

K X-Ray Emission Rates in Superheavy Elements

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K x-ray emission rates in superheavy elements have been calculated by the use of the Dirac-Fock-Slater wave functions with finite-size nuclei. Calculations have been made relativistically and all possible multipoles as well as the retardation effect are taken into account. The x-ray energies are obtained as the difference between the initial and final binding energies of the atomic shells concerned. The importance of contributions from magnetic multipoles and the behavior of emission rates as a function of atomic number are discussed.

KEY WORDS: Superheavy elements/ Inner-shell binding energy/
K x-ray emission rates/

I. INTRODUCTION

In recent years, there have been considerable interest on the inner-shell binding energies and x-ray emission rates in superheavy elements (SHE). This is because the inner-shell x rays may be used as one of techniques for a high-degree certification of SHE. According to recent speculations about the stability of superheavy nuclei, it seems quite reasonable that some superheavy nuclides will be found in nature or at heavy-ion accelerators¹⁻⁴⁾. The most probable candidates are $Z=114$ with $A=298$ and $Z=126$ with $A=354$, where Z and A are the atomic number and the mass number of SHE, respectively. The next possible stability occurs at $Z=164$ and $A=472$. Once these nuclides are produced, they can survive long enough to be observed experimentally because they have double magic number. Some calculations predict also a fairly weak proton-shell closure at $Z=154$.

There is another method to produce SHE for a very short time during ion-atom collisions. When the kinetic energy of incident ions is high enough, the electron clouds of the projectile and the target penetrate with each other and the inner-shell electrons feel a united nuclear charge $Z_1 + Z_2$ in their center, where Z_1 and Z_2 are the nuclear charges of the projectile and the target, respectively. In collisions between heavy atoms, the experimental investigations of inner-shell x rays⁵⁾ and δ electrons⁶⁾ from quasimolecules can provide information about SHE. For example, Mokler *et al.*⁷⁾ observed M-shell x rays of $Z=132, 143$, and 145 in bombardment of 10–60-MeV I ions on Au, Th, and U targets.

The chemical properties and electronic structures of SHE can be predicted theoretically by performing self-consistent-field calculations for atomic electrons⁸⁻¹⁰⁾.

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The results indicate the order of filling the atomic orbitals, the ground-state configurations, the first ionization potentials, and the principal maxima and mean radii of the orbital electron wave functions up to $Z=173$, where the K-shell binding energy drops into the Dirac negative sea and a K-shell vacancy, if it exists, decays by spontaneous positron emission.

The atomic binding energies of inner-shell electrons in SHE and hence K- and L-shell x-ray transition energies have been calculated using the relativistic Hartree-Fock-Slater or Dirac-Fock-Slater (DFS) and the relativistic Hartree-Fock or Dirac-Fock (DF) models. Carlson *et al.*¹¹⁾ calculated the inner-shell binding energies and K x-ray energies for elements between $Z=96$ and 120 by combining the DFS model with the semi-empirical method. Using the same computer program, Lu *et al.*¹²⁾ presented the energy eigenvalues of all shells for elements up to $Z=126$. Fricke and Soff¹³⁾ have extended the similar calculations for elements up to $Z=173$.

On the other hand, Desclaux¹⁴⁾ has used the DF method and calculated energy eigenvalues for elements up to $Z=126$. Calculations of K and L x-ray energies have been made by Carlson and Nestor¹⁵⁾. They used the DF code of Desclaux¹⁴⁾ and the x-ray energies were obtained as the difference between the total energies of ions with a single vacancy in the initial and final states.

In comparison with the x-ray energy calculations, the number of calculations of x-ray intensities in SHE is rather scarce. Lu *et al.*¹⁶⁾ calculated K x-ray intensities for elements from $Z=92$ to 126. They used the DFS wave functions which were obtained previously¹²⁾ and pointed out that in SHE the K-L_I transition, which occurs by magnetic dipole transition, is no longer negligible. The DF calculations of K and L x-ray transition probabilities have been made by Anholt and Rasmussen¹⁷⁾ for several elements between $Z=92$ and 170. They showed that when the transition rates are plotted against Z , the electric multipole transition rates go through a maximum as a function of Z , while the contributions from magnetic multipoles increase with increasing Z .

Recently we have performed the relativistic calculations for M- and N-shell x-ray emission rates by the use of the DFS wave functions^{18, 19)}. In the present work, we have attempted to extend our model to SHE and calculated the K-shell x-ray emission rates. The numerical results for the binding energies and x-ray transition rates are compared with those from other theoretical calculations and the experimental data.

II. METHOD

In the first-order time-dependent perturbation theory, the radiative transition rate for an electron from a state i to a state f can be written by²⁰⁾

$$\Gamma = \frac{\alpha k}{2\pi} \sum_{\text{pol}} \int d\Omega_k |d \langle f | \hat{\alpha} \cdot \hat{\epsilon} \exp(-ik \cdot r) | i \rangle|^2, \quad (1)$$

where k is the photon momentum, α is the fine structure constant, $\hat{\alpha}$ is the Dirac matrix, and $\hat{\epsilon}$ is the polarization vector. Throughout the present work the relativistic units ($\hbar=m_e=c=1$) are used.

The relativistic wave functions for the initial and final states are given by

$$\phi_{n\kappa}^\mu = \frac{1}{r} \left(G_{n\kappa}(r) \chi_\kappa^\mu(\hat{\Omega}) \right), \quad (2)$$

where $G_{n\kappa}(r)$ and $F_{n\kappa}(r)$ are the large and small components of the radial wave function multiplied by radial distance r , $\chi_\kappa^\mu(\hat{\Omega})$ is the spin-angular function, and n is the principal quantum number. The relativistic quantum number κ is defined as $\kappa = \mp(j + 1/2)$ for $j = l \pm 1/2$, where j and l are the total and orbital angular momentum, respectively.

Substituting Eq. (2) into Eq. (1) and using multipole expansion of the radiation field, the angular integration can be done analytically. After summation over the initial substates and average over the final states, Eq. (1) reduces to²¹⁾

$$\Gamma = 2\alpha k^2 \sum_L [f_L(m) + f_L(e)]. \quad (3)$$

Here $f_L(m)$ and $f_L(e)$ are the oscillator strengths corresponding to the L th magnetic and electric multipoles:

$$f_L(m) = B(-\kappa_i, \kappa_f, L) R_L^2(m)/k, \quad (4)$$

$$f_L(e) = B(\kappa_i, \kappa_f, L) R_L^2(e)/k, \quad (5)$$

where $B(\kappa_i, \kappa_f, L)$ is the angular coupling coefficient defined by Scofield²¹⁾ and the subscripts i and f denote the initial and final electron states.

The radial matrix elements for magnetic and electric multipoles are expressed as

$$R_L(m) = (\kappa_i + \kappa_f) \int_0^\infty dr (F_f G_i + G_f F_i) j_L(kr), \quad (6)$$

$$R_L(e) = \int_0^\infty dr \{ [(\kappa_f - \kappa_i)(F_f G_i + G_f F_i) + L(F_f G_i - G_f F_i) j_{L-1}(kr) + L(G_f G_i + F_f F_i) j_L(kr)] \}, \quad (7)$$

where $j_L(x)$ is the spherical Bessel function of first kind of order L .

The radial wave functions G and F are normalized to be

$$\int_0^\infty dr (F^2 + G^2) = 1. \quad (8)$$

These wave functions were calculated using the DFS program, which is equivalent to the HEX code²²⁾. The effect of finite size of the nucleus was taken into consideration by assuming it as a uniformly charged sphere. The nuclear mass was assumed to be that of the stable candidate for $Z=114$, 126, and 164. For other elements, it was obtained from $A = 0.0073Z^2 + 1.3Z + 63.6$ ¹³⁾.

In the present work, we used the so-called *frozen-orbital* approximation, i.e. no allowance for core relaxation was taken into account. The wave functions for neutral atoms, corresponding to the ground-state configurations taken from Fricke and Soff¹³⁾, were used for both initial and final states. The exchange scaling factor for the Slater approximation, α , was taken to be $2/3$, otherwise stated, and the Latter tail correction was

employed.

The x-ray transition energies were estimated from the difference between the binding energies of the atomic shells concerned. It is well known, however, that the energy eigenvalue in the DFS model is different from the electron binding energy²³⁾. According to the method of Slater²⁴⁾, we estimated the approximate binding energy for the atomic shell i from the following expression¹⁸⁾:

$$B_i = -\varepsilon_i + \frac{1}{2} \frac{\partial^2 \langle E \rangle}{\partial q_i^2} \Big|_0 - \frac{1}{6} \frac{\partial^3 \langle E \rangle}{\partial q_i^3} \Big|_0, \quad (9)$$

where $-\varepsilon_i$ is the energy eigenvalue, $\langle E \rangle$ is the total energy of the DFS model, and q_i is the electron occupation number of the atomic shell i . The binding energy in Eq. (9) can be evaluated easily from the results of the DFS calculations.

III. RESULTS AND DISCUSSION

The numerical calculations of K-shell x-ray emission rates for SHE have been performed on the FACOM 380-Q computer at the Institute for Chemical Research, Kyoto University. The electron transitions from L, M, N, and O shells to K shell were considered and the contributions from all possible electric and magnetic multipoles were included. The transitions from N_6 and N_7 shells were calculated, but not presented below because of their small values.

In order to test the present method, we calculated the K-shell x-ray energies for U ($Z=92$). The calculations were made with two choices of the Slater statistical exchange parameters, $\alpha=1$ and $2/3$. Table 1 shows comparison of the K x-ray energies by the use of Eq. (9) with the experimental data. It can be seen that the calculated results with $\alpha=2/3$ are in good agreement with the measured values.

In Table 2, the calculated K-shell binding energies for $Z=110$, 117, and 135 are compared with other theoretical values and with the measured values for the united atom in ion-atom collisions. The energy eigenvalues of the represent calculations are in agreement with those of Lu *et al.*¹²⁾ and Fricke and Soff¹³⁾. It should be noted, however, that the calculations in the present work and those of Fricke and Soff were performed

Table 1 Comparison of calculated K x-ray energies with measured ones for $Z=92$ (keV).

Shell	Measured		Calculated	
	B ^a	BBEH ^b	$\alpha=1$	$\alpha=2/3$
L ₂	94.665	94.656±0.002	95.037	94.702
L ₃	98.439	98.435±0.002	98.900	98.527
M ₂	110.406	110.416±0.003	110.893	110.434
M ₃	111.300	111.300±0.002	111.786	111.314
M ₄	112.01	111.868±0.005	112.339	111.870
M ₅		112.043±0.005	112.521	112.048

^a Ref. 25.

^b Ref. 26.

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Table 2 Comparison of calculated K-shell binding energies with measured ones of united atoms with $Z=110, 117, \text{ and } 135$ (keV).

Z	110	117	135
Calculated			
L ^a	182.825	216.958	
FS ^b	182.029	215.948	337.800
D ^c	183.119	217.165	
EE ^d	182.076	216.100	339.116
BE ^e	183.158	217.343	341.040
Measured			
GU ^f	173 \pm 15		315 \pm 10
GU ^g	172 \pm 15	203 \pm 18	

^a DFS method, Ref. 12.

^b DFS method, Ref. 13.

^c DF method, Ref. 14.

^d Energy eigenvalue, present work.

^e Binding energy from Eq. (9), present work.

^f Ref. 27.

^g Ref. 28.

with $\alpha=2/3$, while Lu *et al.* used $\alpha=1$. This is the reason that the values of Lu *et al.* are slightly larger than others. The binding energies obtained in the present work are larger than the energy eigenvalues in the DFS method and in good agreement with the energy eigenvalues in the DF method¹⁴⁾, where the energy eigenvalue gives a good approximation to the binding energy.

Güttner *et al.*^{27, 28)} measured the δ -electron spectra emitted from Ni, Br, I \rightarrow Pb collisions in coincidence with characteristic K x rays of Pb. From the analysis of the so-called spectral function as a function of the minimum momentum transfer, they obtained the K-shell binding energies seen by the electron at the moment of emission. Their experimental values are close to the calculated values in the united atom limit, but smaller than all theoretical values. This fact suggests that the effective nuclear charge seen by the δ electrons is smaller than the sum of nuclear charges in collisions.

The test for the K x-ray emission rates has been made for U. In Table 3, the K x-ray intensity ratios are compared with the experimental values and with other theoretical calculations. The intensity of the $K_{\alpha 1}$ line is set equal to 100. It is clear that the present results agree well with the experimental values as well as other theoretical values.

In Table 4, the K-shell x-ray emission rates for $Z=126$ are presented in units of eV/\hbar and compared with the calculated results by Lu *et al.*¹⁶⁾ in the DFS method and by Anholt and Rasmussen¹⁷⁾ in the DF method. Since the values of Lu *et al.* were obtained with $\alpha=1$, the present results with $\alpha=1$ are also listed in the table. We have already pointed out that the K x-ray emission rates increase with increasing α ³¹⁾. It can be seen that our values with $\alpha=1$ are in good agreement with those of Lu *et al.* and both values are larger than the present values with $\alpha=2/3$. All the values in the table agree with each other within a few percent.

The K-shell x-ray emission rates for elements from $Z=114$ to 164 are shown in

Table 3 Comparison of calculated K x-ray intensity ratios with measured ones for $Z=92$.

X ray	Shell	Measured		Calculated			
		P ^a	BBEH ^b	S ^c	LMC ^d	AR ^e	Present
K α_2	L ₂	60.8 \pm 2.2	62 \pm 1	62.5	62.5	62.73	62.51
K α_1	L ₃	100	100	100	100	100	100
K β_2	M ₂	11.5 \pm 0.6	11 \pm 1	11.53	11.5	11.60	11.52
K β_1	M ₃	22.0 \pm 1.2	24 \pm 1	22.5	22.6	22.59	22.47
K β_3	M _{4,5}	0.75 \pm 0.05		0.864	0.83	0.860	0.851
K $\beta_{1/2}^1$	N ₂	2.9 \pm 0.2		2.88	2.89	2.86	2.87
	N _{3,4,5}	6.1 \pm 0.4		6.09	6.1	6.09	6.03
	O _{2,3}	2.00 \pm 0.15		2.07	2.07	2.04	2.04

^a Ref. 29.^b Ref. 26.^c DFS method, Ref. 30.^d DFS method, Ref. 16.^e DF method, Ref. 17.

Table 5. It is seen from the table that the transitions from L₁, M₁, N₁, and O₁ shells increase with increasing Z . They are magnetic dipole and forbidden transitions for low- Z elements. For high- Z elements, the contributions from magnetic multipoles increase rapidly and in the case of $Z=164$ the L₁-K transition is the strongest K x-ray line.

Other transition rates have a maximum as a function of Z , i.e. decrease for very

Table 4 K-shell x-ray emission rates in $Z=126$ (ev/h).

Shell	LMC ^a	AR ^b	$\alpha=1^c$	$\alpha=2/3^d$
L ₁	5.104	5.17	5.124	5.045
L ₂	100.26	97.9	100.30	99.77
L ₃	125.91	125.1	125.88	125.08
M ₁			1.615	1.587
M ₂	13.37	13.2	13.37	13.35
M ₃	33.25	33.2	33.25	32.95
M ₄	0.998	1.01	0.996	0.981
M ₅	0.849	0.84	0.848	0.836
N ₁			0.511	0.501
N ₂	3.350	3.26	3.351	3.344
N ₃	10.07	10.1	10.07	9.930
N ₄	0.362	0.362	0.362	0.354
N ₅	0.315	0.313	0.315	0.308
O ₁			0.1676	0.1642
O ₂	0.998		0.9984	0.9948
O ₃	3.119		0.3127	0.3079

^a DFS model, Ref. 16.^b DF model, Ref. 17.^c DFS model with $\alpha=1$, present work.^d DFS model with $\alpha=2/3$, present work.

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Table 5 K-shell x-ray emission rates for elements from $Z=114$ to 164 (eV/h).

Shell	114	135	154	164
L_1	1.354	12.99	74.62	148.81
L_2	67.18	124.40	106.42	62.51
L_3	93.13	145.61	154.34	137.30
M_1	0.4362	3.924	18.84	33.36
M_2	10.94	11.83	0.1608	9.644
M_3	23.09	40.38	48.37	46.68
M_4	0.6019	1.276	1.536	1.359
M_5	0.5801	0.9740	0.8861	0.6726
N_1	0.1359	1.234	5.657	9.652
N_2	2.834	2.701	0.2636	3.207
N_3	6.633	12.59	16.21	16.03
N_4	0.2045	0.4797	0.6240	0.5735
N_5	0.1994	0.3755	0.3731	0.2954
O_1	0.04212	0.4127	1.900	3.239
O_2	0.8087	0.8022	0.1131	1.094
O_3	1.912	4.100	5.332	5.441

high- Z values. Anholt and Rasmussen¹⁷⁾ explained this behavior as follows. For high- Z elements, the K-shell electrons are pulled in closer toward the nucleus (called *relativistic contraction*), while the outer-shell electrons are not pulled in so much. This fact reduces the overlap between the K-shell and outer-shell wave functions, and the integral over the product between the overlap and the spherical Bessel function, Eq. (7), becomes small.

For the transitions from M_2 , N_2 , and O_2 shells, there is also a minimum in transition rates. When Z increases, the transition rates reach to a maximum, decrease to a minimum, and then increase again.

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