Theoretical study on K, L, and M X-ray transition energies and rates of neptunium and its ions*

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The transition energies and electric dipole (E1) transition rates of the K, L, and M lines in neutral Np have been theoretically determined from the MultiConfiguration Dirac–Fock (MCDF) method. In the calculations, the contributions from Breit interaction and quantum electrodynamics (QED) effects (vacuum polarization and self-energy), as well as nuclear finite mass and volume effects, are taken into account. The calculated transition energies and rates are found to be in good agreement with other experimental and theoretical results. The accuracy of the results is estimated and discussed. Furthermore, we calculated the transition energies of the same lines radiating from the decaying transitions of the K-, L-, and M-shell hole states of Np ions with the charge states Np^{1+} to Np^{6+} for the first time. We found that for a specific line, the corresponding transition energies relating to all the Np ions are almost the same; it means the outermost electrons have a very small influence on the inner-shell transition processes.

Keywords: Np ions, K, L, and M X-rays, transition energies, transition rates

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1. Introduction

The long-lived nuclide ^{237}Np is produced in a nuclear reactor when ^{235}U or ^{238}U interact with thermal or fast neutrons; it is one of the most important radiative hazards in the nuclear fuel cycle.[1–4] Its basic nuclear properties are well known, however, the electronic properties are still a subject of intense debate, largely due to the rich and peculiar phenomenology that they display[5,6] and the complicated electronic structure of the 5f state.[7] On the one hand, the experiments are extremely expensive and difficult to perform due to the toxic and radioactive nature. On the other hand, the theory needed to study heavy elements and their ions is necessarily based on complex models, which often need huge computing facilities or much time to be worked out. Therefore, many unanswered questions and arguments still remain unexplained,[8] particularly fundamental questions such as the type of angular momentum coupling and the character of bonding. The theoretical study of Np X-rays is of particular interest in the field of atomic and nuclear physics for it can provide valuable information about electronic structures.

Up to now, many studies about transition energies and rates of neutral Np have been done experimentally and theoretically. Walfson *et al.*[9] obtained electron binding energies of Np from internal conversion spectroscopy in the alpha decay of ^{241}Am. Later, electron binding energies of Np were also studied by others.[10–21] Barreau *et al.*[22] and Ahmad *et al.*[23] measured energies, natural widths and intensities of Np K X-rays, respectively. Jaffe *et al.*[24] and Day[25] analyzed the L X-rays of Np following the decay of ^{241}Am for the first time. Weksler *et al.*[26] measured transition energies of L lines associated with α-decay of ^{241}Am with a high-resolution Si (Li) spectrometer. A recent investigation on these lines was reported by Lépy *et al.*[27] using high-purity Ge and Si (Li) detectors. Krause *et al.*[28] determined energies and widths of $M\alpha_1$, $M\alpha_2$, $M\beta$, $M\zeta_1$, and $M\zeta_2$ X-rays and energies of N_4 to N_7 levels by the photoelectron technique; energies of M and N levels have been calculated by using the Dirac–Fock method. Some other previously experimental works were devoted to the study on the wavelengths of the M X-rays.[29,30] Bhalla[31] calculated the radiative transition rates for a vacancy in the M-shell of Np with the Hartree–Fock–Slater method. Lu *et al.*[32] calculated transition rates of filling a K-shell vacancy with an electron in the L- to P- shells by making use of the same method. Scofield[33] investigated the transition rates for filling the K-shell vacancy with the Hartree–Slater method. Furthermore, to the best of our knowledge, there have been very few works with regard to Np ions, only Desclaux *et al.*,[34] who studied one-electron binding energies of 4d to 5f orbitals, Slater radial, integrals and expected values of r^n for 5f orbital of several Np ions using the Dirac–Fock method. Motivated by the need for accurate results of transition energies and rates in the field of atomic and nuclear physics for Np, there is an

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opportunity here to make actual calculations using the MCDF method.

In this work, transition energies and rates of K, L, and M X-rays of neutral Np and ions with charge states 1+ to 6+ are studied systematically using the atomic structure package GRASP2K based on the MCDF method. In Section 2, the theoretical method is described briefly. In Section 3, the transition energies and rates of neutral Np and its ions are discussed. Finally, some conclusions of this work are given in Section 4.

2. Theoretical method

In this work, the wavefunctions were generated by the widely used atomic structure package GRASP2K[35] based on the MCDF method. Since this method has been discussed extensively elsewhere,[36–38] we only repeat the essential features here. In the MCDF method, the Dirac–Coulomb Hamiltonian of a given atom or ion with N electrons is written as

$$H_{\mathrm{DC}} = \sum_{i=1}^{N} h_{\mathrm{D}}(r_i) + \sum_{i<j}^{N} \frac{1}{r_{ij}}, \tag{1}$$

where h_{D} is the one-electron Dirac operator including the kinetic energy of the electron and its interaction with the nucleus, which can be given by the following expression,

$$h_{\mathrm{D}}(r) = c\boldsymbol{\alpha}.\boldsymbol{p} + \beta c^2 + V_{\mathrm{nuc}}(r). \tag{2}$$

An atomic state function (ASF) of the system can be expressed as a linear combination of configuration state functions (CSFs)

$$|\Psi_\alpha(PJM)\rangle = \sum_{r=1}^{n_{\mathrm{c}}} c_r(\alpha)|\gamma_r(PJM)\rangle, \tag{3}$$

where n_{c} denotes the number of CSF, $c_r(\alpha)$ is the mixing coefficients corresponding to each CSF, which can be obtained by diagonalization of the Hamiltonian matrix, and γ_r represents the configuration and any other quantum numbers required to specify the state, for example, parity, orbital occupancy, coupling scheme and so on. The CSFs are the antisymmetrized linear combinations of the product wavefunctions of relativistic spin-orbitals given by

$$\Phi(r) = \frac{1}{r}\begin{pmatrix} P_{n\kappa}(r)\chi_{\kappa m}(\hat{r}) \\ \mathrm{i}Q_{n\kappa}(r)\chi_{-\kappa m}(\hat{r}) \end{pmatrix}, \tag{4}$$

where n is the principal quantum number; κ and m are the relativistic angular quantum number and its z components, respectively, $\kappa = \pm(j+1/2)$ for $l = (j \pm 1/2)$, with l and j being the orbital and the total angular momenta of the electron. $P_{n\kappa}(r)$ and $Q_{n\kappa}(r)$ are the large and small components of radial wavefunctions and $\chi_{\kappa m}(\hat{r})$ is the spinor spherical harmonic function. This leads to a set of integro-differential equations, which determine the radial wavefunctions for 1s, 2s, 2p, 3s, 3p, 3d, 4s, ... orbitals and the Hamiltonian matrix.

According to the time-dependent perturbation theory, the Einstein spontaneous transition probability for the E1 transition from the upper state β to the lower state α can be given by[39]

$$A_{\beta\alpha} = \frac{2\pi}{2j_\beta + 1} \sum_{M_\beta} \sum_{M_\alpha} |\langle \alpha(P_\alpha J_\alpha M_\alpha)|O^{(1)}|\beta(P_\beta J_\beta M_\beta)\rangle|^2, \tag{5}$$

where $O^{(1)}$ is the electric dipole operator, j_β is the total angular momentum of upper state β, $|\beta(P_\beta J_\beta M_\beta)\rangle$ and $|\alpha(P_\alpha J_\alpha M_\alpha)\rangle$ are the wavefunctions of upper state β and lower state α, respectively.

3. Calculation and discussion

In these calculations, the radial wavefunctions have been obtained by solving the Dirac equations in the Thomas–Fermi potential, followed by screened hydrogenic functions; good starting wavefunctions are provided for the self-consistency procedure, which eventually helps overcoming convergence problems or simply reduces the number of iterations needed for a given set of configurations. Here, we have the ground configurations of each ion as follows: [Rn]$5f^4 6d7s^2$ for neutral Np, [Rn]$5f^4 6d7s$, [Rn]$5f^5$, [Rn]$5f^4$, [Rn]$5f^3$, [Rn]$5f^2$, and [Rn]5f for Np ions with the charge states 1+ to 6+, respectively. The extended optimal level (EOL) scheme[40] is used to determine the initial and final state wavefunctions involved, separately. For example, to calculate the $K\alpha_1$ X-ray transition energy in neutral Np, we set the $1s^{-1}_{1/2}2s^2 2p^6 \cdots (5f^5 6d7s^2)$ as the upper configuration state and the $1s^2 2s^2 2p^{-1}_{3/2} \cdots (5f^5 6d7s^2)$ as the lower configuration state. It is well known that in high accuracy atomic calculations, one may need to go beyond the Coulomb approximation for the two-electron interaction. Based on these wavefunctions, the usual next step is to include the Breit interaction as well as leading QED effects into energy levels as perturbation. In order to emphasize their contributions to the final value, we calculate two kinds of results with and without Breit interaction and QED effects separately. We found that both of these corrections reduce the values of transition energies, which could be evaluated through the given percentage uncertainties. It is important to know that these contributions on K-lines transition energies are more substantial, and then L and M lines, respectively. The contribution of Breit interaction ranges from 0.21% to 0.25% on K X-rays, from 0.04% to 0.40% and 0.08% to 0.29% on L and M X-rays of Np, respectively. Likewise, the QED contributions to the transition energies are as follows: ranges from 0.22% to 0.43% on K X-rays, from 0.32% to 0.40% on L X-rays and from 0.08% to 0.30% on M X-rays. For instance, $K\alpha_1$ is 101055.70 eV, as given in Table 1; its values without QED effects and Breit interaction are calculated as 101494.47 eV and 1010308.00 eV, respectively. That means QED and Breit corrections reduce the

Table 1. Calculated transition energies (eV) and rates (s^{-1}) of K X-rays in neutral Np along with other experimental and theoretical results. The entries in parentheses denote the powers of ten while those in square-brackets refer to the reference number.

Lines	Trans.	Transition energies				Transition rates		
		This work	Expt.[22]	Other Expt.	Calc.	This work	Calc.[32]	Calc.[33]
$K\alpha_1$	K–L_{III}	101055.70	101057±3	101085±4[14]	101054[10]	5.93(16)	6.94(16)	6.94(16)
					101068[13]			
					101056.3[19]			
					101059.80[42]			
$K\alpha_2$	K–L_{II}	97068.18	97069±3	97095±4[14]	97064[11]	4.31(16)	4.36(16)	4.36(16)
				97080±20[23]	97078[13]			
					97069.80[42]			
$K\beta_1$	K–M_{III}	114243.34	114244±3	114240±20[23]	114231.4(56)[41]	6.26(15)	1.57(16)	1.57(16)
$K\beta_2^I$	K–N_{III}	117566.20		117569.32[12]	117581.9(59)[41]	1.88(15)	4.47(15)	4.45(15)
				117510±30[23]				
$K\beta_2^{II}$	K–N_{II}	117352.29		117332.32[12]	117340.3(73)[41]	1.89(15)	2.02(15)	2.01(15)
$K\beta_3$	K–M_{II}	113313.90	113308±4	113300±20[23]	113300.1(54)[41]	7.62(15)	8.03(15)	8.03(15)
				113298±37[21]				

value of $K\alpha_1$ by 438.77 eV and 252.30 eV, respectively; that is, their contributions on the $K\alpha_1$ are about 0.43% and 0.25%, respectively. Meanwhile, we found that including the Breit interaction and QED effect into energy levels, the results are closer to the experiment values, so their influence is of great importance and cannot be neglected for inner-shell transition processes. In any case, these effects merely shift the position of the spectrum in energy and do not affect the profile shape.

The calculated transition energies and rates of K X-rays for neutral Np are listed in Table 1 along with other theoretical results and experimental ones. Overall, a reasonable agreement between these calculated results and other experimental values is found for K X-rays, as can be seen from Table 1. For the transition energies, the largest relative discrepancies between these results and those experimental values[14,22,23] are less than 0.03%, 0.005%, and 0.32%, respectively. Taking the $K\alpha_2$ for example, the transition energy is 97068.18 eV compared with the experimental result 97069±3 eV of Barreau *et al.*[22] For the transition rates of these lines, the results presented are smaller than the other ones, which may due to including the Breit interaction and QED effects in the calculations. In order to compare with the measured spectra,[24] our transition rates of K X-rays are convoluted into a Gaussian line shape of broadening 600 eV and plotted in Fig. 1. As can be seen, they are in very well accordance with the measured spectra of Ahmad *et al.*[23]

In Table 2, we display the transition energies and rates of L X-rays with other available theoretical predictions[33,41,43] and experimental results.[26,27] One can see that the discrepancies in transition energies between our calculations and those available results are relatively small for most of the L X-rays and that the differences are within 5 eV. However, for some lines, such as $L\beta_6$, Ll, $L\eta$, and $L\gamma_4$, the absolute differences in energy from different works are kind of large, comparatively reaching about 20 eV. But the transition energies for

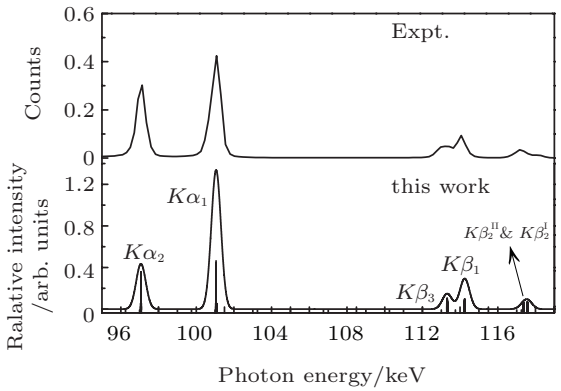


Fig. 1. The calculated K X-rays spectra (bottom) of Np compared with the measured spectra (top) of Ahmad *et al.*[23] The full width at half maximum used is 600 eV.

these lines show good agreement with the experimental values of Lépy *et al.*,[27] the largest percentage uncertainty is less than 0.18%, which is below the relative standard uncertainty (0.8%) of the detector used. With regard to the transition rates of L X-rays, most of them have the same powers of ten as those calculated by Scofield[33] except the lines $L\beta_5$, $L\gamma_1$, and $L\gamma_6$. Also, to compare with the experimental spectra of L X-rays, our transition rates are plotted in Fig. 2 using the same Gaussian line shape of broadening 150 eV as the experimental spectra performed by Johnston.[44]

The transition energies and rates of several M X-rays are presented in Table 3 with the other available data from Bearden *et al.*,[13] Krause *et al.*[28] and Bhalla.[31] It is worth noting that the transition energies are in very good agreement with these results within the experimental error, the largest absolute uncertainty between the MCDF calculations and other experimental values of Ref. [28] is 2.56 eV, which is less than 0.09%. As can be seen, the transition energies of M X-rays are much less than those of K and L X-rays by one and two powers of ten, respectively. In regard of the transition rates of these lines, there are no serious discrepancies being

Table 2. The calculated transition energies (eV) and rates (s^{-1}) of L X-rays in neutral Np along with other experimental and theoretical results. The entries in parentheses denote the powers of ten.

Lines	Trans.	Transition energies					Transition rates	
		This work	Expt.[26]	Expt.[27]	Calc.[41]	Calc.[43]	This work	Calc.[33]
$L\alpha_1$	L_{III}–M_V	13943.86	13944	13950	13943.0(13)	13944.1	2.30(15)	3.84(15)
$L\alpha_2$	L_{III}–M_{IV}	13759.55	13760	13760	13757.2(14)	13759.7	2.08(14)	4.38(14)
$L\beta_1$	L_{II}–M_{IV}	17752.27	17750	17750	17746.9(16)	17750.2	1.15(15)	5.23(15)
$L\beta_2$	L_{III}–N_V	16840.06	16835	16840	16840.3(26)	16840.0	1.03(14)	8.11(14)
$L\beta_3$	L_I–M_{III}	17992.99	17992	17990	17990.8(36)	17989	2.76(14)	1.17(14)
$L\beta_4$	L_I–M_{II}	17061.26	17061	17060	17059.5(34)	17060.7	1.39(15)	1.38(15)
$L\beta_5$	L_{III}–O_{IV}	17503.58	17500	17500		17508.1	4.11(13)	1.74(14)
$L\beta_6$	M_{III}–N_I	16111.15	16109	16110	16110.2(30)	16130	5.86(13)	6.91(13)
$L\gamma_1$	L_{II}–N_{IV}	20778.00	20784	20780	20784.5(28)	20784.4	1.88(14)	1.20(15)
$L\gamma_2$	L_I–N_{II}	21096.56	21099	21100	21099.7(53)	21110	3.75(14)	3.74(14)
$L\gamma_3$	L_I–N_{III}	21340.71	21340	21340	21341.2(39)	21340	1.17(14)	3.65(14)
$L\gamma_4$	L_I–O_{II}	22157.77	22144	22120		22200	6.52(13)	9.18(13)
$L\gamma_5$	L_{II}–N_I	20111.29	20099	20100	20099.9(32)	20120	4.02(13)	4.03(13)
$L\gamma_6$	L_{II}–O_{IV}	21484.12	21491	21490		21488	1.73(13)	2.36(14)
$L\gamma_8$	L_{II}–O_I	21254.51	21262	21260			9.22(12)	1.05(13)
$L\gamma_{13}$	L_I–P_{II}	22400.28		22400			5.46(12)	3.12(13)
Ll	L_{III}–M_I	11863.10	11887	11870	11870.8(21)	11890	1.84(14)	2.69(14)
$L\eta$	L_{II}–M_I	15855.38	15877	15860	15860.5(26)	15876	7.97(13)	8.70(13)

Table 3. The calculated transition energies (eV) and rates (s^{-1}) of M X-rays in neutral Np along with other experimental and theoretical results. The entries in parentheses denote the powers of ten.

Lines	Trans.	Transition energies				Transition rates	
		This work	Expt.[28]	Calc.[13]	Calc.[28]	This work	Calc.[31]
$M\alpha_1$	M_V–N_{VII}	3263.26	3260.7(1)	3261.4	3263.2	3.03(13)	2.96(13)
$M\alpha_2$	M_V–N_{VI}	3252.32	3249.4(4)	3249.2	3250.7	1.49(12)	2.02(12)
$M\beta$	M_{IV}–N_{VI}	3435.70	3434.2(2)	3435.3	3435.8	8.82(13)	4.42(13)
$M\gamma$	M_{III}–N_V	3666.15		3664.4	3667.6	5.67(13)	2.04(13)
$M\zeta_1$	M_V–N_{III}	2579.51	2579.1(8)	2579.0	2575.0	3.56(12)	1.89(12)
$M\zeta_2$	M_{IV}–N_{II}	2520.29	2521.6(1.2)	2522.6	2513.6	7.67(12)	6.02(12)

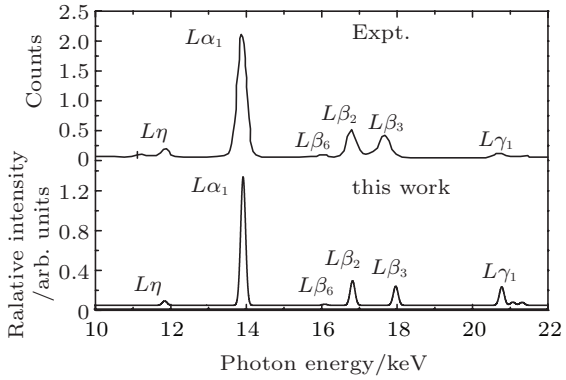


Fig. 2. The calculated L X-ray spectra (bottom) of Np compared with the measured spectra (top) of Johnston.[44] The full width at half maximum used is 150 eV.

found. All the transition rates of M Xx-rays are convoluted into a Gaussian line shape with broadening of 35 eV and plotted in Fig. 3 in order to be compared with future experimental spectra. In Table 4, we compare the binding energy differences of the L_{II}–L_{III} and M_{II}–M_{III} derived from the $K\alpha_1$–$K\alpha_2$ and $K\beta_1$–$K\beta_3$ energy differences with those obtained from the L_{II}, L_{III}, M_{II}, and M_{III} binding energies given by Hagström *et al.*,[10] Bearden *et al.*[13] and Nelson *et al.*[21] A comparison of our results with previous theoretical and experimental ones shows a reasonable accuracy. However, any improvements in theory, such as inclusion of the higher order corrections of the various contributions to the transition energies, would require measurements of feasible accuracy, which yields a good accord between theoretical and experimental values.

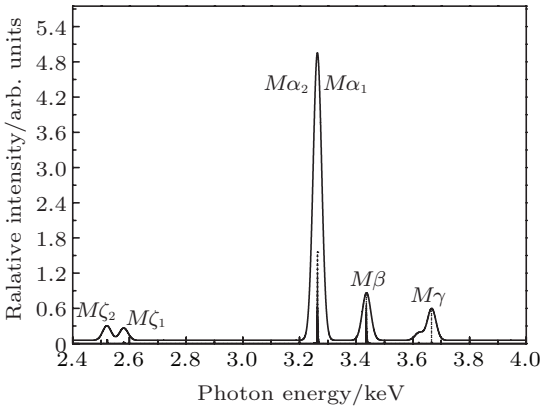


Fig. 3. The calculated M X-rays spectra convoluted with the full width at half maximum of 35 eV.

Table 4. The calculated L_{II}–L_{III} and M_{II}–M_{III} binding energy differences (keV) of Np along with other theoretical[10,13] and experimental[21] results.

Trans.	This work	Calc.[10]	Calc.[13]	Expt.[21]
L_{II}–L_{III}	3.988	3.990	3.990	3.989±0.008
M_{II}–M_{III}	0.929	0.932	0.931	0.953±0.039

On the basis of the above comparisons of transition energies and rates of K, L, and M X-rays emitted by the K-, L- and M-shell hole states in neutral Np, we further calculated transition energies of the same radiative lines from the Np^{1+} to Np^{6+} ions for the first time; the results are listed in Table 5. It is found that for a specific line, the corresponding transition energies related with all the Np ions are almost the same. For the majority of the K, L, and M X-rays, the energies decrease very slightly with increasing ionization state, while the transition energies of the $L\beta_2$, $L\beta_5$, $L\beta_6$, $L\gamma_{13}$, $M\alpha_1$, $M\beta$, and $M\gamma$ lines increase very slowly. For example, for the K X-rays, the largest difference in energy between the same lines emitted from Np^{1+} and Np^{6+} ions is only about 5.0 eV, while the difference is about 13.30 eV for both the L and M X-rays. So we conclude that the outermost electrons have a very small influence on the inner-shell transition processes. According to this, we infer that the transition energies of these lines from the corresponding oxides also have similar features due to the negligible effects of the co-valence electrons.

Table 5. The calculated transition energies (eV) of K, L and M X-rays for the Np^{1+} to Np^{6+} ions.

Lines	Np^{1+}	Np^{2+}	Np^{3+}	Np^{4+}	Np^{5+}	Np^{6+}
$K\alpha_1$	101043.60	101043.20	101042.90	101042.20	101041.90	101041.20
$K\alpha_2$	97050.30	97048.04	97047.96	97047.47	97047.24	97046.67
$K\beta_1$	114206.90	114206.80	114206.50	114205.10	114204.90	114203.30
$K\beta_2^{\rm I}$	117556.80	117556.10	117555.10	117554.60	117554.40	117554.20
$K\beta_2^{\rm II}$	117312.70	117310.20	117309.50	117310.20	117308.20	117307.70
$K\beta_3$	113276.20	113275.40	113274.70	113274.20	113273.20	113272.60
$L\alpha_1$	13938.26	13937.47	13937.39	13937.11	13936.82	13936.44
$L\alpha_2$	13756.94	13753.10	13752.39	13752.25	13752.08	13751.60
$L\beta_1$	17751.75	17748.07	17747.14	17747.00	17746.74	17746.21
$L\beta_2$	16829.21	16829.59	16829.99	16830.01	16830.35	16830.77
$L\beta_3$	17992.67	17992.51	17992.46	17991.67	17991.23	17990.46
$L\beta_4$	17062.32	17061.19	17060.80	17060.62	17059.90	17059.89
$L\beta_5$	17502.11	17502.32	17502.67	17502.88	17505.58	17505.76
$L\beta_6$	16096.54	16096.41	16096.17	16096.12	16096.08	16095.02
$L\gamma_1$	20785.54	20779.87	20779.24	20778.64	20778.50	20778.31
$L\gamma_2$	21098.78	21095.99	21095.65	21095.48	21094.93	21094.87
$L\gamma_3$	21342.39	21342.13	21341.38	21341.11	21340.95	21340.82
$L\gamma_4$	22160.67	22159.85	22158.97	22158.91	22157.48	22157.43
$L\gamma_5$	20088.94	20091.81	20091.54	20091.23	20090.65	20090.62
$L\gamma_6$	21482.70	21484.33	21486.46	21487.90	21490.06	21494.56
$L\gamma_8$	21256.82	21256.73	21256.69	21256.45	21254.70	21254.00
$L\gamma_{13}$	22400.09	22400.53	22401.39	22405.18	22407.36	22413.40
Ll	11856.87	11856.44	11856.23	11856.09	11855.59	11855.18
$L\eta$	15848.72	15851.56	15851.30	15850.92	15850.73	15850.05
$M\alpha_1$	3262.29	3262.63	3262.86	3263.09	3263.76	3267.49
$M\alpha_2$	3251.78	3251.54	3244.86	3243.32	3242.54	3238.44
$M\beta$	3434.92	3432.89	3436.50	3436.65	3436.69	3440.10
$M\gamma$	3666.10	3665.57	3665.84	3666.11	3667.34	3668.63
$M\zeta_1$	2575.33	2575.19	2575.09	2574.96	2574.69	2574.49
$M\zeta_2$	2514.88	2510.51	2513.68	2514.33	2514.75	2514.80

4. Conclusion

Motivated by the need for accurate transition data in a variety of scientific applications, in this work, the transition energies and rates for the K, L, and M X-rays in neutral Np have been calculated using the MCDF method. Throughout the calculations, the contributions from the Breit interaction and QED effects, nuclear finite mass and volume effects are also considered. The influences of the Breit interaction and QED effects are found to be of great importance and cannot be neglected for the inner-shell transition energies. We see that Breit interaction and QED effects are important in precision calculations of heavy atoms. Their contributions are larger in the innermost transition while the trend reverses to be small when one steps toward the outer-shells' orbitals. These results have been compared with other available experimental and theoretical ones and good agreement between them was found. In addition, we further calculated the transition energies of X-rays radiated by the K-, L-, and M-shell hole states of Np ions with the charge states 1+ to 6+ for the first time.

One can find that for a specific line, the corresponding transition energies related with all the Np ions are almost the same. Also, for the majority of the K, L, and M X-rays, the transition energy decreases very slightly with increasing the ionization state, while for the transition energy of the $L\beta_2$, $L\beta_5$, $L\beta_6$, $L\gamma_{13}$, $M\alpha_1$, $M\beta$, and $M\gamma$ lines, it increases very slowly. Therefore, we summarized that the outermost electrons have very small influence on the inner-shell transition processes.

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