Induced Emission of Gamma Radiation from Isomeric Nuclei

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Abstract

We study the possibility to influence the lifetime of nuclear isomeric states with the aid of incident fluxes of photons. We assume that a nucleus initially in an isomeric state $|i\rangle$ first absorbs an incident photon of energy $E_{ni}$ to reach a higher intermediate state $|n\rangle$, then the state $|n\rangle$ decays to a lower state $|l\rangle$. In favorable cases the two-step induced emission rates become equal to the natural isomeric decay rates for incident power densities of the order of $10^{10}$ W cm$^{-2}$.

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1 Introduction

In a recent work it has been shown that the lifetime of isomeric nuclear states can be influenced by X-ray electron-nuclear double transitions (XENDT’s), which are processes in which a transition effected by an inner atomic electron takes place simultaneously with a nuclear electromagnetic transition [1]. The rate of deexcitation of isomeric nuclei induced
by XENDT’s was calculated for the case when the holes in the atomic shells are produced by incident ionizing electrons and it was found that the induced nuclear deexcitation rate becomes comparable to the natural decay rate for ionizing electron fluxes of the order of $10^{14}$ W cm$^{-2}$.

In this work we shall study the possibility to influence the lifetime of nuclear isomeric states with the aid of fluxes of gamma-ray photons. The induced deexcitation of the isomeric nucleus considered in this work is a two-step process. We assume that the nucleus initially in the isomeric state $|i\rangle$ first absorbs a photon of energy $E_{ni}$ so that the nucleus reaches the higher intermediate state $|n\rangle$. The state $|n\rangle$ then decays to a lower state $|l\rangle$ by the emission of a gamma-ray photon having the energy $E_{nl}$ or by internal conversion. In Sec. II we estimate the spectral intensities for which the single-photon induced emission rates become equal to the isomeric decay rates. These single-photon intensities turn out to be extremely large. In Sec. III we introduce the concept of two-step deexcitation of isomeric nuclei induced by incident photon fluxes, and estimate the spectral intensities for which the two-step induced emission rates become equal to the isomeric decay rates. In Sec. IV we list the nuclear isomers for which the required incident power density are within the reach of existing experimental techniques. In favorable cases the two-step induced emission rates become equal to the isomeric decay rates for incident power densities of the order of $10^{10}$ W cm$^{-2}$.

2 Single-photon induced gamma emission rates

We consider an isomeric state $|i\rangle$ of energy $E_i$ and assume that a nucleus in the state $|i\rangle$ decays to a lower state $|l\rangle$ of energy $E_l$. A photon of energy $E_{il}$ may be spontaneously emitted in this isomeric transition. In the presence of a beam of incident photons there will also be a certain rate for the induced emission of photons of energy $E_{il}$. In this section we estimate this single-photon induced gamma emission rate. We describe the incident photon flux by the spectral intensity $N(E)$, defined such that $N(E)dE$ should represent the number of photons incident per unit surface and time and having the energy between $E$ and $E + dE$.
The probability per second for the induced emission of a photon of energy $E$ is

$$w^{(1)}_i = \int_0^\infty \sigma_i(E)N(E)dE.$$  

(1)

The cross section for induced emission $\sigma_i(E)$ is

$$\sigma_i(E) = \frac{\pi c^2 \hbar^4 \Gamma_{il}(\Gamma_i + \Gamma_l)}{2E^2 (E - E_{il})^2 + \hbar^2(\Gamma_i + \Gamma_l)^2/4}.$$  

(2)

In Eq. (2) $\Gamma_{il}$ is the partial width for gamma-ray emission in the transition $|i\rangle \rightarrow |l\rangle$, $\Gamma_i = \ln 2/t_i$ is the natural isomeric decay rate, $t_i$ being the isomeric half-life, and $\Gamma_l$ is the total width of the state $|l\rangle$. If $|l\rangle$ is the ground state, then $\Gamma_l = 0$. The level widths are expressed in this work in units of s$^{-1}$. If the spectral intensity $N(E)$ is a slowly varying function of $E$ the single-photon induced emission rate becomes, from Eqs. (1) and (2)

$$w^{(1)}_i = \frac{\pi^2 c^2 \hbar^3}{E_{il}^2} \Gamma_{il}N(E_{il}).$$  

(3)

The half-life $t_i$ of the isomeric state is related to the width $\Gamma_{il}$ by

$$t_i = \frac{\ln 2 f_{IT}}{(1 + \alpha_{il})\Gamma_{il}},$$  

(4)

where $f_{IT}$ is the fraction of isomeric decays by gamma-ray emission or electron conversion and $\alpha_{il}$ is the internal conversion coefficient for the transition $|i\rangle \rightarrow |l\rangle$.

The single-photon induced-emission rate $w^{(1)}_i$ and the isomeric decay rate $\ln 2/t_i$ become equal for a spectral intensity

$$N_1(E_{il}) = \frac{(1 + \alpha_{il})E_{il}^2}{\pi^2 c^2 \hbar^3 f_{IT}}.$$  

(5)

Numerically we have

$$N_1(E_{il}) = 3.95 \times 10^{29} \frac{(1 + \alpha_{il})E_{il}^2}{f_{IT}},$$  

(6)

where $N_1$ is expressed in photons cm$^{-2}$s$^{-1}$eV$^{-1}$ and $E_{il}$ is expressed in keV. The single-photon spectral intensity in Eq. (6) is extremely large.
3 Two-step deexcitation of isomeric nuclei induced by incident photon fluxes

In this section we analyze the possibility of a two-step deexcitation of a nuclear isomeric state. The first step consists in the absorption by the nucleus in the isomeric state $|i\rangle$ of a photon having the energy $E_{ni}$, the nucleus thus making a transition to a higher intermediate state $|n\rangle$ of energy $E_n$. The state $|n\rangle$ then decays into a lower state $|l\rangle$ by the emission of a gamma-ray photon having the energy $E_{nl}$ or by internal conversion, as shown in Fig. 1. In some cases the state $|l\rangle$ may be situated above the isomeric state $|i\rangle$, and in these cases the transition $|n\rangle \rightarrow |l\rangle$ is followed by a further gamma-ray transition to a lower state $|l'\rangle$.

The multipolarity of the single-photon gamma-ray transition $|i\rangle \rightarrow |l\rangle$ is usually E3, M3, E4 or M4, while the multipolarity of the transitions $|i\rangle \rightarrow |n\rangle$, $|n\rangle \rightarrow |l\rangle$ may be lower. Then the rate of the induced two-step gamma transition $|i\rangle \rightarrow |n\rangle \rightarrow |l\rangle$ may be in some cases higher than the rate of the single-photon induced transition $|i\rangle \rightarrow |l\rangle$, for the same applied spectral intensity. The spin $J_n$ of the nuclear intermediate state $|n\rangle$ must be enclosed between the spins $J_i, J_l$ of the states $|i\rangle, |l\rangle$.

For a flux of incident photons of spectral intensity $N(E)$, the rate of the transition $|i\rangle \rightarrow |n\rangle$ is

$$w_{in}^{(1)} = \int_0^\infty \frac{2J_n + 1}{2J_l + 1} \frac{\Gamma_{ni}\Gamma_n}{2E^2 (E - E_{ni})^2 + 4\Gamma_n^2 \hbar^4 / 4^4} N(E) dE,$$

where $E_{ni} = E_n - E_i, \Gamma_n$ is the total width of the state $|n\rangle$, $\Gamma_n \gg \Gamma_i$, and $\Gamma_{ni}$ is the partial width of the electromagnetic transition $|n\rangle \rightarrow |i\rangle$. The total width $\Gamma_n$ of the intermediate state $|n\rangle$ which decays into the states $|i\rangle, |l\rangle$ and other lower states $|l'\rangle$ is given by

$$\Gamma_n = (1 + \alpha_{ni})\Gamma_{ni} + (1 + \alpha_{nl})\Gamma_{nl} + \sum_{l'} (1 + \alpha_{nl'})\Gamma_{nl'},$$

where $\alpha_{ni}, \alpha_{nl}, \alpha_{nl'}$ are the internal conversion coefficients for the transitions $|n\rangle \rightarrow |i\rangle, |n\rangle \rightarrow |l\rangle, |n\rangle \rightarrow |l'\rangle$. The rate $w_{il}^{(2)}$ for the two-step transition $|i\rangle \rightarrow |n\rangle \rightarrow |l\rangle$ is then

$$w_{il}^{(2)} = w_{in}^{(1)} (1 + \alpha_{nl})\Gamma_{nl}/\Gamma_n,$$

where $\Gamma_{nl}$ is the partial width of the electromagnetic transition $|n\rangle \rightarrow |l\rangle$. If $N(E)$ is a
slowly-varying function of energy, the two-step transition rate is

\[ w_{il}^{(2)} = \frac{2J_n + 1}{2J_i + 1} \frac{\pi^2 c^2 \hbar^3}{E_{ni}^2} (1 + \alpha_{nl}) \frac{\Gamma_{ni} \Gamma_{nl}}{\Gamma_n} N(E_{ni}). \quad (10) \]

The two-step transition rate is thus proportional to the effective width

\[ \Gamma_{eff} = (1 + \alpha_{nl}) \frac{\Gamma_{ni} \Gamma_{nl}}{\Gamma_n}. \quad (11) \]

The two-step induced rate \( w_{il}^{(2)} \) becomes equal to the isomeric decay rate \( \ln 2/t_i \) for a spectral intensity

\[ N_2(E_{ni}) = \frac{2J_i + 1}{2J_n + 1} \frac{E_{ni}^2}{\pi^2 c^2 \hbar^3} \frac{1}{F} \frac{t_n}{t_i}. \quad (12) \]

where

\[ F = (1 + \alpha_{nl}) \frac{\Gamma_{ni} \Gamma_{nl}}{\Gamma_n^2}. \quad (13) \]

In Eq. (13) the width \( \Gamma_n \) is related to the half-life \( t_n \) of the state \( |n\rangle \) by \( \Gamma_n = \ln 2/t_n \). We have \( 1/F \geq 4 \), but usually \( 1/F \gg 4 \).

The ratio \( N_2(E_{ni})/N_1(E_{ni}) \) is

\[ \frac{N_2(E_{ni})}{N_1(E_{il})} = \frac{2J_i + 1}{2J_n + 1} \frac{f_{FT}}{(1 + \alpha_{il})} \frac{E_{ni}^2}{E_{il}^2} \frac{1}{F} \frac{t_n}{t_i}. \quad (14) \]

Although usually \( 1/F \gg 4 \), we have however \( t_n/t_i \ll 1 \), and \( N_2/N_1 \ll 1 \). Thus, the two-step approach to the problem of induced gamma emission requires much lower incident power densities than the single-photon approach.

The number of photons per unit surface and time in the incident beam is of the order of \( N_2(E_{ni})E_{ni} \), and the power density, measured in W cm\(^{-2} \), is of the order of

\[ P_2 = N_2(E_{ni})E_{ni}^2. \quad (15) \]

We have numerically

\[ N_2(E_{ni}) = 3.95 \times 10^{29} \frac{2J_i + 1}{2J_n + 1} \frac{\Gamma_n^2}{(1 + \alpha_{nl}) \Gamma_{ni} \Gamma_{nl} t_i} \frac{t_n}{E_{ni}^2}, \quad (16) \]

\[ P_2 = 6.33 \times 10^{16} \frac{2J_i + 1}{2J_n + 1} \frac{\Gamma_n^2}{(1 + \alpha_{nl}) \Gamma_{ni} \Gamma_{nl} t_i} \frac{t_n}{E_{ni}^4}, \quad (17) \]

where \( N_2 \) is expressed in photons cm\(^{-2} \) s\(^{-1} \) eV\(^{-1} \), \( P_2 \) in W cm\(^{-2} \) and \( E_{ni} \) in keV. For \( \Gamma_n^2/\Gamma_{ni} \Gamma_{nl}=4 \), \( \alpha_{nl} = 0 \), \( t_n/t_i = 10^{-13} \), \( E_{ni} = 30 \) keV, \( J_i = J_n \) we have \( N_2 = 1.42 \times 10^{20} \) photons cm\(^{-2} \) s\(^{-1} \) eV\(^{-1} \),
and \( N_2 E_{ni} = 4.27 \times 10^{24} \text{photons cm}^{-2}\text{s}^{-1} \), which for \( E_{ni} = 30 \text{ keV} \) corresponds to \( N_2 E_{ni}^2 = 2.05 \times 10^{10} \text{ W cm}^{-2} \), which is not exceedingly high. However, as mentioned previously, the quantity \( \Gamma_n^2/(1 + \alpha_{nl})\Gamma_{ni}\Gamma_{nl} \) has values which in reality are much larger than the lower limit of 4.

4 Case study

We have determined the spectral intensity \( N_2 \) and the power level \( P_2 \) at which the induced emission rate becomes equal to the isomeric decay rate for isomeric nuclei having a half-life \( t_i > 10 \text{ min} \) and for which the cascade originating on the state \( |n\rangle \) contains a gamma-ray transition of energy \( E_\gamma > 2E_{ni} \). The latter relation represents a condition of upconversion of the energy of the incident photons. Some of the isomeric nuclei having these properties are listed in Table I. In the analysis of the nuclear properties we have used the Table of Isotopes \([2]\), and the internal conversion coefficients are taken from the BNL data base \([3]\) and from refs. \([4]\) and \([5]\). If \( t_n, t_i \) and the relative gamma-ray intensities \( R_{ni}, R_{nl}, R_{nl'} \) of the downward transitions from the state \( |n\rangle \) are all known, we have calculated \( N_2 \) and \( P_2 \) from Eqs. (12), (13) and (15), using for \( F \) the value

\[
F_R = \frac{(1 + \alpha_{nl})R_{ni}R_{nl}}{[(1 + \alpha_{ni})R_{ni} + (1 + \alpha_{nl})R_{nl} + \sum_{l'}(1 + \alpha_{nl'})R_{nl'}]^2}.
\]

Otherwise we have calculated \( N_2 \) and \( P_2 \) by using the Weisskopf estimates for the radiative widths of the transitions and the half-lives \( t_n, t_i \) appearing in Eqs. (12), (13) and (15). The error of the values of \( N_2 \) and \( P_2 \) calculated with the aid of the Weisskopf estimates is in general of about two orders of magnitude.

We see from Table I that the lowest values of \( P_2 \) are of the order of \( 10^{10} \text{ W cm}^{-2} \), while in most cases the gamma-ray power density \( P_2 \) is of the order of \( 10^{14} \text{ W cm}^{-2} \). In the case of the 6.8 h isomeric nucleus \(^{93}\text{Mo}\), a 4.8 keV photon generates a cascade in which an 1477 keV photon is emitted. In the case of the 3.5 h isomeric nucleus \(^{202}\text{Pb}\), a 38.6 keV photon generates a cascade in which a 960 keV photon is emitted. In the case of the 67.2 min isomeric nucleus \(^{204}\text{Pb}\), a 78.56 keV photon produces the emission of a 990 keV photon. In the case of the 141 y isomeric nucleus \(^{242}\text{Am}\), a 4.3 keV photon produces the emission of a...
52.9 keV photon. In the case of the 2.9 y isomeric nucleus $^{102}$Rh, a 13.7 keV photon produces the emission of a 112.5 keV photon. In the case of the 13.7 h isomeric nucleus $^{69}$Zn, a 92.7 keV photon produces the emission of a 531 keV photon. In the case of the 6.01 h isomeric nucleus $^{99}$Tc, a 38.4 keV photon produces the emission of a 181.0 keV photon. In the case of the 31 y isomeric nucleus $^{178}$Hf, an 126.1 keV photon generates a cascade in which a 574 keV photon is emitted.

The analysis has been restricted in this work to cases of upconversion, for which the cascade originating on the state $|n\rangle$ contains a gamma-ray photon of energy $E_\gamma > 2E_{ni}$. Moreover, the analysis has been restricted to isomeric nuclei for which there is a tabulated intermediate state $|n\rangle$ of known energy and known spin and parity. In the proximity of isomeric states there are also tabulated levels of known energy but unknown or uncertain spin or parity. The assignment of spins and parities and the discovery of new states may lead in time to the finding of isomeric nuclei with favorable intermediate states for which the power level $P_2$ should be lower than the present estimate of $10^{10}$ W cm$^{-2}$.

The power level $P_2$ required for the observation of induced gamma emission is large because the effective width $\Gamma_{eff}$ appearing in Eq. (11) is a very small fraction of the total width of the spectrum of pumping radiation. The use of small-area isomeric samples and of pulsed incident radiation may reduce the total energy of the incident pulse to practical values. An alternative to the broad-band pumping is the use of Mössbauer sources of gamma radiation. Considering an isomeric nucleus of proton number $Z$ and mass number $A$, the nuclei $(Z-1, A)$ or $(Z+1, A)$ may in certain cases populate the intermediate state $|n\rangle$ of the nucleus $(A, Z)$ by $\beta^-$ or respectively $\beta^+$, EC disintegrations. In this way, Mössbauer sources containing the nuclei $(Z-1, A)$ or $(Z+1, A)$ may produce photon fluxes of energy $E_{ni}$ in a narrow interval of width $\hbar \Gamma_n$ around $E_{ni}$. An example of this kind is $^{202}_{82}$Pb, whose 42 ns intermediate state at 2208.4 keV is populated by the decay of the nucleus $^{202}_{83}$Bi, which has a half-life of 1.72 h. On the other hand, while Mössbauer sources can produce large spectral intensities, they cannot produce pulses of radiation. The two-step transition rates obtained from the Mössbauer scattering by isomeric nuclei depends much on the practical details of the experiment.
5 Conclusions

The power levels required by the photon approach to the problem of induced gamma emission studied in this work are comparable to the power levels required by the XENDT approach described in [1]. The choice of isomeric nuclei was restricted in this work by the upconversion condition $E_\gamma > 2E_{ni}$, and it was restricted to intermediate states of known energy and known spin and parity. For the cases considered in this work, the required level of incident power for which the two-step emission rate becomes equal to the natural emission rate of isomeric nuclei is of the order of $10^{10}$ W cm$^{-2}$.

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Fig. 1. Paths for the deexcitation of a nuclear isomeric state. The two possibilities are the spontaneous deexcitation of the isomeric state $|i\rangle$ to a lower state $|l\rangle$, and the two-step deexcitation, when the nucleus first makes a transition from the isomeric state $|i\rangle$ to a higher nuclear state $|n\rangle$ by the absorption of a photon of energy $E_{ni}$, then it emits a photon to reach the lower nuclear state $|l\rangle$. 
Table I. Spectral intensity $N_2$ and power level $P_2$ for which the induced emission rate becomes equal to the natural emission rate from an isomeric nucleus. The isomeric level has the energy $E_i$ and the half-life $t_i$, the incident photons have the energy $E_{ni}$, $E_{nl}$ is the energy of the gamma-ray photon emitted from the state $|n\rangle$ and $E_\gamma$ is the largest photon energy in the cascade from the intermediate state $|n\rangle$. An R in the method column means that $N_2$ and $P_2$ have been calculated from the relative gamma-ray intensities and the measured $t_n$ and $t_i$, and a W in the method column means that $N_2$ and $P_2$ have been obtained from the Weisskopf estimates for the gamma-ray widths and half-lives.
| Nucleus | $E_i$, keV | $t_i$, y | $E_{ni}$, keV | $E_{nl}$, keV | $N_2$, ph./cm$^2$ s eV | $P_2$, W/cm$^2$ | Method | $E_\gamma/E_{ni}$ |
|---------|----------|--------|-------------|-------------|----------------|-----------------|--------|--------------|
| $^{186}$Re | 149.0 | 2.0×10$^5$ | 37.0 | 12.1 | 5.1×10$^{19}$ | 1.1×10$^{10}$ | W | 2.0 |
| $^{152}$Eu | 45.6 | 9.27 | 32.6 | 13.0 | 5.8×10$^{21}$ | 1.0×10$^{12}$ | R | 2.0 |
| $^{52}$Mn | 377.7 | 21.1 | 353.7 | 731.5 | 1.1×10$^{21}$ | 2.1×10$^{13}$ | R | 2.1 |
| $^{152}$Eu | 45.6 | 9.27 | 19.7 | 65.3 | 6.3×10$^{23}$ | 3.9×10$^{13}$ | R | 3.3 |
| $^{178}$Hf | 2446.1 | 31 | 126.1 | 140.3 | 4.4×10$^{22}$ | 1.1×10$^{14}$ | R | 4.6 |
| $^{96}$Tc | 34.3 | 51.5 | 11.1 | 45.3 | 6.8×10$^{24}$ | 1.3×10$^{14}$ | W | 4.1 |
| $^{44}$Sc | 271.1 | 58.6 | 78.7 | 349.8 | 2.4×10$^{23}$ | 2.4×10$^{14}$ | W | 4.4 |
| $^{202}$Pb | 2169.8 | 3.53 | 38.6 | 168.1 | 1.3×10$^{24}$ | 3.1×10$^{14}$ | W | 17.0 |
| $^{99}$Tc | 142.7 | 6.01 | 38.4 | 181.1 | 1.6×10$^{24}$ | 3.7×10$^{14}$ | R | 4.7 |
| $^{178}$Hf | 2446.1 | 31 | 126.1 | 437.0 | 1.5×10$^{23}$ | 3.9×10$^{14}$ | R | 4.2 |
| $^{99}$Tc | 142.7 | 6.01 | 38.4 | 40.6 | 2.2×10$^{24}$ | 5.2×10$^{14}$ | R | 3.7 |
| $^{119}$Te | 261.0 | 4.70 | 99.4 | 40.0 | 6.5×10$^{23}$ | 1.0×10$^{15}$ | W | 3.2 |
| $^{71}$Zn | 157.7 | 3.96 | 128.6 | 286.3 | 4.7×10$^{23}$ | 1.3×10$^{15}$ | W | 2.2 |
| $^{201}$Bi | 846.3 | 59.1 | 240.1 | 195.9 | 4.6×10$^{23}$ | 4.3×10$^{15}$ | R | 3.7 |
| $^{242}$Am | 48.6 | 141 | 4.3 | 52.9 | 2.1×10$^{27}$ | 6.1×10$^{15}$ | W | 12.4 |
| $^{93}$Mo | 2424.9 | 6.85 | 4.8 | 267.9 | 2.8×10$^{27}$ | 1.0×10$^{16}$ | W | 307.7 |
| $^{186}$Re | 149.0 | 2.0×10$^5$ | 37.0 | 86.6 | 9.5×10$^{25}$ | 2.1×10$^{16}$ | W | 2.3 |
| $^{69}$Zn | 438.6 | 13.7 | 92.7 | 531.2 | 2.2×10$^{25}$ | 3.0×10$^{16}$ | W | 5.7 |
| $^{84}$Rb | 463.6 | 20.2 | 151.4 | 615.0 | 9.1×10$^{24}$ | 3.4×10$^{16}$ | W | 4.1 |
| $^{204}$Pb | 2185.8 | 67.2 | 78.6 | 990.3 | 5.0×10$^{25}$ | 5.0×10$^{16}$ | R | 12.6 |
| $^{115}$In | 336.2 | 4.48 | 260.9 | 597.1 | 5.9×10$^{24}$ | 6.4×10$^{16}$ | W | 2.3 |
| $^{191}$Os | 74.4 | 13.1 | 57.6 | 131.9 | 1.6×10$^{26}$ | 8.5×10$^{16}$ | W | 2.3 |
| $^{96}$Tc | 34.3 | 51.5 | 14.9 | 49.2 | 2.5×10$^{27}$ | 9.0×10$^{16}$ | W | 3.3 |
| $^{102}$Rh | 140.8 | 2.9 | 13.7 | 112.5 | 9.4×10$^{27}$ | 2.8×10$^{17}$ | W | 8.2 |
two-step deexcitation

direct deexcitation