Power densities for two-step gamma-ray transitions from isomeric states

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Abstract

We have calculated the incident photon power density $P_2$ for which the two-step induced emission rate from an isomeric nucleus becomes equal to the natural isomeric decay rate. We have analyzed two-step transitions for isomeric nuclei with a half-life greater than 10 min, for which there is an intermediate state of known energy, spin and half-life, for which the intermediate state is connected by a known gamma-ray transition to the isomeric state and to at least another intermediate state, and for which the relative intensities of the transitions to lower states are known. For the isomeric nucleus $^{166m}$Ho, which has a 1200 y isomeric state at 5.98 keV, we have found a value of $P_2 = 6.3 \times 10^7$ W cm$^{-2}$, the intermediate state being the 263.8 keV level. We have found power densities $P_2$ of the order of $10^{10}$ W cm$^{-2}$ for several other isomeric nuclei.

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The induced deexcitation of an isomeric nucleus is a two-step process in which the nucleus, initially in the isomeric state |i⟩, first absorbs a photon of energy $E_{ni}$ to reach a higher intermediate state |n⟩, then the nucleus makes a transition to a lower state |l⟩ by the emission of a gamma-ray photon of energy $E_{nl}$ or by internal conversion. In a previous work [1] it has been required that the cascade originating on the state |n⟩ should contain a gamma-ray transition of energy $E_\gamma > 2E_{ni}$, the latter relation representing a condition of upconversion of the energy of the incident photons, and it was found that 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$^{-2}$. The Weisskopf estimates where used in [1] in cases when tabulated half-lives or partial widths were not available.

In this report we have analyzed, regardless of the upconversion condition $E_\gamma > 2E_{ni}$, the two-step transitions for isomeric nuclei with a half-life greater than 10 min, for which there is an intermediate state |n⟩ of known energy, spin and half-life, for which the intermediate state |n⟩ is connected by a known gamma-ray transition to the isomeric state and to a lower state |l⟩, and for which the relative intensities of the transitions from the intermediate state |n⟩ are known. [2]

We have calculated the incident power density $P_2$ for which the two-step induced emission rate becomes equal to the natural isomeric decay rate. We have found a value of $P_2 = 6.3 \times 10^7$ W cm$^{-2}$ for $^{166}$Ho, which has a 1200 y isomeric state at $E_i=5.98$ keV, and the intermediate state is the $E_n=263.8$ keV level, so that $E_{ni} = 257.8$ keV. We have also found several isomers for which the power density $P_2$ is of the order of $10^{10}$ W cm$^{-2}$.

We assume that the nucleus is initially in an isomeric state |i⟩ of energy $E_i$, spin $J_i$ and half-life $t_i$. By absorbing an incident photon of energy $E_{ni}$, the nucleus makes a transition to a higher intermediate state |n⟩ of energy $E_n$, spin $J_n$ and half-life $t_n$. The state |n⟩ then decays into a lower state |l⟩ of energy $E_l$ and spin $J_l$ 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⟩ may be situated above the isomeric state |i⟩, and in these cases the transition |n⟩ → |l⟩ is followed by further gamma-ray transitions to lower states.

As shown in [2], the rate $w_{il}^{(2)}$ for the two-step transition |i⟩ → |n⟩ → |l⟩ is

$$
w_{il}^{(2)} = \sigma_{ni} \hbar \Gamma_{eff} N(E_{ni}).
$$  (1)
where
\[
\sigma_{ni} = \frac{2J_n + 1 - \pi^2 c^2 \hbar^2}{2J_i + 1} E_{ni}^2
\]  
(2)
is the induced-emission cross section for the transition \(|i\rangle \rightarrow |n\rangle\). The quantity \(\Gamma_{eff}\) is the effective width of the two-step transition,
\[
\Gamma_{eff} = F_R \ln 2/t_n,
\]  
(3)
where the dimensionless quantity \(F_R\) has the expression
\[
F_R = \frac{(1 + \alpha_{ni})R_{ni}R_{nl}}{[(1 + \alpha_{ni})R_{ni} + (1 + \alpha_{nl})R_{nl} + \sum_{l'}(1 + \alpha_{nl'})R_{nl'}]^2},
\]  
(4)
\(R_{ni}, R_{nl}, R_{nl'}\) being the relative gamma-ray intensities and \(\alpha_{ni}, \alpha_{nl}, \alpha_{nl'}\) the internal conversion coefficients for the transitions \(|n\rangle \rightarrow |i\rangle, |n\rangle \rightarrow |l\rangle, |n\rangle \rightarrow |l'\rangle, l' \neq i, l\). In Eq. (2) \(N(E_{ni})\) is the spectral intensity for the incident photon flux, 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 spectral intensity \(N_2\) for which the two-step transition rate \(w_{il}^{(2)}\) becomes equal to the natural decay rate \(\ln 2/t_i\) of the isomeric nucleus is
\[
N_2 = \frac{\ln 2}{\sigma_{int} t_i},
\]  
(5)
where the integrated cross section \(\sigma_{int}\) is
\[
\sigma_{int} = \sigma_n \hbar \Gamma_{eff}.
\]  
(6)
The incident power density \(P_2\) for which the induced emission rate becomes equal to the natural decay rate of the isomeric state can then be estimated as
\[
P_2 = N_2(E_{ni}) E_{ni}^2.
\]  
(7)

We have analyzed two-step transitions for isomeric nuclei with a half-life greater than 10 min, for which there is an intermediate state \(|n\rangle\) of known energy \(E_n\), spin \(J_n\) and half-life \(t_n\), for which the intermediate state is connected by a known gamma-ray transition to the isomeric state \(|i\rangle\) and to a lower state \(|l\rangle\), and for which the relative intensities \(R_{ni}, R_{nl}, R_{nl'}\) of the transitions to lower states are known. The internal conversion coefficients have been calculated by interpolation from refs. [3] and [4]. The cases for which all the previously mentioned quantities were available in
ref. [2] are listed in Table I. If the input values were available for several two-step transitions of a certain isomeric nucleus, we gave the two-step transition having the lowest value of $P_2$. The two-step transitions for the nuclei $^{52}$Mn, $^{99}$Tc, $^{152}$Eu, $^{178}$Hf, $^{201}$Bi, $^{204}$Pb, which fulfil the upconversion condition, have been studied in ref. [1] and are not included in Table I. If the half-life $t_n$ of the intermediate state was given in [2] as less than ($<$) or greater than ($>$) a certain value, we have calculated $\hbar \Gamma_{\text{eff}}$, $\sigma_{\text{int}}$ and $P_2$ corresponding to the given limiting value of $t_n$. In the case of $^{166}$Ho we have neglected the 3.1 keV transition, of unknown intensity, from the 263.8 keV level. In the case of $^{97}$Tc the intensity of the 441.2 keV transition from the 656.9 keV level was given as an upper limit. In the case of $^{121}$Sn we have neglected the weak 56.35 keV transition from the 925.6 keV level. The half-life of the 0.0768 keV isomeric level of $^{235}$U and the intensity of the 637.7 keV transition from the 637.79 keV intermediate state are approximate values. In the case of $^{34}$Cl the intensity of the 2433.8 keV transition from the 2580.2 keV intermediate state was given as an upper limit, as are the intensities of some other transitions from this intermediate state. In the case of $^{123}$Sn we have neglected the weak 284.7 keV transition from the 1130.5 keV intermediate state.

For the isomeric nucleus $^{166m}$Ho, which has a 1200 y isomeric state at $E_i$=5.98 keV, we have found a value of $P_2 = 6.3 \times 10^7$ W cm$^{-2}$, the intermediate state being the $E_n$=263.8 keV level. The relatively low value of $P_2$ is due to the long half-life of this isomer. The largest effective width in Table I is for $^{34m}$Cl, for which $\hbar \Gamma_{\text{eff}} = 2.5 \times 10^{-3}$ eV. The relatively large effective width is due to the short half-life of the intermediate state, $t_n$=3 fs. The energy of the pumping transition is however large, $E_{ni}$=2433.8 keV, and the multipolarity is E2/M1. The large value of $P_2$ for $^{34}$Cl is due to the relatively small half-life of the isomeric state. There are several cases in Table I for which the power density $P_2$ is of the order of $10^{10}$ W cm$^{-2}$. For $^{97}$Tc and $^{95}$Tc the low values of $P_2$ can be attributed to the short half-lives of the intermediate state, whereas for $^{113}$Cd and $^{121}$Sn the values of $P_2$ are a result of the long isomeric half-lives. The two-step transitions for which all the input values were available represent a small fraction of the total number of possible two-step transitions in isomeric nuclei.

Since the number of isomeric nuclei in a sample is limited by the total activity of that sample, a longer isomeric half-life means a larger number of isomeric nuclei in the sample. The two-step induced transitions can be induced by incident photons, as assumed in this work, or directly by
incident electrons. The cross sections for the two-step electron excitation of isomeric states are about two orders of magnitudes lower than the cross sections for two-step photon excitation but the two-step photon excitation rates are also proportional to the efficiency with which the energy of an incident electron beam can be converted into bremsstrahlung.
References

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Fig. 1. Two-step deexcitation of an isomeric nucleus. The nucleus, initially in the isomeric state $|i\rangle$, makes a transition to a higher nuclear state $|n\rangle$ by the absorption of an incident photon of energy $E_{ni}$, then it emits a photon of energy $E_{nl}$ to reach the lower nuclear state $|l\rangle$.

Table I. Power density $P_2$ for which the two-step induced emission rate becomes equal to the natural decay rate of the isomeric state of energy $E_i$ and half-life $t_i$. The intermediate state of the problem is the level of energy $E_n$ and half-life $t_n$. The multipolarities of the transitions of energy $E_{ni}, E_{nl}$ are given in the multipole column.
| Nucleus | $E_i$ (keV) | $t_i$ (s) | $E_n$ (keV) | $t_n$ (s) | $E_{ni}$ (keV) | $E_{nl}$ (keV) | multipole $ni/nl$ | $F_R$ | $h\Gamma_{eff}$ (eV) | $\sigma_{int}$ (cm$^2$ eV) | $P_2$ (W cm$^{-2}$) |
|---------|----------|---------|----------|---------|------------|------------|-----------------|-----|-----------------|-----------------|----------|
| $^{166}$Ho | 6.0 | $3.8 \times 10^{10}$ | 263.8 | $<5.0 \times 10^{-10}$ | 257.8 | 72.9 | M2/E2 | 8.0 $\times 10^{-2}$ | 7.3 $\times 10^{-8}$ | 3.1 $\times 10^{-27}$ | 6.3 $\times 10^{7}$ |
| $^{97}$Tc | 96.6 | 7.8 $\times 10^6$ | 656.9 | $>7.6 \times 10^{-13}$ | 560.3 | 441.2 | E2/E1 | 2.8 $\times 10^{-2}$ | 1.7 $\times 10^{-5}$ | 6.1 $\times 10^{-25}$ | 7.3 $\times 10^{9}$ |
| $^{95}$Tc | 38.9 | 5.3 $\times 10^6$ | 927.8 | $>5.9 \times 10^{-13}$ | 888.9 | 301.1 | E1/M1 | 1.4 $\times 10^{-1}$ | 1.1 $\times 10^{-4}$ | 1.0 $\times 10^{-24}$ | 1.6 $\times 10^{10}$ |
| $^{113}$Cd | 263.6 | 4.4 $\times 10^8$ | 522.3 | $3.2 \times 10^{-10}$ | 258.8 | 206.4 | E2/E1 | 1.1 $\times 10^{-2}$ | 1.6 $\times 10^{-8}$ | 5.9 $\times 10^{-28}$ | 2.8 $\times 10^{10}$ |
| $^{121}$Sn | 6.3 | 1.7 $\times 10^9$ | 925.6 | $2.5 \times 10^{-10}$ | 919.3 | 925.6 | M2/E2 | 3.7 $\times 10^{-2}$ | 6.7 $\times 10^{-8}$ | 2.0 $\times 10^{-28}$ | 2.7 $\times 10^{11}$ |
| $^{58}$Co | 24.9 | 3.3 $\times 10^4$ | 373.9 | $6.0 \times 10^{-13}$ | 349.1 | 320.8 | M1/M1 | 3.2 $\times 10^{-2}$ | 2.4 $\times 10^{-5}$ | 7.6 $\times 10^{-25}$ | 5.4 $\times 10^{11}$ |
| $^{83}$Kr | 41.5 | 6.6 $\times 10^3$ | 562.0 | $6.0 \times 10^{-12}$ | 520.4 | 552.6 | E2/E1 | 1.9 $\times 10^{-1}$ | 1.5 $\times 10^{-5}$ | 6.2 $\times 10^{-25}$ | 7.4 $\times 10^{12}$ |
| $^{131}$Xe | 163.9 | 1.0 $\times 10^6$ | 666.9 | $<5.0 \times 10^{-10}$ | 503.0 | 325.8 | E2/M1 | 2.4 $\times 10^{-1}$ | 2.2 $\times 10^{-7}$ | 2.2 $\times 10^{-27}$ | 1.2 $\times 10^{13}$ |
| $^{189}$Os | 30.8 | 2.1 $\times 10^4$ | 219.4 | $1.9 \times 10^{-10}$ | 188.6 | 219.4 | M1/E2 | 4.1 $\times 10^{-2}$ | 9.9 $\times 10^{-8}$ | 8.6 $\times 10^{-27}$ | 2.2 $\times 10^{13}$ |
| $^{119}$Sn | 89.5 | 2.5 $\times 10^7$ | 787.0 | $1.9 \times 10^{-10}$ | 697.4 | 763.1 | M2/E2 | 4.8 $\times 10^{-3}$ | 1.2 $\times 10^{-8}$ | 6.1 $\times 10^{-29}$ | 3.5 $\times 10^{13}$ |
| $^{235}$U | 0.0768 | 1.5 $\times 10^3$ | 637.79 | $1.3 \times 10^{-11}$ | 637.72 | 637.79 | E1/E2 | 1.2 $\times 10^{-1}$ | 4.1 $\times 10^{-6}$ | 7.8 $\times 10^{-26}$ | 3.9 $\times 10^{14}$ |
| $^{34}$Cl | 146.4 | 1.9 $\times 10^3$ | 2580.3 | $<3 \times 10^{-15}$ | 2433.8 | 2580.2 | E2/M1 | 1.7 $\times 10^{-2}$ | 2.5 $\times 10^{-3}$ | 7.0 $\times 10^{-25}$ | 4.9 $\times 10^{14}$ |
| $^{125}$Te | 144.8 | 5.0 $\times 10^6$ | 642.2 | $<6.0 \times 10^{-10}$ | 497.4 | 606.7 | M2/E2 | 1.3 $\times 10^{-3}$ | 9.7 $\times 10^{-10}$ | 1.0 $\times 10^{-29}$ | 5.5 $\times 10^{14}$ |
| $^{117}$Sn | 314.6 | 1.2 $\times 10^6$ | 711.5 | 9.8 $\times 10^{-10}$ | 396.6 | 553.0 | M2/E2 | 1.7 $\times 10^{-3}$ | 8.0 $\times 10^{-10}$ | 1.3 $\times 10^{-29}$ | 1.1 $\times 10^{15}$ |
| $^{87}$Sr | 388.5 | 1.0 $\times 10^4$ | 2414.5 | 1.3 $\times 10^{-13}$ | 2026.0 | 643.8 | M1/E1 | 5.7 $\times 10^{-3}$ | 2.0 $\times 10^{-5}$ | 3.8 $\times 10^{-26}$ | 1.2 $\times 10^{15}$ |
| $^{124}$Sb | 36.8 | 1.2 $\times 10^3$ | 125.2 | 8.6 $\times 10^{-8}$ | 88.4 | 37.6 | E2/E2 | 5.1 $\times 10^{-2}$ | 2.7 $\times 10^{-10}$ | 1.0 $\times 10^{-28}$ | 7.0 $\times 10^{15}$ |
| $^{123}$Sn | 24.6 | 2.4 $\times 10^3$ | 1155.0 | $<1.0 \times 10^{-10}$ | 1130.5 | 536.4 | E2/E1 | 1.4 $\times 10^{-2}$ | 6.2 $\times 10^{-8}$ | 3.7 $\times 10^{-28}$ | 1.6 $\times 10^{17}$ |
| $^{103}$Rh | 39.8 | 3.4 $\times 10^3$ | 650.1 | $<1.0 \times 10^{-10}$ | 610.3 | 292.7 | M1/E1 | 3.8 $\times 10^{-4}$ | 1.7 $\times 10^{-9}$ | 1.3 $\times 10^{-29}$ | 9.2 $\times 10^{17}$ |
| $^{117}$In | 315.3 | 7.0 $\times 10^3$ | 1891.9 | $4.2 \times 10^{-10}$ | 1576.6 | 840.2 | E1/E2 | 8.6 $\times 10^{-3}$ | 9.4 $\times 10^{-9}$ | 1.4 $\times 10^{-29}$ | 2.7 $\times 10^{18}$ |