Lifetime of Isomer in High Charge States

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Abstract Nuclear lifetime shift of charged atom, due to lack of internal conversion (isomer) or electron capture transition (β decay), is analyzed. Theoretical predict is given, that electron density of inner shell at nuclear surface decreases and energy level also decreases with increasing of charge state, nuclear lifetime increases slightly with previous reason but increases with a jump when energy level drops to a critical value. The shifts of halflife are compared for internal conversion with different nuclear decay energy and different type of transitions. A common used computer code of this calculation created by ourselves, and the calculation results are compared with experiment one.

Key words lifetime of isomer, high charge state, electron energy level, electron density

1 Introduction

The environment variation of outside nuclei affects the nuclear reaction and decay. The α decay rate and fusion of astro-nuclear reactions are greatly sensitive to the difficulty of penetration through the coulomb barrier. This barrier is slightly different for nuclei surrounded by electron cloud from for bare nuclei. The environmental effect of the electron cloud can slightly disturb the barrier and thus affect the decay rate by the order of 1.5×10^{-7} to 4.0×10^{-5} for ^{226}Ra[1]. In low energy nuclear reactions, the effects of environment are obvious[2], such as transfer reaction[3,4] and the fusion reactions[5], particularly in astro-nuclear reactions.

The nuclear process, in which no electron participates directly, is affected by environment through the change of screening for ejected or projectile particles, such as the β^+, β^- decay.

The radioactive decay process, in which atom electrons participate directly, affected by the electron cloud density outside nuclear surface, like the weak interaction involved in electron capture (EC), and level transition involved in the internal conversion (IC). Those transition rates are essentially proportional to the electron density at the nuclear surface where electrons are available, the most direct way to change the rates is thus to change the total electron density at nuclear surface. In few case, it is through the shift of atom level that shell energy value near the transition threshold energy.

The change of electric environment affects the electric potential field, thus affects the electron wave function. However, the Coulomb field near nucleus is very strong; the shape of the electron wave function is independent with environment but disturbed by environment. The ionization process only slightly affect the distribution region of electron wave function and perturbations the level energy of electron shell. The electron capture and the internal conversion are sensitive to the electron density near the nuclear surface, special for the inner sub-shell. For high charge state, the density magnitude tends to zero, the lifetime of unstable nuclear shift rapidly, to the forbidden electron capture and internal conversion, the final value of lifetime only determine by β^+, γ transition, respectively. This is useful in study of high spin isomer beam in RIKEN and CSR experiment in IMP.

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2 Principles and method

2.1 Isomer and internal conversion

A nucleus in an excited state can decay to a low level or ground state by photon emission, or alternatively, by internal conversion (all transition energy directly transfer to inner shell electron, emitted electron, then, the atom decay by special X ray or Auger electron). In some case like high spin bound ground state, and near closure shell of nuclide with small transition energy and with large angular momentum change, the nuclide have long lifetime for the γ transition forbidden, formed so call "isomer". In some isomer states, the internal conversion takes an important role when the atom is at high charge state, lifetime can be increasing to 600 % of neutral atom[5] in which the internal conversion possibility becomes zero.

A common feature of those processes is sensitivity to the electron density near the surface of nucleus. One can give the relationship of internal conversion coefficient (ICC) between the value $\alpha^{(Q)}$ at charge state Q and the value of $\alpha^{(0)}$ at neutral atom:

$$\alpha^{(Q)} = \alpha^{(0)} \lim_{r\to 0}(\rho^{(Q)}(r)/\rho^{(0)}(r)), \qquad (1)$$

here, the ρ is electron distribution density. Any change of the configuration of out electronic shells will modify the screening and the inner electronic wave functions. The electronic density of near the nucleus, and the ICC will be discussed at next section.

The internal conversion rate and gamma-ray emission rate are proportional to the square of the same nuclear matrix element. Therefore, the internal conversion coefficient, the ratio of those two kinds rate, is independent with nuclear matrix element, and can be calculated accurately. Internal conversion is also forbidden for the transition rule in isomer states, however, a low electron density near the nuclear surface or even no electron outside can forbid the internal conversion. It results in a life difference for the nuclei at the charge states and at neutral atom. In the lowest, non-trivial order of perturbation theory the internal conversion coefficient for transition of pure electric multiple order L is given by

$$\alpha_\sigma(EL) = \pi\alpha W \sum_k \frac{(2j_0+1)(2j+1)}{L(L+1)} \begin{Bmatrix} j_0 & j & L \\ 1/2 & -1/2 & 0 \end{Bmatrix}^2 |R_{kk_0}(EL)|^2 \qquad (2)$$

in addition, for a pure magnetic multiple order L by

$$\alpha_\sigma(ML) = \pi\alpha W \sum_k \frac{(2j_0+1)(2j+1)}{L(L+1)}(k+k_0)^2 \times \begin{Bmatrix} j_0 & j & L \\ 1/2 & -1/2 & 0 \end{Bmatrix}^2 |R_{kk_0}(ML)|^2, \qquad (3)$$

here k_0 k, describe the angular quantum of initial state of bound electron and that of final state of the continuum electron, α is the fine structure constant $e^2/\hbar c$, W is the nuclear transition energy, index σ is atomic sub-shell of the ejected electron. The total conversion coefficient is the sum ICC of all possible sub-shell.

The transition matrix elements R, only including the static electric contribution, is defined by

$$R_{kk_0}(EL) = \int_0^\infty dr\left\{Wrh_L^{(1)}(Wr)[g_kf_{k_0} - f_kg_{k_0}] \times \left[\frac{d}{dr}rh_L^{(1)}(rW)\right][g_kg_{k_0} - f_kf_{k_0}]\right\}, \qquad (4)$$

and by

$$R_{kk_0}(ML) = \int_0^\infty dr[g_kf_{k_0} - f_kg_{k_0}]h_L^{(1)}(Wr), \qquad (5)$$

where $h_L^{(1)}(Wr)$ is a spherical Hankel function of first order, g_k, f_k are the Dirac radial wave functions for projectile electron, g_{k_0}, f_{k_0} are the Dirac radial wave functions for initial occupied bound orbit. Calculation of the quantity $g_k(Z,W,r)$, $f_k(Z,W,r)$ requires Dirac equation. For the central potential $V(r)$, the radical components are a set of coupled differential equations[6]

$$\left[\frac{d}{dr} + \frac{(k+1)}{r}\right]g_k - (W+1+V(r))f_k = 0,$$
$$\left[\frac{d}{dr} - \frac{(k+1)}{r}\right]f_k + (W-1+V(r))g_k = 0, \qquad (6)$$

where

$$k = l \text{ for } j = l - 1/2,$$
$$k = -l - 1 \text{ for } j = l + 1/2,$$

l is angular quantum number of the ejected orbit electron. As an application for internal conversion, $V(r)$ should include the effects of nuclear charge distribution and of the screening by the orbital electron. Bambynek[7] reviewed the solution in the screening potential and in the electron exchange potential. For general purpose, the interpolation conveniently gives the values of neutral atom ICC from Hager and Seltzer[8] table and Dragoun[9] table.

Isomer states are formed by forbidding of nuclear transition, but the ICC is determined by the static electric environment, the ICC value of isomer state is the same as that of other energy states.

2.2 Wave function and ICC of charged atom

The WKB screening approximation is the best describing the screening effect of the electron cloud on nuclear coulomb field in β decay and the internal conversion. The method suggested by Rose[10] using electron wave function replacements[11]. For our propose, the function replaced from the screening to no screening by

$$g_k^2(Z,W,r) \to \frac{P'W}{W'P}g_k^2(Z,W,r),$$
$$f_k^2(Z,W,r) \to \frac{P'W}{W'P}f_k^2(Z,W,r), \tag{7}$$

Where $W' = W - V_0'$, here V_0' is the difference between the coulomb potential energy and the exact potential energy of the continuum electron at the nuclear radius R. P is the momentum of internal conversion electron $P = \sqrt{W^2 - 1}$ and P' is the momentum of ejected electron, respectively. $\hbar = c = m_e = 1$. In our case, the wave function is neutral atom of self-consistent potential one in which the screening is included, the correction is a reverse process of Ref. [11] and V_0 replace by V_0' which is the change of potential screening at nuclear surface for ionization.

The study on the WKB screening approximation shows that WKB screening method should be correct for $P^2 \gg WV_0'$[12], and ratio of λ (screened)/λ (unscreened) of β decay, here λ is transition probability, shows that the WKB is overestimates the screening correction for very low energy of β decay and for high nuclear charge Z[13]. Using unscreened Coulomb potential, screening self-consistent potential including the exchange effect of electron and WKB screening approximation, Matese's calculation[14] shows that the WKB is in good agreement with the self-consistent potential in a large range of β transition energy and a large range of nuclear charge number Z.

The potential energy shift in atom is written in $V_0' = a\alpha^2 Z^{4/3}$ according to Thomas-Fermi model, between no screening effect and neutral atom. The Thomas-Fermi parameter a equal to 1.96 to 1.81 for light to heavy nuclei. But a is also a function of Z, a values are taken from Garrett[15] and their interpolation in this work. For the low energy, we use P dependent screening potential of form[16],

$$V_0'(Z,P) = V_0'(Z)\exp\left(-\frac{a_s}{P} - \frac{b_s}{P^2}\right), \tag{8}$$

where P is a momentum of the screened electron, the parameter a_s and b_s are

$$a_s = -0.102 + 0.238 \times 10^{-2} Z + 0.101 \times 10^{-4} Z^2 - 0.111 \times 10^{-6} Z^3,$$
$$b_s = 0.0156 - 0.360 \times 10^{-4} Z - 0.383 \times 10^{-5} Z^2 + 0.242 \times 10^{-7} Z^3.$$

The internal conversion coefficient α is a sum over all sub-shell, and the relationship between the radial electron density and the electron wave function is

$$\rho(r) = \frac{1}{r^2}\sum_\sigma W_\sigma [f_\sigma^2(r) + g_\sigma^2(r)], \tag{9}$$

where the index σ of f and g indicates sub-shell of wave function, the wave function dependent with energy and momentum. We can deduce equation (1) to

$$\alpha^{(Q)} = \sum_\sigma \left[\alpha_\sigma \left(\frac{P'W}{PW'}\right)_\sigma \left(\frac{N_\sigma^{(Q)}}{N_\sigma^{(0)}}\right)\right], \tag{10}$$

where $N_\sigma^{(Q)}$ and $N_\sigma^{(0)}$ are the number of the electrons in σ sub-shell for charged and neutral atom, respectively. If the shape and space distribution of electron in sub-shell cannot be changed, the equation (10) approximately is written into

$$\alpha^{(Q)} = \sum_\sigma \left[\alpha_\sigma \left(\frac{N_\sigma^{(Q)}}{N_\sigma^{(0)}}\right)\right]. \tag{11}$$

2.3 Shell energy shift and ICC

From neutral to charged atom, the change of screening and exchanging potential modify the electron wave function according to equation (6), and give the shell energy a perturbation. Formula (8) gives us the static electric potential change at the nuclear surface before unscreening to screening, and V_0' is the sum of the shell potential change, but it is dominated by the K, L shell potential changes. In some case, V_0' approximately equal to ΔE_K. The nuclear Coulomb potential is described as

$$V(r) = -\frac{Z}{r} + \sum_{\sigma=1}^{\sigma_N}\int \rho_\sigma(r')\frac{dr'}{r-r'} - \left(\frac{81}{8\pi}\right)\sum_{\sigma=1}^{\sigma_N}(\rho(r)_\sigma)^{1/3}$$

here the first term is nuclear electric potential, the second is electron-screening term, and the third is exchanging term after replacing by the Slater's average value. In ionization process, the potential change is contributed by screening and exchanging of ejected outer electron

$$V_0(Z,Q) = \sum_{\sigma_M}^{\sigma_N}\int \rho_\sigma(r')\frac{dr'}{r-r'} - \left(\frac{81}{8\pi}\right)\sum_{\sigma_M}^{\sigma_N}(\rho(r)_\sigma)^{1/3} \tag{13}$$

σ_M, σ_N are inner and outer sub-shell of charged atom. The $V_0'(Z)$ of neutral ${}_{20}\mathrm{Ca}$ is different from the difference between the value ${}_{50}\mathrm{Sn}^{30+}$ and neutral ${}_{50}\mathrm{Sn}$. The V_0 of ${}_{20}\mathrm{Ca}^{20+}$ is different from that of ${}_{20}\mathrm{Ca}^{18+}$. In equation (8), the P dependent part is tiny modification only acting at very low momentum. Another part is proportional to $Z^{4/3}$, Z is the nuclear charge number or the number of electrons for neutral atom. We assume the V_0 for a atom with charged number Q and nuclear charge number Z is $V_0(Z,Q) = V_0(Z) - V_0(Z-Q)$ for P independent part, and energy level shift V_σ approximately are written as

$$V_\sigma = \lim_{r\to 0} V_0(Z,Q,P)\frac{\rho_\sigma(r)}{\sum_\sigma \rho_\sigma(r)}. \tag{14}$$

Energy level shift of atom is important for some case of internal conversion. Internal conversion occurs at the electron which has smallest bound energy. In some case, the transition changes from possible for neutral atom to impossible for charged atom, the nuclear half-life has suddenly changed in ionization process. As nuclear transition energy E is less than V_σ, the internal conversion is forbidden for the subshell σ.

Un-screening energy level of atom obeys the Bohr formula for hydrogen-like level. In this work, every shell and sub-shell energy use a well known table of bound energy of atom, and formula (13),(14).

2.4 Effect of charge states on electron capture

Electron capture process (ε) for proton rich heavy nuclei, like the internal conversion, involves the bound electron. For lacking electron in charged atom, halflife time is increased comparing with the neutral atom. If the Q value of the ε process is Q, bound energy of the electron is E, the neutrino energy (neglecting the atomic recoil energy) is q_x ($q_x = Q - E_x$), linear momentum of the bound electron is p_x, and the electron density near the nuclear surface is ρ and the factor associated electron exchange and overlapping is B_x, the capture probability become

$$\lambda = M_L^2\frac{(2L-2)!!}{(2L-1)!!}\sum_x \frac{n_x p_x^{2(k_x-1)} q_x^{2(L-k_x+1)}\rho B_x}{(2k_x-1)![(2(L-k_x)+1)!]}, \tag{15}$$

where L is the angular momentum of electron capture transition, M_L is transition matrix element.

On one hand, the screening reduction changes the potential field, also changes the electron wave function and reduces the density ρ of the electron near nuclear surface and the rates of the electron capture for the same sub-shell. On the other hand, decreasing electron number n_x in shell x results in proportionally reducing the contribution of the sub-shell. Decreasing the occupation number in the shell causes reducing the terms of sum. If no electron outside nucleus, of course, $\lambda = 0$, all nuclear half-life are determined only by β^+.

2.5 γ transition brunch ratios

Many isomer decays have several brunches, and brunch ratios have to be given by experimental data or by calculation. In our code, if there is no experimental data of the ratios, the theoretical rates of electromagnetic transition are given by the Weisskopf single particle estimates, the decay width for multiple radiation with energy E[17] are

$$\Gamma(\mathrm{E}L) = 21.77\left(\frac{L+1}{L}\right)\left(\frac{3}{3+L}\right)^2 \times \frac{A^{2L/3}}{[(2L+1)!!]^2}\left(\frac{E_\gamma}{164.44}\right)^{2L+1} \tag{16}$$

$$\Gamma(\mathrm{M}L) = 6.69\frac{\Gamma(\mathrm{E}L)}{A^{2/3}}.$$

3 Results and discussion

Internal conversion coefficient (ICC) is the function of transition energy, the type of transition, and charge states. In Fig.1(a), the relationship of half-life shift to the charge states is shown for different transition energy for the ^{149}Dy nucleus with E2 type transition. If the transition energy is small, the half-life shift is large; increasing the transition energy, the half-life shift is decreasing rapidly.

The Fig.1(b) is shown the ^{149}Dy at transition as-

sumed a 50keV energy level, for different transition types. The half-life shift is large at a higher multi-polarity, and the magnetic transitions (M-type) are of stronger effect than the electric transition (E-type) at near bare nuclei. The lifetime of excited nucleus states in highly charged atoms is increased to the maximum, when it is bare, that lifetime determine only by the γ transition and internal conversion is forbidden.

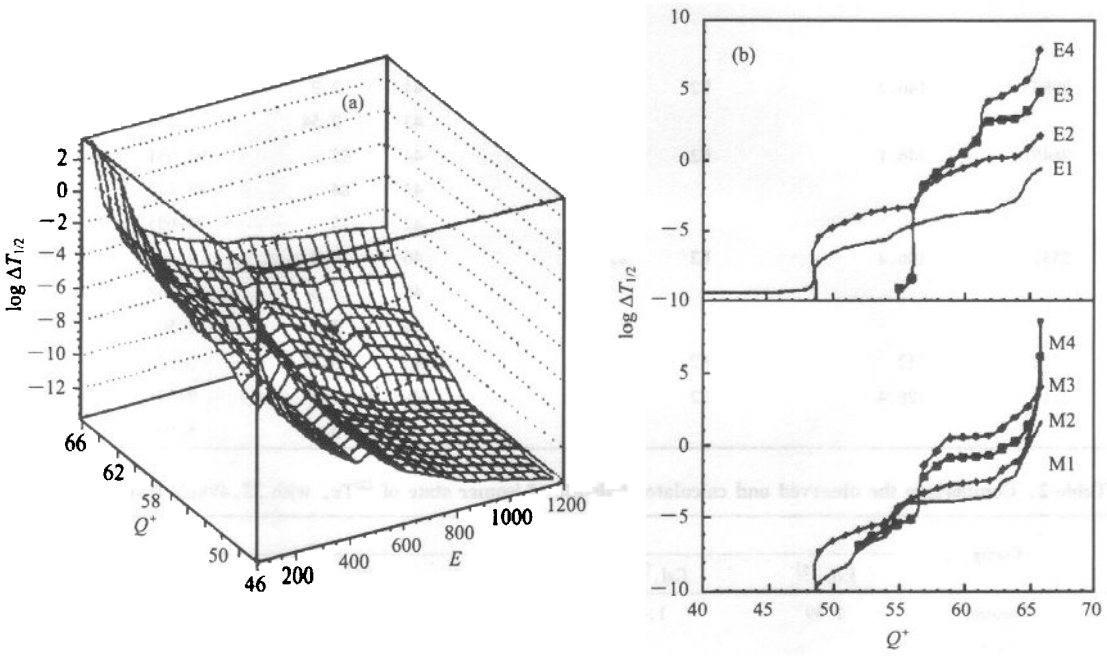

Fig.1. Relationship of the half-life shift with charge states of ^{149}Dy for different energy (E2) (a) and for different transition type (energy = 50 keV) (b).

Table 1. Compare our calculation to Grzywacz calculation and experiment lifetime.

Isomer	E^*/keV	E_T/keV	Type	$T_{1/2}{}^c$/μs	Q^+	$T_{1/2}{}^a$/μs	$T_{1/2}{}^b$/μs	$T_{1/2}{}^c$/μs
^{18}F	112.1	184.16	E2	0.162	9	0.163	0.162	0.162
^{22}Na	583	583	E2	0.244	11	0.244	0.244	0.244
^{63}Ni	87.15	87.15	(M3)	1.67	28	3.34	3.344	3.344
^{66}Cu	1154	563.34	M2	0.596	29	0.598	0.596	0.596
^{71}Se	260.5	260.5	(E1)	19	34	20.9	19.278	19.278
					33	20.1	19.245	19.244
	79.23	79.23	E2	0.34	38	1.4	1.166	1.166
	266	266.3	E2	0.178	39	0.184	0.182	0.182
					38	0.181	0.181	0.181
	2956	98.68	E2	0.457	38	0.946	0.966	0.966
					37	0.670	0.666	0.660
	2888	76.99	E2		40	5.13	4.862	4.862
					39	1.84	2.374	2.324
					38	1.59	1.569	1.527
^{90}Mo	2875	62.9	E2		42	8.3	7.754	7.754
					41	2.4	2.440	2.337
					40	1.40	2.306	1.376
	2034	50.1	E2	3.76	41	57	57.492	57.492
					40	8.8	10.730	9.995
^{92}Nb	2203	115.8	E2	0.167	41	0.28	0.281	0.281
^{92}Mo	2761	147.8	E2	0.190	42	0.246	0.246	0.246

Table 1. (continued)

Isomer	E^*/keV	E_T/keV	Type	$T_{1/2}{}^e$/μs	Q^+	$T_{1/2}{}^a$/μs	$T_{1/2}{}^b$/μs	$T_{1/2}{}^c$/μs
					41	0.221	0.219	0.219
					40	0.201	0.198	0.198
^{92}Tc	270	56.34	E2	1.03	43	11.3	11.243	11.243
					42	2.33	2.659	2.500
					41	1.30	1.505	1.406
^{93}Ru	2083	146.2	E2	2.150	44	2.9	2.878	2.878
					43	2.54	2.533	2.528
^{94}Ru	2645	146.1	E2	71	44	95	94.981	94.981
					43	84	83.625	83.438
					42	75	74.685	74.396
^{96}Pd	2531	106.4	E2	2.2	46	4.7	4.647	4.647
					45	3.3	3.258	3.225
					44	2.5	2.507	2.470
^{100}Rh	112	112	E2	0.130	45	0.25	0.242	0.242
^{105}Cd	2517	126.4	E2	4.5	48	7.4	7.376	7.376
					47	5.9	5.899	5.866

Table 2. Comparison the observed and calculated half-life of isomer state of ^{125}Te, with 35.49keV transition energy.

Charge state	Config.	$T_{1/2}$/ns				$E_b{}^K$/keV	
		Exp.[5]	Cal.[5]	This work[a)]	This work[b)]	Cal.[5]	This work
0	(neutral)	1.49	1.49	1.49	1.49	31.81	31.81
40	$(3s^2)$			2.1	1.50		34.17
44	$(2s^22p^4)$		1.5	2.8	1.53	35.27	35.09
45	$(2s^22p^3)$	< 2	8.0	3.3	1.54	35.58	35.33
46	$(2s^22p^2)$	< 2	8.3	3.9	1.58	35.91	35.56
47	$(2s^22p^1)$	6 ± 1	8.5	15.1	10.0	36.26	35.81
48	$(2s^2)$	11 ± 2	10.3	40.3	10.2	36.60	36.04
49	$(2s^1)$	13.9[19]		55.8	13.9		36.28
50	$(1s^2)$			90.3	21.9		36.53

a) This work, M1 and E2 mixture type transition.

b) This work, M1 type transition.

Internal conversion occurs for the electron at the surface of nucleus, and the probability is proportional to the electron density at surface of nuclei. Increasing the number of the charge state, the contributions of reducing the density are termination of outer orbit and shift of inner orbits, but the density is dominated by inner orbits. The shifts of lifetime are mainly contributed by 1s and 2s orbits.

The half-life time of Grzywacz[18] calculation (row a), our simple calculation with equation (11) (row b), with wave function shift equation (10) (row c) and experimental one (row e) shown in table 1. The experimental half-life time, possible transition types, excited energy and other parameters take from[18] and its references, or from NNDC nuclear database. It is shown that all our results agree with Grzywacz calculation very well, and our simple way can give the rather correct value.

In table 2, the first excited state of ^{125}Te is an isomer with a half-life of 1.49 ns for neutral atom, the excited energy of the isomer is 35.49 keV, the transition type is E2 and M1 mixture, which refer from NNDC, E2 is about 88 % and M1 is 12 %. Comparing the calculated half-life of charged atom from equation (16) using different possible transition types with experimental value, we find that the calculation in Ref.[5] could not gives a half-life time jump at charge state 47, our calculations (row a and row b) give a reasonable jump at charge state 48, here we use the mixture transition in row a, and pure M1 transition in row c, the values of half-life time suggest that the transition should be pure M1 transition. Only the ICC li-

fetime for pure M1 transition is 13.9μs, the same as the experimental value[19] for charge state 49. This is good example of available in neutral but blocking in the high charge state for K-shell of internal conversion, so the lifetime has a jump when charge number greater than 46. It is very sensitive to the shell energy/or the threshold of internal conversion.

For an isomer beam with energy greater than 10MeV/nucleon, where the atoms are nearly bare, some isomer states for a special transition type may have a long lifetime[5]. At beam energy lower than 10MeV/nucleon or a transition with higher energy than 400 keV, the shift of the isomer lifetime is relatively small, it is the same for EC decay process. Although the shifts of nuclear lifetime cannot be measured in normal experiments, this effect will become very important for the beam storage in the CSR with long time or the nuclear particle is in the plasma states in environment of astro-nuclear reactions.

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高电荷态同质异能态的寿命

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摘要　分析了带电原子由于缺少内转换(同质异能态)或电子俘获跃迁(β衰变)引起的核寿命的变化. 理论预言了随着电荷态的增加,内壳的电子在核表面的电子密度降低,内壳电子能级也降低,不稳定原子核的寿命由于前一原因而缓慢增加,但当内壳电子能级降到特殊能量时产生剧烈增加. 比较了由于不同的跃迁能量和跃迁类型对内转换过程的半衰期的影响,并将计算的理论值与一些实验测量值进行了比对. 并开发了进行这类计算的公共计算程序.

关键词　同质异能态寿命　高电荷态　电子能级　电子密度

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