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Nuclear Excitation by Electron Transition and Its Application to Uranium 235 Separation

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A new mechanism for nuclear excitation is studied theoretically by considering the deexcitation of electronic states of atom. When an electron of inner shells is kicked off by the bombarded electron or X ray, an electron of the adjacent shells immediately jumps into the vacancy. The energy corresponding to the difference of the binding energies for these two shells, E_1-E_2, is usually carried away by the emitted characteristic X ray or the Auger electron which is ionized from an outer shell. There is, however, another possibility of this energy release by exciting a nuclear state. That is, the nuclear ground state is excited to the higher energy level by receiving the excess energy of the electronic state by means of electromagnetic interactions between nucleus and electrons. A theory is made for this process, and it is found that the probability of this process is extremely small in most of the cases, but it will be appreciable, if the interaction energy is approximately equal to the energy mismatch $E_N-(E_1-E_2)$, where E_N is the nuclear excitation energy. An example of ^{235}U is discussed in connection with a possible separation of ^{235}U from uranium isotopes.

§ 1. Introduction

If an atom with several electrons is excited in that an orbital electron of a lower level is raised to a higher level, an X ray is usually emitted, while an electron of the adjacent shells jumps into the vacancy. Instead, the interactions with electrons in the higher shells lead to the ejection of latter electrons into a continuous spectrum. This process competes with the emission of the photon, and it is called the Auger electron emission. In this paper, we wish to point out that there is a third process of the deexcitation mechanism, which we call the nuclear excitation by electron transition. The possibility of this process may be understood as follows: In the process of deexcitation of the atomic system, an electron emits a photon. If this is a real photon, it is the X-ray emission. If the photon is virtual and it is absorbed by an electron of an outer shell, this electron is ejected to the continuous energy state, and it is called the Auger process. Under a certain condition, a virtually emitted photon is absorbed by a nucleus, and the nucleus is excited.

This is shown in a more picturesque way in Fig. 1, where a line refers to the nuclear level and the lines with circles refer to electron levels. The line with two solid circles represents symbolically the closed shell with $2j+1$ electrons (j being the total angular momentum of the orbital electron), and the open circle indicates an electron hole. In Fig. 1(a) there is an electron hole with

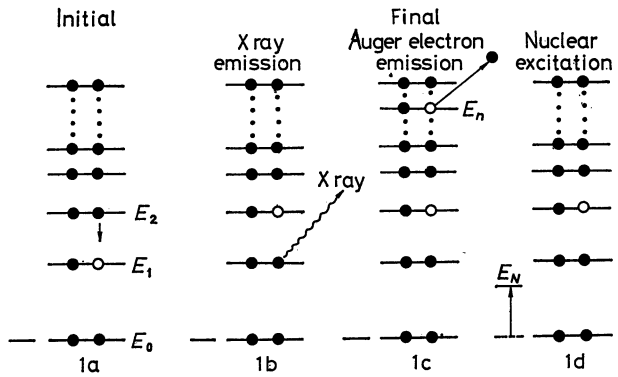

Fig. 1. Deexcitation mechanism of the electron system. A line refers to the nuclear level and the lines with circles refer to the electronic levels. The line with two solid circles represents symbolically the closed shell with $2j+1$ electrons, except for the top level which may not be completely filled, and the open circle indicates the electron hole. In the initial state in Fig. 1(a), there is an electron hole in an inner shell. There are three final states: X-ray emission in Fig. 1(b), Auger electron emission in Fig. 1(c) and nuclear excitation in Fig. 1(d).

binding energy E_1 which can be made, for example, by bombarding the atom by X rays or electrons. In the process of the deexcitation where the orbital electron with binding energy E_2 jumps down to E_1 level, there is an emission of the X ray in Fig. 1(b), an emission of the Auger electron in Fig. 1(c), or the nuclear excitation in Fig. 1(d). (In principle, two levels with E_1 and E_2 need not to be adjacent.) In these three processes, the energy conservation should be satisfied. Therefore, the X-ray energy is equal to (E_1-E_2) in the X-ray emission. In the Auger process, this energy is equal to the sum of the ionization energy and the kinetic energy of the electron. In order to realize the nuclear excitation, the excitation energy E_N should be equal to (E_1-E_2). (More precisely, the energy difference $E_N-(E_1-E_2)$ should not be too large compared with the interaction energy of nuclear and electronic states. This will be seen later.) Besides this energy conservation, the spin and parity of the resultant system should be conserved.

As a similar problem, the nuclear fine structure in the muonic atom was studied theoretically,[1),2)] and the nuclear excitation due to deexcitation of the muonic levels has been observed already in the muonic X-ray experiments.[3)] According to a theory of the nuclear excitation by electron transition which will be developed in this paper, the probability of this process is extremely small in most of the cases, but it will be appreciable, if the required conditions are reasonably fulfilled.

Experimental verification of this process involves two physical implications: One is to open a new field of spectroscopy which may be called the nucleo-atomic spectroscopy, in addition to the conventional atomic and nuclear spectro-

scopies, since our study concerns with deexcitation of the electronic level and the excitation of the nuclear level. As a consequence we can study the interactions between the nucleus and electrons and the nuclear structure problems. The other problem, which may be of more practical interest, concerns with purification of certain isotopes, such as ^{235}U. In the deexcitation of the nuclear first excited state the ^{235}U atom emits an internal conversion electron. After emission, this atom is in a chemical form different from that of the other uranium atoms. This is because one of the valence electrons is internally converted. A possibility of chemical separation of ^{235}U from the other uranium isotopes has been pointed out by Otozai, if we can excite the ^{235}U nuclei.[4] The nuclear excitation by electron transition can, in principle, supplies us with excited states of ^{235}U by bombarding the mixture of uranium isotopes with X rays or electrons.

Now we will construct a theory of nuclear excitation by electron transition. First we discuss the mixing of the atomic levels by perturbation due to the interaction between the nucleus and electrons in § 2. Next we calculate the probability of formation of the nuclear excited state in § 3. An example of ^{235}U is numerically studied in § 4, and discussions are given in § 5.

§ 2. Configuration mixing of quantum states due to perturbation

In this section we make a theory of nuclear excitation by electron transition. In this particular phenomenon, the nucleus and the orbital electrons are involved. Therefore, we have to study the resultant system of the nucleus and electrons, and the electromagnetic transition between different states of this resultant system. In the formulation we assume that the branching ratio of this process compared with the X-ray and Auger electron emissions is so small that we can adopt various approximations which make the theory simpler.

In the following we have ψ_1 for the wave function of the ground state of nucleus and ψ_2 for that of the excited state with excitation energy E_N. The wave function of $(Z-1)$ electrons is denoted by

$$\phi_i(1, 2, \cdots, Z-1),$$

which is antisymmetrized with respect to $(Z-1)$ electrons. The subscript i refers to the state where the electron of the i-th orbit in the Z-electron system is vacant. Therefore, the energy of ϕ_i is (mc^2-E_i) less than the ground-state energy E_Z of the Z-electron system. Here mc^2 is the rest mass energy of the electron, and E_i is the binding energy of the i-th orbit. For simplicity, we omit (E_Z-mc^2), and the energy of the state ϕ_i is written as E_i where

$$E_0>E_1>E_2>\cdots>0\,.$$

The wave function Ψ of the resultant system of the nucleus and electrons is assumed to be the product of individual wave functions

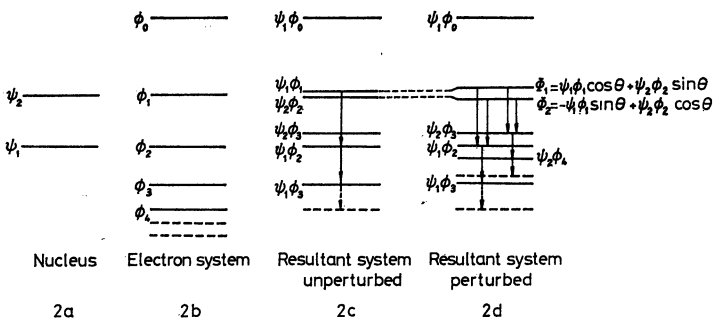

Fig. 2. Energy levels.

$$\Psi = \psi_1\phi_0, \psi_1\phi_1, \psi_1\phi_2, \cdots, \psi_2\phi_0, \psi_2\phi_1, \psi_2\phi_2, \cdots .$$

In Fig. 2, energy levels are shown. The energy levels for nucleus and electrons are given in Figs. 2(a) and 2(b), while the resultant system Ψ has the energy level given in Fig. 2(c).

The electromagnetic transitions between different Ψ states in Fig. 2(c) are the X-ray emission and the Auger process in the lowest approximations. Therefore, these transitions are represented by $\psi_1\phi_i \to \psi_1\phi_j$. (For the Auger process, we have to use $\psi_i\phi_j'$ instead of $\psi_i\phi_j$. Here ϕ_j' refers to the state where the electron hole jumps up to the j-th orbit and one of the outer-shell electrons moves out in the continuum state. For simplicity, these two states are written by the same symbol ϕ_j.) Similarly there are transitions $\psi_2\phi_i \to \psi_2\phi_j$. We are discussing the case where the electron transition is considerably faster than the nuclear gamma decay $\psi_2\phi_i \to \psi_1\phi_i$, or the internal conversion process $\psi_2\phi_i \to \psi_1\phi_j$ (or more precisely, $\psi_1\phi_j'$).

In the case which we are interested in, the nuclear excitation energy is nearly equal to the difference of binding energies of electrons,

$$E_1 - E_2 \approx E_N .$$

In this case the sum of the unperturbed energies of the nuclear state and the electron system is given by

$$E^{(0)}(\Psi_1) = E_1 \qquad \text{for the state } \Psi_1 = \psi_1\phi_1$$

and

$$E^{(0)}(\Psi_2) = E_2 + E_N \qquad \text{for the state } \Psi_2 = \psi_2\phi_2 .$$

Here Ψ_1 refers to the state where the nucleus is in the ground state and the electron hole is in the orbit with E_1, Ψ_2 refers to the state where the nucleus is in the excited state and the electron hole is in the orbit E_2. (see Fig. 2(c).) In the case where the energies of Ψ_1 and Ψ_2 are nearly degenerated, these two states are no longer the eigenstates of the energy. We have to take into account

the interactions between Ψ_1 and Ψ_2, and the true eigenstates are two linear combinations of Ψ_1 and Ψ_2, which are obtained by the diagonalization of the energy matrix. Figure 2(d) shows a schematical energy level of the resultant system of the nucleus and electrons which is perturbed by interactions.

According to the standard perturbation calculation, we have eigenfunctions by the diagonalization of the 2×2 energy matrix, as the following linear combinations of the unperturbed wave functions Ψ_1 and Ψ_2:

$$\begin{aligned}\Phi_1 &= \Psi_1 \cos\theta + \Psi_2 \sin\theta\,,\\ \Phi_2 &= -\Psi_1 \sin\theta + \Psi_2 \cos\theta\,.\end{aligned} \tag{1}$$

The mixing ratio can be determined by angle θ as

$$\cos\theta \sin\theta = \frac{(\Psi_2|H'|\Psi_1)}{E(\Phi_1)-E(\Phi_2)} \approx \frac{(\psi_2\phi_2|H'|\psi_1\phi_1)}{E(\Psi_1)-E(\Psi_2)} = \frac{E'}{E(\Psi_1)-E(\Psi_2)}\,. \tag{2}$$

Here $E(\Phi_1)$ denotes the energy for the state Φ_1, etc. The interaction Hamiltonian H' is assumed to be very small, and therefore we have

$$E(\Phi_i) \approx E(\Psi_i) \approx E^{(0)}(\Psi_i), \qquad i=1,2\,.$$

We will evaluate the mixing angle θ for a possible nuclear excitation by assuming the Coulomb interaction as H',

$$H' = -\frac{e^2 Z}{|\boldsymbol{r}_N - \boldsymbol{r}_e|}\,. \tag{3}$$

Here $\boldsymbol{r}_N$ is the position vector of the nucleus and $\boldsymbol{r}_e$ is that of the electron which is relevant for the transition.

Before we perform an explicit calculation of Eq. (2), we shall discuss the probability of formation of the nuclear excited state ψ_2.

§ 3. Probability of formation of nuclear excited state

In the presence of perturbation H', we have obtained eigenstates Φ_1 and Φ_2 in Eq. (1). If there are no other degenerated states, we have a series of eigenstates for resultant atomic system, $\Psi_0, \Phi_1, \Phi_2, \Psi_3, \Psi_4, \cdots$. Here Ψ_j is one of the states in Fig. 2(d) which have the form $\psi_k\phi_l$. Transitions take place from the higher energy states to the lower ones, in this series. The transitions among these states are again emissions of X ray or Auger electron, and the electron hole jumps up until it reaches to the utmost outer shell. Our problem is to calculate how many percent of the nuclear states remains in its excited state at the end of the electron transitions.

Now let us start the discussion from the electrically neutral atom with atomic number Z. First an electron of the closed shell is kicked off by the bombarded electron or X ray. In this instance, the wave function of the $(Z-1)$ electrons is denoted by ϕ_i. Therefore, the resultant system of the nucleus and $(Z-1)$

electrons is given by $\Psi_1=\psi_1\phi_1$ or

$$\Psi_1=\Phi_1\cos\theta-\Phi_2\sin\theta\,, \tag{4}$$

where we have used Eq. (1). That is, the creation of the electron hole in the orbit with E_1 corresponds to the production of the eigenstates Φ_1 and Φ_2 in a ratio $\cos^2\theta/\sin^2\theta$. (The interaction should be completed before the state Ψ_1 decays into lower states. This condition is satisfied, since we are discussing the case where the interaction energy is larger than the line width of the state ϕ_1.) As is shown in Fig. 2(d), these two states decay by emitting the X rays and Auger electrons. These transitions are again the following types of transitions:

$$\psi_1\phi_1\longrightarrow\psi_1\phi_j\qquad(j>1)$$

and

$$\psi_2\phi_2\longrightarrow\psi_2\phi_k\,.\qquad(k>2) \tag{5}$$

The nuclear deexcitation

$$\psi_2\phi_2\longrightarrow\psi_1\phi_2$$

is again assumed to be slower than the processes (5). The higher-order processes

$$\psi_2\phi_2\longrightarrow\psi_1\phi_k\qquad(k>2)$$

are also omitted. If we write Eq. (1) explicitly, we have

$$\begin{aligned}\Phi_1&=\psi_1\phi_1\cos\theta+\psi_2\phi_2\sin\theta\,,\\ \Phi_2&=-\psi_1\phi_1\sin\theta+\psi_2\phi_2\cos\theta\,.\end{aligned} \tag{6}$$

That is, both two states contain the $\psi_2\phi_2$ component so that the transitions of these states to $\psi_2\phi_k$ are allowed by means of the process (5). For example, the transition amplitude for Φ_1 is given by

$$\begin{aligned}(\psi_2\phi_k|H_{\rm int}|\Phi_1)&=\sin\theta\,(\psi_2\phi_k|H_{\rm int}|\psi_2\phi_2)\\ &\approx\sin\theta\,(\phi_k|E1|\phi_2)=M_2\sin\theta\,.\end{aligned} \tag{7}$$

The interaction Hamiltonian $H_{\rm int}$ is one for the radiative and nonradiative transitions. The main part of the transition amplitude is presumably the electric dipole component, which is symbolically represented by a matrix element M_2. The subscript 2 stands for the nuclear excited state ψ_2. The state $\psi_2\phi_k$ decays successively to lower states until the electron hole reaches to the utmost outer shell, where the electronic system is stable. During this period of the electronic transitions, the nucleus is still in the state ψ_2. That is, the nucleus is produced in its excited state. This state decays later by emitting a gamma ray or a conversion electron and goes back to the ground state ψ_1. Therefore, the probability of the formation of the excited state is determined by the decay rates of $\psi_2\phi_2$ components in Φ_1 and Φ_2.

The production rate of the nuclear excited state ψ_2 is proportional to the

decay probability times the initial state density. Since the state Φ_1 is produced with a ratio $\cos^2\theta$ initially and its decay rate is proportional to the square of Eq. (7), the production rate of ψ_2 through Φ_1 is

$$|M_2|^2 \sin^2\theta \cos^2\theta .$$

In the same way, the production rate through Φ_2 is

$$|M_2|^2 \cos^2\theta \sin^2\theta .$$

(If these transitions $\Phi_1 \to \psi_2\phi_3$, $\Phi_2 \to \psi_2\phi_3$, etc., take place, additional X rays should be observed, as is shown in Fig. 2(d). Experimentally this is not the case. This, however, does not mean the prohibition of these transitions. Instead, the transitions rates for these processes are extremely small compared with regular transitions, such as $\psi_1\phi_1 \to \psi_1\phi_j$.)

In a similar consideration we have the production rates of the ground-state nucleus in the transitions $\Phi_1 \to \psi_1\phi_j$ and $\Phi_2 \to \psi_1\phi_j$, respectively, given by

$$|M_1|^2 \cos^4\theta \qquad \text{and} \qquad |M_1|^2 \sin^4\theta$$

with

$$M_1 = (\phi_j | E1 | \phi_1),$$

where the subscript 1 for the matrix element refers to the nuclear ground state ψ_1.

Finally the production rate of the excited state of the nucleus versus that of the ground state is given by

$$P = \frac{|M_2|^2}{|M_1|^2} \frac{2\cos^2\theta \sin^2\theta}{\cos^4\theta + \sin^4\theta} . \tag{8}$$

As a matter of fact, we have to consider the summation over k for the different final states ϕ_k, and similarly the summation over j for the different final states ϕ_j. This means that $|M_2|^2/|M_1|^2$ in Eq. (8) should read

$$\sum_k |M_2|^2 / \sum_j |M_1|^2 .$$

For simplicity we keep the expression (8) unchanged. As a consequence, $|M_i|^2$ ($i=1$ or 2) should be the probability of annihilation of the state ϕ_i (or equivalently the probability of annihilation of the electron hole in the orbit with binding energy E_i). The ratio $|M_2|^2/|M_1|^2$ can, in principle, be calculated, and it is also obtainable experimentally. If we put $\tan^2\theta = x$, we can simplify Eq. (8) as follows:

$$P = \frac{2x}{1+x^2} \frac{|M_2|^2}{|M_1|^2} \approx 2x \frac{|M_2|^2}{|M_1|^2} \qquad \text{for } x \ll 1 . \tag{9}$$

Now let us explain the above process of the formation of the nuclear excited state again by the schematic diagrams in Fig. 3. In Fig. 3(a), an orbital elec-

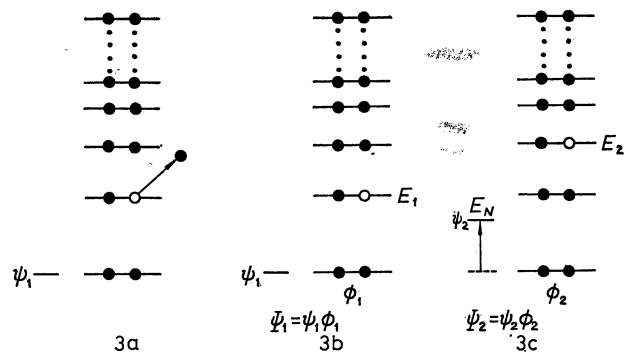

Fig. 3. Energy levels.

tron is kicked off by the bombarded electron or X ray. As a consequence, the state $\Psi_1=\psi_1\phi_1$ is produced in Fig. 3(b). Since the energy of Ψ_2 state in Fig. 3(c) has almost the same energy as that of Ψ_1, these two states interact with each other, and the energy eigenstates should be the linear combinations of the two states of Figs. 3(b) and 3(c). The Ψ_1 component in Φ_1 or Φ_2 decays into the state Ψ_i or into Ψ_j by emitting an Auger electron or X ray, and the electron hole is successively moving up to an appropriate orbit which is not necessarily the adjacent level to the initial level of the electron hole. The hole may skip several levels and it may move up to any higher level which has an appropriate value of the spin and parity for the allowed electromagnetic transitions. Successive transitions of the Ψ_2 component are indicated in Figs. 4(a) and 4(b), and they terminate when the electron hole reaches to the top level. Finally, the nuclear excited state ψ_2 emits the gamma ray and the nucleus is in the ground state ψ_1, as is shown in Fig. 4(c).

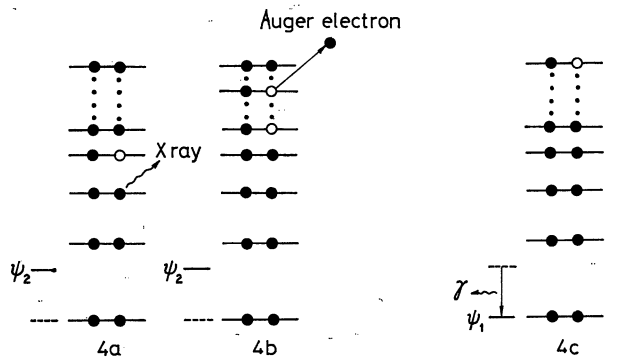

Fig. 4. Successive transitions of Ψ_2 component in Φ_1 and Φ_2.

§ 4. Numerical results for ^{235}U

This sort of nuclear excitation mechanism is already seen in the muonic atom. The muon mass is 207 times the electron mass so that the muon is lo-

cated at 207 times nearer distance from the nucleus than the electron. Therefore, the Coulomb interaction E' in Eq. (2) is quite strong, and the mixing of the unperturbed states in Eq. (1) is large enough to produce a strong dynamic nuclear excitation in certain nuclei. The energy which is released from the muon on the occasion of the muonic transition between two muonic orbits is transferred to the nucleus with a high probability.

In our case, the energy which is released from the electron on the occasion of the transition of the electron hole is transferred to the nucleus. Normally this transformation takes place with a negligibly small probability, since the electrons are located at a distance far from the nucleus and the Coulomb energy in Eq. (2) is vanishingly small, and we cannot observe the nuclear excitation by this electron transition. However, if the atomic number Z becomes larger, the radius of the electron orbit will be smaller, and we can expect, at least, a possibility of nuclear excitation.

In this section, we show a possible nuclear excitation by electron transition in ^{235}U. First of all we point out two examples of energy level matching:

$$E_1 - E_2 \approx E_N .$$

The energy level for ^{235}U nucleus is given in Fig. 5(a) where the first excited state is the 26 min state at (30 ± 3) eV, and the second excited state is the 13.1 keV state. The energy of the first excited state is given by Mazaki and Shimizu.[5] The nuclear excitation energy of the $\frac{1}{2}^+$ state approximately coincides with the energy difference of the electronic $6d_{3/2}$ and $6p_{3/2}$ levels[6] (see Fig. 5(b)).

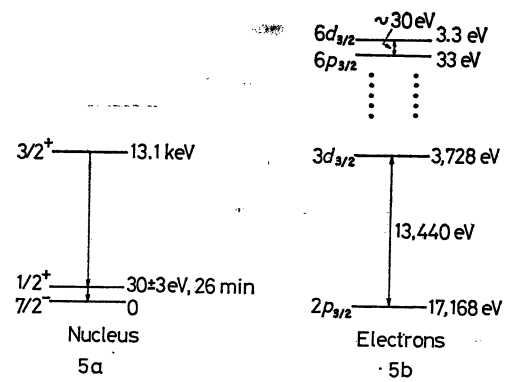

Fig. 5. Nuclear and electronic levels in ^{235}U.

$$E_N = (30 \pm 3)\,\text{eV},$$
$$E_1 - E_2 = (33 - 3.3)\,\text{eV} = 29.7\ \text{eV}.$$

Although the energy matching is good and the condition for the spin and parity (which will be given below) is satisfied, this case may not be useful. This is because the electrons in the $n=6$ orbit are located at the distance far from the nucleus, and therefore the Coulomb interactions between the nucleus and electrons are too small.

The energy difference of electronic $3p_{3/2}$ and $3d_{3/2}$ levels in Fig. 5(b) almost coincides with the nuclear excitation energy of the $\frac{3}{2}^+$ state in Fig. 5(a).

$$E_N = 13.1\ \text{keV},$$
$$E_1 - E_2 = (17{,}168 - 3{,}728)\,\text{eV} = 13{,}440\ \text{eV}.$$

We shall discuss this case as one of the favorable cases to produce possible nuclear excitation by electron transition. In this case, the condition for spin-parity is also fulfilled. The condition is as follows: In order to have nonzero value of the Coulomb interaction, $(\Psi_2|H'|\Psi_1)$ in Eq. (2), Ψ_1 and Ψ_2 should have the same spin and parity. Now let us denote I_1 and I_2 for the nuclear spins of the states ϕ_1 and ϕ_2, and J_1 and J_2 for the spins of the electron systems ϕ_1 and ϕ_2, respectively. We assume a case where J_1 is equal to the spin of the electron hole. The atom is, therefore, in the angular momentum state F.

$$F_1 \text{ for the state } \Psi_1=\phi_1\phi_1$$

and

$$F_2 \text{ for the state } \Psi_2=\phi_2\phi_2 .$$

That is, we have

$$\boldsymbol{I}_1+\boldsymbol{J}_1=\boldsymbol{F}_1 \qquad \text{and} \qquad \boldsymbol{I}_2+\boldsymbol{J}_2=\boldsymbol{F}_2 , \tag{10a}$$

and consequently the condition

$$\boldsymbol{I}_1+\boldsymbol{J}_1=\boldsymbol{I}_2+\boldsymbol{J}_2 \tag{10b}$$

is required. In our case,

$$I_1=\tfrac{7}{2}^-, \quad J_1=\tfrac{3}{2}^- \qquad \text{and} \qquad I_2=\tfrac{3}{2}^+, \quad J_2=\tfrac{3}{2}^+.$$

From Eq. (10a) we have

$$F_1=2, 3, 4, 5 \qquad \text{and} \qquad F_2=0, 1, 2, 3 .$$

That is, Ψ_1 is a mixture of the 2^+, 3^+, 4^+ and 5^+ states and Ψ_2 is a mixture of the 0^+, 1^+, 2^+ and 3^+ states. Therefore, we have the common states of 2^+ and 3^+ for Ψ_1 and Ψ_2, and the spin-parity condition is satisfied. Although we point out the spin-parity condition here, we do not develop the Racah algebra for our calculation. Instead, we shall show an order of magnitude calculation for the probability of formation of the 13.1 keV state of ^{235}U, since the wave functions of the nucleus and electrons are not well known for our purpose.

The off-diagonal matrix elements of the Coulomb interaction can be derived as follows: The Coulomb interaction in Eq. (3) is expanded into multipole components,

$$H'=-e^2Z\sum_{lm}\frac{4\pi}{2l+1}\frac{r_N{}^l}{r_e{}^{l+1}}Y_{lm}^*(\theta_e, \phi_e)\,Y_{lm}(\theta_N, \phi_N). \tag{11}$$

For simplicity, we assume $r_N<r_e$, and the contribution from the $r_N>r_e$ part is neglected in the following calculation. E' in Eq. (2) is given by

$$E'=(\Psi_2|H'|\Psi_1)=(\phi_2\phi_2|H'|\phi_1\phi_1).$$

Now let us denote the single-electron wave function of the orbit where electron hole exists by φ_i. Integrating over the electron variables except for those which

belong to the electron jumping down,

$$E' = (\psi_2\varphi_1|H'|\psi_1\varphi_2)$$
$$= -4\pi e^2 Z \sum_{lm} \frac{1}{2l+1} \int_0^R \psi_2^* Y_{lm}(\theta_N, \phi_N) r_N{}^l \psi_1 d\Omega_N r_N{}^2 dr_N$$
$$\times \int_0^\infty \varphi_1^* Y_{lm}^*(\theta_e, \phi_e) r_e{}^{-(l+1)} \varphi_2 d\Omega_e r_e{}^2 dr_e . \tag{12}$$

Here the electron wave function φ_1 is for the orbit where the electron hole originally exists, and φ_2 for the orbit where the electron hole jumps up.

For our particular example, the electric octupole has a nonzero value, and it is given by

$$E' = -\frac{4\pi}{7}\alpha Z \langle r_N{}^3\rangle \langle r_e{}^{-4}\rangle mc^2 \tag{13}$$

with

$$\alpha Z = \frac{92}{137} \qquad \text{and} \qquad mc^2 = 0.51 \times 10^6 \text{ eV} .$$

Here $\langle r_N{}^3\rangle$ is the off-diagonal matrix element of $r_N{}^3$, and $\langle r_e{}^{-4}\rangle$ is the off-diagonal matrix element of $r_e{}^{-4}$. These two quantities are given in units of the third and inverse fourth powers of the electron Compton wave length, respectively.

Next we adopt the simplest approximation for the matrix elements:

$$\langle r_N{}^3\rangle \sim R^3 \qquad \text{and} \qquad \langle r_e{}^{-4}\rangle \sim r_n{}^{-4}. \tag{14}$$

Here R is the nuclear radius and r_n is the radius of the n-th Bohr orbit. (If we compute $\langle r_e{}^{-4}\rangle$ by adopting the electron wave functions for a hydrogen-like atom, we have nearly the same result as above.) Furthermore, we assume that the electric octupole matrix element is enhanced with a factor 10 because of the nuclear deformation character. Numerically, we have

$$r_2 \sim 6 \qquad \text{and} \qquad R \sim 2 \times 10^{-2}$$

and consequently

$$E' \sim -0.04 \text{ eV} . \tag{15}$$

The mixing ratio in Eq. (2) is given by

$$\sin\theta \cos\theta = \frac{E'}{E(\Psi_1) - E(\Psi_2)} = \frac{-0.04 \text{ eV}}{(13{,}440 - 13{,}100)\text{ eV}}$$
$$= -1.1 \times 10^{-4}$$

and

$$x = \tan^2\theta \approx 1.2 \times 10^{-8}.$$

The probability of formation of nuclear excited state is given by Eq. (9):

$$P=\frac{\text{(probability of annihilation of } 3d_{3/2} \text{ electron hole)}}{\text{(probability of annihilation of } 2p_{3/2} \text{ electron hole)}}\times 2x$$

$$\sim\frac{1}{10}\times 2\times 1.2\times 10^{-8}\sim 2.4\times 10^{-9}. \tag{16}$$

Here the factor 1/10 is simply assumed for the first factor in the first line of Eq. (16). Finally we have a very small probability of nuclear excitation by electron transition compared with the X-ray or Auger electron emission.

§ 5. Discussion

We have obtained an extremely small value of $P\sim 2\times 10^{-9}$ for the nuclear excitation of the 13.1 keV state in ^{235}U. Although this is a very rare event, it is not impossible to detect it since the decay of this state creates the 26 min metastable state which is subsequently deexcited by the internal conversion process.

Since there is a possibility of chemical separation of ^{235}U nuclei which are in the excited states,[7] the nuclear excitation by electron transition is useful if P is appropriately large. At the present stage of investigation, the excitation of the 13.1 keV state is not appropriate for the purpose of mass production of enriched ^{235}U. It may, however, be useful for producing a small amount of the 100% pure ^{235}U, which is not available from conventional enriching procedures. The process is also useful for a calibration of the concentration of ^{235}U if the process is established. For this experiment, we have to bombard the uranium atoms with 17.2 keV X rays or electrons.

The direct excitation of the 30 eV state by the electron transition would become interesting if we can adjust the electronic energy level distances by means of chemical forms. The broadening of the width and the red shift of X-ray line under high pressures and temperatures may help the energy matching condition. We have still a difficulty that the level width of the 26 min state is extremely sharp.

We have investigated the Coulomb interaction as the interaction mechanism. The other electromagnetic interactions should also be taken into consideration. For example, the magnetic quadrupole excitation should be studied for the 13.1 keV state in ^{235}U.

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Note Added in Proof: The nuclear excitation by electron transition may be called the inverse internal conversion process.