Bull. Inst. Chem. Res., Kyoto Univ., Vol. 66, No. 6, 1989

M-Shell X-Ray Emission Rates for Dy in Dirac-Fock Approximation

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Received December 26, 1988

X-ray emission rates for vacancies in various M subshells have been calculated for Dy atom. The relativistic Hartree-Fock atomic model with finite-size nucleus was used and the difference of the atomic potentials between initial and final states due to the vacancy transfer was taken into consideration. Contributions from all multipoles were included and the retardation effect was taken into account. The calculated results are compared with those from the frozen-orbital approximation with the relativistic Hartree-Fock and Hartree-Fock-Slater models.

KEY WORDS: M-x-ray emission rates/ Hartree-Fock model/ Relaxed-orbital approximation/

I. INTRODUCTION

When an atomic inner-shell vacancy is produced, it is generally filled by radiative or radiationless transitions. In the former case, a characteristic x ray is emitted, while in the latter an Auger-electron emission takes place. Experimental studies on x-ray spectra can provide much important information in various fields of pure and applied physics, such as atomic and molecular physics, atomic collisions, plasma physics, and solid state physics. Many theoretical calculations of x-ray emission rates with realistic wave functions have been reported and discussed in the recent review of Crasemann.[1]

However, most of these calculations deal with K- and L-shell x-ray emission and the works on M-shell x rays are rather scarce. This is mainly because only limited number of experiments for M-shell x rays have been performed due to experimental difficulties originated from the complex nature of M-x-ray spectra.

With recent advence in high-energy-resolution detectors for low-energy x rays, we can observe various components of M-x-ray spectra separately and precise measurements for M-shell x-ray transitions become possible. Recently Arai *et al.*[2] have measured M-shell x-ray spectra from rare earth compounds bombarded with charged particles and studied the chemical effect on the M x rays. Furthermore, M-shell x-ray emission rates in Dy atom have received a special attention in connection with the measurement of the neutrino rest mass in electron capture decay of ^{163}Ho.[3]

For M-shell radiative transition probabilities, the effect of relativity is significant because such transitions take place only for medium and heavy elements. There have been reported only three relativistic calculations of M-shell x-ray emission rates with self-consistent-field wave functions. Bhalla[4] has calculated M-x-ray emission rates with

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the Dirac-Fock-Slater (DFS) (or sometimes called relativistic Hartree-Fock-Slater) wave functions. He included contributions from all multipoles as well as the retardation effect. His results cover six elements between Z=48 and 93. Similar calculations have been made by Mukoyama and Adachi[5] for rare-earth elements from Z=60 to 72. They also used the DFS models, but evaluated the x-ray transition energies as the difference between the binding energies obtained from the DFS method instead of the experimental values. Chen and Crasemann[6] have computed M-x-ray emission rates in the Dirac-Fock (DF) model for ten elements with atomic numbers $48 \leq Z \leq 92$. They compared the DF values with the DFS values and comparison between the results in the Coulomb gauge and those in the length gauge was also made.

It should be noted that all these calculations are besed on the *frozen-orbital* approximation, i.e. the same atomic potential is used both for the initial state and for the final state. However, it would be more realistic to consider that in the initial state a vacancy exists in an M subshell and the hole moves to an outer shell in the final state. This corresponds to the use of different atomic potentials for the initial and final states. Such a model is called the *relaxed-orbital* approximation.

In the present work, we present numerical results of the M-shell x-ray emission rates for Dy atom in the relaxed-orbital approximation. The reasons why we choose this element are that M-x-ray spectra in rare earth region are extensively studied and that the atomic data for Dy are of great importance in estimation of the neutrino rest mass. The calculations have been made relativistically by the use of the DF wave functions. The contributions from all multipoles and the retardation effect are taken into account. The calculated results are compared with the DFS and the DF values in the frozen-orbital approximation.

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 by[7]

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

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

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

$$\Psi^{\mu}_{n\varkappa} = \frac{1}{r}\begin{pmatrix} G_{n\varkappa}(r)X^{\mu}_{\varkappa}(\hat{\Omega}) \\ \mathrm{i}F_{n\varkappa}(r)X^{\mu}_{-\varkappa}(\hat{\Omega}) \end{pmatrix}, \tag{2}$$

where $G_{n\varkappa}(r)$ and $F_{n\varkappa}(r)$ are the large and small components of the radial wave function multiplied by radial distance r, $X^{\mu}_{-\varkappa}(\hat{\varphi})$ is the spin-angular function, and n is the principal quantum number. The relativistic quantum number $\varkappa$ is defined as $\varkappa = \mp(j+\frac{1}{2})$ for $j = l \pm \frac{1}{2}$, where j and l are the total and orbital angular monentum, 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 to[8]

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

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

$$f_L(\mathrm{m}) = B(-k_i,\ k_f,\ L)\ R_L^2(m)/k, \tag{4}$$

$$f_L(\mathrm{e}) = B(k_i,\ k_f,\ L)\ R_L^2(e)/k, \tag{5}$$

where $B(-\varkappa_i,\ \varkappa_f,\ \mathrm{L})$ is the angular coupling coefficient defined by Scofield[8] 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

$$R_L(m) = (\varkappa_i + \varkappa_f) \int_0^\infty \mathrm{d}r (F_f G_i + G_f F_i) j_L(kr), \tag{6}$$

$$R_L(e) = \int_0^\infty \mathrm{d}r \{ [(\varkappa_f - \varkappa_i)(F_f G_i + G_f F_i) + L(F_f G_i - G_f F_i) j_{L-1}(kr)] + L(G_f G_i - F_f F_i) j_L(kr) \}, \tag{7}$$

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

The radial weve functions were calculated by the DF method.[9] The effect of the finite nuclear size was taken into consideration by assuming the nucleus as a uniformly charged sphere.

In the present work, the so-called *relaxed-orbital* approximation was used. The energy eigenvalues and wave functions in the initial state were obtained for the configuration where a vacancy exists in an M subshell. In the final state, the change in the atomic potential resulting from the rearrangement of atomic electrons due to the electron transition in x-ray emission was taken into account and the separate DF calculations were made for the configuration with a hole in the shell from which the electron fills an M-shell vacancy. The x-ray transition energy was obtained from the difference in the energy eigenvalues of the corresponding electron between the initial and final states.

In the relaxed-orbital approximation, the electron wave functions in the initial and final states are not orthogonal because of different atomic potentials. This fact means that the exchange and overlap effects would be important.[10] However, no theoretical studies of these effects on M-x-ray emission rates have been reported. For simplicity, we neglected the exchange and overlap effects in the present work.

III. RESULTS AND DISCUSSION

The numerical calculations of M-shell x-ray emission rates for Dy atom have been performed on the FACOM M-380Q computer of Institute for Chemical Research, Kyoto University. The electron transitions for filling M-subshell vacancy from all possible shells were considered and the contributions from all electric and magnetic multipoles according to the selection rule were included. For transitions from an open shell, the average emission rates were obtained by scaling the closed-shell rate with the number of available

Table 1. M_1-shell x-ray emission rates in Dy (eV/$\hbar$).

Shell	DFS[a]	DF[b]	Present[c]
M_2	7.807(−5)[d]	8.783(−5)	8.537(−5)
M_3	1.150(−3)	1.185(−3)	1.170(−3)
M_4	4.383(−7)	5.054(−7)	4.836(−7)
M_5	8.914(−7)	1.005(−6)	9.651(−7)
N_1	1.029(−9)	1.003(−9)	2.828(−9)
N_2	5.366(−3)	4.971(−3)	4.826(−3)
N_3	6.043(−3)	5.611(−3)	5.372(−3)
N_4	3.253(−5)	3.138(−5)	3.175(−5)
N_5	4.539(−5)	4.400(−5)	4.446(−5)
N_6	9.846(−8)	1.018(−7)	1.249(−7)
N_7	6.543(−8)	6.520(−8)	7.803(−8)
O_1	2.797(−10)	2.549(−10)	4.996(−10)
O_2	7.923(−4)	6.865(−4)	6.663(−4)
O_3	8.944(−4)	7.534(−4)	7.048(−4)
P_1	2.364(−11)	1.799(−11)	2.407(−11)

[a] Dirac-Fock-Slater model in frozen-orbital approximation (Ref. 5).
[b] Dirac-Fock model in frozen-orbital approximation.
[c] Dirac-Fock model in relaxed-orbital approximation.
[d] 7.807(−5) means 7.807×10^{-5}.

Table 2. M_2-shell x-ray emission rates in Dy (eV/$\hbar$).

Shell	DFS[a]	DF[b]	Present[c]
M_3	3.008(−8)[d]	3.073(−8)	3.507(−8)
M_4	2.371(−3)	2.372(−3)	2.293(−3)
M_5	2.670(−10)	3.293(−10)	3.264(−10)
N_1	2.007(−3)	1.935(−3)	1.800(−3)
N_2	3.330(−10)	3.274(−10)	7.084(−10)
N_3	8.471(−6)	8.201(−6)	7.746(−6)
N_4	1.001(−2)	9.124(−3)	8.957(−3)
N_5	2.997(−8)	3.083(−8)	3.303(−8)
N_6	6.125(−5)	5.849(−5)	6.423(−5)
N_7	6.622(−11)	7.401(−11)	1.053(−10)
O_1	3.351(−4)	3.031(−4)	2.840(−4)
O_2	6.853(−11)	6.354(−11)	1.597(−10)
O_3	1.266(−6)	1.194(−6)	7.141(−7)
P_1	2.657(−5)	1.874(−5)	2.593(−5)

[a] Dirac-Fock-Slater model in frozen-orbital approximation (Ref. 5).
[b] Dirac-Fock model in frozen-orbital approximation.
[c] Dirac-Fock model in relaxed-orbital approximation.
[d] 3.008(−8) means 3.008×10^{-8}.

electrons.

The numerical results for radiative transition rates when a vacancy is in the M subshell are listed in Tables 1–5. The emission rates are expressed in units of eV/ħ. The present values are compared with the DFS and DF values in the frozen-orbital approximation.

The DFS values were taken from the table of Mukoyama and Adachi.[5] On the

Table 3. M_3-shell x-ray emission rates in Dy (eV/ħ).

Shell	DFS[a]	DF[b]	Present[c]
M_4	6.873(−5)[d]	7.114(−5)	6.757(−5)
M_5	8.823(−4)	8.889(−4)	8.486(−4)
N_1	2.626(−3)	2.524(−3)	2.394(−3)
N_2	4.524(−6)	4.395(−6)	4.227(−6)
N_3	3.941(−6)	3.836(−6)	3.653(−6)
N_4	1.233(−3)	1.132(−3)	1.119(−3)
N_5	9.903(−3)	9.103(−3)	8.965(−3)
N_6	7.375(−6)	7.106(−6)	7.711(−6)
N_7	2.201(−5)	2.039(−5)	2.234(−5)
O_1	4.309(−4)	3.861(−4)	3.813(−4)
O_2	6.638(−7)	6.494(−7)	5.033(−7)
O_3	5.729(−7)	5.400(−7)	3.866(−7)
P_1	3.410(−5)	2.386(−5)	3.360(−5)

[a] Dirac-Fock-Slater model in frozen-orbital approximation (Ref. 5).
[b] Dirac-Fock model in frozen-orbital approximation.
[c] Dirac-Fock model in relaxed-orbital approximation.
[d] 6.873(−5) means 6.873×10^{-5}.

Table 4. M_4-shell x-ray emission rates in Dy (eV/ħ).

Shell	DFS[a]	DF[b]	Present[c]
M_5	2.868(−10)[d]	3.135(−10)	3.501(−10)
N_1	4.678(−7)	4.300(−7)	3.583(−7)
N_2	8.751(−4)	8.194(−4)	7.206(−4)
N_3	1.177(−4)	1.109(−4)	9.251(−5)
N_4	1.499(−6)	1.402(−6)	1.232(−6)
N_5	6.141(−7)	5.766(−7)	4.984(−7)
N_6	1.373(−2)	1.236(−2)	1.267(−2)
N_7	4.103(−9)	4.258(−9)	5.381(−9)
O_1	9.082(−8)	9.647(−8)	2.009(−7)
O_2	1.120(−4)	1.024(−4)	1.319(−4)
O_3	1.452(−5)	1.306(−5)	1.689(−5)
P_1	7.359(−9)	5.747(−9)	1.281(−8)

[a] Dirac-Fock-Slater model in frozen-orbital approximation (Ref. 5).
[b] Dirac-Fock model in frozen-orbital approximation.
[c] Dirac-Fock model in relaxed-orbital approximation.
[d] 2.868(−10) means 2.868×10^{-10}.

Table 5. M_5-shell x-ray emission rates in Dy (eV/$\hbar$).

Shell	DFS[a]	DF[b]	Present[c]
N_1	4.342(−7)[d]	4.107(−7)	3.492(−7)
N_2	4.196(−10)	4.129(−10)	4.199(−10)
N_3	7.634(−4)	7.342(−4)	6.270(−4)
N_4	4.140(−7)	3.954(−7)	3.524(−7)
N_5	1.584(−6)	1.521(−6)	1.340(−6)
N_6	6.441(−4)	5.841(−4)	6.003(−4)
N_7	6.389(−3)	5.557(−3)	5.747(−3)
O_1	8.389(−8)	8.447(−8)	1.815(−7)
O_2	6.525(−11)	7.471(−11)	2.283(−10)
O_3	9.410(−5)	8.345(−5)	1.083(−4)
P_1	6.790(−9)	5.219(−9)	1.176(−8)

[a] Dirac-Fock-Slater model in frozen-orbital approximation (Ref. 5).
[b] Dirac-Fock model in frozen-orbital approximation.
[c] Dirac-Fock model in relaxed-orbital approximation.
[d] 4.342(−7) means 4.342×10^{-7}.

other hand, Chen and Crasemann[6] have not listed their results for Dy. Therefore, we have calculated the DF values in the frozen-orbital approximation in the manner similar to the relaxed-orbital values, except that we used the same atomic potential corresponding to the ground-state configuration both for the initial and final states.

It can be seen from the tables that in most cases the present results are in agreement with the values in the forzen-orbital approximation. However, there is about factor-of-two difference in the following cases; the transitions from N_1 and O_1 shells in M_1-shell x rays, from N_2, N_7, and O_2 shells in M_2-shell x rays, from O_1 and P_1 shells in M_4-shell x rays, and from O_1, O_2, and P_1 shells in M_5-shell x rays. It is interesting to note that in all these transitions the contributions from magnetic multipoles are significant.[5] On the other hand, as has been shown previously,[5] in most cases the lowest possible multipoles are dominant in M-shell x-ray emission rates and the contributions from magnetic multipoles are negligible. In these cases, the difference between the frozen-orbital approximation and the relaxed-orbital approximation is small.

We used Eq. (7) as the matrix element for electric multipole transitions. This is based on the Coulomb gauge which corresponds to the dipole velocity form for electric dipole transitions in the nonrelativistic limit. Chen and Crasemann[6] have compared the M-shell x-ray emission rates in the Coulomb gauge with those in the length gauge, which leads to the dipole length form in the nonrelativistic limit. They listed only the values in the length gauge, but from their figures it can be seen that the latter resulsts exceed the former ones by 15–20% for Dy.

In the present work, we neglected the exchange and overlap effects in x-ray emission. According to the results of Scifield, [10] the intensity of $K\beta$ x rays increases considerably by taking into account these effects. However, no calculation has been performed for M-shell x rays. It is interesting to estimate the exchange and overlap effects on the M-shell x-ray emission rates. Such a study is being in progress.

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