Induced emission of γ radiation from isomeric nuclei

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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 $|n\rangle$ and 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}. [S0556-2813(98)01606-9]

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I. 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 γ-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 γ-ray photon having 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 a 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 is 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}.

II. SINGLE-PHOTON INDUCED γ 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 γ 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 energy between E and $E+dE$.

The probability per second for the induced emission of a photon of energy E is

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

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

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

In Eq. (2), Γ_{il} is the partial width for γ-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 Γ_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),

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

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

$$t_i=\frac{\ln 2 f_{IT}}{(1+\alpha_{il})\Gamma_{il}}, \tag{4}$$

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

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

$$N_1(E_{il})=\frac{(1+\alpha_{il})E_{il}^2}{\pi^2c^2\hbar^3 f_{IT}}. \tag{5}$$

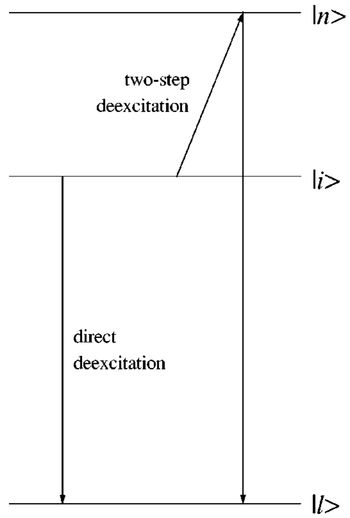

FIG. 1. Paths for the deexcitation of a nuclear isomeric state. The two possibilities are the spontaneous deexcitation of the isomeric state $|i\rangle$ to a lower state $|l\rangle$ and the two-step deexcitation; when the nucleus first makes a transition from the isomeric state $|i\rangle$ to a higher nuclear state $|n\rangle$ by the absorption of a photon of energy E_{ni}, it then emits a photon to reach the lower nuclear state $|l\rangle$.

Numerically we have

$$N_1(E_{il})=3.95\times 10^{29}\frac{(1+\alpha_{il})E_{il}^2}{f_{IT}}, \tag{6}$$

where N_1 is expressed in photons $\mathrm{cm}^{-2}\ \mathrm{s}^{-1}\ \mathrm{eV}^{-1}$ and E_{il} is expressed in keV. The single-photon spectral intensity in Eq. (6) is extremely large.

III. 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 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 γ-ray photon having 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 γ-ray transition to a lower state $|l'\rangle$.

The multipolarity of the single-photon γ-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 γ 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

$$w_{in}^{(1)}=\int_0^\infty\frac{2J_n+1}{2J_i+1}\frac{\pi c^2\hbar^4}{2E^2}\frac{\Gamma_{ni}\Gamma_n}{(E-E_{ni})^2+\hbar^2\Gamma_n^2/4}N(E)dE, \tag{7}$$

where $E_{ni}=E_n-E_i$, Γ_n is the total width of the state $|n\rangle$, $\Gamma_n\gg\Gamma_i$, and Γ_{ni} is the partial width of the electromagnetic transition $|n\rangle\rightarrow|i\rangle$. The total width Γ_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

$$\Gamma_n=(1+\alpha_{ni})\Gamma_{ni}+(1+\alpha_{nl})\Gamma_{nl}+\sum_{l'}(1+\alpha_{nl'})\Gamma_{nl'}, \tag{8}$$

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

$$w_{il}^{(2)}=w_{in}^{(1)}(1+\alpha_{nl})\Gamma_{nl}/\Gamma_n, \tag{9}$$

where Γ_{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

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

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

$$\Gamma_{\mathrm{eff}}=(1+\alpha_{nl})\Gamma_{ni}\Gamma_{nl}/\Gamma_n. \tag{11}$$

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

$$N_2(E_{ni})=\frac{2J_i+1}{2J_n+1}\frac{E_{ni}^2}{\pi^2c^2\hbar^3}\frac{1}{F}\frac{t_n}{t_i}, \tag{12}$$

where

$$F=(1+\alpha_{nl})\Gamma_{ni}\Gamma_{nl}/\Gamma_n^2. \tag{13}$$

In Eq. (13) the width Γ_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 $N_2(E_{ni})/N_1(E_{nl})$ is

$$N_2(E_{ni})/N_1(E_{il})=\frac{2J_i+1}{2J_n+1}\frac{f_{IT}}{(1+\alpha_{il})}\frac{E_{ni}^2}{E_{il}^2}\frac{1}{F}\frac{t_n}{t_i}. \tag{14}$$

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 γ 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

$$P_2=N_2(E_{ni})E_{ni}^2. \tag{15}$$

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 energy E_i and half-life t_i, the incident photons have energy E_{ni}; E_{nl} is the energy of the γ-ray photon emitted from the state $|n\rangle$ and E_γ 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 γ-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 γ-ray widths and half-lives.

Nucleus	E_i (keV)	t_i	E_{ni} (keV)	E_{nl} (keV)	N_2 [photons/(cm^2 s eV)]	P_2 (W/cm^2)	Method	E_γ/E_{ni}
^{186}Re	149.0	2.0×10^5 yr	37.0	12.1	5.1×10^{19}	1.1×10^{10}	W	2.0
^{152}Eu	45.6	9.27 h	32.6	13.0	5.8×10^{21}	1.0×10^{12}	R	2.0
^{52}Mn	377.7	21.1 min	353.7	731.5	1.1×10^{21}	2.1×10^{13}	R	2.1
^{152}Eu	45.6	9.27 h	19.7	65.3	6.3×10^{23}	3.9×10^{13}	R	3.3
^{178}Hf	2446.1	31 yr	126.1	140.3	4.4×10^{22}	1.1×10^{14}	R	4.6
^{96}Tc	34.3	51.5 min	11.1	45.3	6.8×10^{24}	1.3×10^{14}	W	4.1
^{44}Sc	271.1	58.6 h	78.7	349.8	2.4×10^{23}	2.4×10^{14}	W	4.4
^{202}Pb	2169.8	3.53 h	38.6	168.1	1.3×10^{24}	3.1×10^{14}	W	17.0
^{99}Tc	142.7	6.01 h	38.4	181.1	1.6×10^{24}	3.7×10^{14}	R	4.7
^{178}Hf	2446.1	31 yr	126.1	437.0	1.5×10^{23}	3.9×10^{14}	R	4.2
^{99}Tc	142.7	6.01 h	38.4	40.6	2.2×10^{24}	5.2×10^{14}	R	3.7
^{119}Te	261.0	4.70 d	99.4	40.0	6.5×10^{23}	1.0×10^{15}	W	3.2
^{71}Zn	157.7	3.96 h	128.6	286.3	4.7×10^{23}	1.3×10^{15}	W	2.2
^{201}Bi	846.3	59.1 min	240.1	195.9	4.6×10^{23}	4.3×10^{15}	R	3.7
^{242}Am	48.6	141 yr	4.3	52.9	2.1×10^{27}	6.1×10^{15}	W	12.4
^{93}Mo	2424.9	6.85 h	4.8	267.9	2.8×10^{27}	1.0×10^{16}	W	307.7
^{186}Re	149.0	2.0×10^5 yr	37.0	86.6	9.5×10^{25}	2.1×10^{16}	W	2.3
^{69}Zn	438.6	13.7 h	92.7	531.2	2.2×10^{25}	3.0×10^{16}	W	5.7
^{84}Rb	463.6	20.2 min	151.4	615.0	9.1×10^{24}	3.4×10^{16}	W	4.1
^{204}Pb	2185.8	67.2 min	78.6	990.3	5.0×10^{25}	5.0×10^{16}	R	12.6
^{115}In	336.2	4.48 h	260.9	597.1	5.9×10^{24}	6.4×10^{16}	W	2.3
^{191}Os	74.4	13.1 h	57.6	131.9	1.6×10^{26}	8.5×10^{16}	W	2.3
^{96}Tc	34.3	51.5 min	14.9	49.2	2.5×10^{27}	9.0×10^{16}	W	3.3
^{102}Rh	140.8	2.9 yr	13.7	112.5	9.4×10^{27}	2.8×10^{17}	W	8.2

We have numerically

$$N_2(E_{ni})=3.95\times10^{29}\frac{2J_i+1}{2J_n+1}\frac{\Gamma_n^2}{(1+\alpha_{nl})\Gamma_{ni}\Gamma_{nl}}\frac{t_n}{t_i}E_{ni}^2, \quad (16)$$

$$P_2=6.33\times10^{16}\frac{2J_i+1}{2J_n+1}\frac{\Gamma_n^2}{(1+\alpha_{nl})\Gamma_{ni}\Gamma_{nl}}\frac{t_n}{t_i}E_{ni}^4, \quad (17)$$

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, and $J_i=J_n$ we have $N_2=1.42\times10^{20}$ photons cm^{-2} s^{-1} eV^{-1}, and $N_2E_{ni}=4.27\times10^{24}$ photons cm^{-2} s^{-1}, which for $E_{ni}=30$ keV corresponds to $N_2E_{ni}^2=2.05\times10^{10}$ 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.

IV. 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 $t_i>10$ min and for which the cascade originating on the state $|n\rangle$ contains a γ-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 database [3] and from Refs. [4] and [5]. If t_n, t_i and the relative γ-ray intensities R_{ni}, R_{nl}, and $R_{nl'}$ 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

$$F_R=\frac{(1+\alpha_{nl})R_{ni}R_{nl}}{[(1+\alpha_{ni})R_{ni}+(1+\alpha_{nl})R_{nl}+\Sigma_{l'}(1+\alpha_{nl'})R_{nl'}]^2}. \quad (18)$$

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} W cm^{-2}, while in most cases the γ-ray power density P_2 is of the order of 10^{14} W cm^{-2}. In the case of the 6.8 h isomeric nucleus ^{93}Mo, a 4.8 keV photon generates a cascade in which a 1477 keV photon is emitted. In the case of the 3.5 h isomeric nucleus ^{202}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}Pb, a 78.56 keV photon produces the emission of a 990 keV photon. In the case of the 141 yr isomeric nucleus ^{242}Am, a 4.3 keV photon produces the emission of a 52.9 keV photon. In the case of the 2.9 yr 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 yr isomeric nucleus ^{178}Hf, a 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 γ-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 γ emission is large because the effective width $\Gamma_{\rm eff}$ 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 broadband pumping is the use of Mössbauer sources of γ radiation. Considering an isomeric nucleus of proton number Z and mass number A, the nucleus $(Z-1,A)$ or $(Z+1,A)$ may in certain cases populate the intermediate state $|n\rangle$ of the nucleus (A,Z) by β^- or, respectively, β^+, electron capture (EC) disintegrations. In this way, Mössbauer sources containing the nucleus $(Z-1,A)$ 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 depend much on the practical details of the experiment.

V. CONCLUSIONS

The power levels required by the photon approach to the problem of induced γ 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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