeV, and at 91.5 MeV, three or four.

RATES OF TWO-ELECTRON ONE-PHOTON TRANSITIONS

As noted above, the $K\alpha\alpha$ and $K\alpha\beta$ transitions treated in this paper are frequently referred to as "two-electron (one photon) transitions."[1-5] However, it is well known that true two-electron (one-photon) transitions are strictly forbidden because the operators responsible for radiative electronic transitions are only one-electron operators. In spite of this, *apparent* two-electron transitions are well established in optical spectroscopy.[12,13] Such transitions, including the $K\alpha\alpha$ and $K\alpha\beta$ transitions, depend upon the interelectronic electrostatic interactions which couple together the motions of the electrons. Each of these transitions represents a jump from one many-electron state to another many-electron state, satisfying the overall (many-electron) selection rules on total parity, and total angular momentum. Consequently, it should be recognized that these transitions are fundamentally many-electron processes which, unlike the "normal" (one-electron) transitions, cannot be described in terms of one-electron or independent particle models alone.

As an electric dipole transition, the rate of a $K\alpha\alpha$ transition is given by the dipole-length formula as

$$A_R(\alpha\alpha) = \tfrac{4}{9}\alpha^3\omega_{\alpha\alpha}^3 |R_N(\alpha\alpha)|^2 , \tag{2}$$

where R_N is an N-electron dipole-length transition moment. To a good approximation, R_N is given by

$$R_N(\alpha\alpha) = S_{12}R_1(2p, 1s) + \sum_I c_I(i)R_1(I_1, I_2) + \sum_J c_J(f)R_1(J_1, J_2) , \tag{3}$$

where $S_{12} = \langle 2s(i) | 1s(f) \rangle$ is the overlap of the initial (upper) state $2s$ orbital with the $1s$ orbital of the final (lower) state; $c_I(i)$ and $c_J(f)$ are the mixing coefficients for configuration I in the initial state and configuration J in the final state; and $R_1(a, b)$ is the one-electron transition moment for orbital a in the initial state and orbital b in the final state.

The first term in Eq. (3) represents the extent to which the upper state $2s$ orbital already contains a (small) component of the lower state $1s$ orbital. In a frozen orbital approximation, the $1s$ and $2s$ orbitals would be orthogonal and S_{12} would vanish. Because of the changes in shielding which accompany the transition, there is a small nonzero value for S_{12}. A simple approximation to S_{12} can be found by considering the overlap of a hydrogenic $2s$ orbital with a shielded hydrogenic $1s$ orbital. In this case, $S_{12} \simeq \langle 1s_H(Z-s) | 2s_H(Z) \rangle \simeq (32\sqrt{2/81})(s/Z)$, where Z is the atomic number, and s is the $1s$ shielding constant. Thus, S_{12} should vary as $1/Z$, and for $Z = 28$ and $s = \frac{5}{16}$, S_{12} should have a value of approximately 0.006 24. This compares favorably with the value of $S_{12} = 0.00823$ obtained from the RHF orbitals in the $(1s^0 2s^2 2p^6 3s^2\ {}^1S) \rightarrow (1s^2 2s 2p^5 3s^2\ {}^1P^o) K\alpha\alpha$ transition.

Unlike the first term in Eq. (3), which can be interpreted in terms of average interactions between electrons (i.e., shielding), the second and third terms represent the instantaneous interactions between the electrons (i.e., electron correlation effects) in the initial and final states of the transition. The coefficients in the second term are given by

$$c_I(i) = \langle \Phi_0(i) | H | \Phi_0(I) \rangle / [E_0(i) - E_0(I)] , \tag{4}$$

where $\Phi_0(i)$ is the dominant configuration in the initial state and $\Phi_0(I)$ is a virtual configuration (in the initial state) which differs from the dominant configuration of the final state by the substitution of orbital I_2 for orbital I_1. The coefficients in the third term are given by similar

formulas.

The importance of the second and third terms in Eq. (3) can be demonstrated by noting that the rate of the corresponding $K\alpha^h$ transition is given by

$$A_R(\alpha) = \tfrac{4}{9}\alpha^3\omega_\alpha^3 |R_N(\alpha)|^2 \simeq \tfrac{4}{9}\alpha^3\omega_\alpha^3 |R_1(2p, 1s)|^2 , \tag{5}$$

Therefore, if the second and third terms in Eq. (3) are neglected, as is done in any independent-particle approximation, one finds

$$A_R^{(0)}(\alpha\alpha) = (\omega_{\alpha\alpha}/\omega_\alpha)^3 S_{12}^2 A_R(\alpha) , \tag{6}$$

where $\omega_{\alpha\alpha} \simeq 2\omega_\alpha$. These rates, however, can also be expressed in terms of the corresponding dipole-velocity and dipole-acceleration formulas[14] as

$$A_V(i,f) = \tfrac{4}{9}\alpha^3\omega_{if} |V_N(i,f)|^2 \tag{7a}$$

$$A_A(i,f) = \tfrac{4}{9}\alpha^3\omega_{if}^{-1} |A_N(i,f)|^2 , \tag{7b}$$

where V_N and A_N are the dipole-velocity and dipole-acceleration transition moments, respectively. In principle, the length, velocity, and acceleration formulas all represent equivalent formulations of the same quantity $A(i,f)$ and if the exact wave functions are used for both the initial and final states, all three formulas yield the same result. In this case, however, if the steps leading to Eq. (6) are repeated for the velocity and acceleration formulas, one finds

$$A_V^{(0)}(\alpha\alpha) = (\omega_{\alpha\alpha}/\omega_\alpha) S_{12}^2 A_V(\alpha) \tag{8a}$$

and

$$A_A^{(0)}(\alpha\alpha) = (\omega_{\alpha\alpha}/\omega_\alpha)^{-1} S_{12}^2 A_A(\alpha) . \tag{8b}$$

Consequently, if it is assumed that

$$A_R(\alpha) = A_V(\alpha) = A_A(\alpha) = A(\alpha) ,$$

it is found that

$$A_R^{(0)}(\alpha\alpha) \simeq 4A_V^{(0)}(\alpha\alpha) \tag{9a}$$

and

$$A_V^{(0)}(\alpha\alpha) \simeq 4A_A^{(0)}(\alpha\alpha) . \tag{9b}$$

For any independent-particle approximation, therefore, the length, velocity, acceleration, etc., formulas for the rate of the $K\alpha\alpha$ transition yield results differing by successive factors of approximately four. This difficulty occurs because apparent two-electron transitions such as these are basically concerted processes which are directly dependent upon the instantaneous interactions between the electrons. Independent-particle approximations, however, are fundamentally one-electron models in which the instantaneous interactions between the electrons are neglected. Consequently, it is not possible for such approximations to provide physically realistic descriptions of fundamentally many-electron processes such as the $K\alpha\alpha$ transitions.

The importance of the second and third terms in Eq. (3) can also be demonstrated by estimating the magnitudes of the contributions to these terms. The most important contributions to the second terms come from virtual configurations in the initial state which differ from the dominant configuration of the initial state by the occupancy of two electrons (i.e., two spin orbitals), and from the dominant configuration of the final state by the occupancy of a single spin orbital. The most important contributions to the third term involve similar relationships. (This assumes that the dominant configuration in each state is represented by its Hartree-Fock wave function, and therefore, due to Brillouin's theorem,[15] single excitations from the dominant configuration are very unimportant.) Consequently, the numerators of the coefficients of these configurations, as shown in Eq. (4), are given by

$$\langle \Phi_0(i) | H | \Phi_0(I) \rangle = \langle ab | 1/r_{12} | kl \rangle - \langle ab | 1/r_{12} | lk \rangle , \tag{10}$$

where virtual configuration $\Phi_0(I)$ is related to dominant configuration $\Phi_0(i)$ by replacing spin orbitals a and b by spin orbitals k and l.

For example, the $2s^2$ pair of electrons in the initial state of a $K\alpha\alpha$ transition (configuration $1s^0 2s^2 2p^m \ldots$) can be replaced by a $1s^2$ pair to yield the configuration $1s^2 2s^0 2p^m \ldots$ which is related to the final state in the $K\alpha\alpha$ transition (configuration $1s^2 2s 2p^{m-1} \ldots$) by a $2p$ to $2s$ transition. In this case, the denominator of Eq. (4) corresponds to the energy of a $2s^2$ to $1s^2$ transition, which is roughly that of the $K_{\alpha\alpha}(2s2p \text{ to } 1s^2)$ transition itself, or approximately 15 keV. Thus, the contribution of this configuration to Eq. (3) is given by

$$c_I(i) R_1(2p, 2s) \approx (\langle 2s^2 | 1/r_{12} | 1s^2 \rangle / 15 \text{ keV}) \langle 2p | r | 2s \rangle .$$

The two-electron integral in the numerator of this expression can be directly evaluated, yielding the result of approximately 14 eV. The coefficient $c_I(i)$, therefore, has a value of roughly 1×10^{-3} in the first term in Eq. (3). The $\langle 2p | r | 2s \rangle$ transition moment, however, is roughly four times as large as the $\langle 2p | r | 1s \rangle$ transition moment. Consequently, the magnitude of this contribution alone is roughly half as large as the first term in Eq. (3). Furthermore, there are additional contributions of comparable magnitude within the second term of Eq. (3), as well as contributions from the third term.

In addition, it should be noted that the denominators in Eq. (4) vary as the square of the atomic

number, whereas the numerators vary linearly with the atomic number. Therefore, the coefficients $c_I(i)$, like the orbital overlap factors S_{12}, are approximately proportional to $1/Z$. Furthermore, the transition moments R_1 are proportional to $1/Z$ in all three terms of Eq. (3). Consequently, the relative importance of each of the terms in Eq. (3) is approximately independent of atomic number. In addition, it also follows that, in agreement with the prediction of the shake-down model,[4] the $K\alpha^h$ to $K\alpha\alpha$ branching ratio should be approximately proportional to the square of the atomic number.

APPLICATION OF MANY-ELECTRON THEORY

The preceding considerations demonstrate that electron correlation plays a crucial role in determining the rates of $K\alpha\alpha$ transitions. As specified by the non-closed-shell many-electron theory (NCMET) of Sinanoğlu,[16-18] the correlation effects (configurations) which are important for the calculation of transition probabilities are the internal and semi-internal correlations. The internal correlation effects include all possible $i,j \rightarrow k,l$ substitutions within a specified set of spin orbitals defined as the "Hartree-Fock sea," where i and j are occupied members of this set, and k and l are vacant members of this set. The semi-internal correlation effects include all substitutions of the type $i,j \rightarrow k,\hat{f}_{ij;k}$, where i and j are occupied members of the Hartree-Fock sea, k is a vacant member of this set, and $\hat{f}_{ij;k}$ is a semi-internal function outside this set (i.e., orthogonal to all spin orbitals contained in the Hartree-Fock sea).

Using Sinanoğlu's system of atomic-structure programs ATOM,[19] we have calculated transition probabilities for the $K\alpha^h$ and $K\alpha\alpha$ transitions from the $1s^0 2s^2 2p^6 3s^{2\,1}S$ state of the nickel +18 ion. Results were calculated using length, velocity, and acceleration formulas, and the lack of orthogonality between initial- and final-state orbitals was treated fully using the method of Westhaus and Sinanoğlu.[17]

In the $K\alpha^h$ transition, the final state (in L-S coupling) is the $1s2s^2 2p^5 3s^{2\,1}P^0$ state of Ni^{+18}. Simple one-electron out-of-shell ($\Delta n \neq 0$) transitions such as this are well described by the RHF approximation. Consequently, the length, velocity, and acceleration results for this transition (1.88×10^6 nsec^{-1}, 1.83×10^6 nsec^{-1}, and 1.87×10^6 nsec^{-1}, respectively) are all in good agreement. Even a simple hydrogenic approximation can describe this transition fairly well, yielding a transition probability of 8.0 $(2^{10}Z^4/3^7)$ nsec^{-1}. For $Z=28$, this hydrogenic formula yields a transition probability of 2.3×10^6 nsec^{-1}, in qualitative agreement with the RHF results.

The final state in the $K\alpha\alpha$ transition is the $1s^2 2s2p^5 3s^{2\,1}P^0$ state of Ni^{+18}. In the RHF approximation, we obtain length, velocity, and acceleration results of 925, 240, and 64 nsec^{-1} respectively. As indicated previously, these results each differ by successive factors of approximately four.

In order to include the effects of internal correlation, it is necessary to add four configurations to the initial-state RHF wave function. These configurations are obtained by making $2s^2$ to $1s^2$, $2p^2$ to $1s^2$, $3s^2$ to $1s^2$, and $2s3s$ to $1s^2$ substitutions in the dominant configuration. The RHF plus internal wave function for this state was then calculated as the *highest* root in the resulting 5×5 configuration interaction (CI) calculation. There are no important internal configurations in the final states of these $K\alpha\alpha$ and $K\alpha^h$ transitions.

Use of the resulting (five-configuration) initial-state wave function and the original-single-configurational (RHF) final-state wave function to calculate the rate of the $K\alpha\alpha$ transition yields length, velocity, and acceleration results of 3, 238, and 64 nsec^{-1}, respectively. Thus, in terms of the length formulation, it is seen that the orbital overlap (S_{12}; RHF) contribution, and the contribution from internal correlation effects are nearly equal in magnitude but opposite in sign, resulting in nearly complete cancellation. The velocity and acceleration results, however, show practically no change from the preceding (RHF-RHF) calculations. This is caused by the fact that most of the internal contributions are dependent on the $2s$-$2p$ transition moments. In the length form, as indicated previously, the $2s$-$2p$ transition moment is approximately four times as large as the corresponding $1s$-$2p$ transition moment. The velocity and acceleration forms of the $2s$-$2p$ transition moment, however, are *small* compared to the corresponding $1s$-$2p$ transition moments, and they vanish entirely in the limit of hydrogenic $2s$ and $2p$ orbitals. This is demonstrated by noting that

$$\langle 2s_H | v | 2p_H \rangle = (\epsilon_{2s} - \epsilon_{2p}) \langle 2s_H | r | 2p_H \rangle = 0 , \qquad (11)$$

because $\epsilon_{2s} = \epsilon_{2p}$ for hydrogenic ions.

It should be noted that recalculation of the $K\alpha^h$ transition probability using this five-configuration wave function yielded practically no change at all in comparison to the RHF results given above.

These results indicate that semi-internal correlation effects as well as internal correlation effects are necessary to calculate the rate of a $K\alpha\alpha$ transition. Of all the possible semi-internal configurations in the initial state, only those which

result from $2s2p \rightarrow 1s\hat{f}_p$ substitutions, where $\hat{f}_p$ is a p-symmetry semi-internal orbital (orthogonal to the $2p$ orbital), can make significant contributions to the $K\alpha\alpha$ transition dipole. The resulting $1s\ 2s\ 2p^5 3s^2\hat{f}_p$ configuration is counted as two because it forms two terms of ^1S symmetry. This yields a total of seven configurations for the upper-state wave function. In the lower state there are two possible semi-internal configurations which make direct contributions to the transition moment. These arise from $1s^2 \rightarrow 2s\,\hat{f}_s$ and $1s^2 \rightarrow 2p\,\hat{f}_p$ substitutions and they result in a three-configuration wave function for the lower state in this transition. All other configurations obtained by pair wise substitutions in either state differ from the dominant configurations of the other state by more than one electron and consequently cannot make significant contributions to the transition moment.

Calculation of the $K\alpha\alpha$ transition probability using the resulting seven-configuration initial-state wave function and three-configuration final-state wave function yields 276, 279, and 94 nsec^{-1}, respectively. Although the length and velocity agree to within 1%, the acceleration result differs by a factor of three. However, it is not unusual for the acceleration result to differ from the length and velocity results by one or even two orders of magnitude.[17] Consequently, although the acceleration result must be rejected as inaccurate, the fact that it differs from the length and velocity results by only a factor of three is actually relatively good. In spite of the excellent agreement between the length and velocity results, however, they may be uncertain by as much as $\pm 50\%$ because of the importance of cancellations within the transition moment. (Because such cancellations occur only in the length formulation, and not in the velocity form, this uncertainty estimate may be somewhat pessimistic.)

Based on the geometric means of the length and velocity results, we obtain a $K\alpha^h/K\alpha\alpha$ branching ratio of 6680 ± 3300. For comparison, we note that Kelly,[5] using the method of many-body perturbation theory, has obtained a branching ratio of 5740 for Fe. Assuming that this ratio is proportional to Z^2, this value corresponds to a ratio of 6660 for Ni in remarkably good agreement with our result. Like our calculation, the work of Kelly also includes the effects of electron correlation in both initial and final states. However, Kelly's calculation, on the +2 ion of Fe, includes $3p$, $3d$, and $4s$ electrons, whereas the calculations of this work were performed on the +18 ion of nickel in which all $3p$, $3d$, and $4s$ electrons were stripped away. Consequently, the closeness of the agreement of our result to Kelly's must be regarded as largely accidental, especially considering the degree of uncertainty in the results. Nevertheless, comparison of our results to Kelly's provides evidence that, as would be expected, $3p$, $3d$, and $4s$ electrons have very little influence on the rates of $K\alpha^h$ and $K\alpha\alpha$ transitions. In addition, comparison of these results may also suggest that the estimated uncertainty of $\pm 50\%$ attributed to our result may be somewhat pessimistic.

These results are also in fairly good agreement with the experimental ratio of 5000 ± 600 recently determined by Stoller *et al.*[20] In making a comparison between theoretical and experimental values, however, one must note that because of (i) the importance of correlations among the L-shell electrons, and (ii) large cancellations within the $K\alpha\alpha$ transition dipole, this ratio may be very sensitive to the L-shell occupancy. If so, the measured ratio would be strongly dependent on the projectile energy, and may differ systematically from calculations which assume an initially closed L shell.

CONCLUSIONS

In summary, the results of this work, both experimental and theoretical, all support the interpretation of the features in the x-ray spectra first reported by Wölfli *et al.*[1] as apparent two-electron one-photon transitions. Experimental data reported here show that the (peak) positions of these features are dependent upon the projectile velocity. Comparison of experimental measurements with theoretical calculations indicates that the observed variation of peak positions with projectile velocity is caused by changes in the number of L-shell vacancies as functions of projectile velocity.

Examination of the rate of the $K\alpha\alpha$ x-ray transition demonstrates that this type of transition is fundamentally a many-electron process directly dependent upon the concerted motions of the electrons. As such, it is not possible to obtain physically realistic descriptions of these transitions in terms of independent-particle models alone.

Calculations of the rates of $K\alpha\alpha$ and $K\alpha^h$ transitions based on the non-closed-shell many-electron theory of Sinanoğlu yield a $K\alpha^h/K\alpha\alpha$ branching ratio in excellent agreement with results recently obtained by Kelly using many-body perturbation theory. These results are also in good agreement with recent experimental measurements of Stoller *et al.*[20]

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