An Upper Limit for the Probability of Nuclear Excitation by Electron Transition in ^{235}U

Atsushi Shinohara,[#] Tadashi Saito,[*] Kazuo Taniguchi,[†] Kiyoteru Otozai,[##] Shigero Ikeda,[###] and Hiroshi Baba
Department of Chemistry and Laboratory of Nuclear Studies, Faculty of Science, Osaka University, Toyonaka, Osaka 560
[†]*Department of Solid State Electronics, Osaka Electro-Communication University, Neyagawa, Osaka 572*

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We have attempted to observe nuclear excitation by electron transition (NEET) in ^{235}U with Mo K X-ray irradiation and off-line radioactivity measurement. Nuclear excitation of the 13-keV level was measured through the isomer radioactivity. An upper limit for the NEET probability was obtained as $P < 9 \times 10^{-10}$.

A vacancy created in an inner atomic shell will be immediately filled with an electron jumping from an outer orbit. This electronic transition accompanies the energy release as a form of an X-ray photon or an Auger electron. In addition, the energy can be transferred to the nucleus, if atomic and nuclear transitions satisfy the special quasi-resonance conditions, namely, nearly degenerated energies and the same multipolarity. Thus nuclear excitation may take place in particular nuclides. This process, referred to as nuclear excitation by electron transition, abbreviated as NEET, was first considered theoretically[1] and since then confirmed by several experiments.[2–10] Various theoretical approaches have also been taken up toward the understanding of the NEET mechanism.[11–18]

The nucleus ^{235}U has an isomer[19] with a half-life of 26 min lying at 0.077 keV.[20] In the original paper[1] concerning NEET, ^{235}U was exemplified in connection with a possible isotope separation proposed by one of the authors (K.O.). Direct excitation of the isomer was out of consideration, because the nucleus and electron cooperating in NEET are so distant that the Coulomb interaction between them should be too small. A candidate is the second excited state lying at 13.038 keV which will be raised in accordance with L_3M_4 or L_3M_5 electronic transition. The atomic and nuclear transitions have a common E3 component and a small energy difference, $\Delta = 0.402$ or 0.578 keV, respectively. The 13-keV level instantaneously decays with a branching ratio of 100% to the isomer, whose 26-min radioactivity can be used as an indication of the NEET of the former nuclear level. The NEET probability, P, was estimated to be as small as $P \sim 10^{-9}$ per created L_3 vacancy. Excitation of the 13-keV level by NEET has not been observed so far.

The direct route to the isomer ^{235m}U through NEET was considered to be feasible in a laser-induced hot plasma in which an ionized electron shell captures a free electron in the continuum.[10] In the restraint of the electron transitions between two bound states such as O_4P_3 giving $P \sim 10^{-10}$ in total, pellet implosion by intense laser or charged-particle beam as used in inertial confinement fusion may also reach to the direct production of an appreciable amount of the isomer by NEET.[12] In another theoretical study,[13] it was estimated that the NEET from the continuum dominates in the isomer production, while at higher plasma temperatures inelastic scattering of plasma electrons gains predominance. Experimental evidence was reported that ^{235m}U was populated by NEET from natural uranium in a laser-produced plasma.[4] The mechanism for the isomer production has, however, been still controversial[14,18] and the experimental confirmations were done in vain.[18,21]

Here we report our experimental attempt to observe the NEET excitation of the 13-keV level which is expected to have a larger P value than the isomer. The experiment was composed of the X-ray irradiation of highly enriched ^{235}U for ionization and the succeeding measurement of the radioactivity of ^{235m}U produced by the NEET process.

The targets used were uranium enriched to 99.83% in ^{235}U. The following isotopes were also included: ^{233}U, 5×10^{-4}%; ^{234}U, 6.2×10^{-2}%; ^{236}U, 3.7×10^{-2}%; and ^{238}U, 6.9×10^{-2}%. The solely significant radioactive impurity was ^{234}U whose specific activity was about twice that of ^{235}U. Metallic uranium was electroplated on stainless-steel disks. Three ^{235}U targets were purchased from CEA, France, as the form of α-reference sources. Their thicknesses were 2.0, 10.2, and 41.4 μg/cm^2 each and the active diameter was 35 mm.

An X-ray tube of the fixed anode type was specially designed and constructed for the effective irradiation. The photon-producing anode of the tube was metallic molybdenum electroplated on a Cu substratum. Mo K X-rays (17.4–20.0 keV) are suitable for ionizing the U L_3 subshell (17.2 keV). The X-ray tube had a large Al window of 36 mm inner diameter to allow the irradiation of the target in a close distance of about 30 mm from the anode. The anode, window, and vessel of the tube were cooled by running water. The irradiations of the targets with X-rays were carried out for one hour each with a tube voltage of 50 kV and a tube current of 80 mA.

The internal conversion electrons emitted from ^{235m}U have extremely low energies of mainly several eV to 75 eV.[20] A 2π windowless Q-gas-flow Geiger-Müller counter was used to measure the induced radioactivity. The detection of the radiations from ^{235m}U by the counter was confirmed by using a source of ^{235m}U recoils from the α-decay of ^{239}Pu.[22] Measurements were started in a few min after the end of the irradiation and continued for 4 h.

The two-component decay analysis with a 26-min component (^{235m}U) and the constant background (^{235}U and ^{234}U) was attempted for each measured decay curve. However, a statistically meaningful amount of the component with the half life of 26 min could not be extracted. The results are summarized in Table 1. An upper limit of the NEET probability was then deduced by taking into consideration the detection limit L_D of each measurement. The L_D values at 95% confidence level were deduced from the errors $\sigma(C_\mathrm{m})$ associated with net counts C_m of ^{235m}U by using the relation $L_\mathrm{D} = 3.29\sigma(C_\mathrm{m})$ according to Currie.[23]

The direct route of producing the isomer via the $O_{4,5}$ vacancies should make a negligible contribution as compared with that resulting from the L_3 vacancy. Therefore, the counting rate of the conversion electrons of ^{235m}U can be ascribed only to the 13-keV-level excitation and expressed by

$$C = \epsilon N x H(\mathrm{L}_3) P B G I (1 - e^{-\lambda t}), \qquad (1)$$

where ϵ is the efficiency for detection of the conversion electrons, N the atomic density, x the target thickness, B the branching ratio of the 13-keV level to the isomer, G the effective solid angle of the irradiation (= 0.57 sr), I the electron number equivalent to the tube current, λ the decay constant of the isomer, and t the irradiation time. The factor $H(\mathrm{L}_3)$ is

Tabel 1. Upper limits for P deduced from L_D obtained by the present experiment. C_m and C_B are the count rates of the isomer at the end of irradiation and the constant background, respectively.

Target no.	Nx/atoms cm^{-2}	C_m/s^{-1}	C_B/s^{-1}	L_D/s^{-1}	C_γ/s^{-1}	ϵ	P
1	5.0×10^{15}	-0.1 ± 0.2	6.55 ± 0.04	0.6	1.2×10^{-5}	0.115	$< 0.9 \times 10^{-9}$
2	2.62×10^{16}	-0.8 ± 0.4	28.0 ± 0.09	1.3	1.7×10^{-5}	0.028	$< 1.5 \times 10^{-9}$
3	1.06×10^{17}	1.4 ± 0.8	100.7 ± 0.2	2.6	2.4×10^{-5}	0.012	$< 1.7 \times 10^{-9}$

the effective cross section per primary tube electron for photoionization of the L_3 subshell in a U atom and given by

$$H(L_3) = \int [\sigma_3(E) + \sigma_2(E) f_{23} + \sigma_1(E)(f_{12}f_{23} + f_{13})] n(E) dE, \tag{2}$$

where σ_i is the cross section for L_i-subshell ionization by a photon with energy E,[24] whose spectral intensity being $n(E)$, and f_{ij} is the Coster-Kronig transition probability for shifting a vacancy from the i-th subshell to a higher j-th subshell.[25] The photon spectrum from the X-ray tube, $n(E)$, was estimated with the aid of the DIBRE bremsstrahlung calculation code,[26] which we revised to include the calculation of the Mo K X-ray intensities by taking the geometrical conditions into account. Thus we obtained $H(L_3) = 5.38$ b/sr for a 50-keV electron.

The detection efficiency ϵ was estimated by the Monte Carlo method. The calculation is based on the electron mean free path $\Lambda(E)$ in the material and includes the effect of single scattering only, where it is assumed that the energy of a singly scattered electron is calculated from the stopping power dE/dx and the path length x and further that the scattering is isotropically elastic. Employed for $\Lambda(E)$ was the escape depth of low energy electrons[27] and for dE/dx the semiempirical equation[28] for low energy electrons derived from the LSS theory.[29] The estimated ϵ values and deduced upper limits of P are summarized in Table 1.

In the present experiment, the γ-ray nuclear resonance absorption was a competitive process against NEET. The count rates of the isomer attributable to this process, C_γ, were estimated for nuclear levels at 0.077, 13.0, and 46.2 keV by using the Breit-Wigner single level formula and the $n(E)$ calculated above. As shown in Table 1, the contributions are obviously negligible compared with the detection limits L_D obtained in this experiment.

An upper limit of the NEET probability was deduced to be $P < 9 \times 10^{-10}$ in the present work. This is somewhat smaller than those estimated theoretically ($P \sim 2.4 \times 10^{-9}$ or 1×10^{-9}).[1,12] Our results are not inconsistent with the result for the direct isomer production which was discussed[4] to be smaller by an order of magnitude than the theoretical estimate.[12] It may hence be necessary to revise the theoretical estimates for NEET in ^{235}U. The use of synchrotron radiation will enable us to make a further improved experiment. For example, we can estimate $H'(L_3) = 66$ b/mrad, $G' = 1$ mrad, and $eI = 500$ mA for photons from the wiggler at the photon factory of KEK.[8] This will readily bring the reduction of the detection limit by two orders of magnitude. A crude estimate using eq.(6) of ref.[3] can be given as $P = 4.5 \times 10^{-12} F^2$ for the L_3 vacancies in ^{235}U, where F is the correction factor for the nuclear collective motion. If the collective effect is working significantly, the detection of NEET will become feasible, and some information on NEET which does not depend on the external factors such as the plasma temperatures can be obtained.

References and notes

\# Present address: *Department of Chemistry, Faculty of Science, Nagoya University, Chikusa, Nagoya 464-01.*

\#\#Present address: *Faculty of Home Economics, Kobe Women's University, Suma, Kobe 654.*

\#\#\# Present address: *Department of Material Chemistry, Faculty of Science and Technology, Ryukoku University, Otsu, Shiga 520-21.*

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