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

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Abstract. **This paper is a continuation of our two previous publications under the same title and is devoted to the features of a chain reaction proceeding upon nonequilibrium radiation of a plasma in the presence of nonradiative decay channels of the trigger level and other processes. The required accuracy of the resonance of nuclei with a line spectrum of ions in the plasma and the criterion for a positive energy effect in the case of pulsed combustion reaction are analysed and an example of the numerical estimate of the reaction parameters for the isomeric $^{242}_{95}$Am nucleus is considered.**

Keywords: *gamma transitions, isomeric nuclei, combustion reaction.*

1. Introduction

The possibility of a new type of chain nuclear reaction in a system of long-lived isomeric nuclei and a hot dense plasma was considered in our previous papers [1, 2]. The energy stored in the metastable states of isomeric nuclei is liberated as a result of an anti-Stokes transition to an auxiliary upper trigger state (bypassing the strongly forbidden direct downward transition) followed by a spontaneous decay of this state. The transition to the upper trigger state occurs upon absorption of X-ray photons emitted by the plasma, which are in resonance with the trigger transition. The closure of the energy cycle of the reaction is provided by the fact that the nuclear energy released upon anti-Stokes transitions (in the form of gamma quanta, conversion electrons, etc.) is partially absorbed by the plasma and causes its heating, while the spectral components of plasma radiation, which are in resonance with the trigger upward transition in the nuclei, induce the anti-Stokes process. In this paper, we continue to study various aspects of this nuclear reaction.

2. Rate balance equations for plasma radiation deviating from equilibrium

In the phenomenological model of the combustion reaction [1, 2], we used the equilibrium Planck distribution $j_{\rm Pl}$ of the black-body radiation as the zero-order approximation for the plasma radiation spectrum $j(\omega)$ in the reaction zone. However, it is well known that the spectrum $j(\omega)$ of plasma radiation with the electron temperature T does not coincide at all with the distribution $j_{\rm Pl}$ for the same temperature and is a complex function of the ionic composition of the plasma and specific experimental conditions. Nevertheless, if we use the zero-order approximation (i.e., the Planck distribution $j_{\rm Pl}$ as a reference distribution), the next approximation to the actual spectrum of plasma radiation can be represented in the form

$$j(\omega) = \psi(\omega) j_{\rm Pl}, \tag{1}$$

where $\psi(\omega)$ is the dimensionless coefficient depending on frequency ω. In particular, the trigger spectral component of the plasma at frequency ω_0 is defined by the coefficient $\psi(\omega_0) \equiv \psi_0$. Similarly, the integrated radiation intensity is

$$\int_0^\infty \hbar\omega\psi(\omega) j_{\rm Pl} {\rm d}\omega = \Psi\sigma_{\rm St} T^4, \tag{2}$$

where $\sigma_{\rm St}$ is the Stefan constant.

It is known that due to intrinsic self-absorption in a dense plasma, the intensity of its integrated radiation can be many orders of magnitude lower than the intensity predicted by the Stefan – Boltzmann law; i. e., Ψ can be much smaller than unity. These considerations are confirmed by numerical examples [3]. For example, the integrated intensity of plasma radiation with concentration 10^{16} cm^{-3} and temperature 10 keV actually corresponds not to this temperature, but to the black-body temperature which is equal only to 2 keV; i.e., $\Psi \approx 1.6 \times 10^{-19}$. Another example: the addition of 1 % (by concentration) of oxygen to a hydrogen plasma with concentration 10^{13} cm^{-3} and temperature 10 eV increases the integrated radiation intensity by a factor of 10^4 due to the characteristic oxygen line.

Thus, the diversity of the spectral composition of radiation emitted by a real plasma is reflected phenomenologically by introducing coefficients $\psi(\omega)$ and Ψ, and the balance equations (15) and (16) from Ref. [2] for the normalised electron temperature $\theta \equiv k_{\rm B}T/\hbar\omega_0$ of the plasma and the concentration n of metastable nuclei for the normalised time $\kappa \equiv t/\tau_0$ take the form

$$\frac{{\rm d}\theta}{{\rm d}\kappa} = b\frac{n^*}{n_0}\left[-\left(\Psi\theta^4 - \theta_0^4\right) + \psi_0\frac{n}{n^*}\left(\exp\frac{1}{\theta} - 1\right)^{-1}\right], \tag{3}$$

$$\frac{{\rm d}n}{{\rm d}\kappa} = -\frac{\pi}{2}\psi_0 n\left(\exp\frac{1}{\theta} - 1\right)^{-1} + \frac{\tau_0}{V}\Phi, \tag{4}$$

where θ_0 is the normalised temperature of the reactor shell;

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according to expression (18) from Ref. [1], Φ is the rate of the inflow of metastable nuclei from the external source per unit time (Φ is exactly equal to the rate of total outflow of nuclei from the reaction zone, so that the total concentration of nuclei $n_0 = \mathrm{const}$ remains unchanged);

$$n^* = \frac{2\sigma_{\mathrm{St}}\tau_0 S}{\pi\eta\hbar\omega_{\mathrm{mg}}V}\left(\frac{\hbar\omega_0}{k_{\mathrm{B}}}\right)^4 \tag{5}$$

is the normalisation of the concentration of metastable nuclei[1];

$$b = \frac{\pi}{2}\frac{k_{\mathrm{B}}}{\chi}\frac{\hbar\omega_{\mathrm{mg}}}{\hbar\omega_0}\eta; \tag{6}$$

χ is the effective heat capacity of the plasma per particle; $\hbar\omega_{\mathrm{mg}}$ and $\hbar\omega_0$ are the energies of the metastable state and a trigger photon, respectively; V and S are the volume and surface area of the reaction zone; η is the ratio of the energy absorbed in the reaction zone to the energy released by metastable nuclei; k_{B} is Boltzmann constant;

$$\tau_0 = \frac{2\pi\hbar}{1+\alpha_{\mathrm{tg}}}\frac{\Gamma_{\mathrm{m}}+\Gamma_{\mathrm{t}}}{\Gamma_{\mathrm{tm}}\Gamma_{\mathrm{tg}}} \tag{7}$$

is the characteristic time of the anti-Stokes transition (see expression (10) from Refs [1] and also [4]); Γ_{tm} and Γ_{tg} are the radiative widths of the $\mathrm{t} \to \mathrm{m}$ and $\mathrm{t} \to \mathrm{g}$ transitions, respectively; $\Gamma_{\mathrm{t}} = \Gamma_{\mathrm{tm}}(1+\alpha_{\mathrm{tm}}) + \Gamma_{\mathrm{tg}}(1+\alpha_{\mathrm{tg}})$ и $\Gamma_{\mathrm{m}} = \Gamma_{\mathrm{mg}}(1+\alpha_{\mathrm{mg}})$ are the total widths of levels t and m; and α_{tm}, α_{tg} and α_{mg} are the coefficients of the internal electron conversion of the corresponding transitions. If a metastable level is long-lived, as usual, we can put $\Gamma_{\mathrm{m}} = 0$; in this case,

$$\tau_0 \approx \frac{2\pi\hbar}{\Gamma_{\mathrm{tm}}}\left(1 + \frac{\Gamma_{\mathrm{tm}}}{\Gamma_{\mathrm{tg}}}\frac{1+\alpha_{\mathrm{tm}}}{1+\alpha_{\mathrm{tg}}}\right), \tag{8}$$

and the characteristic time τ_0 is mainly determined by the radiative width Γ_{tm} of the trigger transition if the numerator and the denominator in the parentheses are of the same order of magnitude.

3. Steady-state solutions of the balance equations

The steady-state solution of Eqns (3) and (4) are given by

$$\left(\theta^4 - \frac{\theta_0^4}{\Psi}\right)\left(\exp\frac{1}{\theta} - 1\right) = \frac{\psi_0}{\Psi}\frac{n}{n^*} \equiv \frac{n}{n^*_{\mathrm{eff}}}, \tag{9}$$

$$\Phi = \frac{\pi}{2}n^*\frac{V}{\tau_0}\left(\Psi\theta^4 - \theta_0^4\right). \tag{10}$$

The first of these equations differs significantly from the corresponding steady-state solution (15) from Ref. [1]: the effective normalisation $n^*_{\mathrm{eff}} = n^*\Psi/\psi_0$ of the concentration n of metastable nuclei, which is used instead of n^* in the right-hand side of expression (9) and on the abscissa axis of the S-shaped diagram of states [1], may be much smaller than n^* if $\Psi/\psi_0 \ll 1$ (see Section 2). This clearly indicates how one should form the ionic composition and, hence, the plasma spectrum in the experiments (e.g., by introducing ions with emission lines lying in the energy range of trigger photons $\hbar\omega_0$ for increasing coefficient ψ_0). A decrease in the effective normalisation n^*_{eff} as compared to n^* may play an important role, facilitating the decrease in the threshold concentration $n_{\mathrm{B}} = 0.21n^*_{\mathrm{eff}}$ of metastable nuclei (formula (16) from Ref. [1]). It should be noted here that a change in the term θ_0^4/ψ in (9) as compared to the corresponding term in Eqn (15) from Ref. [1] does not affect significantly the form of the steady-state solution because $\theta^4 \gg \theta_0^4$.

The stability of the steady-state solution at the upper branch of the S-shaped curve follows immediately from an analysis of the signs of the derivatives upon a deviation of the representation point from the curve in any direction.

4. Criterion for positive energy effect in pulsed combustion reaction

Each combustion pulse in which the energy

$$W = \hbar\omega_{\mathrm{mg}}\Delta N, \tag{11}$$

of metastable states is released is initiated by an external ignition with the energy deposition [2]

$$\Delta W = \hbar\omega_0\frac{\chi}{k_{\mathrm{B}}}(\theta_2 - \theta_1)\Delta N, \tag{12}$$

where ΔN is the number of nuclei in which an anti-Stokes transition has occurred and $\theta_2 - \theta_1 < 0.25$ is the difference between the normalised temperatures of the middle threshold branch and the lower stable branch of the S-shaped curve corresponding to the steady-state solution of the balance equations [1, 2]. Obviously, a positive physical energy effect is achieved for $W > \Delta W$, i.e., under the condition

$$\frac{\chi}{k_{\mathrm{B}}} < 4\frac{\hbar\omega_{\mathrm{mg}}}{\hbar\omega_0} < \frac{\hbar\omega_{\mathrm{mg}}}{\hbar\omega_0}(\theta_2 - \theta_1)^{-1}, \tag{13}$$

which can be easily fulfilled in many cases. Naturally, inevitable losses may make this criterion more stringent.

5. Resonance accuracy for nuclei with the line spectrum of ions in a plasma

It was mentioned above that the ratio ψ_0/Ψ can be increased considerably by deliberately introducing into plasma the alien ions with emission lines coinciding with the trigger transition in an isomeric nucleus. Unfortunately, the available tabulated values of the energy of nuclear transitions and wavelengths of characteristic X-ray lines have a relative error approximately equal to 0.001 in most cases, so that it is difficult to reliably determine the exact resonances of nuclei and ions (atoms).

This difficulty is partially alleviated by the high plasma temperature. The relative Doppler broadening of nuclear and ionic (atomic) lines in the high-temperature plasma

$$\frac{\hbar\omega_{\mathrm{D}}}{E_{\mathrm{mt}}} \approx 8\times10^{-5}\left(\frac{T}{A}\right)^{1/2} \tag{14}$$

(here, A is the number of nucleons in a nucleus; $E_{\mathrm{mt}} = \hbar\omega_0$; T is measured in electronvolts), which facilitates the overlap of resonance curves, may become almost equal to the errors in the tabulated values for the corresponding

[1]In formula (13) in Ref. [1], expressions for n^* and q contain misprints: the factor η is missing in the denominators.

transitions (e.g., ~ 0.0001 at a temperature of 100 eV). Note that the displacements of the lines due to a recoil, which are smaller than the above-mentioned values by several orders of magnitude, may be ignored altogether.

6. Combustion reaction in case of nonradiative decay channels

In a number of cases, when the probability of nonradiative decay of the trigger level t becomes comparable with the probability of the t $\rightarrow$ g gamma transition, the corresponding nonradiative processes (primarily, the internal electron conversion) should be taken into account. The existence of this decay channel, in which the energy $E_t - E_g$ is released in the form of the kinetic energy of an electron, makes it possible for the plasma to absorb efficiently this energy upon the application of an external magnetic field preventing the escape of electrons from the reaction zone of a small size. This removes the limitations imposed on the size of the reaction zone, which were considered in Section 1 in Ref. [2], and simultaneously makes it possible to increase the coefficient η to its maximum possible value $\eta_{max} = (1 + \alpha^{-1})^{-1}$, where α is the coefficient of internal electron conversion; we assume here that the absorption of gamma quanta emitted along with conversion electrons is absent.

By the way, the presence of fast internal conversion electrons leaving a magnetic trap (e.g., along the magnetic field lines) suggests that the energy of isomer combustion reaction may be directly transformed into the electric energy through the deceleration of these electrons by an external electric field.

7. Isomeric nucleus $^{242}_{95}$Am (approximate calculation of possible reaction parameters)

The isomeric nucleus $^{242}_{95}$Am [5] has attracted the attention of researchers owing to its easy availability. In the energy range of interest, this nucleus possesses three energy levels: the ground level with the angular momentum $J_g = 1^-$, metastable level with the energy $E_m = 48.63$ keV, $J_m = 5^-$ and the total lifetime $\tau_{mg} = 141$ years, and trigger level with $E_t = 52.9$ keV and $J_t = 3^-$. Thus, the energy of a trigger photon is $\hbar\omega_0 = 4.27$ keV and the multipolarities of the m $\rightarrow$ g, t $\rightarrow$ m and t $\rightarrow$ g transitions are E4, E2, and E2, respectively. The ground state is unstable and decays with a half-period of 19.02 h (beta emission and electron capture). Unfortunately, other experimental results concerning this nucleus were not available and were estimated from tables and nomograms from Ref. [6] as $\Gamma_{tm} \approx 4.6 \times 10^{-16}$ eV, $\alpha_{tm} \approx 6000$, $\Gamma_{tg} \approx 2.3 \times 10^{-11}$ eV, $\alpha_{tg} \approx 3000$; as a result, the characteristic time (7) of the anti-Stokes transition is $\tau_0 \approx 10$ s.

Such a long characteristic time raises hopes of realising the continuous combustion regime. The normalisation of plasma concentration (in cm^{-3}) $n^* \approx 2.65 \times 10^{34} S/V$ is quite large (we assume here that $\eta = 1$, which is quite plausible according to the results presented in Section 6 in view of the large value of the internal electron conversion coefficient $\alpha_{tg} \approx 3000$). This normalisation may be compensated by the smallness of the ratio Ψ/ψ_0; for example, for $\Psi/\psi_0 = 10^{-17}$ (see Section 2), the effective normalisation (in cm^{-3}) is $n^*_{eff} = 2.65 \times 10^{17} S/V$. This enables one to maintain the steady-state temperature $\theta = 0.57$ (i.e., $k_B \approx 2.45$ keV), say, for $V/S = 1$ cm and $n/n^*_{eff} = 0.5$. Accordingly, the time of complete combustion of isomeric nuclei (expression (10) from Ref. [1]) and, hence, the average desirable residence time Δt_{mtg} for metastable nuclei in the reaction zone are found to be 30 s.

The regime of continuous nuclear combustion with a power of 3.4 kW requires that the reaction zone be replenished with metastable nuclei at the rate $\Phi = nV/\Delta t_{mtg}$, which amounts, for example, to $\sim 4.4 \times 10^{17}$ nuclei per second for $V = 100$ cm^3. If such a replenishment is realised in the form of an ionic flow, the total ionic current of the order of 70 mA is found to be sufficient for this purpose. The output energy buildup is determined by potentialities for increasing the reaction volume V and by limitations associated with plasma instability. The construction of the reactor and the system of magnetic confinement of the plasma as well as the theoretical and experimental optimisation of its emission spectrum (minimisation of the ratio Ψ/ψ_0) are the subject of further analysis. The main nuclear parameters (the level widths and coefficients of internal electron conversion), which were adopted above as theoretical estimates, also require the experimental refinement. Taking these circumstances into account, the $^{242}_{95}$Am nucleus offers a considerable promise for preliminary experimental investigations.

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