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Remarks on the trigger induction of the radiative decay of metastable states of isomeric nuclei

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***Abstract*. The conditions of known experiments on the trigger acceleration of the radiative decay of isomeric nuclear states are compared. It is shown that in the case of quasi-monochromatic inducing sources, the use of Mössbauer targets can increase the efficiency of the process by a few orders of magnitude. It is pointed out that the asymptotic behaviour of the current value of the absorption cross section requires the comparability of the exposure time with upper-level lifetime of the transition.**

Keywords: *metastable state, isomeric nuclei, forced decay.*

1. Numerous experimental efforts aimed at the acceleration of the decay of long-lived metastable nuclear states – one of the central directions in modern quantum nucleonics, involve the use of the external trigger induction of anti-Stokes transitions in isomeric nuclei in solid targets (see, for example, comprehensive analytic review [1]). This two-step process involves absorption of an external gamma quant and the transition of a nucleus from a metastable (m) to the upper-lying auxiliary state (a) accompanied by a rapid spontaneous decay of the latter. The most popular and virtually almost the only object of experiments is the $^{178\mathrm{m}}$Hf isomer with the metastable-state lifetime of 31 years.

This process is not apparently principally forbidden by fundamental physical laws. Therefore, taking into account the opinion stated sometimes [1] that the results of known experiments are still not convincing, it is useful to consider at least some possible reasons for their quantitative inadequacy. The simplest remarks presented below are especially actual because the X-ray sources used in experiments (for example, from a stomatology X-ray apparatus [2] to relativistic undulator sources [3]) drastically differ from each other in the photon flux density Φ (cm^{-2} s^{-1}), its spectral density $\mathrm{d}\Phi/\mathrm{d}\omega$ (cm^{-2}), pulse duration Δt, etc. As for a nuclear target, its important characteristic is the parameter $\beta = \Delta\omega_\gamma/\Delta\omega_{\mathrm{tot}}$, where $\Delta\omega_\gamma$ is the natural radiative width of the a $\to$ m transition and $\Delta\omega_{\mathrm{tot}}$ is the full inhomogeneous width of the transition.

2. In the case of external induction using the anti-Stokes scheme, the rate of radiative decay of excited states is determined by the rate of the first stage of the process and depends on the spectral characteristic of a radiation source. In the case of a broadband source with the spectral density $(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{bb}}$ specified in the frequency interval $\Delta\omega \geqslant \Delta\omega_{\mathrm{tot}}$, this relative rate $|\mathrm{d}n_{\mathrm{m}}/\mathrm{d}t|n_{\mathrm{m}}^{-1}$ is

$$-\frac{1}{n_{\mathrm{m}}}\frac{\mathrm{d}n_{\mathrm{m}}}{\mathrm{d}t} = \frac{\lambda^2}{2\pi}\frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1}\left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{bb}}\Delta\omega_\gamma, \tag{1}$$

while the total flux density of inducing photons required to obtain this rate is

$$\Phi_{\mathrm{bb}} = \left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{bb}}\Delta\omega_{\mathrm{tot}}, \tag{2}$$

where λ is the resonance wavelength of the gamma transition; n_{m} is the concentration of metastable nuclei; J_{m} and J_{a} are the angular moments of the metastable and auxiliary levels, respectively; and t is time. This gives

$$-\frac{1}{n_{\mathrm{m}}}\frac{\mathrm{d}n_{\mathrm{m}}}{\mathrm{d}t} = \frac{\lambda^2}{2\pi}\frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1}\Phi_{\mathrm{bb}}\beta. \tag{3}$$

The ratio β of linewidths in the experiments is usually much lower than unity.

In the alternative case of a special suppression of the inhomogeneous broadening to the natural width, when $\Delta\omega_{\mathrm{tot}} \to \Delta\omega_\gamma$ and $\beta \to 1$, the rate (3) of the process is determined by the total photon flux density Φ_{nb}, and the required spectral density of a monochromatic source with the linewidth coinciding with $\Delta\omega_\gamma$ is

$$\left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{bb}} = \frac{\Phi_{\mathrm{nb}}}{\Delta\omega_\gamma}. \tag{4}$$

Therefore, by assuming that sources of two different types producing the same relative induction rate have equal spectral densities, the ratio of the total flux densities of inducing photons will be

$$\frac{\Phi_{\mathrm{bb}}}{\Phi_{\mathrm{nb}}} = \beta^{-1} \gg 1, \tag{5}$$

and vice versa, in the case of equal total densities $\Phi_{\mathrm{bb}} = \Phi_{\mathrm{nb}}$, the spectral density ratio is

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$$\frac{(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{bb}}}{(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{nb}}} = \beta \ll 1, \tag{6}$$

i.e.,

$$\frac{\Phi_{\mathrm{bb}}}{\Phi_{\mathrm{nb}}} \frac{(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{bb}}}{(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{nb}}} = 1.$$

These differences can be considerable because, as pointed out above, when the inhomogeneous broadening is not suppressed, the ratio $\beta \ll 1$ can be many orders of magnitude smaller than unity. Note by the way that the gains $\mathrm{d}\Phi/\mathrm{d}z$ of the photon flux in stimulated emission upon suppression of the inhomogeneous broadening and without it differ by many times because $\beta^{-1} \gg 1$.

The efficiency of the induced decay of metastable states can be estimated by the quality parameter Q, the ratio of the decay rate in the presence of external inducing gamma radiation to the rate of the spontaneous process:

$$Q = -\frac{\tau_{\mathrm{m}}}{n_{\mathrm{m}}} \frac{\mathrm{d}n_{\mathrm{m}}}{\mathrm{d}t}. \tag{7}$$

This gives the quality parameters for the two alternative inducing radiation sources:

$$\begin{aligned} Q_{\mathrm{bb}} &= \frac{\lambda^2}{2\pi} \frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1} \left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{bb}} = \frac{\lambda^2}{2\pi} \frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1} \Phi_{\mathrm{bb}} \tau_{\mathrm{m}} \beta, \\ Q_{\mathrm{nb}} &= \frac{\lambda^2}{2\pi} \frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1} \left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{nb}} = \frac{\lambda^2}{2\pi} \frac{2J_{\mathrm{a}}+1}{2J_{\mathrm{m}}+1} \Phi_{\mathrm{nb}} \tau_{\mathrm{m}}, \end{aligned} \tag{8}$$

i.e.,

$$\frac{Q_{\mathrm{bb}}}{Q_{\mathrm{nb}}} = 1 \ \text{for} \ \left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{bb}} = \left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right)_{\mathrm{nb}} \tag{9}$$

for sources with equal spectral densities of gamma radiation and

$$\frac{Q_{\mathrm{bb}}}{Q_{\mathrm{nb}}} = \beta \ll 1 \ \text{for} \ \Phi_{\mathrm{bb}} = \Phi_{\mathrm{nb}} \tag{10}$$

for sources with equal total flux densities of gamma quanta.

Therefore, it follows from (5), (6), (9), and (10) that the use of a sufficiently monochromatic radiation source and the suppression of the inhomogeneous broadening of the transition line can increase the efficiency of the trigger process by many orders of magnitude for the same total density of the photon flux but at the expense of the same increase in the required spectral density.

The elimination of the inhomogeneous line broadening (caused mainly by the thermal motion of atoms containing nuclei) down to the natural linewidth allows one to hope, according to (10), to obtain in some cases a rather high release of the isomer energy even upon a direct induced transition from the metastable to ground state, not using any auxiliary levels [4]. One can see, however, that in all the cases the expediency of suppression of the inhomogeneous broadening of the transition line for increasing the rate of the induced decay of excited states is completely determined by their consistency with the spectral properties of the available gamma-quantum source.

The advantage of the anti-Stokes scheme is that the energy of inducing photons, as a rule, is lower than the released energy of the metastable state (unlike the scheme of a direct stimulated transition [4], where these energies coincide). Another advantage of this scheme is less stringent requirements to the monochromatism of inducing radiation because, for the specified total flux density Φ, the spectral density of the external gamma-quantum flux

$$\left(\frac{\mathrm{d}\Phi}{\mathrm{d}\omega}\right) = \frac{\Phi}{\Delta\omega_{\mathrm{m}} + \Delta\omega_{\mathrm{a}\downarrow}} \tag{11}$$

is determined in fact by the linewidth $\Delta\omega_{\mathrm{a}\downarrow}$ of the auxiliary spontaneous transition, which is, as a rule, much greater than the metastable-level width $\Delta\omega_{\mathrm{m}}$ ($\Delta\omega_{\mathrm{a}\downarrow} \gg \Delta\omega_{\mathrm{m}}$).

3. Let us make some quantitative estimates. Consider the anti-Stokes scheme based on a long-lived isomer with the metastable-state energy 250 keV and the higher-lying auxiliary level with the energy 260 keV. The lifetime of the radiative transition to the metastable state is $\tau_{\mathrm{a}\to\mathrm{m}} = 10^{-8}$ s. Then, if we assume that $Q = 0.01$, we should have $(\mathrm{d}\Phi/\mathrm{d}\omega)_{\mathrm{bb}} \approx 300$ phot cm^{-2} and $Q_{\mathrm{bb}} \approx 10^{15}$ phot cm^{-2} s^{-1}, which exceeds the possibilities of broadband sources by many orders of magnitude. In this case, the energy of inducing photons amounts to only 4 % of the metastable-state energy. The moderate time $\tau_{\mathrm{a}\to\mathrm{m}}$ allows one to use the Mössbauer method to narrow the m → a transition line down to the natural linewidth, by embedding the isomer to a cooled solid matrix. In this case, the cross section σ of the m → a transition from the metastable state to the upper auxiliary level proves to be quite high, $\sigma \approx 2.3 \times 10^{-17}(2J_{\mathrm{a}}+1)(2J_{\mathrm{m}}+1)^{-1}$ cm^2, i.e., approximately six orders of magnitude higher than that in the non-Mössbauer case [we assume here that the coefficient of internal electronic conversion is small ($\alpha \ll 1$) and neglect the Debye–Waller factor], and therefore the required value of Φ_{nb} decreases by the same ratio. When the time $\tau_{\mathrm{a}\to\mathrm{m}}$ noticeably exceeds a microsecond, it is unlikely that the Mössbauer method will be efficient. The alternative approach to the induction of anti-Stokes transitions in isomers could be based on the use of a drastic narrowing of gamma-transition nuclear lines in atoms in a Bose–Einstein condensate [5] if such substantially different experimental methods could be combined.

4. Finally, the asymptotic behaviour of the stimulated emission cross section plays an important role limiting the experimental possibilities. Upon pulsed irradiation of an isomer by an external source, the current value of the transition cross section is a function of time and increases from zero at the initial instant by approaching a stationary value only for the time comparable to the spontaneous decay time of the upper level [4]. This phenomenon, which does not play any noticeable role at short lifetimes (for example, in the optical range), sometimes determines in fact the rate $(-\mathrm{d}n_{\mathrm{m}}/\mathrm{d}t)$ of the induced decay of metastable states, requiring a rather long exposure of nuclei to external radiation, which is comparable to the lifetime of the state. As an example, we recall, experiments on the trigger induction of isomers by series of hard quanta emitted by relativistic electrons accelerated in microwave accelerators. The duration of each of these pulses lies usually in the subnanosecond range, i.e., as a rule, it is many orders of magnitude shorter than the lifetimes of nuclear states, which is not compensated in the least by a long total exposure time of the whole series.

All the above-said restricts the required pulse duration from below, which should be taken into account not only in

Table 1.

Nuclide	E_m/keV	E_a/keV	$E_m/(E_a - E_m)$	τ_m	$\tau_{a\downarrow}$/ns	J_m	J_a	$\sigma/10^{-17}$ cm^2
$^{93}_{42}$Mo	2425	4.8	510	9.8 hours	5.05	$21^+/2$	$17^+/2$	8.8
$^{99}_{43}$Tc	142.7	38.4	3.7	8.55 hours	5.15	$1^-/2$	$5^+/2$	5.05
$^{110}_{47}$Ag	117.6	1.13	104	355 days	52.5	6^+	3^+	104
$^{113}_{48}$Cd	263.6	34.9	7.6	20 years	0.042	$11^-/2$	$3^+/2$	6.65
$^{152}_{63}$Eu	45.6	31.66	1.45	13.3 hours	54.5	0^-	3^-	9.7
$^{179}_{72}$Hf	1105.8	0.077	14400	36 days	< 1	$25^-/2$	$7^+/2$	1270
$^{189}_{76}$Os	30.81	5.39	5.8	8.3 hours	0.76	$9^-/2$	$1^-/2$	1.7
$^{210}_{83}$Bi	271.3	48.4	5.6	4.6×10^6 years	0.0052	9^-	2^-	0.031

Note: E_m is the metastable-state energy; E_a is the energy of the higher-lying auxiliary state; σ is the asymptotic cross section of the trigger m → a transition.

the case of a direct induction of the metastable-state decay [4] but also in the analysis of the first stage of the anti-Stokes process, where the corresponding delays can achieve the values comparable to the time $\tau_{a \to m}$. This time is taken to be 10^{-6} s for a model estimate, which is a few orders of magnitude longer than the above-mentioned subnanosecond duration of hard inducing radiation pulses generated by relativistic electrons. The necessity of a long continuous exposure to inducing radiation comparable to the upper-state lifetime means that $W = \Phi\tau$ in expression (8) for the quality parameter Q is in fact the measure of the total energy of an external source spent for inducing. Of course, to obtain the pragmatic result, the energy W should be considerably lower than the released energy of isomers. This means that narrowband sources have the obvious advantage.

5. The list of possible nuclides suitable for these studies is quite broad. Table 1 presents some of them.

6. In summary, several remarks can be made about the analysis of experiments performed earlier and planning of future experiments on the trigger induction of the decay of the metastable states of isomeric nuclei:

(i) If quasi-monochromatic radiation sources resonant with the trigger transition are available, the use of Mössbauer targets with isomers in cooled crystal matrices (or other methods for suppressing the inhomogeneous broadening of the m → a transition line) can increase the efficiency of the process by a few orders of magnitude.

(ii) This approach used with broadband inducing sources does not promise a noticeable increase in the efficiency.

(iii) To achieve the efficient induction, the continuous exposure to inducing radiation should be comparable to the upper-state lifetime of the transition.

(iv) It is reasonable to expand considerably the list of isomers for experiments (along with nuclides listed in Table 1, for example, the $^{242\mathrm{m}}$Am isomer with the metastable-state lifetime 141 years can be used).

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