Effects of atomic electrons on nuclear stability and radioactive decay

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Main ideas of our approach

1. In nuclear processes, nuclear and atomic physics are not detached, as it is commonly believed.
2. The change *of atomic electronic states* may influence the rate of *nuclear* decay and the condition of *nuclear* stability, and may redistribute the channels of *nuclear* decay. This is not an exotic. It is confirmed with experiments. It is necessary to search for ways to use it.
3. The changes of atomic electronic states may be caused by application of a strong magnetic field..

1 The topic of the present report

1. Experimental observation of violation of the thorium-234 secular equilibrium in electric explosion of metallic foil in a liquid.
2. The impact of β-decay into bound atomic electron states upon the fraction of delayed neutrons.
3. Control of the nuclear reactor.

The present report is aimed at revisiting the problem of, as is commonly believed, weak coupling of nuclear and atomic physics in the nuclear processes. Despite the energy-space-time scale of nuclear processes considerably differ from that of atomic ones, a lot of experimental evidences for a strong effect of atomic electrons on nuclear process is documented.

For nuclear processes with large transition energy (of the order of ~ 10 MeV and larger) the influence of atomic electrons may be really neglected. The latter is not true, however, for nuclear processes with transition energy below ~ 0.5 MeV. Such processes include

(i) various nuclear decays with participation of electro-weak interaction (e.g., $\beta^{\pm}$-decays, K-capture), and

(ii) practically all the nuclear transitions in the long-lived metastable isomers (usually these isomers are born when the excited state of the nucleus

possesses small enough excitation energy, of the order of 100 keV, and the value of its spin substantially differs from that in the ground state).

The de-excitation of isomers is well known, especially for small excitation energies (almost for all isomers), to be essentially influenced by the electrons of internal conversion. The internal conversion mechanism opens a new channel of de-excitation, additional one to emission of the quantum of γ- radiation. Significantly, the probability of conversion electron's emission depends of the atomic electron's wave function in the point of nucleus location. Therefore, this probability changes under deformation of atomic electron's density profile to influence the lifetime of the nucleus.

The above may be illustrated with an excellent experimental work by Bainbridge K.T., Goldhaber M., carried out in 1951 [1]. The authors managed to detect variability of the half-lifetime of ^{99}Tc in various chemical compounds (e.g. in the salt $KTcO_4$ and metallic state of Tc). The coefficient of conversion, α, in the element $^{99}Tc^m$ (of lifetime 6.01 hours) for the transition E3 with transition energy ~2 keV attains the value of 10^{10}

2 The bound-state β-decay

The theory of β-decay with the capture of ejected β-electron into the bound state in the atom (i.e. when β-electron doesn't escape from the atom and occupies a bound atomic state) was developed in [2 – 5]. Such a decay is obviously an inverse process with respect to K-capture. It was noticed that such a decay broadens the volume, in the phase space, of the final state and, hence, increases the probability of the decay. As far as β-decay transforms the nucleus toward an increase of its electric charge Z, an increase of the probability of β-decay results in a shift of the condition of nuclear stability toward bigger values of Z.

The calculation of the ratio of probabilities for β-decay into, respectively, bound and free state of ejected β-electron is similar to conventional calculation of the ratio of probabilities of K-capture and positronic β^+-decay [6]. Relying on the known results, it is easy to derive that the appearance of unoccupied electron state in the atom increases the value of λ, the constant of β-decay, by the value Δλ (in atomic units $\hbar = c = m_e = 1$):

$$\frac{\Delta\lambda}{\lambda} \sim 60\pi \left(\frac{\alpha Z}{E}\right)^3,$$

where α=1/137 is the fine structure constant, Z, electric charge of a nucleus.

It is seen that the value $\Delta\lambda/\lambda$ increases with decreasing energy E

3 Distortion of Secular Equilibrium of ^{234}Th

The effect of the electric explosion of titanium foils on a liquid dielectric was studied in a series of experiments in which a solution of uranyl sulfate (UO_2SO_4) in the distilled water was used as a dielectric [7] In this experiment, a decrease of intensity of the line <1001 keV> of ^{234}U, which is a daughter product of $^{234}Pa^m$, relatively to intensity of doublet lines <92.38|92.80 keV> of the mother nucleus $^{234}Pa^m$, was observed.

Figure 1 shows the diagram of the decays of ^{234}Th which is a product of α-decay of ^{238}U. The metastable state of ^{234}Pa (of lifetime 1.17 minutes) possesses the excitation energy 73.92. The excited nuclear states of $^{234}Pa^m$ are populated via β-decay of ^{234}Th with subsequent depopulation via emission of γ-quantum. The decay of the excited state of $^{234}Pa^m$ includes two groups of channels: via radiative decay to ground state $^{234}Pa^m(0^-) \rightarrow {}^{234}Pa^m(3^+) \rightarrow {}^{234}Pa(4^+)$ (0.16%) and via β-decays in ^{234}U (99.84%) with subsequent emission of γ-quanta

Figure 2 shows the ratio of intensities of spectral line <1001>, I_{1001} , and doublet spectral line <92>, I_{92}. This ratio is normalized by the same ratio for the undisturbed solution, as measured before explosion:

$$R = \frac{\left(I_{1001}/I_{92}\right)_{exp}}{\left(I_{1001}/I_{92}\right)_{Control}}$$

If nothing happens with the solution, this ratio is equal to unity. This ratio does not depend on the density of atoms in the solution, and of fluctuations of this density, because these spectral lines are due to the chain of transformation of the same mother nucleus.

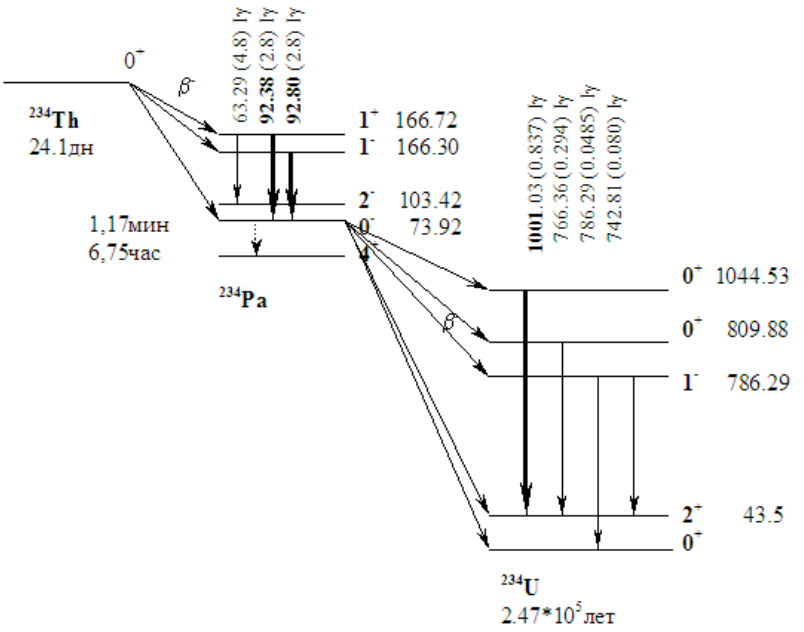

Fig. 1. The diagram of decays of ^{234}Th

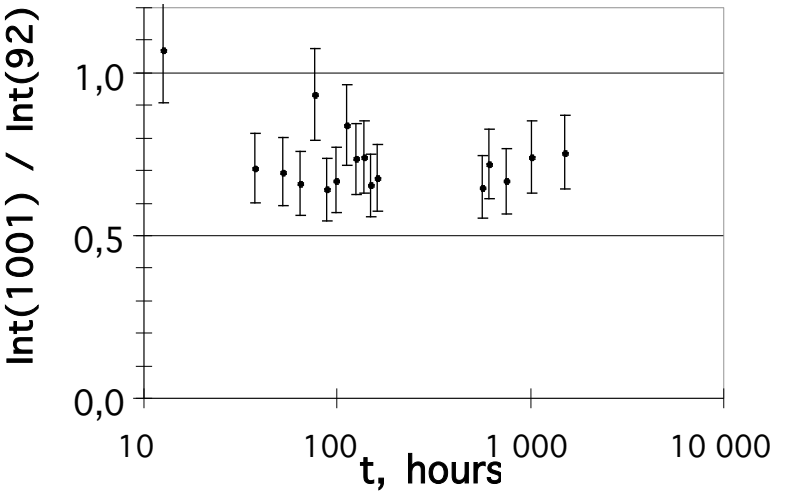

Fig.2 The time dependence of the ratio of line intensities $I_{1001}/I_{92,}$ normalized by this ratio for undisturbed sample.

To interpret the observed distortion we suggest the following hypothesis. Assume the appearance, for any reason, of unoccupied electron state in the β-active atoms of ^{234}Th and, probably, ^{234}Pa^{m}. In such a case, as noted above, the decay constant for all the allowed transitions is increased. As far as the energies of β-decay of ^{234}Th and ^{234}Pa^{m} amount to, respectively, ~ 100 keV and ~ 1 MeV, it is seen that the change of decay constants for ^{234}Pa^{m} is small as compared to those for ^{234}Th:

$$\left.\frac{\Delta\lambda}{\lambda}\right|_{Pa} \sim 10^{-3} \left.\frac{\Delta\lambda}{\lambda}\right|_{Th} << \left.\frac{\Delta\lambda}{\lambda}\right|_{Th}$$

In the equilibrium state the intensity I_{1001} of the line <1001> of uranium ^{234}U, which is a daughter product of ^{234}Pa^{m} , is determined by the quantity of protactinium atoms, N_{Pa} , and the decay constant λ_{Pa} for atom ^{234}Pa^{m}: $I_{1001} \propto N_{Pa} \cdot \lambda_{Pa.}$ The number of atoms, N_{Pa} , is determined by the following balance equation:

$$N_{Pa} \cdot \lambda_{Pa} = N_{Th} \cdot (\lambda_1 + \lambda_2),$$

where N_{Th} is the number of Th atoms, λ_1 , the decay constant for the decay of Th to the ground state of $^{234}Pa^m$ ($E_1 \cong 200$ keV), λ_2 , the decay constant for the decay of Th to the excited state of $^{234}Pa^m$ ($E_2 \cong 100$ keV). The intensity of the line of <92.38|92.80> of protactinium is equal to $I_{92} \propto N_{Th} \cdot \lambda_2$. Using the estimate for $\Delta\lambda/\lambda$, it is easy to derive from above equation, with allowance for the smallness of $\Delta\lambda_{Pa}/\lambda_{Pa}$, that the intensity of the line <1001> decreases relatively to that of the line <92> (as compared with similar ratio in the undisturbed sample). Indeed, because of inequality $E_2 < E_1$ and of its consequence, $\Delta\lambda_2/\lambda_2 > \Delta\lambda_1/\lambda_1$, we obtain:

$$R = \frac{(I_{1001} + \Delta I_{1001}) \Big/ (I_{92} + \Delta I_{92})}{I_{1001} \Big/ I_{92}} = \frac{1 + \Delta N_{Pa} \Big/ N_{Pa}}{1 + \Delta\lambda_2 \Big/ \lambda_2} \cong 1 - \frac{\lambda_1}{\lambda} \cdot \left(\frac{\delta\lambda_2}{\lambda_2} - \frac{\delta\lambda_1}{\lambda_1} \right) < 1$$

where $\lambda = (\lambda_1 + \lambda_2)$. It is such a sign of the change of the ratio of spectral line intensities that has been observed in the experiment.

4 The impact of bound-state β-decay upon the fraction of delayed neutrons

The decay of ^{235}U gives large number of daughter nuclei of atomic weight in the range from A=72 to A=160. The distributions of daughter nuclei in their mass and electric charge have been investigated in the literature in detail. The majority of daughter nuclei are unstable due to an excess of neutrons. A part of these nuclei (~50 nuclei) is capable of emitting the delayed neutrons. The diagram of decay is shown in Fig. 3.[8] As noted above, the presence of unoccupied states of atomic electron moves the nuclear stability threshold towards decreasing the neutron-proton ratio.

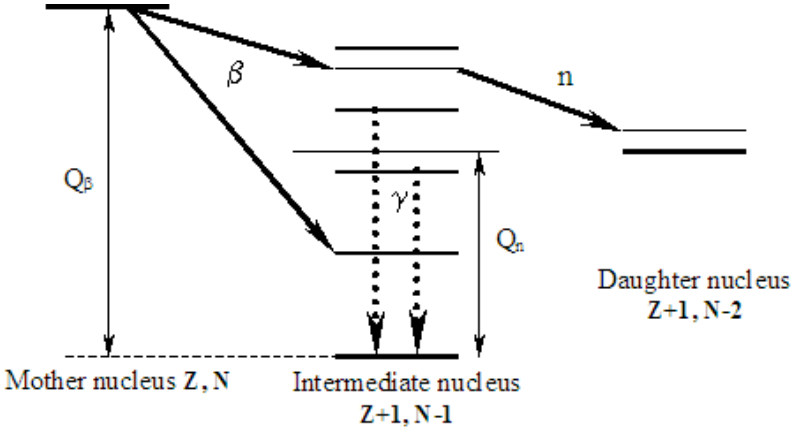

Fig. 3. The diagram of decay of an atom emitting the delayed neutron.
Q_β is maximal energy of β-decay,
Q_n , the binding energy of the neutron in the intermediate nucleus

Hence, the ionized nuclei with an excess of neutrons may release the neutrons easier. **Below we discuss this issue in more detail.**

The β-decay of mother nucleus (i.e. of the emitter of delayed neutron) via the channel with the lower energy of β-transition gives the intermediate nucleus in the excited nuclear state. For the excitation energy exceeding the value of Q_n , the binding energy of the neutron in the nucleus, the intermediate nucleus emits the neutron. This emission takes place practically instantaneously, so that the delay time is fully determined by the lifetime of the mother nucleus. Note that the fraction of delayed neutrons is determined by the β-decays with small transition energy while their intensity for all the emitters of delayed neutrons doesn't exceed 10%.

For the majority of intermediate nucleus, one has $Q_n \sim 5 \div 7$ MeV. As far as the energy of β-decay with the neutron yield, $(Q_\beta - Q_n)$, is appreciably smaller than Q_β, it follows that if the channel of the bound-state β-decay is open, the ratio $\Delta\lambda_n/\lambda_n$ for the neutron channel with small energy E has to appreciably exceed $\Delta\lambda_\beta/\lambda_\beta$ of neutronless decay to the lower-lying levels:

$$\frac{\Delta\lambda_n}{\lambda_n} > \frac{\Delta\lambda_\beta}{\lambda_\beta}$$

The fraction of delayed neutrons, β, is proportional to the following ratio:

$$\beta \propto \frac{\lambda_n}{\lambda_n + \lambda_\beta}.$$

The relative change of β may be easily derived:

$$\frac{\Delta\beta}{\beta} = \frac{\lambda_\beta}{\lambda} \cdot \left(\frac{\Delta\lambda_n}{\lambda_n} - \frac{\Delta\lambda_\beta}{\lambda_\beta} \right) > 0,$$

where $\lambda=\lambda_n+\lambda_\beta+\Delta\lambda_n+\Delta\lambda_\beta$. This allows to formulate the following conclusion.

The appearance of unoccupied electron state in an atom capable of emitting the delayed neutron, leads to an increase of the fraction of delayed neutrons.

5 Control of the nuclear reactor

The state of nuclear reactor is used to be described with kinetic equations. These allow for the densities of only those nuclei emitting the delayed neutrons, which underwent the β-decay via neutron channel whereas the daughter nuclei which underwent the β-decay via neutronless channel are thought to be lost for the chain reaction. In fact, the neutrons which cause the production of daughter nuclei undergoing the neutronless β-decay, were taken into account as an effective loss of neutrons, i.e. in decreasing the reactor's excess reactivity ρ.

To analyze the behaviour of reactor under changing constant λ of β-decay, we consider kinetic equations in the single-group approximation for delayed neutrons, with allowance for all the nuclei emitting the delayed neutrons (including those nuclei whose decay doesn't release the neutron):

$$\frac{dn}{dt} = \frac{\rho - \beta}{T} n + \lambda_n \cdot C, \qquad \frac{dC}{dt} = \frac{\beta_t \cdot n}{T} - (\lambda_n + \lambda_\beta) \cdot C,$$

where

- n, neutron density,
- ρ, excess reactivity of nuclear reactor,
- β, the fraction of delayed neutrons,
- $T = 10^{-3}$ s, the lifetime of one generation of instantaneous neutrons,

- C, density of nuclei emitting the delayed neutrons, including those nuclei whose β-decay doesn't release the neutron;
- λ_n , the constant of β-decay with subsequent release of neutrons,
- λ_β , the constant of β-decay without subsequent release of neutrons,
- $\beta_t=\beta\cdot(\lambda_n+\lambda_\beta)/\lambda_n$

Let us consider reactor in a steady-state regime, i.e. under reactivity ρ=0, with the reactivity caused by the instantaneous neutrons being equal to a constant, $\rho_{inst} = -\beta_b$ (β_b is the initial value of the fraction of delayed neutrons). Consider the variations $\lambda_\beta \rightarrow (\lambda_\beta+\Delta\lambda_\beta)$ and $\lambda_n \rightarrow (\lambda_n+\Delta\lambda_n)$ which obey the condition

$$\frac{\Delta\lambda_n}{\lambda_n} > \frac{\Delta\lambda_\beta}{\lambda_\beta}.$$

Assuming the above variations to happen instantaneously (i.e. in a time interval << T), we derive the following relation from the above equation:

$$\frac{d^2n}{dt^2} + \left[\frac{\beta_b}{T} + (\lambda_n + \lambda_\beta + \Delta\lambda_n + \Delta\lambda_\beta)\right]\cdot\frac{dn}{dt} - \frac{\beta_b}{T}n\cdot\left[\Delta\lambda_n\cdot\frac{\lambda_\beta}{\lambda_n} - \Delta\lambda_\beta\right] = 0,$$

which, in the first order in Δλ, describes an instability with the following increment:

$$k = \tfrac{1}{2}\left[\frac{\beta_b}{T} + \lambda\right]\left[\sqrt{1 + 4\left[\Delta\lambda_n\cdot\frac{\lambda_\beta}{\lambda_n} - \Delta\lambda_\beta\right]\frac{\beta_b}{T}\cdot\left[\frac{\beta_b}{T} + \lambda\right]^{-2}} - 1\right],$$

where $\lambda = (\lambda_n+\lambda_\beta+\Delta\lambda_n+\Delta\lambda_\beta)$. Under condition $T\cdot\lambda << \beta_b$ one has:

$$k = \lambda_\beta\cdot\left(\frac{\Delta\lambda_n}{\lambda_n} - \frac{\Delta\lambda_\beta}{\lambda_\beta}\right).$$

It follows from this equation that an impact of unoccupied states of electron in the atoms whose nuclei emit the delayed neutrons, may enhance the power of nuclear reactor without the change of the contribution of instanta-

neous neutrons to the reactivity. As known, major contribution to production of delayed neutrons stems from daughter nuclei of Z~35÷37. Besides, for the neutronless channel the transition energy is estimated to be $E_\beta >> E_n \sim 1$ (in the units of electron's rest mass). This allows to obtain the estimate:

$$k \sim \lambda \cdot \frac{\Delta\lambda_n}{\lambda_n} \sim \lambda \cdot 60\pi \left(\frac{\alpha \cdot 35}{1} \right)^3 \sim \pi \cdot \lambda$$

where λ is the constant of β-decay of nuclei emitting the delayed neutrons, for the case of a neutral atom.

6 Conclusions

The clearance of electron atomic states in a certain number of atoms whose nucleus emits the delayed neutron (that opens the bound-state β-decay channel in such atoms) may increase the fraction of delayed neutrons.

This opens, in principle, the possibility of designing a reactor based on a new strategy of running the reactor: a reactor is designed to be certainly subcritical and capable of being switched on and controlled due to an increase of delayed neutrons. Conversely, the reactivity caused by instantaneous neutrons is retained to be a constant.

Recall that the above-mentioned clearance of electron atomic states may be produced not only by the ionization of atoms but by a strong magnetic field as well.

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