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Double Vacancy Formation in L Shell Accompanying the Internal Conversion of ^{234}U 43.48 keV Transition

Nobutsugu IMANISHI and Ichiro FUJIWARA

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A double vacancy formation in L shell has been studied experimentally for the internal conversion of the ^{234}U 43.48 keV transition. A "hypersatellite" of L_α X rays was observed first at an energy 230±60 eV higher than that of normal X rays. This energy shift gives a value of 0.78±0.14 as an effective screening parameter ΔZ. The probability per L shell conversion forming the double vacancy in L shell and ending in the emission of the L_α hypersatellite was measured to be $(2.1 \pm 0.4) \times 10^{-4}$. The result is compared with theoretical predictions.

KEY WORDS: Internal conversion/ Double vacancy/ L shell/ Hypersatellite/ ^{234}U/ Si(Li) detector/

I. INTRODUCTION

Following nuclear decay the electronic cloud is forced to change itself. To search phenomena caused by the change has been one of the important and stimulating problems, since Curie and Joliot[1] discovered the emission of X rays following α decay. To understand this phenomenon theoretically, Migdal[2] and Feinberg[3] proposed independently a "shakeoff type process", in which an electron is excited or ejected following the sudden change of nuclear charge in the nuclear decay. Many experimental studies[4] stimulated by the theory have been done especially for the case of β decay, and the picture is becoming clearer for that case.

The excitation or ejection following the internal conversion, however, would be more complicated than that in β decay. That is, in addition to the following processes similar to those found in β decay:

1) "shakeoff" type process, and
2) direct collision between the converted and unconverted L electrons,

the following higher-order electronic transitions can give rise to a double vacancy formation:

3) double conversion process, and
4) internal conversion of an internal Compton photon.

So far, only a few nuclei with atomic number $Z \simeq 50$ have been studied experimentally,[4,5] and the accumulation of data is required for understanding the phenomena.

In the present work, the probability forming a double vacancy was measured for the internal conversion transition of the 43.48 keV in ^{234}U, as an examples of high atomic number. This state is formed following the α decay of ^{238}Pu ($T_{1/2}$=87.74 y). Therefore,

今西信嗣, 藤原一郎: Research Section of Nuclear Fuel, Institute of Atomic Energy, Kyoto University Uji, Kyoto 611.

in addition to the above four processes, the following process will be expected:

5) shake off process in the α decay.

In the first to the fourth processes, two vacancies are formed in the L shell at the same time, while in the fifth process, two vacancies are produced in cascade, one is at the time of α decay and the other is in the internal conversion process. Therefore, the first to the forth processes can be distinguished from the fifth by measureing the "hypersatellite" X rays which woud be emitted when a double L shell vacancy is present. The hypersatellite[5,6] is produced by an electron making a transition to one of a double vacancy which causes the X rays to be shifted to a higher energy.

II. EXPERIMENTAL PROCEDURE

1. Source

A little amount of ^{238}Pu was separated from a ^{237}Np sample purchased from Ork Ridge National Laboratory in U. S. A. and was purified with a procedure.[7] Then a 3 mm diameter ^{238}Pu source was prepared by means of electro-deposition on a 50 μg/cm^2 thick aluminum foil which was evaporated on a 6 mg/cm^2 thick mylar backing mounted

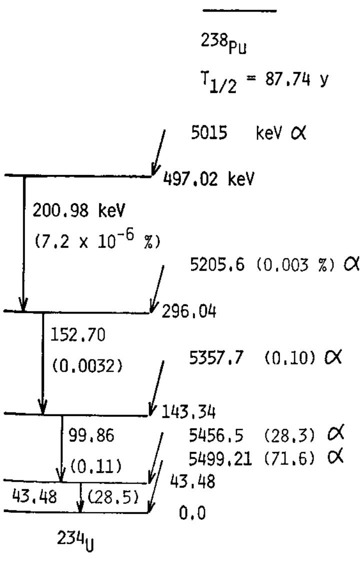

Fig. 1. Decay scheme of ^{238}Pu and ^{234}U 43.48 keV state.

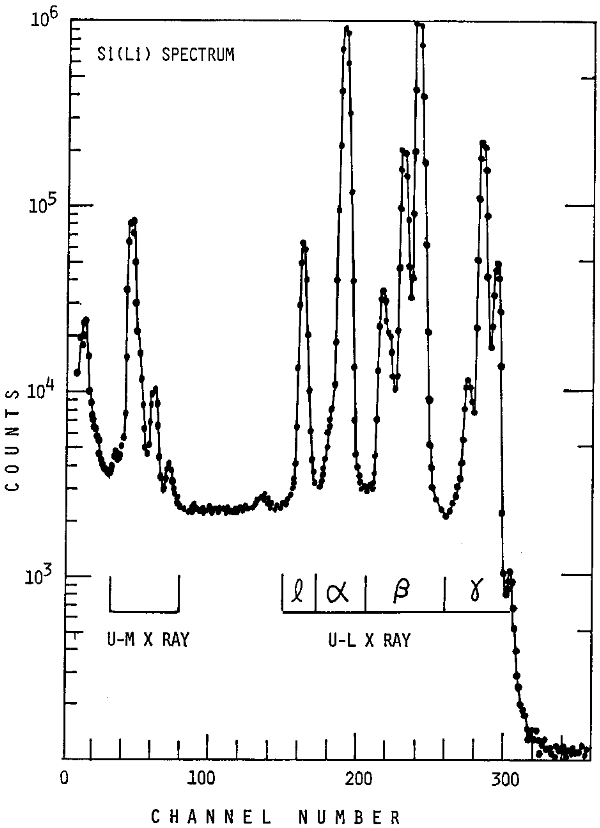

Fig. 2. A Si(Li) singles spectrum of a ^{238}Pu source used in the coincidence measurement.

on a 2 mm thick acrylic resin plate with a hole of 20 mm in diameter at the center. The decay scheme of ^{238}Pu and ^{234}U (43. 48 keV state) is shown in Fig. 1. The absolute intensity of the source was determined as 90.8 ± 1.0 kBq by use of a conventional α- and X-rays coincidence technique. Si(Li) and Ge(Li) spectra of the source were recorded in order to find possible contaminants which could give rise to ^{234}U L_α X ray-^{234}U L X ray coincidences which will be mentioned below. No special contaminant was observed as shown in Fig. 2.

2. Detectors and Coincidence System

A 6 mm diameter by 5. 15 mm thick ORTEC Si(Li) X-ray detector with a 0. 0125 mm thick Be window was used in this work. The resolution was about 364 eV in full width at half maximum at 13. 61 keV of ^{234}U L_α X ray. The Si(Li) detector was connected with a 51 mm$\phi \times$5 mm NaI(Tl) scintillation detector with a beryllium window in a conventional fast-slow coincidence mode. The NaI(Tl) detector generated gate-signals, when it detected photons on the energy of 11 to 21 keV, which covered the ^{234}U L X rays. Fast and slow coincidence resolving times were set at values of 80 ns and 4 μs, respectively. The fast coincidence efficiency ε_c was determined by comparing counts of coincidence events of a weak ^{238}Pu source recorded in the fast-slow coincidence system and in the slow system at the same geometrical configuration. When the coincidence efficency for the slow system was taken to be unity, then the value of ε_c is determined as 0.95 ± 0.05. The reliability of this particular method was given previously.[5]

3. Data Taking and Evaluation of the Probability

Four L_α X ray spectra were taken with the Si(Li) detector in the above coincidence system. Each run was continued for about seven days. Singles spectra of the L_α X ray were recorded once a day, and no drift of the peak position nor broadening of the peak width was observed during the measurement of the coincidence.

The probability per L shell conversion, $W_{L\alpha L}$, forming a double vacancy in the L shell and ending in the emission of the L_α hypersatellite could be obtained from the following two simple relations:

$$W_{L\alpha L} = \frac{b_1 \cdot \eta_\alpha \cdot N_{hyp} \cdot D}{g \cdot \varepsilon_c \cdot N_1 \cdot N_2}, \text{ and} \quad (1)$$

$$W_{L\alpha L} = \frac{2 \cdot b_2 \cdot \eta_\alpha \cdot \alpha_{2L} \cdot (1+\alpha_1) \cdot N_{hyp}}{b_1 \cdot g \cdot \alpha_{1L} \cdot (1+\alpha_2) \cdot N_{12}}, \quad (2)$$

where N_{hyp} = coincidence rate of "hypersatellite" for L_α X ray in the Si(Li) detector and a L X ray in the NaI(Tl) detector,

N_{12} = coincidence rate between a normal Si(Li) L_α X ray and NaI(Tl) L X ray pulses,

N_1 and N_2 = Si(Li) L_α X ray and NaI(Tl) L X ray count rates, respectively,

D = absolute decay rate of the source,

b_1 and b_2 = transition rates of the 43. 48 keV- to ground state and the 143. 34 keV- to 43. 48 keV-state per α decay, respectively,

g = the weighted sum of the probability that, given a double vacancy, a L_α

hypersatellite and a L X ray would be emitted and detected in coincidence,

η_α =the fraction that a L_α X ray would be emitted when a vacancy was formed in the L shell,

α_1 and α_2=conversion coefficients of the 43.48 keV and 99.86 keV transitions, respectively,

α_{1L} and α_{2L}=L shell conversion coefficients of the 43.48 keV and 99.86 keV transitions, respectively, and

ε_c =coincidence circuit efficiency.

III. RESULTS AND DISCUSSION

Si(Li) singles and coincidence spectra of the L_α X ray of ^{234}U are shown in Fig. 3.

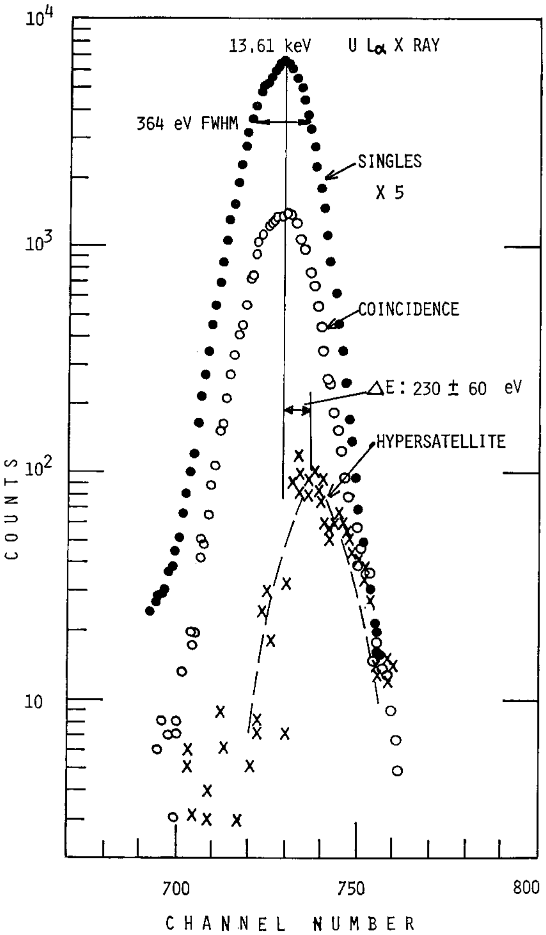

Fig. 3. Si(Li) singles and coincidence spectra of the L_α X ray of ^{234}U. A "hypersatellite" L_α X ray peak was obtained by subtracting the normalized singles spectrum from that of the coincidence. A dashed line is a singles line and is drawn to aid the eye.

The singles spectrum is the sum of 30 spectra taken once a day and is normalized to the coincidence spectrum at the peak position. Background has been subtracted from both peaks. The X-ray peak in the singles spectrum is originated mainly from the internal conversion of the 43.48 keV transition and partly from the 99.86 keV and higher transitions, and the most part of the coincidence peak is due to the 99.86 keV and 43.48 keV cascade transitions. However, the broadening of the coincidence spectrum at the high energy side of the peak is the evidence of the hypersatellite. A spectrum denoted as "hypersatellite" was obtained by subtracting the normalized singles spectrum from that of the coincidence. A dashed line is a normalized singles line and is drawn to aid the eye. The peak of the hypersatellite shifts up in energy by about 230±60 eV from the normal peak position, but is as narrow as the normal one.

This is the first observation of a L_α hypersatellite. So far no other experiment nor theory has been published for the case of L_α hypersatellites. However, some measurements were done for the cases of K_α hypersatellites,[5,6] and the energy shifts were compared with several calculaltions by Briand *et al.*[6] and others.[8,9] They calculated first the effective screening parameter ΔZ of Bergstrom and Hill,[10] and then obtained the value of the energy shifts. Good agreement between the experimental and calculated results is obtained for a predicted value of ΔZ around 0.625.

In the present report, the value of ΔZ for L_α hypersatellite is deduced in similar way to the case for K_α hypersatellites. Then, the effective screening parameter ΔZ is expressed as,

$$\Delta Z=\frac{B_{\mathrm{LL}}(Z)-B_{\mathrm{L}}(Z)}{B_{\mathrm{L}}(Z+1)-B_{\mathrm{L}}(Z)}, \qquad (3)$$

where $B_{\mathrm{L}}(Z)$ and $B_{\mathrm{L}}(Z+1)$ are the binding energies of a L electron for neutral atoms Z and $Z+1$, respectively, and $B_{\mathrm{LL}}(Z)$ is the energy needed to extract an L electron in an atom previously ionized in the L shell. Assuming a ΔZ value for the $L^{-1}M^{-1}$ state equal to unity (as for a $K^{-1}L^{-1}$ state) one can obtain the following relation:

$$\Delta Z=\frac{E^{\mathrm{h}}_{\mathrm{L}\alpha}+[B_{\mathrm{M}}(Z+1)-B_{\mathrm{M}}(Z)]}{B_{\mathrm{L}}(Z+1)-B_{\mathrm{L}}(Z)}, \qquad (4)$$

where, $E^{\mathrm{h}}_{\mathrm{L}\alpha}$ is the energy shift of the L_α hypersatellite and $B_{\mathrm{M}}(Z)$ and $B_{\mathrm{M}}(Z+1)$ are the binding energies of an M electron for neutral atoms Z and $Z+1$, respectively. For the ^{234}U L_α case, the values of B_{L} and B_{M} are (17.1663 keV, 17.6100 keV) and (3.5517 keV, 3.6658 keV) for the Z and $Z+1$ atoms, respectively.[11] Then, the value of ΔZ is obtained as 0.78±0.14. The value is close to the case of K_α hypersatellites.

The probability per L shell conversion, $W_{\mathrm{L\alpha L}}$, forming a double vacancy in L shell and ending in the emission of the L_α hypersatellite has been obtained from Eqs. (1) and (2). The results and data used for the determination of $W_{\mathrm{L\alpha L}}$ are presented in Table I. The values of $W_{\mathrm{L\alpha L}}$ obtained from Eq. (2) contains a large error, because it is taken into account that the recommended value of the branching fraction of 99.86 keV transition is just a mean value of 0.068 and 0.13% spreaded far from each other. The weighted mean value of $W_{\mathrm{L\alpha L}}$ from Eqs. (1) and (2) is $(2.1\pm0.4)\times10^{-4}$.

In the present work, L_β and L_γ hypersatellites have not been studied, therefore, it

Table I. The values of $W_{L\alpha L}$ and data used for the determination of $W_{L\alpha L}$. Symbols defined in text.

	Eq. (1)		Eq. (2)	Weighted mean value
$W_{L\alpha L}$	$(1.9\pm0.4)\times10^{-4}$		$(2.6\pm0.7)\times10^{-4}$	$(2.1\pm0.4)\times10^{-4}$
N_{hyp}	$(8.1\pm1.5)\times10^{-4}$/s		N_{12}	$(1.25\pm0.01)\times10^{-2}$/s
N_1	(44.5 ± 0.2)/s		N_2	(1354.1 ± 6.8)/s
D	$(9.08\pm0.10)\times10^{4}$/s		ε_c	0.95 ± 0.05
b_1	0.285[a]		b_2	0.0011[a]
g	0.68[a]		η_α	0.362[a]
α_1	745[a]		α_{1L}	550[a]
α_2	13.8[a]		α_{2L}	10[a]

[a] See Ref. 12.

is not possible to deduce the respective probablities per internal conversion, W_I, W_{II} and W_{III}, of forming a double vacancy in the L_I, L_{II} and L_{III} subshells. However, the value of $W_{L\alpha L}$ can be written in terms of W_I, W_{II} and W_{III} in the following way:

$$W_{L\alpha L}=[W_{III}+W_{II}\cdot f_{23}+W_I\cdot(f_{13}+f_{12}\cdot f_{23})]\cdot F_{3\alpha}, \qquad (5)$$

where f_{12}, f_{13} and f_{23} are the values of the Coster-Kronig (C-K) yields[13], and $F_{3\alpha}$ represents the fraction of radiative transitions[14] in the L_α peak connected with filling a vacancy in the L_{III} subshell, respectively.

Then, a comparison can be made between the measured value of $W_{L\alpha L}$ and existing theoretical estimates. Carlson *et al.*[15] suggest that the probability for the shakeoff type process in internal conversion would relate to L shakeoff in β decay by,

$$W_i(\mathrm{IC})=W_i(\beta)\cdot[\Delta Z/\Delta Z_\beta]^2, \qquad (6)$$

where W_i(IC) and $W_i(\beta)$ is the probabilities for L_i shakeoff in internal conversion and in β dacay, and ΔZ and ΔZ_β are the effective screening parameter in internal conversion and β decay, respectively. Using $W_i(\beta)$ given by Carlson *et al.*, $\Delta Z_\beta=1$ and $\Delta Z=0.78$ determined in the present work, and applying Eq. (5), one obtains the value of $W_{L\alpha L}$ as 2.34×10^{-4}, where the used values of f_{12}, f_{13} and f_{23} are 0.069, 0.575 and 0.1, respectively, $W_I(\beta)$, $W_{II}(\beta)$ and $W_{III}(\beta)$ are 2.47×10^{-4}, 2.50×10^{-4} and 4.36×10^{-4}, respectively, and $F_{3\alpha}$ is 0.732. Prediction by Carlson *et al.*,[15] when it is corrected for the screening factor obtained from the energy shift of the L_α hypersatellite, reproduces well the experimental result. This implies that the shakeoff process plays a predominant role in forming a double vacancy in L shell.

For the direct collision process, Feinberg[16] has estimated its relative probability to the shakeoff in β decay to be BE/E_0 in the case of $BE \ll E_0$, where BE is the binding energy of the electron subsequently ejected and E_0 is the kinetic energy of the colliding electron. But it is not applicable to the case of the internal conversion in the ^{234}U 43.48 keV transition, because the value of BE/E_0 is very close to unity. Meanwhile, the experimental result[17] of the inner shell ionization by electron has shown that the ionization cross section decreases quite rapidly with decreasing energy of incident electron below $E_0 \simeq 3 \times BE$, and is vanishingly small at $E_0 \simeq BE$. Therefore, the probability of a double vacancy due to the direct collision should be quite less than that of the shakeoff process.

For the higher-order electronic transitions, that is, the double conversion process and the internal conversion of the internal Compton radiation, no quantitative theory has been presented.

In conclusion, the shakeoff process can apparently reproduce the experimental result for the double vacancy formation in the L shell, but more rigid theoretical calculations and measurements of the L_β and L_γ hypersatellites and the energy spectra of the ejected electrons are needed to better understand the double vacancy formation mechanism.

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REFERENCES

(1) I. Curie and F. Joliot, *J. Phys. (Paris)* **7**, 20 (1931).
(2) A. Migdal, *J. Phys. USSR*, **4**, 449 (1941).
(3) E.L. Feinberg, *J. Phys. USSR*, **4**, 423 (1941).
(4) M.S. Freedman, *Annu. Rev. Nucl. Sci.*, **24**, 209 (1974).
(5) H.J. Nagy, G. Schupp, and R.R. Hurst, *Phys. Rev.* **C11**, 205 (1975).
(6) J.P. Briand *et al.*, *J. Phys.* **B9**, 1055 (1976).
(7) D.C. Hoffman, Collected Radiochemical Procedures LA-1721, 4th Ed. Compiled by J. Kleinberg and H.L. Smith, (Los Alamos), 1979, pp. 107.
(8) J.P. Desclaux, C. Briancon, J.P. Thibaud, and R.J. Wallen, *Phys. Rev. Lett.* **32**, 447 (1974).
(9) D.J. Nagel, A.R. Knudson, and P.G. Burkhalter, Proc. 4th Int. Conf. on Vacuum Ultraviolet Radiaton Physics, Hamburg pp. 162.
(10) I. Bergsirom and R.D. Hill, *Ark. Fys.* **8**, 21 (1954).
(11) J.A. Bearden and A.F. Burr, *Rev. Mod. Phys.* **39**, 125 (1967).
(12) Y.A. Ellis, *Nucl. Data Sheets*, **21**, 549 (1977).
(13) W. Bambynek *et al. Rev. Mod. Phys.* **44**, 716 (1972).
(14) S.I. Salem, S.L. Panossian, and R.A. Krause, *Atom. Data. Nucl. Data Tables*, **14**, 91 (1974).
(15) T.A. Carlson, C.W. Nestor, Jr, T.C. Tucker, and F.B. Malik, *Phys. Rev.* **169**, 27 (1968).
(16) E.L. Feinberg, *Sov. J. Nucl. Phys.* **1**, 438 (1965).
(17) C.J. Powell, *Rev. Mod. Phys.* **48**, 33 (1976).