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Self-sustained exothermic reaction of anti-Stokes gamma transitions in long-lived isomeric nuclei. I

L A Rivlin

Abstract. The conditions for implementing a self-sustained exothermic nuclear (combustion) reaction in a system comprising long-lived metastable isomers and quasi-equilibrium high-temperature black-body radiation are examined. In this system the radiative decay of the metastable state is a result of an anti-Stokes process bypassing a strongly forbidden isomeric transition. The anti-Stokes transition in the reaction zone is triggered by the corresponding resonance spectral components of the quasi-equilibrium radiation, the temperature of which, sufficient to close the energy reaction cycle, is in its turn maintained by the absorption of hard photons, emitted by nuclei, in the reaction zone.

1. Introduction

The possibility of using long-lived nuclear isomers to obtain energy [1] is based on the existence of nuclei with fairly long lifetimes of excited metastable states and a specific energy content of the order of tens of megajoules per gramme. This value is approximately two orders of magnitude smaller than the energy content of fissionable nuclides but exceeds by three orders of magnitude the heat generating capacity of a hydrocarbon fuel.

One of the ways of releasing the energy stored in the excited states of isomeric nuclei may involve the implementation of the anti-Stokes transitions $\mathrm{m} \to \mathrm{t} \to \mathrm{g}$ (Fig. 1), bypassing the strongly forbidden $\mathrm{m} \to \mathrm{g}$ transition involving the absorption of external radiation photons with the energy $\hbar\omega_0$. Such energy triggers a more energetic spontaneous transition $\mathrm{t} \to \mathrm{g}$ with the energy $E_{\mathrm{t}} - E_{\mathrm{g}} > \hbar\omega_0$. The energy gain of this process increases with increase in the 'strength' of this inequality.

The list of examples (by no means exhaustive) for certain long-lived (τ_i) isomers with possible anti-Stokes transitions is presented in Table 1. (It is noteworthy that the possibility of using Stokes transitions for the same purpose is limited by the low probability of stimulating the triggering downward transition from a long-lived metastable state.)

There are numerous postulates and theoretical studies concerning nuclear anti-Stokes transitions, including those designed to construct a nuclear gamma-laser (see, for example, Refs [2 – 10]). Experiments undertaken to observe anti-Stokes transitions in isomers are also known [1, 11 – 13].

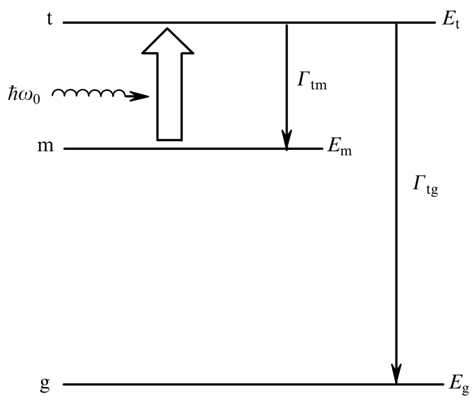

Figure 1.

Table 1. Certain metastable isomers with possible anti-Stokes transitions

Isomers	$E_{\mathrm{tg}}/\mathrm{keV}$	$E_{\mathrm{tm}}/\mathrm{keV}$	$\tau_{\mathrm{i}}/\mathrm{years}$
$^{97}\mathrm{Tc}$	215.7	119.2	90.1
$^{113}\mathrm{Cd}$	298.5	34.9	0.04
$^{115}\mathrm{Cd}$	229.1	48.1	44.6
$^{125}\mathrm{Te}$	321.1	176.4	0.16
$^{129}\mathrm{Te}$	180.8	75.3	0.1
$^{177}\mathrm{Lu}$	1049.5	79.3	0.44
$^{179}\mathrm{Hf}$	1105.9	0.077	0.07
$^{210}\mathrm{Bi}$	319.7	48.4	8.2×10^3
$^{242}\mathrm{Am}$	59.2	4.3	141

Evidently, the rational production of energy by using anti-Stokes transitions in long-lived metastable isomers playing the role of a fuel requires the implementation of a closed cycle of self-sustained nuclear transitions (combustion reactions) in which the fraction of the released nuclear energy is directed towards the excitation of the trigger level t. Such a cycle may be regarded in a certain sense as a chain reaction. The aim of the present study is to examine one of the possible versions of this kind of energy cycle.

2. Phenomenological model

Suppose that the isomeric nuclei are located in a reaction zone representing a black-body cavity at a temperature T (Fig. 2). The radiation of the spectral component of the black body, the energy of which coincides with the energy

L A Rivlin Moscow State Institute of Radio Engineering, Electronics, and Automation (Technical University), prospekt Vernadskogo 78, 117454 Moscow, Russia, E-mail: rla@superlum.msk.ru

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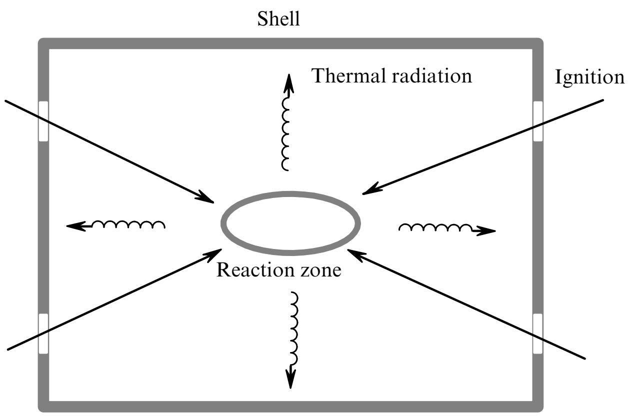

Figure 2.

$\hbar\omega_0$ of the m $\to$ t triggering transition, excites the trigger state t and leads to the spontaneous emission of photons with the energy $E_t - E_g > \hbar\omega_0$. Each such step liberates the difference energy $E_m - E_g$, absorbed by the black body, which maintains the temperature of the latter necessary to stimulate new m $\to$ t triggering transitions.

The self-sustained combustion reaction in the isomeric nuclei – black body system occurs under conditions such that the energy absorbed by the black body covers or exceeds all possible losses, including the losses in the emission of radiation into the external medium (in Fig. 2, this medium is shown as a shell absorbing thermal radiation and maintained at a temperature T_0). Such losses represent in essence the useful energy yield of the whole process.

It is possible to imagine various ways of the physical implementation of a reaction zone filled by high-temperature quasi-equilibrium radiation, in particular by employing a hot plasma with a magnetic or inertial confinement of some kind and/or the use of a 'hohlraum' cavity similar to that employed in experiments on inertial confinement fusion (see, for example, recent results [14 – 17]). Evidently, a combustion reaction of this type cannot start from a 'cold' state and hence requires a preliminary operation involving the heating of the zone ('ignition' — see Fig. 2).

3. Cross section and probability of an anti-Stokes transition

The m $\to$ t $\to$ g anti-Stokes transition with the structure of levels illustrated in Fig. 1 takes place under the influence of an external photon field and consists of the m $\to$ t transition involving the absorption of a photon having the energy $\hbar\omega_0$ and excitation of the nucleus to the trigger state t and of the t $\to$ g transition involving the spontaneous emission of a photon having the energy $E_t - E_g$. Following Ref. [7], the cross section of such an anti-Stokes transition may be estimated from the Breit – Wigner formula

$$\sigma_{\mathrm{mtg}} = \frac{\lambda^2}{8\pi}\frac{\tilde{\Gamma}_{\mathrm{tm}}\tilde{\Gamma}_{\mathrm{tg}}}{(\Gamma_t/2)^2 + (\hbar\Delta\omega)^2}\,, \tag{1}$$

where $\lambda = 2\pi c/\omega_0$ is the wavelength of the external exciting radiation; $\Gamma_t = \Gamma_{\mathrm{tm}} + \Gamma_{\mathrm{tg}} + \ldots$ is the total width of the level t including the widths of the t $\to$ m and t $\to$ g transitions, as well as the widths of the transitions due to all other possible decay channels, including, for example, the internal electron conversion; $\hbar\Delta\omega = E_t - E_m - \hbar\omega_0$ is the 'detuning' of the triggering-photon energy from precise resonance with the m $\to$ t transition;

$$\tilde{\Gamma}_{\mathrm{tm}} = \Gamma_{\mathrm{tm}}\left(1 - \frac{\hbar\Delta\omega}{E_t - E_m}\right)^{2L_{\mathrm{tm}}+1}\,; \tag{2}$$

$$\tilde{\Gamma}_{\mathrm{tg}} = \Gamma_{\mathrm{tg}}\left(1 - \frac{\hbar\Delta\omega}{E_t - E_g}\right)^{2L_{\mathrm{tg}}+1}\,; \tag{3}$$

where L_{tm} and L_{tg} are the multipolarities of the t $\to$ m and t $\to$ g transitions, respectively.

Hence we obtain the cross section

$$\sigma_{\mathrm{mtg}} = \frac{\lambda^2}{8\pi}\frac{\Gamma_{\mathrm{tm}}\Gamma_{\mathrm{tg}}}{(\Gamma_t/2)^2 + (\hbar\Delta\omega)^2}\left(1 - \frac{\hbar\Delta\omega}{E_t - E_m}\right)^{2L_{\mathrm{tm}}+1} \times\left(1 - \frac{\hbar\Delta\omega}{E_t - E_g}\right)^{2L_{\mathrm{tg}}+1} \approx \frac{\lambda^2}{8\pi}\frac{\Gamma_{\mathrm{tm}}\Gamma_{\mathrm{tg}}}{(\Gamma_t/2)^2 + (\hbar\Delta\omega)^2} \times\left[1 - \left(\frac{2L_{\mathrm{tm}}+1}{E_t - E_m} + \frac{2L_{\mathrm{tg}}+1}{E_t - E_g}\right)\hbar\Delta\omega\right]\,, \tag{4}$$

where the approximate equality within the limits of the Lorentzian width is valid by virtue of the small detuning from resonance:

$$|\hbar\Delta\omega| \ll E_t - E_m\,, \quad |\hbar\Delta\omega| \ll E_t - E_g\,. \tag{5}$$

The probability of an anti-Stokes transition with the cross section σ_{mtg} is

$$w_{\mathrm{mtg}} = \int_{-\infty}^{\infty}\sigma_{\mathrm{mtg}}\, j(\omega)\mathrm{d}\omega\,, \tag{6}$$

where the integral is evaluated over all frequencies ω and the corresponding spectral density $j(\omega)$ of the stimulating radiation. Since the width of the continuous spectrum of the quasi-equilibrium radiation in the reaction zone is significantly greater than the Lorentzian width, one can assume that its spectral density $j(\omega)$ is constant near resonance. The probability of the anti-Stokes transition is then

$$w_{\mathrm{mtg}} = \frac{\lambda^2}{8\pi\hbar}\frac{\Gamma_{\mathrm{tm}}\Gamma_{\mathrm{tg}}}{\Gamma_t}j(\omega)\,. \tag{7}$$

We may note that in the derivation of expression (7), a small correction to the cross section σ_{mtg}, which depends explicitly on the multipolarities L_{tm} and L_{tg}, was omitted from the integrand of expression (6).

If the spectral radiation density in the reaction zone is fitted by the Planck function for the equilibrium radiation of an absolutely black body at a temperature T, i.e.

$$j_{\mathrm{Pl}} = \frac{2\pi/\lambda^2}{\exp(\hbar\omega/kT) - 1} \tag{8}$$

(k is the Boltzmann constant), then the probability of the anti-Stokes transition is

$$w_{\mathrm{mtg}} = \frac{\Gamma_{\mathrm{tm}}\Gamma_{\mathrm{tg}}}{4\hbar\Gamma_t}\left(\exp\frac{\hbar\omega_0}{kT} - 1\right)^{-1}\,, \tag{9}$$

whereas the average lifetime of a long-lived metastable isomer in the reaction zone is estimated as

$$\Delta t_{\mathrm{mtg}} \equiv \frac{1}{w_{\mathrm{mtg}}} = \frac{2}{\pi}\tau_0\left(\exp\frac{\hbar\omega_0}{kT} - 1\right)\,, \tag{10}$$

where $\tau_0 = \tau_{\mathrm{tm}}\tau_g/\tau_t$ is the characteristic time of the transition; $\tau_{\mathrm{tm}} = 2\pi\hbar/\Gamma_{\mathrm{tm}}$, $\tau_{\mathrm{tg}} = 2\pi\hbar/\Gamma_{\mathrm{tg}}$, $\tau_t = 2\pi\hbar/\Gamma_t$ are the lifetimes of states in relation to the t $\to$ m and t $\to$ g transitions and the total lifetime of the state t, respectively.

If $\Gamma_t = \Gamma_{tm} + \Gamma_{tg}$, then

$$\Delta t_{mtg} = \frac{2}{\pi}(\tau_{tm} + \tau_{tg})\left(\exp\frac{\hbar\omega_0}{kT} - 1\right) . \tag{11}$$

An idea about the possible scale of Δt_{mtg} is provided by the following example: $\Delta t_{mtg} = 3.4$ µs if $\hbar\omega_0 = 100$ eV, $kT = 25$ eV, and $\tau_0 = 100$ ns.

4. Rate equation in the reaction zone

The rate of change of the radiation energy Q stored in the reaction zone is specified by the rate equation

$$\frac{dQ}{dt} = -\sigma(T^4 - T_0^4)S + w_{tmg}\hbar\omega_{mg}nV\eta , \tag{12}$$

where $\sigma = 5.6 \times 10^{-12}$ W K^4 cm^{-2} is Stefan's constant; T is the temperature of the quasi-equilibrium radiation in the zone; T_0 is the temperature of the outer shell surrounding the zone and absorbing the thermal radiation from this zone (Fig. 2); n is the volume concentration of the metastable nuclei; V and S are the volume and area of the external surface of the reaction zone; η is the fraction of photons with the energy $\hbar\omega_{tg}$ absorbed in the reaction zone; t is the time. The first term on the right-hand side of Eqn (12) describes the radiative energy exchange between the reaction zone and the outer shell and the second term describes the energy released in this zone as a result of the anti-Stokes process.

It must be emphasised that an adequate efficiency η of the absorption and retention of the emitted photons with the energy $\hbar\omega_{tg}$ in the zone is a very important factor which may be one of the bottlenecks of the whole system. In terms of the dimensionless variables

$$\theta = \frac{kT}{\hbar\omega_0}, \quad \theta_0 = \frac{kT_0}{\hbar\omega_0}, \quad \varkappa = \frac{t}{\tau_0}, \quad q = \frac{2Q}{\pi V\hbar\omega_{mg}n^*},$$
$$n^* = \frac{2\sigma}{\pi}\frac{S}{V}\left(\frac{\hbar\omega_0}{k}\right)^4\frac{\tau_0}{\hbar\omega_{mg}} \tag{13}$$

the rate equation (12) assumes the following form:

$$\frac{dq}{d\varkappa} = -(\theta^4 - \theta_0^4) + \frac{n}{n^*}\left(\exp\theta^{-1} - 1\right)^{-1} . \tag{14}$$

An idea about the possible scale of the normalised constant n^* is provided by the following example: $n^* = 8.4 \times 10^{18}$ cm^{-3} if $\hbar\omega_0 = 100$ eV, $\hbar\omega_{mg} = 500$ keV, $\eta = 1$, $\tau_0 = 100$ ns, and $V/S = 1$ cm.

The steady-state $(d/dx = 0)$ solution of the rate equation is given by the transcendental equation

$$(\theta^4 - \theta_0^4)(\exp\theta^{-1} - 1) = \frac{n}{n^*} . \tag{15}$$

The $\theta(n)$ curve, reflecting the steady state, is S-shaped (Fig. 3) with two characteristic points A and B at which $d\theta/dn \to \infty$,

$$\theta_A \approx \theta_0(1 + \theta_0), \quad n_A \approx 4n^*\theta_0^5\exp\theta_0^{-1} ,$$
$$\theta_B \approx 0.25, \quad n_B \approx 0.21n^* \tag{16}$$

(the variables at point A were calculated on the assumption that $\theta_0 \ll 1$ and those at point B on the assumption that $\theta_0^4 \ll \theta^4$).

The first of the characteristic points A is infinitely far along the abscissa axis so that $n_A/n^* = 4 \times 10^{84}$ for

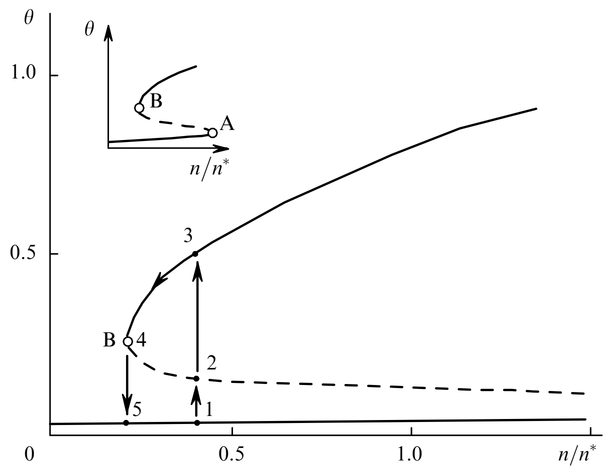

Figure 3.

$\theta_0 = 0.025$ (pint A is shown in the inset in Fig. 3 without maintaining the scale). On the other hand, the second point (point B) has fully realisable coordinates and serves as the transition point from the unstable branch of the curve with $d\theta/dn < 0$ (shown by a dashed line), which plays the role of the threshold or initial reaction curve, to the upper stable branch of the stable regime with $d\theta/dn > 0$.

5. Course of combustion reaction

The course of combustion reaction may be represented schematically in the following form. The initial state is specified by point 1 (Fig. 3) on the lower stable branch of the S-shaped curve with the initial concentration of the metastable nuclei $n/n^* > n_B/n^* = 0.21$ at a temperature $\theta = \theta_0$ of the surrounding medium. Under these conditions, there are neither anti-Stokes transitions nor the combustion reaction. The temperature in the zone is then rapidly raised from the outside to $\theta_0 < \theta < \theta_B$ and the mapping point moves upwards to the unstable threshold branch of the curve at point 2.

This process may be called the 'ignition' of the reaction because, owing to the instability of point 2, a jump to point 3 then takes place. It is accompanied by an avalanche-like increase in temperature to $\theta > \theta_B$. Point 3 is stable, provided that the initial concentration of the metastable nuclei n remains unaltered, but the nuclei decay during the combustion reaction. In order to maintain under these conditions a steady-state reaction with the parameters mapped by point 3 and with the initial power

$$\mathscr{P} = \frac{\pi}{2}\hbar\omega_{mg}\frac{n^*V}{\tau_0}(\theta^4 - \theta_0^4) \tag{17}$$

a constant supply of new metastable nuclei to the reaction zone at the rate

$$\Phi = \frac{\mathscr{P}}{\hbar\omega_{mg}} \tag{18}$$

is essential.

At a rate of arrival of new metastable nuclei in the reaction zone exceeding Φ, the mapping point moves from position 3 to the right along the steady-state branch of the curve and back again. On the other hand, if $\Phi = 0$, i.e. the store of the metastable nuclei decaying during the combustion reaction is not made good, the mapping point moves rapidly along the steady-state branch of the curve to the left, up to the characteristic point B (or 4), where an abrupt drop to the lower steady-state branch at point 5 takes place, i.e. avalanche-like cooling occurs and the reaction, which is pulsed in this case, terminates.

6. Certain quantitative estimates

The estimates below were made for an arbitrarily selected but fully realistic set of parameters of the pulsed version of the combustion reaction:

Energy $\hbar\omega_{\mathrm{mg}}$ released in an elementary anti-Stokes step	500 keV
Triggering-photon energy $\hbar\omega_0$	100 eV
Characteristic time of the transition τ_0	100 ns
Ratio of the volume to the surface of the zone V/S	1 cm
Fraction η of photons with the energy $\hbar\omega_{\mathrm{tg}}$ absorbed in the zone	1
Zone volume V	100 cm^3
Normalised concentration of nuclei n^*	$8.4 \times 10^{18}\ \mathrm{cm}^{-3}$
Initial concentration of nuclei n	$2.5 \times 10^{18}\ \mathrm{cm}^{-3}$
Temperature of outer shell $T_0(\theta_0)$	2.5 eV (0.025)
'Ignition' temperature $T_2(\theta_2)$	17 eV (0.17)
Maximum temperature in the reaction zone $T(\theta)$	42 eV (0.42)
Concentration Δn of the nuclei 'consumed' per pulse	$7.6 \times 10^{17}\ \mathrm{cm}^{-3}$
Lifetime of a metastable nucleus in the reaction zone Δt_{mtg}	630 ns
Energy released in the pulse	6.1 MJ
Power of the radiation in the pulse (estimate made on the assumption that the pulse duration corresponds to 5 characteristic transition times)	1.9 TW

7. Conclusions

The above study and certain simple quantitative estimates indicate internal self-consistency (at least at the level of a phenomenological model) of the concept of a self-sustained exothermic reaction of anti-Stokes gamma-transitions in long-lived isomeric nuclei as a method for the production of energy in pulsed, repetitively pulsed, and possibly cw regimes.

One cannot rule out the possibility of applying this concept to problems of the construction of sources of x rays with a high brightness and total intensity, in particular for experiments on inertial confinement fusion, for the pumping of x-ray and nuclear gamma-lasers [10], etc.

The proposed general approach requires further analysis and a more detailed specification designed to solve the following problems: the physical implementation of high-temperature reaction zones of different types (magnetically or inertially confined plasma and/or black-body cavity — 'hohlraum', etc.); the achievement of rapid 'ignition' of the reaction with the aid of the radiation from a dense plasma generated at the focus of a high-power optical laser (here it is appropriate to mention the successful experiments on the excitation of low-lying nuclear levels in a laser plasma [18]) or in the exploding wire Z-pinch (a radiative temperature of the order of hundreds of electron-volts can be attained by the last method in a secondary 'hohlraum' — see, for example, Refs [14–17] etc.); the analysis of the absorption of photons emitted into the reaction zone, the dynamics and stability of the processes occurring in it, etc.

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References

1. Collins C B, Carroll J J Hyperfine Interact. **107** 3 (1997)
2. Baklanov E V, Chebotaev V P Pis'ma Zh. Eksp. Teor. Fiz. **21** 286 (1975) [JETP Lett. **21** 131 (1975)]
3. Baklanov E V, Chebotaev V P Kvantovaya Elektron. (Moscow) **3** 634 (1976) [Sov. J. Quantum Electron. **6** 345 (1976)]
4. Rivlin L A Kvantovaya Elektron. (Moscow) **4** 676 (1977) [Sov. J. Quantum Electron. **7** 380 (1977)]
5. Arad B, Eliezer S, Paiss Y Phys. Lett. A **74** 395 (1979)
6. Ishkhanov B S, Piskarev I M Yad. Fiz. **32** 593 (1980) [Phys. At. Nucl. **32** 305 (1980)]
7. Becker W, Schlicher R R, Scully M O Phys. Lett. A **106** 441 (1984)
8. Eliezer S, Martinez-Val J M, Paiss Y, Velarde G Kvantovaya Elektron. (Moscow) **22** 1140 (1995) [Quantum Electron. **25** 1106 (1995)]
9. Eliezer S, et al. Laser Phys. **5** 323 (1995)
10. Rivlin L A Kvantovaya Elektron. (Moscow) **27** 189 (1999) [Quantum Electron. **29** 467 (1999)]
11. Collins C B, et al. Phys. Rev. C **37** 2267 (1988)
12. Collins C B, et al. Laser Phys. **9** 8 (1999)
13. Collins C B, et al. Phys. Rev. Lett. **82** 695 (1999)
14. Porter J L Jr, et al. Abstracts of Papers presented at the First International Conference on Inertial Fusion Sciences and Applications (IFSA'99), Bordeaux, 1999, p. 18
15. Sanford T W L, et al. Abstracts of Papers presented at the First International Conference of Inertial Fusion Sciences and Applications (IFSA'99), Bordeaux, 1999, p. 23
16. Lash J S, et al. Abstracts of Papers presented at the First International Conference on Inertial Fusion Sciences and Applications (IFSA'99), Bordeaux, 1999, p. 141
17. McFarlane J J Abstracts of Papers presented at the First International Conference on Inertial Fusion Sciences and Applications (IFSA'99), Bordeaux, 1999, p. 216
18. Andreev A V, Volkov R V, Gordienko V M, et al. Pis'ma Zh. Eksp. Teor. Fiz. **69** 343 (1999) [JETP Lett. **69** 371 (1999)]