

CRITICAL CONCENTRATION FOR STIMULATED AMPLIFICATION OF GAMMA RAYS

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Received 28 June 1991

Abstract. In this paper it is demonstrated that the critical concentration of excited nucleus and the development of powerful enough gamma-laser are possible in the near future. At the present level of technology, the amplification of gamma-lasers could be of more than 10 orders of magnitude, and the power of coherent radiation — more than one kilowatt. The shape and amplitude of the laser pulse are derived as a function of the active elements cooling rate. The differences between gamma-lasers and the visible light lasers are stated. Notwithstanding the high amplification and the single-time action, the stimulated emission of some real radionuclides can last long enough for all scientific experiments and technological applications. The new relations between the Einstein's coefficients and new cross-sections for stimulated emission (absorption) help to deeper understanding of stimulated emission, stars atmospheres, plasma physics, quantum theory of radiation and many other physics processes.

Резюме. В статье показано, что достижение критической концентрации возбужденных ядер и создание достаточно мощных гамма-лазеров возможно в ближайшем будущем. При современном уровне технологии усиление гамма-лазеров может быть более чем на 10 порядков, а мощность когерентного излучения более одного киловатта. Форма и амплитуда лазерного импульса получены как функция скорости охлаждения активного элемента. Установлены отличия от световых лазеров. Несмотря на высокое усиление и однократность действия, стимулированное излучение ряда реальных радионуклидов может быть достаточно долгим для научных экспериментов и технологических применений. Новые соотношения между коэффициентами Эйнштейна и новые значения сечения для стимулированного излучения помогают более углубленному пониманию стимулированного излучения, физики плазмы, звездных атмосфер, квантовой теории излучения.

1. Introduction

A paper was published in 1965 [1], where it was intuitively shown that stimulated emission of recoilless gamma-quanta between two excited levels of the atomic nucleus could be observed under the following conditions: the upper excited level m must have a long lifetime $\tau_m \gg 1$ s, while the lower level n must be short-living with $\tau_n < 10^{-6}$ s. The advantages of such nuclides are several: high population inversion of the active transition, sufficiently large linewidth ($\Gamma_m + \Gamma_n$) which is not affected by the crystalline structure of the substance. Furthermore, it was pointed out [1], that the energy of the $m \rightarrow n$ gamma-transition should satisfy the condition $E_{mn} < 150$ keV so that the recoilless emission probability be big enough. The isomer ^{125m}Te was quoted as an example (its scheme of decay is shown in Fig. 1).

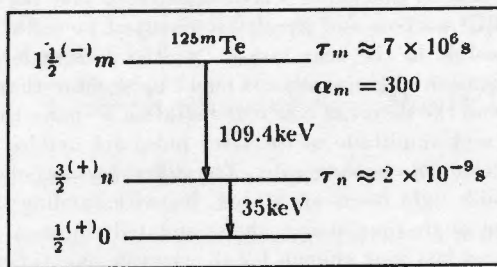


Fig. 1. Scheme of decay of ^{125m}Te with the necessary parameters

A series of articles discussed in [2] appeared later maintaining that stimulated emission of long-living isomers cannot be observed at all. Such wrong assertion is due to the incorrect calculation of the stimulated emission cross-section σ . The authors [2] consider that the cross-section is described by the expression:

$$\sigma = \frac{\lambda^2}{2\pi} \cdot \frac{f\Gamma_{m\gamma}}{\Gamma} \approx 10^{-36} \text{ cm}^2 \quad (\text{for } ^{125m}\text{Te}). \quad (1)$$

This is negligibly small as compared with the cross-section σ_0 of the total absorption and re-emission with the same energy E_{mn} . In (1) λ stands for the transition wavelength; $\Gamma_{m\gamma}$ is the partial width of the upper level for emission of gamma-quanta; $\Gamma = \Gamma_m + \Gamma_n$ is the total gamma linewidth, and f is the portion of recoillessly emitted gamma-quanta. Again in [2], the conclusion made is wrong, i.e. if τ_m is large, then both primary and secondary gamma-quanta will not travel simultaneously because of the time-dependence of the cross-section σ and will be detected as two independent quanta within an average time interval τ_m .

In 1986 Russian authors [3,4] set an experiment with the proposed in [1] ^{125m}Te after the design described in [5]. In these experiments [3,4] for the first time were observed stimulated couples of gamma-quanta travelling in the same direction, detected simulatenously as a quantum with double energy $E_\gamma = 2E_{mn}$. The experimentally obtained cross-section of the stimulated emission $\sigma_{\text{exp}} = (8.4 \pm 1.7) \times 10^{-19} \text{ cm}^2$ is calculated precisely enough in [4]. It differs by about 18 orders of magnitude from the value derived after expression (1). The experiments

[3,4] were analyzed in [6,7] and correct theoretical cross-sections $\sigma_{\gamma\gamma}$ were found for a series of isomers similar to the one from Fig. 1. It was demonstrated that the cross-sections of stimulated emission at the resonance maximum ($\nu = \nu_0$) are defined by the expression:

$$\sigma_{\gamma\gamma} = \frac{\lambda^2}{2\pi} \cdot \frac{(g_m f \Gamma_{m\gamma} + g_n \Gamma_n)}{g_m (\Gamma_m + \Gamma_n)} \quad (2)$$

where g_m and g_n are the corresponding statistical weights of levels m and n . Since $\sigma_{\gamma\gamma}$ is the cross-section of coherent emission of gamma-quanta, a conclusion was drawn that in the experiments [3,4] gamma-quanta emitted with recoil by the secondary nuclei have been also detected. Therefore $\sigma_{\gamma\gamma} = f_2 \sigma_{\text{exp}}$, where f_2 is the probability of stimulated and recoilless emission from the secondary nucleus. The latter is understood already from Einstein's works [8,9]. The recoilless fraction $f_2 = f$ in the experiments [3,4] can be derived both from the experimental and the theoretical cross-section, $f_2 = \sigma_{\gamma\gamma} / \sigma_{\text{exp}} = 0.12$, which very well coincides with the expected value [1,3,4]. The cross-section of the stimulated emission $\sigma_{\gamma\gamma}$ indicates that the development of a gamma-laser is entirely possible. The most important special features of such gamma-laser is that the achievement of the population inversion and the radiation amplification are separated in time. Inverted population can be achieved, for example, by irradiation in a nuclear reactor, and only after that the active element (a long and thin needle) is cooled in order to reach stimulated amplification. Those new cross-sections for stimulated emission (absorption) can help to deeper understanding of different domains of science: stellar atmospheres, plasma physics, quantum theory of radiation.

The objective of the present work is to find out how the shape and the amplitude of the stimulated amplification change with the time of cooling the active element (raising f) and how this depends on the source parameters.

2. Radiation Amplification

For the specific radionuclide ^{125m}Te on which we shall concentrate all further investigations, $f \Gamma_{m\gamma} < \Gamma_m \ll \Gamma_n$ and the total cross-section of recoilless coherent emission is:

$$\sigma_{\gamma\gamma} = \frac{g_n}{g_m} \cdot \frac{\lambda^2}{4} \cdot \frac{\Gamma_n}{\Gamma_m} = 1.03 \times 10^{-19} \text{ cm}^2 \quad (\text{for } ^{125m}\text{Te}). \quad (3)$$

It is much bigger than the cross-section of total absorption and scattering σ_0 for the transition energy E_{mn} ($\sigma_0 = 3.8 \times 10^{-22} \text{ cm}^2$). As it is well known, the inequality $\sigma_{\gamma\gamma} > \sigma_0$ leads to the possibility the linear amplification factor $\beta = f \sigma_{\gamma\gamma} n$ to be greater than the linear total absorption factor $\mu = \sigma_0 n_0$. n is the number of excited nuclei per unit volume, and n_0 is the number of all atoms per unit volume of the active element. As it can be seen, the amplification factor β depends on the recoilless fraction f and therefore on the source temperature T .

The active element is outlined in Fig. 2 — it is a long cylinder composed of BeTe (as in the experiments [3,4]). The population inversion of levels m and n is defined by the ratio of the corresponding lifetimes τ_m and τ_n :

$$\frac{\eta_m}{\eta_n} = \frac{\tau_m}{\tau_n} \approx 3.5 \times 10^{15}. \quad (4)$$

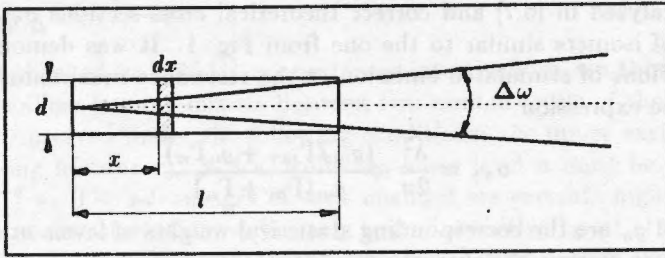


Fig. 2. Layout of the active element of the source

Such population inversion is impossible in the optical domain. Let us assume that the condition $d \ll l$ is fulfilled. The initial specific volume activity is a_0 . The radiation leaving a thin transverse layer dx passes through the distance $(l-x)$ and interacts with the atoms and nuclei. We are interested only in the fraction falling into a solid angle $\Delta\omega = d^2/16l^2$ (along the cylinder's axis). Hence the number of gamma-quanta per unit time as a function of x will be:

$$\frac{dN_{\gamma\gamma}(x)}{dt} = \frac{fa_0\Delta\omega\Delta s dx}{(1+\alpha_m)} e^{(\beta-\mu)(l-x)} + \frac{(1-f)a_0\Delta\omega\Delta s dx}{(1+\alpha_m)} e^{-\mu(l-x)}. \quad (5)$$

Integration over x yields:

$$\frac{dN_{\gamma\gamma}}{dt} = \frac{fa\Delta\omega\Delta s}{(1+\alpha_m)(\beta-\mu)} (e^{(\beta-\mu)l} - 1) + \frac{(1-f)a\Delta\omega\Delta s}{(1+\alpha_m)\mu} (1 - e^{-\mu l}). \quad (6)$$

Here α_m is the inner conversion factor of the $m \rightarrow n$ transition; Δs is the active element's section. The first term corresponds to the intensity of primary gamma-quanta emitted without recoil, and the second one — to those emitted with recoil. At high temperatures $f \rightarrow 0$ and the radiation in both directions of the cylinder's axis becomes:

$$\frac{dN}{dt} = \frac{a_0\Delta\omega\Delta s}{(1+\alpha_m\mu)} (1 - e^{-\mu l}). \quad (7)$$

We shall define the amplification in two ways:

$$M = \frac{dN_{\gamma\gamma}}{dt} / \frac{dN}{dt} = \frac{f\mu(e^{(\beta-\mu)l} - 1)}{(\beta-\mu)(1 - e^{-\mu l})} + 1 - f \quad (8)$$

and

$$M_0 = \frac{dN_{\gamma\gamma}}{dt} / \frac{dN_0}{dt} = \frac{a}{a_0} \cdot \frac{f\mu(e^{(\beta-\mu)l} - 1)}{(\beta-\mu)(1 - e^{-\mu l})} + \frac{a}{a_0}(1 - f). \quad (9)$$

M is the amplification as referred to the diminishing with time activity, and M_0 is the amplification with respect to the initial moment when the activity is a_0 . Since activity decreases with time due to spontaneous decay e^{-t/τ_m} as well as to the intensity of stimulated emission, while a_0 is a fixed constant, M will be always greater than M_0 . We shall define the relative concentration of excited nuclei in state m in the following manner:

$$K = \frac{n}{n_0}; \quad n = Kn_0. \quad (10)$$

3. Dependence of f on Time upon Cooling

For low temperature the following expression is valid

$$f(T) = \exp[-(B + CT^2)]; \quad T \ll \theta \quad (11)$$

where $B = 3E_r/2k\theta$ and $C = \pi^2 E_r/k\theta^3$; E_r is the energy of recoil, k is Boltzmann's constant, θ — the Debye temperature. We assume that temperature decreases after an exponential law:

$$T = T_0 e^{-t/t_0} + T_{\text{He}}. \quad (12)$$

Here T_0 is the initial temperature (high), and $1/t_0$ is the temperature decrement rate. Therefore f as a function of the cooling time t becomes:

$$f(t) = \exp[-(B + C(T_0 e^{-t/t_0} + T_{\text{He}})^2)]. \quad (13)$$

T_{He} is the temperature of liquid helium. At $t \rightarrow \infty$ [3,4] $f = 0.12$ which makes it possible to define the parameters $B = 2.12$; $C = 7.7 \times 10^{-5}$; $\theta = 425$ K for BeTe.

4. Variation of the Excited Nuclei Concentration

Beside spontaneous decay, the excited nuclei concentration decrease will be accelerated by stimulated emission:

$$K(t) = K_0 e^{-t/\tau_m} - \frac{2}{n_0 W} \int_0^t \frac{dN_{\gamma\gamma}(t)}{dt} dt \quad (14)$$

where K_0 is the concentration at the initial moment $t = 0$. The integral presents all decayed nuclei (in the entire volume) due to stimulated emission $dN_{\gamma\gamma}/dt$. The decay takes place in both directions along the cylinder's axis, which yields the factor 2 and in order to express the relative concentration decrease, the denominator $n_0 W$ is introduced. W is the active element's volume, $W = \pi d^2 l/4$. The specific volume activities a and a_0 can be replaced everywhere by the expressions:

$$a = \frac{n_0 K(t)}{\tau_m}; \quad a_0 = \frac{n_0 K_0}{\tau_m}. \quad (15)$$

5. Amplification Factor

The linear amplification factor β does not noticeably depend on the resonant absorption for the $n \rightarrow m$ transition because of its enormous population inversion:

$$\beta = f(t) \sigma_{\gamma\gamma} n_0 K(t) - f(t) \frac{g_m}{g_n} \sigma_{\gamma\gamma} (K(t) n_0 \frac{\tau_n}{\tau_m} + \frac{2\tau_n}{W} \cdot \frac{dN_{\gamma\gamma}(t)}{dt}). \quad (16)$$

The expression in small brackets of the second term is the volume concentration of nuclei in state n at time t . Under the condition $\tau_m \gg \tau_n$ its value stays smaller than $K(t)n_0$ by many orders of magnitude for all possible amplifications and the second term will be further omitted. Obviously, amplification will take place only

if the inequality $\beta_{\gamma\gamma} > \mu$ is fulfilled. The critical concentration K_0 above which amplification is expected is derived from the equality:

$$\beta - \mu = f(\text{He})\sigma_{\gamma\gamma}K_0n_0 - \sigma_0n_0 = 0; \quad K_0 = \frac{\sigma_0}{f(\text{He})\sigma_{\gamma\gamma}}. \quad (17)$$

For BeTe $K_0 = 0.0307$.

6. Results of Calculations

For the numerical calculations expressions (6-9) were used and the integral in (14) was replaced by a sum:

$$\int_0^t \frac{dN_{\gamma\gamma}(t)}{dt} dt = \Delta t \sum_i \frac{dN_{\gamma\gamma}(i)}{dt} \quad (18)$$

where Δt is the time increment. The initial condition $t = 0$ leads to $f(t) = 0$ and $K(t) = K_0$, which yields expression (7) for the radiation along the cylinder's axis.

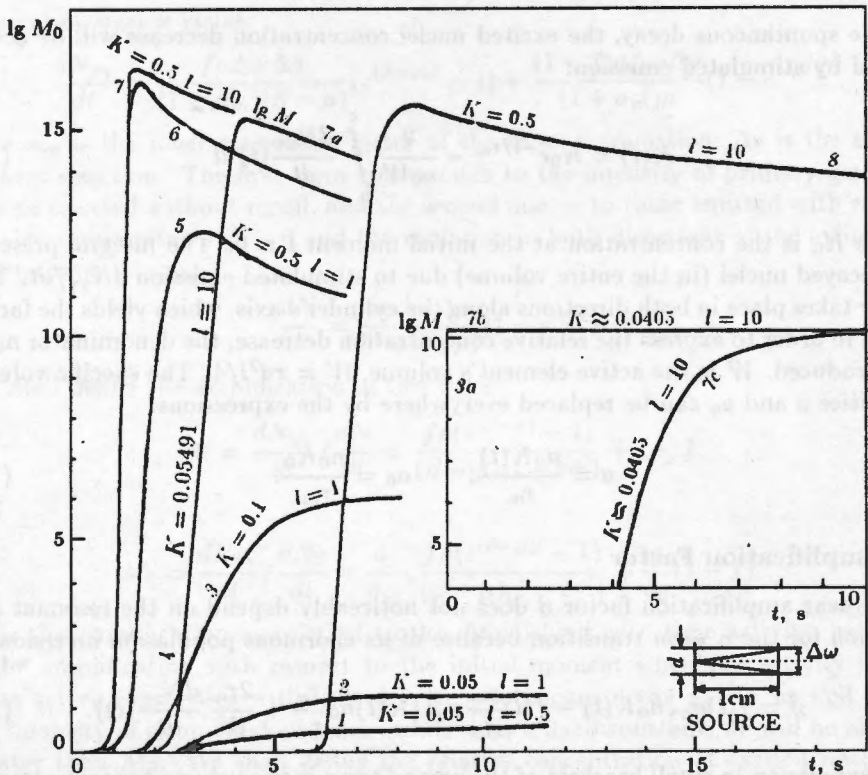


Fig. 3. Dependence of $\lg M_0$ on time of cooling. The explanations are in the text

Temperature decreases with time and $f(t)$ increases. All calculations are performed for BeTe, the active element's length l and the initial concentration K_0 are shown in the Figures. Everywhere $d = 0.1$ cm. The measurement of time in seconds is arbitrary since its replacement with hours or minutes does not alter anything.

The dependence of $\lg M_0$ on t for source of various length l and initial concentration K_0 is plotted in Fig. 3. The parameter t_0 equals 2 s everywhere with the exception of curve 8, where $t_0 = 10$ s. The rest parameters are: $\sigma_{\gamma\gamma} = 1.03 \times 10^{-19}$ cm²; $\sigma_0 = 3.8 \times 10^{-22}$ cm²; $n_0 = 2 \times 10^{22}$; $f = 0.12$ at helium temperature (4 K). As it is evident, at entirely accessible supercritical concentration from $K = 0.05$ to $K = 0.5$ the amplification can be as high as $M_0 = 5$ to $M_0 = 10^{12}$ even for relatively short sources with $l = 0.5$ cm to $l = 1$ cm.

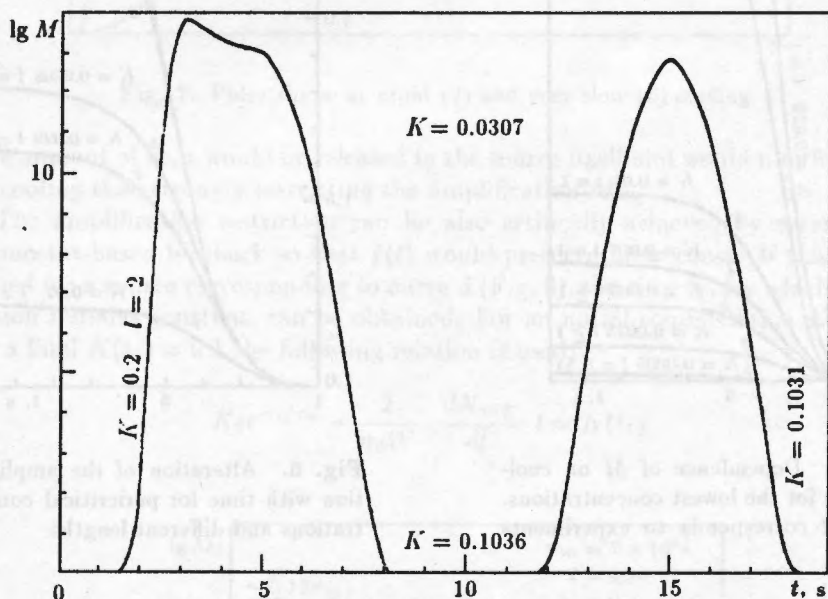


Fig. 4. Two sequential pulses with intermediate heating and cooling of the source

$M \approx 10^{16}$ for curve 6 where $l = 10$ cm. The amplifications 6 and 7 differ only in the definition (expressions (6) and (9)). (Curves 7(a, b) are for $\lg M$ and they are always higher than curve 6 ($\lg M_0$)). The amplification 7 is cut off at the 4th second (sudden heating) and the concentration is $K = 0.0549$. At this concentration the source starts cooling down with the same rate, which corresponds to 7(a). The amplification after 10^3 s (7b) is shown in Fig. 3a. The curve remains practically horizontal for 10 s but the concentration becomes $K = 0.0405$ (7b). At a new sudden heating $\lg M = 0$ and after another cooling curve 7c appears. Thus the same source may be heated and cooled repeatedly. The amplification diminishes gradually but remains sufficiently high. Two pulses are shown in Fig. 4 for which cooling and heating are held at the same rate, $t_0 = 2$ s. For the first pulse heating begins at the 5th s and cooling for the second one — at the 10th s.

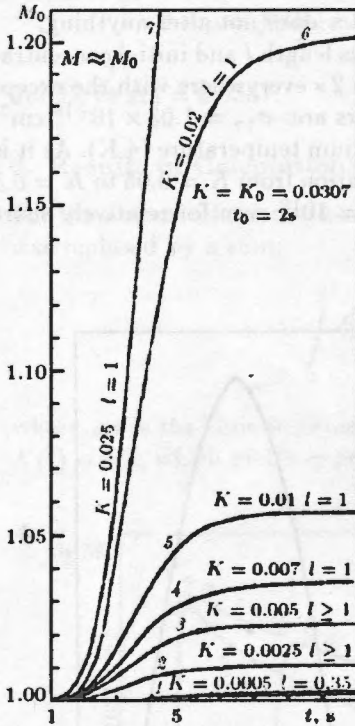


Fig. 5. Dependence of M on cooling time for the lowest concentrations. Curve 1 corresponds to experiments [3,4]

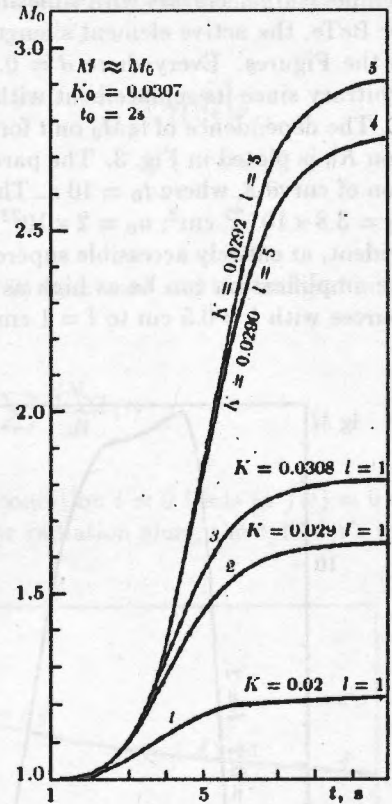


Fig. 6. Alteration of the amplification with time for pericritical concentrations and different lengths

The amplification for subcritical concentrations $K < K_0 = 0.0307$ are shown in Figs 5 and 6. $M \approx M_0$ for such amplifications. It is interesting to note that curve 1 in Fig. 5 corresponds approximately to the experimental results [3,4]. The amplification after reaching helium temperatures can be calculated according to [4]: $M \approx M_0 = 1.002$. The thickness of curve 1 corresponds to the experimental errors. It can be stated, that at a sudden (rapid) cooling the curve is of the type shown in Fig. 7(1), while at a slow cooling it is of type 2 (Fig. 7). The areas under the curves are equal.

7. Power of the Coherent Radiation

As an example we shall dwell upon curve 5 from Fig. 3, for which $K = 0.5$ and $l = 1$ cm, $d = 0.1$ cm. At an amplification $M \approx 10^9$ the coherent radiation power within the angle $\Delta\omega$ is approximately 1 W, and at amplification 10^{12} the power is approximately 10^3 W. Obviously, at powers as high as that, the approximately

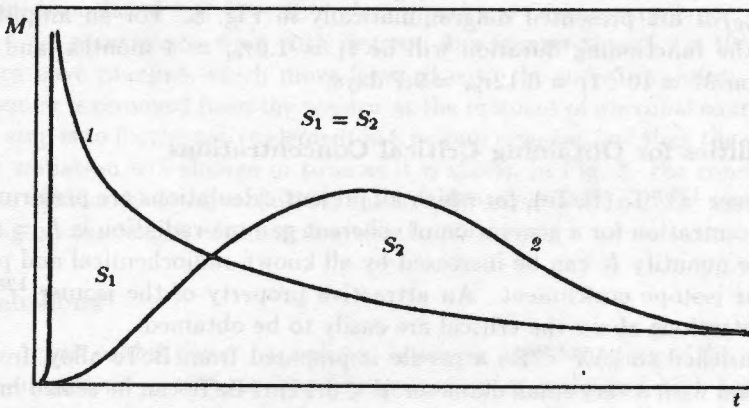


Fig. 7. Pulse shape at rapid (1) and very slow (2) cooling

same amount of heat would be released in the source itself and would interfere with the cooling thus strongly restricting the amplification M .

The amplification restriction can be also artificially achieved by arranging a thermostat-based feedback so that $f(t)$ would preserve $M = \text{const}$. If this is performed for a source corresponding to curve 5 (Fig. 3), the time t_1 , for which amplification remains constant, can be obtained. For an initial concentration $K_0 = 0.5$ and a final $K(t_1) = 0.1$ the following relation is used:

$$K_0 e^{-t/\tau_m} - \frac{2}{n_0 W} \cdot \frac{dN_{\gamma\gamma 0}}{dt} \cdot t = K(t_1). \quad (19)$$

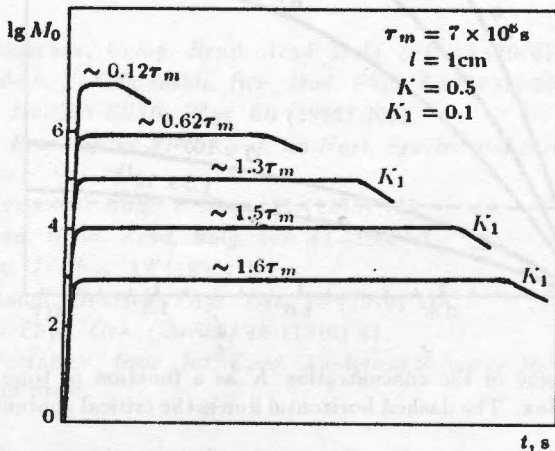


Fig. 8. Artificial restriction of the amplification $\lg M$ and functioning duration of the coherent radiation source

The left-hand side gives concentration alteration with time. $dN_{\gamma\gamma 0}/dt = \text{const}$ at constant amplification. Such pulses, restricted at different amplification levels $M \sim dN_{\gamma\gamma 0}/dt$ are presented diagrammatically in Fig. 8. For an amplification $M = 10^3$ the functioning duration will be $t_1 = 1.6\tau_m = 4$ months, and for an amplification $M = 10^7$, $t_1 = 0.12\tau_m = 9.7$ days.

8. Possibilities for Obtaining Critical Concentrations

For our isomer ^{125m}Te (BeTe), for which all present calculations are performed, the critical concentration for a generation of coherent gamma-radiation is $K = n/n_0 = 0.0307$. The quantity K can be increased by all known radiochemical and physical methods for isotope enrichment. An attractive property of the isomer ^{125m}Te is that concentrations above the critical are easily to be obtained.

From enriched to 95% ^{124}Te , a needle is prepared from BeTe alloy, from 1 to 5 cm long and with a very small diameter, $d < 0.1$ cm. BeTe can be sealed in quartz or other capillary. The needle is irradiated with thermal neutrons in a reactor for time t . The concentration $K(t)$ of ^{125m}Te is calculated by the expression:

$$K(t) = \frac{n(t)}{n_0} = \frac{\phi \sigma_n \theta \tau_m}{(1 - \phi \sigma_\alpha \tau_m)} \left[e^{-\phi \sigma_\alpha \tau_m (t/\tau_m)} - e^{-t/\tau_m} \right]. \quad (20)$$

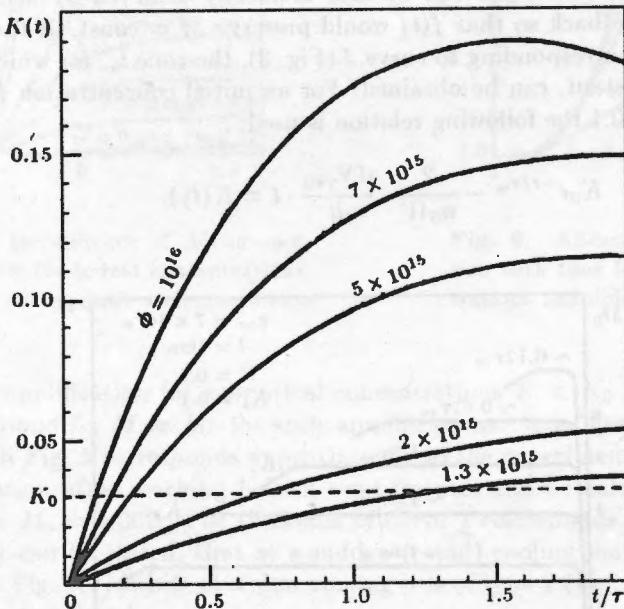


Fig. 9. The change of the concentration K as a function of time of irradiation with different neutron flux. The dashed horizontal line is the critical concentration $K_0 = 0.0307$

Here ϕ ($\text{cm}^{-2}\text{s}^{-4}$) is the neutron flux, σ_n (cm^2) is the cross-section for the production of ^{125m}Te , θ is the isotope enrichment of ^{124}Te , τ_m is the mean lifetime of the isomer level m , the concentration of which is the relevant one, σ_α is the cross-section for neutron capture, ($\sigma_\alpha = 6.8b$, $\sigma_n = 5b$).

The results calculated according to (20) for different neutron flux are plotted in Fig. 9.

For neutron flux $\phi \geq 1.12 \times 10^{15} \text{ n/cm}^2\text{s}^{-1}$ critical concentrations are achievable and there are research reactors with neutron flux greater than $1.2 \times 10^{15} \text{ n/cm}^2\text{s}^{-1}$. All curves have maxima which move forward with the increasing of ϕ . If the obtained isomer is removed from the reactor at the moment of maximal concentration, the next step is to fix the active element in a helium cryostat and then the stimulated coherent radiation will change in time as it is shown in Fig. 3. For concentrations above the critical $K > K_0 = 0.0307$, the amplification will increase exponentially depending on the length of the active element l .

9. Conclusions

The main result of this paper, we believe, is deeper understanding of the stimulated emission of gamma-quanta, spectral investigation of stars and high temperature plasma, quantum theory of radiation. From the exposed above is evident, that the creation of the source of powerful coherent gamma-radiation is entirely accessible at the current state of experimental technology. If no efforts for separation of the excited nuclei from all the rest are made, the supercritical concentrations $K > K_0$ can be reached by irradiation of ^{124}Te in a neutron flux $\phi \approx 1 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ for time t_{max} . For the same time, at normal neutron fluxes $\phi \approx 3 \times 10^{13} \text{ cm}^{-2}\text{s}^{-1}$ the amount of ^{124}Te to be irradiated should be about 1.5 grams and subsequent radiochemical or similar work is necessary to separate the excited nuclei from the rest of the substance.

In all cases, difficulties (sometimes insurmountable) noted and discussed in [2] seem to us now an inexplicable delusion. The present and other [10] investigations make us confident that an amplification $M \geq 10^9$ is entirely real and such a powerful source of coherent gamma-radiation will appear in short time.

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