Energetically forbidden internal conversion processes ignited by intense radiation fields

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Initially energetically forbidden internal conversion processes induced by a high intensity radiation field of appropriate frequency are discussed in the case of ^{99m}Tc and ^{105m}Ag. Hindering effects of saturation and power broadening are taken into consideration as well. Results of a recent paper are also disputed. [S0556-2813(98)02406-6]

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It is well established by now that the γ decay rate of radioactive isotopes cannot be altered directly by an external radiation field [1] of the highest intensity available nowadays. Some time ago, however, it was shown that, at least in theory, it is possible to block the process of internal conversion (IC) by removing electrons from those shells that significantly contribute to it [2]. Perhaps even more importantly, we later showed theoretically that the rate of IC can be modified—viz., it can be enhanced—in strong electromagnetic fields. If IC is originally forbidden energetically, i.e., if the transition energy of the nucleus simply is not enough to kick out tightly bound electrons, then in an intense radiation field the absorption of one or more photons makes it allowed for these electrons, as well. The theoretical background of this so-called induced internal conversion was investigated in several papers [3–5].

Here we discuss two types of induced IC. First, we investigate the *normal* IC process where the electron, assisted by an intense radiation field, makes a direct transition into a free state. In the second case, we consider the *discrete* IC process where the transition energy of the nucleus plus the energy of the absorbed photon(s) becomes resonant with an electronic transition, i.e., when it is equal to the energy difference of the binding energy of two atomic shells. In both of these cases we use the model of a previous paper by one of us [6]. Calculations are performed on the basis of this model and numerical results for the ^{99m}Tc and ^{105m}Ag isotopes are presented. In addition, we also give a brief review of this topic, pointing out the hindering effects of electron stripping [7], power broadening [8,9], and saturation [4,5,8]. Indeed, in order to reach a measurable effect, as it turns out, an extremely high photon flux is required, where these hindering effects become significant and neglecting them may lead to totally false numerical predictions. We compare the transition rates for various induced IC processes discussed in order to find the dominating one. We also deal with the huge numerical difference appearing between our results and a former one [10]. Finally, we consider the experimental possibilities in the light of our results.

First, following the formalism of Ref. [6], we employ a simple Coulomb form of electron-nucleus electromagnetic interaction and a one-electron hydrogenlike Hamiltonian. The S-matrix element of the process can be written as

$$S_{fi}=\frac{1}{i\hbar^2}\langle f|H_\alpha|i\rangle 2\pi\delta(\epsilon_{ab}+\hbar\omega-\Delta\epsilon). \tag{1}$$

Here ϵ_{ab} is the energy difference between the initial and final nuclear states, $\hbar\omega$ is the energy of the incoming photon, and $\Delta\epsilon>0$ denotes the energy difference of the energy eigenvalues of the atomic states. It can be given by $\Delta\epsilon=\epsilon_2-\epsilon_1$ in the discrete case and by $\Delta\epsilon=\varepsilon-\epsilon_1$ in the continuous case, where ϵ_1 is the energy eigenvalue of the electron shell from which the IC takes place, ϵ_2 is the energy eigenvalue of the excited shell, and $\varepsilon=q^2\hbar^2/2m$. In addition, m is the rest mass and q is the wave number of the outgoing free electron. The operator H_α has the form

$$H_\alpha=-\boldsymbol{\alpha}\cdot\nabla_{\mathbf{r}}\sum_{p=1}^{Z}\frac{e^2}{|\mathbf{r}-\mathbf{x}_p|}, \tag{2}$$

with

$$\boldsymbol{\alpha}=\frac{e}{m}i\sum_{\mathbf{k},\nu}\left(\frac{2\pi\hbar\omega}{V}\right)^{1/2}\frac{1}{\omega^2}a_{\mathbf{k},\nu}\boldsymbol{\varepsilon}_\nu. \tag{3}$$

Here e is the elementary charge, ω is the angular frequency of the external radiation field, V is the volume of normalization, $\boldsymbol{\varepsilon}_\nu$ is the polarization vector, and $a_{\mathbf{k},\nu}$ is the photon destruction operator of the quantized electromagnetic field. $\mathbf{r}$ denotes the electron and $\mathbf{x}_p$ the proton coordinates. Finally, Z is the number of protons in the nucleus. The final and initial states are $|f\rangle=|\mathbf{q}\rangle\otimes|b\rangle\otimes|0\omega,\mathbf{k},\nu\rangle$ and $|i\rangle=|1\rangle\otimes|a\rangle\otimes|1\omega,\mathbf{k},\nu\rangle$, where $|a\rangle$ and $|b\rangle$ represent the excited and deexcited states of the nucleus, respectively. $|\mathbf{q}\rangle$ is the free Coulomb state and $|1\rangle$ describes the bound state of the electron. The remainder correspond to the states of the quantized electromagnetic field, where $\mathbf{k}$ is the wave number vector, ν denotes the two states of polarization, and $0\omega,1\omega$ refer to the number eigenstates of photons with frequency ω.

In the following we briefly recall the necessary steps to derive the transition probability per unit time. Equation (2) is expanded in terms of $\mathbf{x}_p$ [see Eqs. (13) and (14) in [6]] and near the threshold, i.e., for very low kinetic energies, the radial part $R_{q,l}$ of the free Coulomb wave function $\langle\mathbf{x}|\mathbf{q}\rangle$ (p. 240 in [11]) is approximated [12] as $\lim_{q\to0}R_{q,l}$

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$=(4\pi q/r)^{1/2}J_{2l+1}[(8Zr/a_0)^{1/2}]$. Here J_{2l+1} is the Bessel function of the first kind of order $2l+1$.

Since we would like to compare eventually the rate of the induced process to the natural decay rate, we introduce a quasi cross section $\sigma^*=W_{fi}/[(c/V)T_\gamma]$, where $T_\gamma=\{8\pi(L+1)/L[(2L+1)!!]^2\hbar\}(\omega_{ab}/c)^{2L+1}B(EL,I_a\rightarrow I_b)$ denotes the transition probability per unit time of the electric multipole transition of multipolarity L [13], c is the velocity of light, $B(EL,I_a\rightarrow I_b)$ stands for the reduced transition probability associated with radiative transition of multipole order EL, and I_a and I_b are the initial and final angular momenta of the nucleus. The quantity σ^* has a dimension of cm^2 s. Multiplying it by the photon flux of the radiation field we obtain a dimensionless quantity (α_{ind}), which is the IC coefficient (ICC) of the induced process of the given shell. σ^* can be given as

$$\sigma_n^*=\frac{\alpha_f\hbar^2}{3m^2a_0}\frac{1}{\omega^3}\frac{L[(2L+1)!!]^2}{2L+1}(2l+1)\times\begin{pmatrix}L+1 & l_1 & l\\ 0 & 0 & 0\end{pmatrix}^2 I^2\left(\frac{\omega_{ab}}{c}\right)^{-(2L+1)}, \tag{4}$$

where $I=\int_0^\infty r^{3/2}J_{2l+1}[(8Zr/a_0)^{1/2}]r^{-L-2}R_{n_1,l_1}dr$. Here the shielding effect of the other electrons is taken into account by substituting for Z its effective value defined by $|\epsilon_1|=R_yZ^2/n_1^2$, where R_y is the Rydberg energy. α_f is the fine structure constant, a_0 is the Bohr radius, and R_{n_1,l_1} is the radial part of the atomic wave function of the initially bound electron.

Let us consider now the discrete IC process for ^{99m}Tc, investigated previously by others [10]. In order to carry out the calculation, we have to take into account the energy distribution of the excited electron state. For this reason we consider a Lorentzian line shape and in the final state we make the following substitution: $|\mathbf{q}\rangle\rightarrow|2\rangle$. Here $|2\rangle$ means the excited state of the electron with quantum numbers $n=6$ or greater typically. Adapting our formulas to this special case we get

$$\sigma_d^*=\alpha_f\hbar^4\frac{\pi}{3m^3a_0}\frac{1}{\omega^3}\frac{L[(2L+1)!!]^2}{2L+1}(2l_2+1)\times\begin{pmatrix}L+1 & l_1 & l_2\\ 0 & 0 & 0\end{pmatrix}^2\frac{(I_{l_1,l_2}^{n_1,n_2})^2(\omega_{ab}/c)^{-(2L+1)}\Gamma}{(\epsilon_{ab}+\epsilon_1+\hbar\omega-\epsilon_{20})^2+\Gamma^2/4}, \tag{5}$$

where l_1 and l_2 are the orbital quantum numbers of the electron shells, Γ is the width of the excited state, and ϵ_{20} is its central energy; $I_{l_1,l_2}^{n_1,n_2}=\int_0^\infty r^2R_{n_1,l_1}r^{-(L+2)}R_{n_2,l_2}dr$, where R_{n_2,l_2} is the radial part of $\langle\mathbf{x}|2\rangle$. The subscripts of σ^* refer to the type of induced IC.

At high intensities it is mandatory to take into account hindering effects such as stripping, saturation, and power broadening. Here we want just to recall the most important aspects that are necessary for our considerations. For more details see [3–5,9], where two types (bound and free Volkov) of dressed solution were used for electron states in intense radiation fields. Near the threshold we can approximate

$$\alpha_{\mathrm{ind}}=\alpha_{\mathrm{th}}T, \tag{6}$$

where $T=\Sigma_{N>\xi}T(b_N)$ with $T(b_N)=\int_0^1J_N^2(b_Nx,-d/4)dx$. The assisting radiation is treated as a linearly polarized laser field. $J_N(a,b)$ stands for the generalized Bessel functions. We have defined the following quantities: $b_N=eE_0q_N/m\omega^2=b_0(N+\xi)^{1/2}$, where E_0 stands for the amplitude of the electric field of the radiation field; $q_N=[(N+\xi)\hbar\omega/R_y]^{1/2}/a_0$; and $\xi=\Delta/\hbar\omega-d/2$ with $\Delta=\epsilon_1+\hbar\omega_{ab}$ and $d=e^2E_0^2/2m(\hbar\omega)^3$ where d is the so-called ponderomotive potential. Furthermore, $b_0=1.07\times10^{-6}\ I^{1/2}(\hbar\omega)^{-3/2}$, where I is the laser intensity in $\mathrm{W/cm}^2$ and $\hbar\omega$ is in units of eV. The number N represents the number of photons participating in the multiphoton process. We have introduced α_{th} which is the threshold ICC. This quantity can be considered as the ICC of a fictitious nucleus which could emit a gamma quantum the energy of which is just enough to lift the given electron into a free state without the help of any external photon. This quantity was calculated nonrelativistically in Eq. (18) of Ref. [5] as well as by others [14] on a fully relativistic basis.

By comparing α_{th} to the induced ICC of the examined shell, we may establish a criterion which can help us to determine the limits of validity of the first model. Namely, if $\alpha_{\mathrm{ind}}\simeq\alpha_{\mathrm{th}}$, i.e., if $T\simeq1$ is satisfied, the influence of saturation is about to become significant. Furthermore, the intensity where $T=1$ is satisfied is called threshold intensity I_{th}. If the intensity exceeds this level, our first, simple, model can no longer be used and one must employ Eq. (6), which can properly account for saturation. The behavior of the quantity T has been investigated in [4,15] where it was plotted vs b_0. (At high intensities the hindering effect of the ponderomotive potential has to be taken into account, too [15].)

If the intensity satisfies the condition $b_0\ll1$, i.e., if we have an intensity that is right below the threshold level, then we can use the small argument expressions of the Bessel functions. With this simplification we can write $T(b_K)=(b_K/2)^{2K}/[(K!)^2(2K+1)]$, where K denotes the minimum number of photons necessary to ignite the originally energetically forbidden process. This small intensity limit of Eq. (6) gives us a chance to compare it to the results of the upper derived *normal* process. By this way with $K=1$, we have obtained a reasonable agreement (in the same order) between the numerical results of the two completely different models at intensities which are far below the threshold level.

Let us now mention the effect of power broadening which gives rise to a strong hindering of induced *discrete* IC itself. Owing to the strong radiation field the width of the considered level is $\Gamma=\Gamma_0+\Gamma_{if}(I)$, where Γ_0 is the natural linewidth and $\Gamma_{if}(I)$ corresponds to the emerging laser intensity dependent broadening (for details see [9]). For intensities higher than $\sim10^{12}$ $\mathrm{W/cm}^2$ this intensity-dependent part can enhance the original natural linewidth, resulting in a decrease of the induced ICC [9].

Applying our formulas directly to ^{99m}Tc and ^{105m}Ag we estimate the requested flux that can give rise to a measurable

effect, as well as the intensity which is necessary to ''halve the lifetime'' of these isomers.

In the special case of ^{99m}Tc the metastable level has a lifetime of 6.01 h and decays by an $E3$ transition into a very-short-lived level. The released γ energy is 2.1726 keV [17]. Normally, IC is allowed from $3s$ and outer shells. We consider now the $2p_{1/2}$ (binding energy is 2676.9 eV) and the $2p_{3/2}$ shells (binding energy is 2793.2 eV). The energy defects, i.e., the applied photon energies, are 504.3 eV on the $2p_{1/2}$ shell and 620.6 eV on the $2p_{3/2}$ shell, respectively.

First we present numerical results for the *normal* case for ^{99m}Tc. With the aid of Eq. (4) we come to the final results $\sigma_n^*(\mathrm{Tc},2p_{3/2})=5.5\times10^{-30}$ cm^2 s and $\sigma_n^*(\mathrm{Tc},2p_{1/2})=3.6\times10^{-30}$ cm^2 s. To reach the value of $\alpha_{\mathrm{ind}}=1$ we need intensities of 1.5×10^{13} W/cm^2 and 2.8×10^{13} W/cm^2, respectively. For ''halving'' the metastable lifetime of ^{99m}Tc, one should reach $\alpha_{\mathrm{ind}}=\alpha_{\mathrm{tot}}=1.637\times10^7$ where α_{tot} is the total ICC. The corresponding intensities are $I_{1/2}(\mathrm{Tc},2p_{3/2})=2.5\times10^{20}$ W/cm^2 and $I_{1/2}(\mathrm{Tc},2p_{1/2})=4.6\times10^{20}$ W/cm^2 [16]. But at such high intensities one has to address the question of the appearance of hindering effects. The intensity that belongs to the threshold value is $I_{\mathrm{th}}=1.3\times10^{21}$ W/cm^2. We have that $I_{\mathrm{th}}>I_{1/2}$ and this is the very reason why we may accept the results obtained from our first, simple model and why it is sensible at all to speak about halving the lifetime. Although it appears as though we were able to halve the lifetime of isomers merely applying the required intensity, we have to be aware of the fact that at extremely high intensities the atoms get strongly ionized. This electron stripping causes the Tc atom to partially lose those electrons that contribute significantly to IC [8]. This is why halving remains a fiction in spite of our results above. Moreover, we have to mention that ^{99m}Tc represents a rather exceptional case. It is more typical that $\alpha_{\mathrm{th}}\ll\alpha_{\mathrm{tot}}$ and in these cases it is impossible to perform the halving because saturation sets in. Therefore if in a calculation of the intensity for halving the lifetime the hindering effects are arbitrarily neglected, one may get a totally false result.

To show the difficulties let us look at an example (^{105m}Ag) for this latter situation. Here the triggered transition is from the $1s$ shell which has an electron binding energy of 25.4140 keV and the $E3$ transition energy is 25.465 keV [17]. Thus the energy defect is about 51 eV. The corresponding result is $\sigma_n^*(\mathrm{Ag},1s)=1.7\times10^{-34}$ cm^2 s. To satisfy $\alpha_{\mathrm{ind}}=1$, an intensity of $I=4.8\times10^{16}$ W/cm^2 should be provided. The threshold intensity is $I_{\mathrm{th}}=1.3\times10^{18}$ W/cm^2 and α_{th} is less than 1000th of α_{tot}. This is why halving becomes totally meaningless in this case.

Next, we proceed with an estimation of *discrete* IC for ^{99m}Tc, using Eq. (5) as a starting point. Assuming $\Gamma_0=6.56\times10^{-3}$ eV (see [18]), $\epsilon_{20}=-8.2$ eV, the quantities that we are looking for are $\sigma_d^*(\mathrm{Tc},2p_{3/2})=5.5\times10^{-31}$ cm^2 s and $\sigma_d^*(\mathrm{Tc},2p_{1/2})=3.0\times10^{-31}$ cm^2 s at the resonant incoming photon energies of 496.1 eV and 612.4 eV, respectively. To obtain this result we considered only the dominant $n_2=6$ case. We also note that the normal process has a somewhat larger probability than the discrete one. The intensities required to reach $\alpha_{\mathrm{ind}}=1$ are $I(\mathrm{Tc},2p_{3/2})=1.4\times10^{14}$ W/cm^2 and $I(\mathrm{Tc},2p_{1/2})=3.3\times10^{14}$ W/cm^2, respectively.

One cannot but admit that the above intensities are extremely high and there is no monochromatic source in this energy range which can provide intensities in this range. Indeed, the recent x-ray lasers with photon energies of about 0.5 keV, needed for ^{99m}Tc, or about 51 eV, for ^{105m}Ag, are right below the requested intensities. Applying available laser sources multiphoton processes can only take place. But the larger the number of the photons participating in the process is, the larger is the intensity necessary to achieve the same effect [5]. Apart from lasers, however, there are other radiation sources that may provide the appropriate intensity. That might have been the fact which inspired others [10] to investigate the discrete IC process in a continuous spectrum, i.e., in synchrotron radiation. They claim that recent undulators are able to provide the intense radiation field required to halve the lifetime of ^{99m}Tc. Because we found their resulting intensities strikingly low, we decided to scrutinize their results entirely. So in the following, for the sake of a sensible comparison, we apply our results to the case of synchrotron radiation. As is customary in the terminology of synchrotrons we are going to determine the brilliance of the radiation needed, measured in photons/(cm^2 s mrad^2 0.1% BW), according to our resulting intensities. To attain this goal let us integrate the product of Eq. (5) and the spectral photon flux $J(\omega)$ over a range around the central energy of the resonance curve:

$$\alpha_{\mathrm{ind}}=\int_{\omega_0-\Delta\omega}^{\omega_0+\Delta\omega}\sigma_d^*(\omega)J(\omega)d\omega, \tag{7}$$

where $J(\omega)$ is measured in units of photons/(cm^2 s Hz) and we used $\hbar\Delta\omega=1$ eV. It is allowed to drag $J(\omega)$ out of the integration since the radiation spectra of synchrotrons can be considered nearly constant within the short interval we have used [19]. For the $2p_{3/2}$ shell we obtain $\int\sigma_d^*(\omega)d\omega=8.6\times10^{-18}$ cm^2. Because of the sharp edge of resonance, this value actually is not sensitive to changing the limits of the integral by a small amount. Calculating the brilliance that corresponds to our results we get $F(p_{3/2})=8.9\times10^{31}$ photons/(cm^2 s 0.1% BW). (It is obtained by integrating the brilliance over the whole solid angle instead of about 10^{-5} rad which is the source divergence of recent undulators [19].) In order to obtain this result we have used the 0.1% BW$=7.66\times10^{14}$ Hz relation. If one would like to halve the lifetime of ^{99m}Tc, about $F=1.5\times10^{39}$ photons/(cm^2 s 0.1% BW) should be provided. This value is several orders of magnitude higher than the flux of even the most recent undulator systems [19].

Comparing the final results of Ref. [10] to ours we have to recognize the huge numerical discrepancy between the two papers. Namely, on the basis of our calculation one needs a brilliance of about 14 orders greater for halving than the one predicted by [10], where the given final result is summed up for all the shells with principal quantum numbers $n\geqslant6$. But this difference between the two calculations cannot be held responsible for the hugh numerical disagreement [20]. Nevertheless, we note that synchrotrons and plasma radiation sources, as the authors of [10] pointed out properly, are worthy of attention.

In the light of the above results, we come to the conclusion that even the recently available radiation sources cannot

produce high enough intensities to reduce the lifetime of the discussed isomers significantly. Although halving the lifetime seems to be out of reach, synchrotrons may provide a promising prospect for this field. We claim that the available sources already are in a range that can give rise to measurable effects as compared to the decay rates due to the purely electromagnetic transition of these isomers. Thus, the effect can be observed experimentally by measuring the emitted x-ray line or the outgoing Auger electrons accompanying the induced IC process. The accompanying x-ray radiation is at about 5 Å for ^{99m}Tc and, much harder, at about ~ 0.5 Å for ^{105m}Ag. Fortunately, both are well measurable, e.g., by using an energy-dispersive setup with the appropriate semiconductor detector. Moreover, the energies of these x-ray transitions are well separable from the frequency of the incoming assistant radiation and differ from other lines that can be measured on these isomers ordinarily.

Although the high intensities, needed in a possible experimental realization, may cause a real difficulty, we are sure that practical tests of the above results are worth investigating, considering the experimental and theoretical benefits.

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