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Nuclear isomers as a tool for studying the influence of zero-point fluctuations of an electromagnetic field on the probability of spontaneous electromagnetic transitions

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Abstract. A review of works on the influence of zero-point fluctuations of the electromagnetic (EM) field (ZPFs) on EM transitions in atomic nuclei inside metals is presented. In a metal, as well as in a resonator of extremely small dimensions, ZPFs are suppressed in the region of low frequencies ω , for which the reflection of EM waves from the metal surface is still significant ($\hbar\omega$ is less than ~ 1 keV). Based on the concept of the stimulation of spontaneous EM transitions of energy $\hbar\omega_0$ by resonant ZPFs of frequency ω_0 , one could expect suppression of transitions of energy up to ~ 1 keV for excited nuclei in a metal matrix. In experiments with nuclear isomers, such an effect was indeed found for conversion transitions of 76, 910, and 2173 eV energy, which cannot be explained only by the deformation of the electron shells of isomeric atoms or by the scattering of conversion transitions with a decrease in the ZPFs energy density in metals at the transition frequencies.

Keywords: nuclear isomers, probability of nuclear transitions, zero-point fluctuations of the electromagnetic field

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Ядерные изомеры как инструмент для исследования влияния нулевых колебаний электромагнитного поля на вероятность спонтанных электромагнитных переходов

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Аннотация. Проведен обзор экспериментальных и теоретических работ по влиянию нулевых колебаний электромагнитного (ЭМ) поля (НКП) на вероятность ЭМ переходов малой энергии в атомных ядрах внутри металлов. В металле, как и в резонаторе предельно малых размеров, НКП подавлены в области малых частот ω , для которых еще существенно отражение ЭМ волн от поверхности металла ($\hbar\omega$ меньше ~ 1 кэВ). Исходя из представления о стимуляции спонтанных ЭМ переходов энергии $\hbar\omega_0$ резонансными НКП частоты ω_0 , можно было ожидать подавления ЭМ переходов энергии до ~ 1 кэВ для возбужденных ядер в матрице металла. В экспериментах с ядерными изомерами действительно был обнаружен такой эффект для конверсионных переходов энергии 76, 910 и 2173 эВ, который не может быть объяснен только деформацией электронных оболочек изомерных атомов или рассеянием конверсионных электронов на атомах матрицы. Качественно эффект соответствует подавлению конверсионных переходов при уменьшении в металле плотности энергии НКП на частоте перехода.

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Ключевые слова: ядерные изомеры, вероятность ядерных переходов, нулевые колебания электромагнитного поля

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Introduction

Immediately after the creation of quantum electrodynamics, the idea arose of stimulating the spontaneous emission of photons by zero-point fluctuations of an electromagnetic (EM) field (ZPFs) [1] (see also references in [2]). This idea was based on the description of the interaction of charged particles according to perturbation theory, when the Hamiltonian of a system of particles and an EM field is divided into the Hamiltonians of particles and fields (photons and ZPFs), that do not interact with each other, and the energy of interaction between them. Then, if we do not consider the processes induced by photons, the interaction of particles is caused by their interaction with ZPFs. The ZPFs energy density $dW_{ZPF}(\omega)$ in the frequency range $d\omega$ is given by the formula (see, for example, [3])

$$dW_{\rm ZPF}(\omega) = \theta^2(\omega) \frac{\hbar\omega^3}{\pi^2 c^3} d\omega, \qquad (1)$$

where *c* is the light speed, \hbar is Planck's constant divided by 2π . For the convenience of further analysis, a real function $\theta(\omega)$ is introduced into formula (1), in the free space $\theta(\omega) = 1$.

The matrix element $S^{\text{ph}}(t)$ of spontaneous emission of a photon of energy $\hbar\omega_0$ at moment t is proportional to the following integral (see, for example, [4])

$$S^{\rm ph}(t) \propto \int_{-\infty}^{\infty} d\omega \, \omega^{3/2} \, \theta(\omega) \, J_1(\omega) \underbrace{\int_{t_0}^{t} dt_1 \, e^{i \, (\omega_0 - \omega) \, t_1}}_{\delta(\omega_0 - \omega)} \propto \theta(\omega_0), \tag{2}$$

where t_0 is the moment of formation of the excited state, $J_1(\omega)$ is the space integral of the particle transition current. The average photon emission time is of the order of the half-life of the excited state $T_{1/2}$ and $t - t_0 >> 1/\omega_0$. Then the integral over time gives the δ -function and the emission probability is proportional to the $dW_{\rm ZPF}(\omega_0)$. In a cavity with a diameter of D with ideally reflecting walls (in a resonator), ZPFs do not contain low frequencies $\omega < c/D$. If an emitter is placed in the resonator of $D < c/\omega_0$, then the emission of a photon of energy $\hbar\omega_0$ will be suppressed, which was observed for excited atoms (see references in [2]).

Dependence of the probability of a nuclear conversion transition on the ZPFs spectrum

It is interesting to study the stimulation of spontaneous EM transitions by ZPFs in a quantum system located inside a medium with photon absorption, for example, inside a metal. For this, however, the emission of photons by atoms is inconvenient, since photons of low energy do not emerge from the metal, and the lifetimes of the atom's excited states are short. For research, it is more convenient to isomeric nuclear states that decay by a low-energy conversion transition, when nuclear excitation is transferred to the electron shell of an atom with electron emission due to inelastic scattering of an atomic electron by an excited nucleus [4]. It was shown in [2] that ZPFs stimulate the nuclear conversion transition in the same resonant manner as the transition with photon emission. Indeed, the matrix element $S^{c}(t)$ of the conversion transition of energy $\Delta E_{\rm N} = \hbar \omega_{0}$ is obtained in the second order of the perturbation theory and has the form

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$$S^{c}(t) \propto \int_{-\infty}^{\infty} d\omega J_{2}(\omega) \omega \theta^{2}(\omega) \int_{t_{0}}^{t} dt_{1} e^{i(\omega_{c}+\omega)t_{1}} \int_{t_{0}}^{t_{1}} dt_{2} e^{i(\omega_{0}-\omega)t_{2}} \propto \theta^{2}(\omega_{0}), \qquad (3)$$

where $J_2(\omega)$ is the spatial integral of the product of the transition currents of the nucleus and the conversion electron, $\hbar\omega_e$ is the change in the electron energy, the limits of integration over time t and t_0 are the same as in formula (2). The interaction duration $t_1 - t_0$ is of the order of $T_{1/2}$ of the isomeric state and always $\Delta E_N (t_1 - t_0) >> \hbar$. Then, in formula (3), the integral over time t_2 gives the δ -function, which provides the resonance in the stimulation of the conversion transition by ZPFs. In the case of inelastic scattering of free particles, the condition $\Delta E (t_1 - t_0) >> \hbar$ is not satisfied and there is no resonance in the transfer of energy ΔE .

Zero-point fluctuations of the EM field (ZPFs) inside the medium

For nuclei in the resonator, where $\theta(\omega_0) = 0$, according to formula (3), the probability of conversion transitions of energy $\hbar\omega_0$ is equal to zero. For nuclei in a metal, such an effect is possible at $\hbar\omega_0$ less than ~1 keV, when reflection from the metal is still significant for EM waves with a frequency ω_0 . Metal is the limiting case of a small collapsing resonator and due to the Doppler effect upon reflection of ZPFs from the approaching walls of the initial resonator, the frequency of ZPFs increases and the energy density $dW_{ZPF}(\omega)$ of low frequency ZPFs inside the metal decreases [5].

For a transparent medium, $dW_{\text{ZPF}}(\omega)$ is given by formula (1) at $\theta(\omega) = n^{3/2}$ [3], the optical refractive index $n \leq 1$ [6]. In a medium with absorption of EM radiation, $\theta(\omega) \leq 1$ as well [3, 5]. Metals seem to play a special role, since ZPFs cannot be absorbed by the medium due to transitions between stationary states of atoms (see, e.g., [7]). But $dW_{\text{ZPF}}(\omega)$ can decrease upon interaction with electrons in a continuous energy spectrum, in particular, with conduction electrons in a metal. The absorption of ZPFs in a metal is weaker than the absorption of real photons, and edge effects for ZPFs can affect deeper layers of the metal than for real photons.

Another way to reduce $dW_{\rm ZPF}(\omega)$ inside a medium is possible by increasing the energy of interaction between ZPFs and the electrons of the atoms of the medium, if the region of free states in the continuous electron energy spectrum expands during phase transitions in the medium [8]. Thus, when a crystal is formed from free atoms, the number of allowed electronic states increases when continuous energy bands appear for electrons instead of discrete levels of low energy for free atoms. After the phase transition, the ZPFs spectrum changes with time due to the diffusion of ZPFs into the medium from the free space.

In all considered cases, in the metal $\theta(\omega) \le 1$ and $\theta(\omega) \to 1$ with increasing ω . Then, according to formula (3), one can expect suppression of conversion transitions of energy up to ~ 1 keV for nuclei in a metal. This effect was studied at the Khlopin Radium Institute on several nuclear isomers.

Isomeric transition 76 eV of ^{235m}U nuclei in silver

The ^{235m}U isomer is formed in the α -decay of ²³⁹Pu and decays into the ²³⁵U ground state by a conversion transition of energy 76 eV. This transition was studied by many authors; the ^{235m}U recoil nuclei emitted from the ²³⁹Pu layer were collected in an Ar-atmosphere or in a vacuum onto backings of different metals. It was shown that $T_{1/2} \approx 26$ min and varies within 5% due to deformation of the electron shells of isomer atoms or due to scattering of conversion electrons on backings atoms. The decay of ^{235m}U in a metal at a depth of d > 100 Å was studied only in [9, 10] using the following method. 1) The 1000 Å thick Ag-layer was deposited on the W-backing, then ^{235m}U recoil nuclei were deposited on this Ag-layer. To improve the structure of the U – Ag system, the preparation was annealed and then divided it into several identical samples. 2) Silver was removed from Sample 1 by heating it in vacuum so that the uranium atoms remained on the surface of the W-backing, and the decay curve of Sample 1 was measured by detecting ^{235m}U conversion electrons with a channel electron multiplier. At this time, ^{235m}U in other samples remained "buried" in the Ag-layers. 3) Then the silver was removed from Sample 2 and the decay curve

was also measured for it with the same detector. If the isomeric transition is suppressed inside the Ag-layer, then a "step" ΔN is formed between the decay curves of the samples, caused by different residence times of ^{235m}U in silver for the samples. The course of the experiments was checked by examining the structure of the deposited Ag-layers and the integral spectrum of conversion electrons from the samples. Together with ^{235m}U, in the same way ²³⁷U nuclei from the ²⁴¹Pu layer were applied to the preparations as an isotopic tag; its activity in the samples was determined by ²³⁷U γ -quanta.

Dozens of experiments were carried out according to the scheme described above. All of them showed an excess activity of ^{235m}U for the compared samples, in which ^{235m}U was in silver longer. But with the same difference in Δt in the exposure time of Samples 1 and 2 in silver, the magnitude of this excess varied, apparently due to insufficient reproducibility of the structure of the U–Ag system. The greatest effects were obtained when ^{235m}U nuclei were deposited on the preparations in an Ar-atmosphere (Fig. 1), which was possibly caused by a change in the ZPFs spectrum during phase transitions in the Ag–U oxide layers at the boundary of the Ag-layers upon annealing of the preparations, as was noted in Section 3. When four samples were prepared at the same time, for them the accumulation of the "step" effect was observed with time, however, this accumulation gradually ceased over a time of about 100 min. Whether this saturation was due to the diffusion of ^{235m}U nuclei onto the surface of the Ag-layers, or, as discussed in Section 3, it was due to the diffusion of ZPFs into the interior of the Ag-layer with ^{235m}U nuclei after the phase transition upon annealing of the Ag-layer, this question remained unclear and needs further research.



Fig. 1. Example of 235m U decay curves in Samples 1 and 2. *N* is the electron count per 60 s, *t* is the time after the end of 235m U deposition. Asterisks indicate the moments of silver removal from the samples. Step ΔN between curves gives $T_{1/2} > 230$ min for 235m U inside silver. The normalization error for *N* in Sample 2 (dashed lines) is caused by the 237 U activity measurement in the samples. Here and below, the errors are at the level of one standard deviation

Isomeric transition 2173 eV of ^{99m}Tc nuclei in metals

The ^{99m}Tc isomer with a period $T_{1/2} \approx 6.02$ h decays via a conversion transition of an energy of 2173 eV to the underlying state and then to the ground state of the nucleus, emitting a 140 keV γ -quantum. $T_{1/2}$ can be measured with an accuracy of 0.01%, $T_{1/2}$ variations within 0.1% were observed due to deformation of the electron shell of ^{99m}Tc atoms. In Ref. [11], an increase in $T_{1/2}$ of the ^{99m}Tc isomer was found as it deepens into the matrix of Cu, Ag, Sn, Au, and Pb metals. In [11], matrices with ^{99m}Tc uniformly distributed over them were used. The ^{99m}Tc matrices of different sizes were compared in terms of the change in the intensity of 140 keV γ -quanta counting with time, using a setup that made it possible to alternately measure γ -spectra from four sources. There always has been ^{99m}Tc pertechnetate as the reference source. In all cases, the observed $T_{1/2}$ for ^{99m}Tc increased with an increase in the matrix size and, accordingly, with an increase in the depth of ^{99m}Tc in the matrix. The decay curves of ^{99m}Tc in Pb-matrices are shown in Fig. 2.



Fig. 2. Decay curves of ^{99m}Tc in Pb-matrices. $Y = \ln[N(t) \exp(\ln 2 \cdot t/t_o)]$, *t* is the time, $t_o = 6.02$ h, N(t) is the count of 140 keV γ -quanta from sources per 3000 s. *d* is the average depth from which ^{99m}Tc decay γ -quanta are visible. For the smaller matrix (d = 0.19 mm), $T_{1/2} = 6.028 \pm 0.002$ h, for the larger matrix (d = 0.23 mm) $T_{1/2} = 6.043 \pm 0.004$ h

Transition 910 eV of ^{154m}Eu nuclei in tin

The 910 eV transition raising from the decay of the ^{154m}Eu isomer ($T_{1/2} = 46$ min) obtained from Sm in the (p, n) reaction at the cyclotron is promising for studying the matrix effect [12]. This transition is at the beginning of one of the competing cascades of ~100 keV γ -transitions, via which the ^{154m}Eu isomer decays. By measuring the relative yield of these γ -quanta, one can measure 910 eV transition probability for ^{154m}Eu nuclei directly inside the matrix. Another advantage of this transition is that its energy is less than the energy of the ^{99m}Tc isomeric transition, and the matrix effect for 910 eV transition should be greater than for ^{99m}Tc transition. In experiment [12], it actually turned out that when thin flat pieces of an Sm-metal containing ^{154m}Eu isomers are dissolved in Sn-metal, the probability of 910 eV transition decreases by approximately 20% compared to the probability for ^{154m}Eu chloride.

Study of other nuclear isomers in metals

At the Khlopin Radium Institute the ^{229m}Th, ^{234m}Pa, and ^{244m}Np isomers were also studied, but they turned out to be inconvenient for experiments. The isomeric transition in the ^{229m}Th nucleus has a very low energy $\Delta E \approx 8$ eV, and the transition probability strongly depends on the chemical environment of the isomer; against this background, it is difficult to distinguish the ZPFs effects. For ^{234m}Pa ($T_{1/2} = 1.17$ min), it turned out that the isomeric transition has a very high energy $\Delta E \approx 2.5$ keV, and the matrix effects are too small to observe. For the ^{240m}Np isomer ($\Delta E < 18$ keV, $T_{1/2} = 7.22$ min), an excess of $T_{1/2}$ for nuclei in the Ag-metal was observed compared to the Np-chloride, but the complexity of the experiment prevented systematic studies.

Conclusion

The results of the experiments indicate the suppression of conversion transitions of nuclei in metals, which cannot be explained only by the deformation of the electron shells of atoms or by the scattering of conversion electrons in metals. Qualitatively, the effect corresponds to the suppression of these transitions with a decrease in the ZPFs energy density in metals at the transition frequency. Further studies are needed to more accurately elucidate the nature of this effect.

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