# Acceleration of the decay of $^{235m}$U by laser-induced resonant internal conversion

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The 26-min isomer $^{235m}$U is used to demonstrate that nuclear transitions can be accelerated by laser radiation. An unorthodox mechanism is proposed for internal conversion with the excitation of an electron to a discrete level. The resonance defect between electronic and nuclear transitions is corrected for by suitably choosing the laser radiation frequency.

## 1. INTRODUCTION

The transformation and decay of particles acquire certain characteristic features when they occur in the field of intense electromagnetic radiation. However, although currently available laser intensities do not directly affect the rates of decay of elementary particles[1] or of nuclear reactions,[2] they can have a significant effect on nuclear decay by interacting with atomic electrons. For example, $K$-capture in light atoms was used in Ref. 3 to show that the hyperfine level populations can be altered by an electromagnetic wave.

A unique example is provided by the $^{235m}$U nucleus whose metastable state has a half-life of 26 min and an energy of only 76 eV (Ref. 4), which is comparable with the binding energies of outer-shell electrons. The $^{235m}$U nucleus returns to the ground state by converting $E3$ transitions in the $5p$, $6p$,and $7f$ shells.[5] The internal conversion coefficient $\alpha(E3)$ is very high, and formal calculations show that $\alpha(E3)\approx 10^{19}$ (Ref. 5), but this figure does not take into account the possibility of $\gamma$-rays due to electronic bridges,[6] which reduces the size of $\alpha$ by approximately five orders of magnitude.[7] These electronic bridges were examined experimentally in Ref. 8 in the case of the 30.7-keV transition in $^{93}$Nb. Internal conversion coefficients calculated without these electronic-bridge corrections are usually referred to as "tabulated coefficients."[9]

It was noted in Ref. 10 that the nuclear transition in $^{235m}$U lies close to the resonance with the electronic $5f\rightarrow 6s$ $E3$-transition, so that a study was made of the probability of excitation of the nuclear isomeric state for the given electron transition.

Our aim here is to observe that the closeness of the nuclear and electronic transition energies, and also the high-internal-conversion coefficients can be exploited to achieve a considerable acceleration of nuclear decay by laser radiation. In particular, by cancelling the resonance defect by the emission or absorption of laser photons in the atomic electron shell, it is possible to increase the decay rate of $^{235m}$U to its ground state by a few orders of magnitude.

Since this conversion mechanism relies on the transition of an electron to a discrete level, and not, as usual, to the continuum, we shall refer to it concisely as "discrete conversion." Moreover, since its probability is directly related to the detuning between nuclear and atomic transition frequencies, we shall also use the phrase "resonant conversion."

The effect of discrete conversion on nuclear decay was examined in Ref. 11 for $^{189}$Os, but the effect was found to be small because of the large detuning from resonance. A similar scheme was considered in Ref. 12 for a muon in the $K$-orbit of a fragment from nuclear fission induced by a radiationless transition of the muon in a $\mu$-atom. In this case, the cascade deexcitation of the fragment may include conversion with the excitation of the muon to the $2p$ level, followed by the emission of a mesic x-ray photon. According to the predictions of Ref. 12, the $\gamma$-ray spectrum of the fragments should then contain characteristic mesic x-ray lines. A large effect can be achieved because, not surprisingly, the probability for discrete conversion is much higher than the usual probability.

Since our basic idea relies on resonant internal conversion, which is not well represented in the literature, Sec. 2 is devoted to the theory of this process. It gives definitions and derivations of relations that extend the ordinary internal conversion mechanism to discrete electron transitions. Section 3 examines the influence of a laser field on resonant conversion. Numerical estimates are given in Sec. 4 for the deexcitation of the isomeric state of $^{235m}$U. The concluding section summarizes our results and examines possible future research.

## 2. RESONANT INTERNAL CONVERSION

The internal conversion coefficient $\alpha(\tau,L)$, where $\tau$ and $L$ are, respectively, the type and multipolarity of the transition, is defined as the ratio of conversion and radiative transition probabilities for a given nuclear transition. Conversion coefficients can be calculated by existing and well known methods (see for example, Ref. 9). However, the direct extension of the theory of internal conversion to the resonant case is difficult for two reasons. First, the formal determination of the conversion coefficient from the same relations gives a quantity with the dimensions of energy, so that the physical interpretation of the coefficient as a probability ratio is lost. This happens because of the normalization of the final-state wave function. Second, the usual formalism does not take into account the resonant character of the process. The theory must therefore be extended to the discrete case.

This is not difficult to do because the atomic state $n'$ formed during resonant conversion has finite width, and decays along a particular channel $k$ with partial width $\Gamma_k$ (Fig. 1), where

$$\sum_k \Gamma_k=\Gamma_a,$$

in which $\Gamma_a$ is the total width of the level $n'$.

If we confine our attention in the spectral expansion of the electron Green's function in Fig. 1 to a single resonant

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level $n'$, we can write the amplitude for the process in the form

$$F=F_cF_k/(\Delta+i\Gamma_a/2), \tag{1}$$

where $\Delta=\hbar(\omega_n-\omega_a)$ is the difference between the nuclear and electronic transition energies, $F_c$ is the resonant conversion amplitude, and $F_k$ is the partial amplitude for the decay of the state $n'$ along the channel $k$.

Summing over all the possible decay channels, we obtain the following expression for the width of the process:

$$\Gamma=\frac{\Gamma_c\Gamma_a}{2\pi(\Delta^2+\Gamma_a^2/4)}, \tag{2}$$

where $\Gamma_c$ is the analog of the conversion width

$$\Gamma_c=\frac{2\pi}{2I_i+1}\sum_{mm'M_iM_f}|F_c|^2. \tag{3}$$

In (2) and (3), we have carried out (1) the standard averaging over $M_i$, (2) summation over $M_f$, i.e., over the projections of the nuclear spins $I_i$ and $I_f$ in the initial and final states, respectively, and (3) summation over $m, m'$, i.e., over the projections of the electron angular momenta $j, j'$ in the initial ($n$) and the intermediate ($n'$) states, respectively. It was assumed in this procedure that the electron shell $n$ was completely filled and the shell $n'$ was completely empty. The extension to the case of partially filled shells can be carried out in an obvious manner.

The ratio $\alpha_b=\Gamma_c/\Gamma_\gamma$ would be the tabulated internal conversion coefficient if the state $n'$ were in the continuous spectrum.[1)] We shall retain the designation "conversion coefficient" for this quantity, but for the sake of clarity, we shall add the subscript $d$. The coefficient will now have the dimensions of energy. On the other hand,the usual internal conversion coefficient will be extended to the discrete case by introducing the conversion factor $R=\Gamma/\Gamma_\gamma$, defined as the ratio of probabilities of discrete conversion and emission of a photon by the nucleus. It is readily shown from (2) and (3) that

$$R=\frac{\alpha_d(\tau,L)\Gamma_a}{2\pi(\Delta^2+\Gamma_a^2/4)}. \tag{4}$$

In our particular problem, we can neglect relativistic corrections that are usually taken into account in calculations of internal conversion coefficients. If we now pass to the limit in the well known formulas,[9] we obtain ($\hbar=c=1$)

$$\alpha_d(EL)=\frac{\alpha\pi L}{L+1}(2j+1)(j^1/_2L0|j'^1/_2)^2[(2L-1)!!\mathfrak{M}]^2\omega_n^{-2L-1}, \tag{5}$$

$$\mathfrak{M}=\langle n'|r^{-L-1}|n\rangle=\int_0^\infty g'(r)g(r)r^{1-L}\,dr,$$

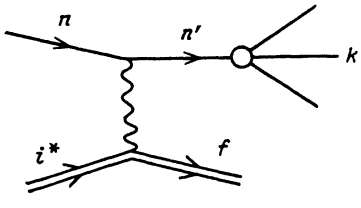


FIG. 1. Feynman diagram for resonant internal conversion. Double line represents a nuclear transition.

where $\alpha$ is the fine structure constant and $g$ and $g'$ are the nonrelativistic radial wave functions of the electron in the initial and final states, respectively.

### 3. RESONANT CONVERSION IN A LASER FIELD

Formula (4) can be extended without particular difficulty to the case of conversion in a laser field. It is essential to take into account the atomic shift and broadening, and to multiply (4) by the square of the amplitude for the state to which the electron undergoes the transition in the total wave function of the atom in the final state.

Figure 2 shows the simplest Feynman diagram for the proposed scheme. To make things clearer, from now on we shall confine our attention to a particular transition in $^{235m}$U. The nuclear $E3$ photon produces a virtual transition of the $6s$ electron to the $n'f$ level, which we shall take to be the $5f_{5/2}$ state.[10] To remove the resonance defect, this level is mixed by the laser radiation with the $n''d$ or $n''g$ level by absorption or emission of a laser photon, where both processes have equal probability for stimulated electromagnetic transitions. The deexcitation of the nucleus, accompanied by the electron transition $6s\to n''d, n''g$, and the formation of a hole in the $6s$ shell, is thus possible in the laser field. The decay of the hole requires separate examination. In particular,it can be used diagnostically as an indication of a particular nuclear decay scheme; for example, we can record a photon or an Auger electron from the $6p\to 6s$ transition. We note that the electromagnetic decay of the hole in the $6s$ shell can occur, at least in principle, in a coherent manner, and can thus lead to stimulated x-ray emission by the ensemble of uranium atoms with metastable nuclei.

We shall suppose that the most likely candidate for the final $n'd$ state is the $6d_{3/2}$ level at about 3 eV above the $5f_{5/2}$ level.[10] Retaining the components that are of interest to us here, we can write the final-state atomic wave function in the form

$$\Psi=(|6d_{3/2}\rangle+\beta|5f_{5/2}\rangle e^{-i\omega t})e^{-tEt/\hbar}, \tag{6}$$

where $E$ is the energy of the $6d$ level of the atom under normal conditions, $\omega$ is the frequency of the laser radiation, and the quantity

$$\beta=\Delta^{-1}\langle 5f_{5/2}|H_L|6d_{3/2}$$

determines the admixture of the $5f$ state to the $6d$ state in the laser field in first-order perturbation theory. $H_L$ is the Hamiltonian for the interaction between the electron and the radiation field which, in the electric dipole approximation, can be expressed in terms of the electric field $\mathscr{E}$: $H_L=-e\mathscr{E}z$.

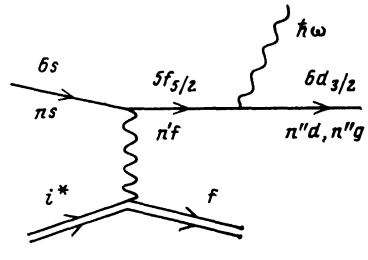


FIG. 2. Simple Feynman diagram for resonant conversion in laser radiation.

We emphasize that the function given by (6) is not the usual wave function of the two-level system in the resonant field because the quantity $\hbar\omega$ is not necessarily close to the separation between the $5f$ and $6d$ levels. However, even when these quantities are close to one another, (6) takes into account the antiresonant term that is significant for our problem. The validity of this "antiresonant approximation" in the present case is due to the fact that the dipole-coupled $5f$ and $6d$ states are close to one another in comparison with all other atomic states.

It follows from (6) that the $5f_{5/2}$ component appears in the wave function of the final state with the energy $E+\hbar\omega$. Hence the conversion factor in the presence of the laser radiation, obtained from (4), takes the form

$$R=\alpha_d(E3)\,|\beta|^2\frac{\Gamma_a/2\pi}{(\Delta-\Delta'-\hbar\omega)^2+(\Gamma_a/2)^2}, \tag{7}$$

where $\Delta'$ is the energy difference between the $6d$ and $5f$ atomic states. The sign in front of $\hbar\omega$ shows that the resonance occurs when there is stimulated emission of a laser photon by the atom, since $\Delta>\Delta'>0$. for the above particular level scheme of the uranium atom.

#### 4. RESULTS OF CALCULATIONS

We shall use the results reported in Refs. 5 and 10 to estimate the size of the above effect. Accordingly, we assume that the $5f_{5/2}\to 6s_{1/2}$ atomic transition energy is $\sim 69$ eV and that the energy difference $\Delta'$ between the $5f_{5/2}$ and $6d_{3/2}$ atomic levels is about 3 eV. Next, according to Ref. 10, the average value of the matrix element in (5), i.e., $\langle 5f_{5/2}|r^{-4}|6s_{1/2}\rangle$, evaluated by the Thomas-Fermi-Dirac or the Hartree-Fock-Slater method is $\sim 60a_0^{-4}$, where $a_0$ is the Bohr radius. The corresponding matrix element for the $5f_{7/2}$ state is smaller by two orders of magnitude,[10] so that the contribution of this state need not be taken into account.

The width $\Gamma_a$ of the atomic state is determined by radiative and Auger transitions, and by also the probability of ionization by laser radiation. The radiative width $\Gamma_r$ of the $6p_{1/2}\to 6s_{1/2}$ transition is $0.5\times 10^{10}$ s$^{-1}$, whereas the result for the $6p_{3/2}\to 6s_{1/2}$ transition is $\Gamma_r\approx 1.8\cdot 10^{10}$ s$^{-1}$ (Ref. 5). As far as we know, there are no published calculations of the Auger width $\Gamma_A$ for these transitions. However, if we use the general results for Auger processes[13,14] we find that they are of the order of the radiative probabilities. Moreover, the ionization of an atom significantly reduces the Auger transition probability even if it does not remove it altogether. Actually, sixfold ionization removes electrons from the outer $5f$, $7s$, and $6d$ shells, and the only energetically allowed processes are the near-threshold Coster-Kronig processes $P_{\rm I}P_{\rm II}P_{\rm III}$ (Ref. 15). In the case of tenfold ionization, the $6p_{3/2}$ shell disappears, and this removes the Auger transition problem and substantially reduces the radiative width. Nevertheless, for qualitative estimates, we assume that $\Gamma_r\approx\Gamma_A\approx 2\cdot 10^{10}$ s$^{-1}$.

There is no ionization broadening in the absence of laser radiation, and $\Gamma_a=\Gamma_r+\Gamma_A\approx 4\cdot 10^{10}$ s$^{-1}$. The decay of the nucleus is then possible via discrete conversion, i.e., by the $6s\to 5f$ excitation of an electron, followed by the decay of the $6s$ hole. In the case of a large resonance defect $\Delta\gg\Gamma_a$ we find from (4) that

$$R=\alpha_d(E3)\,\Gamma_a/2\pi\Delta^2. \tag{8}$$

Using the above numerical estimate, we find that $R\approx 2\cdot 10^{12}$, i.e., even in the absence of the laser field, this mechanism is more probable by twelve orders of magnitude than the direct radiative transition. However, it does not affect the nuclear decay because $R$ is the smaller than the internal conversion coefficient $\alpha(E3)$ by seven orders of magnitude.[10]

We note that, if the hole decays along the elastic channel, the process of discrete conversion (see the diagram in Fig. 1) becomes a special case of the electronic bridge when resonance in the intermediate state ensures that the main contribution is provided by one discrete level. In our case, however, the elastic channel is highly suppressed because it corresponds to the atomic $E3$ transition.

We now turn to discrete conversion in the laser field. For moderate intensities $I\sim 10^{12}$ W/cm$^2$, we can tune the laser frequency to resonance, $|\Delta-\Delta'-\hbar\omega|\ll\Gamma_a$, and find from (7) that

$$R=2\,\alpha_d(E3)\,|\beta|^2/\pi\Gamma_a. \tag{9}$$

The atomic matrix element in the expression for the parameter $\beta$ can be obtained in the quasiclassical approximation by the quantum defect method,[16] and is found to be $\sim 0.3a_0$. This takes into account the fact that such fields can produce multiphoton stripping of the two outer $7s$ electrons (see Ref. 17 which discusses this process in optical fields and the rare-earth atoms Ba, Sr, and Ca). Intensities of this order correspond to a field strength of about $2\times 10^7$ V/cm, so that $\beta\approx 3\cdot 10^{-3}$.

The ionization broadening of the $5f$ and $6d$ levels due to the multiphonon photoelectric effect $\Gamma_i$ is then much less than $\Gamma_r$ (see Ref. 17). The dynamic Stark level shift $\Delta_s$ is determined by the same matrix element as the parameter $\beta$:

$$\frac{\Delta_s}{\Delta}\approx|\beta|^2\left|\frac{\Delta}{\Delta'-\hbar\omega}\right|. \tag{10}$$

It is readily verified that this quantity can also be neglected.

We therefore have $R\approx 7\cdot 10^{18}$ in these fields, which is comparable with the conversion coefficient for the outer shells. Consequently, for laser intensities of a few terawatts per square centimeter, the lifetime of the nucleus $^{235m}$U is reduced by a factor of several units.

Of course, when laser pulses are employed, the "integrated" reduction in the lifetime is not as great. For standard laser pulses $\sim 10^{-8}$ s long, and pulse repetition frequencies of about 1 kHz, the relative reduction in the lifetime is about $10^{-5}$, which is, nevertheless, accessible to measurement by existing methods (see, for example, Ref. 18).

In stronger fields, it is much more difficult to perform such estimates. This is so, first, because multiply charged uranium ions may be formed by multiphoton ionization. Tenfold-ionized uranium atoms were observed in Ref. 19. They were produced by ultraviolet laser radiation of wavelength 193 nm, pulse length of about $10^{-11}$ s, and intensity $I\approx 10^{14}$ W/cm$^2$. The data reported in Ref. 19 can be used to estimate the ionization broadening $\Gamma_i$ of the $5f$ and $6d$ levels in which we are interested here. This is found to be $\sim 10^{10}$ s$^{-1}$, which is comparable with the radiative width reproduced above. For these intensities, $R\approx 7\cdot 10^{20}$. Moreover, ionization of the atom leads to a significant increase in the matrix element $m$ in (5), which explains the localization of the electron wave function near the nucleus of the ion as

compared with the case of the neutral atom. This effect is well known in the theory of internal conversion. It explains the increase in the internal conversion coefficient with increasing atomic number ($\sim Z^3$), and also with the increase in the energy of the conversion electron.[9] This is why the value of $R$ quoted above will in reality be even higher. Since this value of $R$ is much greater than the coefficient of ordinary conversion in the outer shells, the lifetime of $^{235m}$U will be reduced by two or three orders of magnitude for intensities on the order of $10^{14}$ W/cm$^2$.

As far as the Stark level shift in such fields is concerned, estimates based on (10) show that the resonant change in the relative frequency is then less than 5% However, we have to take into account the fact that, for pulsed laser radiation, and also for radiation with a finite aperture, the quasicrossing of the quasienergy atomic harmonics[20] ensures that practically any frequency within this 5% interval will give rise to discrete resonant conversion.

Unfortunately, it does not seem possible to extrapolate the above data to higher intensities because, at present, there is no generally accepted model for the formation of multiply charged ions, so that the width $\Gamma_i$ is difficult to estimate.

The calculations presented above do not take into account the renormalization of the tabulated internal conversion coefficients due to the electronic bridges. The point is that the electronic bridges produce a change only in the nuclear radiative-transition probability, and do not affect the conversion probability.[6,7] On the other hand, our aim in this paper has been to compare the probabilities of ordinary and discrete conversion. For this, it is sufficient to compare the corresponding internal conversion coefficients $\alpha$ and the discrete conversion factors $R$, and the result of the comparison does not depend on whether or not the electronic-bridge correction to the radiative nuclear-transition probability has been taken into account, since this correction appears in the same way in the expressions for $\alpha$ and for $R$.

## 5. CONCLUSION

Our main results may be summarized as follows.

1. Our estimates demonstrate the fact that, at least in principle, the rate of decay of $^{235m}$U to the ground state can be controlled by means of laser radiation.

2. This also applies to the reverse process, whereby the nucleus is excited during an electron transition.

3. The magnitude of the effect is a linear function of the laser intensity and $|\beta|^2$ up to $I_{\text{crit}} \approx 10^{14}$ W/cm$^2$.

It is important to note that we have examined the electron conversion process $6s \to 5f \to 6d$ which, according to Ref. 5, is the closest to resonance. Similar conclusions are obtained by considering other possibilities, for example, $6p \to 6d \to 5f$. A particular scheme for experimental investigation must be chosen on the basis of a given model, the possibilities of available laser facilities, and the energy of the electron transitions.

We note particularly that resonant conversion can be used as a method of producing vacancies in the atomic shell. As already noted, radiative filling of these vacancies can be exploited, at least in principle, for the generation of coherent radiation at the corresponding frequency.

There is a number of points that require further attention. This applies above all to the electron transition energies, and also to the matrix elements and widths of hole states in the neutral uranium atom and its ions.

Discrete resonant conversion in fields of intensity greater than $I_{\text{crit}}$ is of fundamental interest. We recall that $I_{\text{crit}}$ is determined by the condition that the ionization broadening of electron states excited during discrete conversion must be equal to their radiative width. The above value of $I_{\text{crit}}$ was deduced from experimental data reported in Ref. 19 and, as already noted, it does not seem possible to extrapolate the results to intensities $I > I_{\text{crit}}$.

Nevertheless, qualitative analysis suggests that the resonant conversion factor $R$ will reach maximum somewhere near $I \approx I_{\text{crit}}$. The reduction in this factor for $I > I_{\text{crit}}$ is due to the rapid (faster than linear) intensity dependence of ionization broadening.[17] Moreover, in the case of 13-fold ionization of uranium, the process of discrete resonant conversion ceases because both $6s$ electrons have already been removed.

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[1)] Real internal conversion coefficients can differ substantially from the tabulated values if the latter are much larger than unity (cf. remarks in the Introduction and in Sec. 4 in connection with electronic bridges).

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