Nuclear Gamma-Ray Laser of Optical Range.

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A possibility of amplification of the 7.6 eV (3.5 eV) γ-radiation by the stimulated γ-emission of the ensemble of the ^{229m}Th isomeric nuclei in a host dielectric crystal with a large band gap is proved theoretically. This amplification is a result of the following three factors: 1) the excitation of a great number of the ^{229m}Th isomers by laser radiation; 2) the creation of the inverse population of nuclear levels in a cooled sample placed in magnetic field; 3) the emissions/absorption of the optical photons by thorium nuclei in the crystal without recoil (the Mössbauer effect in the optical range).

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Nuclear gamma-ray laser is a known challenge of the modern physics. To succeed in the field, one has to overcome at least two basic problems, namely: to accumulate a sufficient amount of isomeric nuclei in a sample, and to narrow down the emission gamma-ray line to its natural radiative width. These and other problems, as well as early unsuccessful attempts of their solution are discussed in detail in reviews [1–3] (see also list of previous reviews in [2]). After reading these works one naturally comes to the conclusion that the problem of gamma-ray laser working on nuclear transitions cannot be solved by traditional methods [3].

Nevertheless, there is a plausible way to overcome all difficulties and to develop a gamma-ray laser working on magnetic dipole ($M1$) transition in the optical range between the first excited level and the ground state of the ^{229}Th nucleus. The energy, E_{is}, of the abnormally low-lying level in ^{229}Th is 7.6±0.5 eV according to the recent study [4]. Earlier measurements gave $E_{is} = 3.5 \pm 1.0$ eV [5]. The work is now on the way to determine the energy of the $J^{\pi}_{is} = 3/2^+$ isomeric level with a high precision. There are some experimental schemes, which allow experimentalists to make such measurements. For example, the ^{229m}Th(3/2$^+$, 7.6 eV) isomeric nuclei can be generated by a direct photoexcitation from the ^{229}Th nuclei in the ground state $J^{\pi}_{gr} = 5/2^+$. A laser of the vacuum ultraviolet (VUV) region can serve as a source of photons, if its line of radiation is tuned at the nuclear transition [6, 7]. Other promising sources are synchrotron radiation in the VUV region [8] and resonant part of the radiation spectrum of special lamps [6]. It is very important that for both estimations of the isomeric energy E_{is} one can find a working compound with an energy gap of the forbidden electronic states (Δ), such that $\Delta > E_{is}$. For example, for $E_{is} = 3.5 \pm 1.0$ eV the suitable candidate is the ThO_2 crystal, where the $^{229}Th^{4+}$ ions are "separated" from the conduction band by the 6 eV energy gap [9, 10]. If the correct estimation is $E_{is} = 7.6 \pm 0.5$ eV then according to [8] ^{229}Th nuclei can be implanted in $LiCaAlF_6$. There, the $^{229}Th^{4+}$ ions substitute for the Ca^{2+} ions and lead to the formation of the Th:$LiCaAlF_6$ alloy. The expected energy gap for Th:$LiCaAlF_6$ is 10 eV [8].

Thus there is a unique situation for low-energy nuclear spectroscopy both in $^{229}ThO_2$, and in ^{229}Th:$LiCaAlF_6$. There, a photon is expected to interact directly with the atomic nucleus thereby bypassing the interaction with the electron shells [9], i.e. the internal conversion and electronic bridge [7, 10]. This opens up many interesting perspectives and possible applications. Below we name some of them: the gamma-ray emission of the excited ^{229}Th nuclei in the optical range called the "nuclear light" [7]; a check of the dependence of the isomeric level half life on the refractive index n [9, 10]; the nuclear metrological standard of frequency [6] called the "nuclear clock" [11]; the Mössbauer effect in the optical range [6] with an extremely small ratio of the line width to the gamma-ray energy; the investigation of relative effects of the variation of the fine structure constant e^2 and the dimensionless strong interaction parameter m_q/Λ_{QCD} [12]; a check of the exponentiality of the decay law of an isolated metastable state at long times [13] and so on. The main subject of consideration in the present work is the nuclear gamma-ray laser in the optical range, which is the most interesting and nontrivial application of the ^{229m}Th isomer.

The most important characteristic of the gamma-ray laser is its amplification factor. The amplification coefficient χ of gamma radiation caused by stimulated emission in a medium is given [1–3] by

$$\chi = L\left(\frac{\lambda_{is}^2}{2\pi}\frac{\Gamma_{rad}}{\Delta\omega_{tot}}\frac{1}{1+\alpha}\left(n_{is} - \frac{n_{gr}}{g}\right) - \mu\right). \quad (1)$$

Here we work in the system of units $\hbar = c = k = 1$, L is the length of the sample, $\lambda_{is} = 2\pi/E_{is}$ is the wavelength of the isomeric transition, Γ_{rad} is the radiative width of the nuclear transition from the isomeric level to the ground state (or the natural width of the transition line), $\Delta\omega_{tot}$ is the total width of the transition line which accounts for all the types of the homogeneous and inhomogeneous broadening, α is the coefficient of the electronic conversion, n_{is} is the density of isomeric nuclei, n_{gr} is the density of nuclei in the ground state, $g = g_{gr}/g_{is}$ where $g_{gr(is)} = 2J_{gr(is)} + 1$ is the statistical weight of the ground (isomeric) state, and μ is the linear attenuation coefficient for resonant photons with the energy E_{is} in the medium in all processes excluding the resonant excitations of the nuclear isomer from the ground state. It is evident that the amplification of the gamma radiation occurs if $\chi > 1$.

The key parameter in formula (1) is the radiating width Γ_{rad} which requires the evaluation of the nuclear matrix element of the $M1$ transition $3/2^+(E_{is}) \to 5/2^+(0.0)$. One can calculate this matrix element using the Alaga rules and experimental data [14–16] for the $M1$ transition $9/2^+$(97.13 keV)$\to 7/2^+$(71.82 keV) [17] (these states together with the ground and isomeric ones belong correspondingly to the same rotational bands of the ^{229}Th nucleus). Recalculation of the experimental data obtained in [14–16] gives respectively the following values for the reduced probability of the nuclear isomeric transition in terms of the Weisskopf units $B_{W.u.}(M1; is \to gr) = 0.038$, 0.024, and 0.014. The data spread is considerable. The Coriolis interaction between rotational bands of the ground and isomeric states enhances the $is \to gr$ transition probability, but only marginally: by a factor of 1.2-1.3 [17]. That is why one can use the Alaga rules for the preliminary estimation of the reduced probability of the transition with its average value $B_{W.u.}(M1; is \to gr) \approx 0.025$.

In the following we take $E_{is} = 7.6 \pm 0.5$ eV and $\lambda_{is} = 163 \pm 11$ nm for the numerical estimations (generalization to the case $E_{is} = 3.5 \pm 1.0$ eV is trivial). The average value of the radiative width of the $M1$ transition $3/2^+$(7.6 eV)$\to 5/2^+$(0.0) is $\Gamma_{rad}(M1; is \to gr) \approx 3 \times 10^{-17}$ eV.

In the ^{229}Th:LiCaAlF$_6$ crystal the Th^{4+} ions have the closed electron shell structure of Radon [8]. Since the band gap in this crystal exceeds the energy of the isomeric transition in ^{229}Th, the internal conversion of the $3/2^+$(7.6 eV) level is forbidden and $\alpha = 0$. The probability of the electron bridge [18] (a third order term in the electromagnetic interaction constant e) is very small [9] in comparison with the probability of the gamma radiation and can be neglected.

In the ^{229}Th:LiCaAlF$_6$ crystal the line width $\Delta\omega_{tot}$ of the transition, formula (1), is determined mainly by the interaction of the nuclear magnetic moments [8]. It is well known that the magnetic moment of the ground state $\mu_{gr} = \mu_N$, where μ_N is the nuclear magneton. The magnetic moment of the isomeric level μ_{is} was calculated in [17] on the basis of the available experimental data at that time. New experimental data [15] for the difference $|g_R - g_K|$ of the rotational gyromagnetic ratio g_R and the intrinsic gyromagnetic factor g_K for the rotational band of the isomeric level $K^\pi = 3/2^+$ are in good agreement with the results of earlier measurements and lead to the following estimation: $\mu_{is} = (-0.07 \pm 0.03)\mu_N$. With these values for μ_{gr} and μ_{is} the upper value for the broadening of spectral line of the isomeric transition as 10 kHz [8] is approximated by $\Delta\omega_{tot} < 7 \times 10^{-12}$ eV.

Now we consider the conditions for the two-step creation of the inverse population in the system of the ^{229}Th nuclei. The first step is obtaining a considerable number of the isomeric nuclei. The most effective way is to excite the ^{229}Th nuclei at the isomeric level $3/2^+(7.6 \pm 0.5$ eV) by VUV laser irradiation of the transparent target, which contains the ^{229}Th nuclei. The equations for n_{is} and n_{gr} as functions of irradiation time are given by

$$\begin{aligned} dn_{is}/dt &= \sigma\varphi n_{gr} - \Lambda_{is} n_{is} - g\sigma\varphi n_{is}, \\ dn_{gr}/dt &= -\sigma\varphi n_{gr} + \Lambda_{is} n_{is} + g\sigma\varphi n_{is}. \end{aligned} \tag{2}$$

Here σ is the resonance cross section of nuclear photo excitation by laser radiation with the wavelength $\lambda_L = \lambda_{is}$ and the line width $\Delta\omega_L$: $\sigma = (\lambda_{is}^2/2\pi)\Gamma_{rad}(gr \to is)/\Delta\omega_L$. Λ_{is} is the decay constant of the isomeric level: $\Lambda_{is} = \ln(2)/T_{1/2}^{is}$, where $T_{1/2}^{is}$ is the level half-life. In our case $\Lambda_{is} = \Gamma_{rad}(M1; is \to gr)$ and $T_{1/2}^{is} \approx 15$ s (for this preliminary estimation we take $n = 1$ for the refractive index (details see in [9, 10]). The third terms on the right parts of Eqs. (2) describe the gamma emission of the isomeric nuclei stimulated by laser photons. Below we will demonstrate that both emission and absorption occur without recoil. Therefore if the laser radiation is in resonance with the nuclear transition $gr \to is$, it will be also in resonance with the return transition $is \to gr$.

The solutions of Eq. (2) with the initial conditions $n_{gr}(t = 0) = n_0$ and $n_{is}(t = 0) = 0$ are

$$\begin{aligned} n_{is} &= n_0 \frac{\sigma\varphi(1 - e^{-(\Lambda_{is} + (1+g)\sigma\varphi)t})}{\Lambda_{is} + (1+g)\sigma\varphi}, \\ n_{gr} &= n_0 \frac{\Lambda_{is} + \sigma\varphi(g + e^{-(\Lambda_{is} + (1+g)\sigma\varphi)t})}{\Lambda_{is} + (1+g)\sigma\varphi}. \end{aligned} \tag{3}$$

From Eqs. (3) it follows that it is impossible to obtain *real* inverse population, i.e. to fulfill the condition $n_{is} - n_{gr} > 0$ in our two level system with $g \equiv g_{gr}/g_{is} > 1$ by the method of laser pumping. The asymptotic value at $t \to \infty$ for $n_{gr}(t)/n_{is}(t)$ is $\Lambda_{is}/\sigma\varphi + g$. Notice that it is larger than one. It is impossible to fulfill the condition $n_{is} - n_{gr}/g > 0$ too. This more simple condition is necessary for the amplification of the gamma radiation (see formula (1)). On the other hand, by laser radiation pumping one can excite a considerable number of nuclei on the isomeric level. As an initial density of the ^{229}Th nuclei in the Th:LiCaAlF$_6$ crystal we take $n_0 = 10^{18}$ cm^{-3} [8], which amounts to 10^{-4} of the LiCaAlF$_6$ crystal density. Such a small number of the Th^{4+} ions (substituting for Ca^{2+} ions) in the LiCaAlF$_6$ crystal is not expected to change considerably the electronic band structure of the LiCaAlF$_6$ crystal.

For practical estimations we now consider a crystal of ^{229}Th:LiCaAlF$_6$ with the 0.1×0.1 cm^2 cross-section and

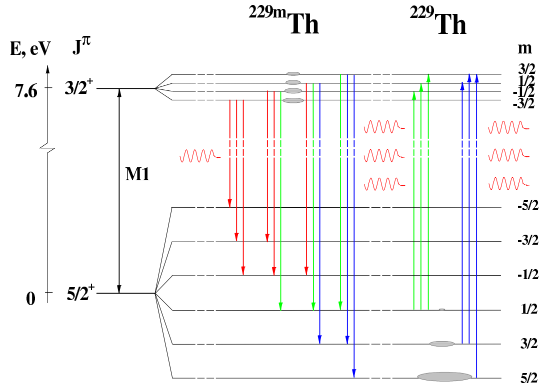

Figure 1: Color online. The transitions between the Zeeman sublevels of the ^{229}Th nucleus in magnetic field (see text for details). The gray ovals are proportional to partial level populations.

$L = 1$ cm length. Then for a VUV laser with the average power $P = 3$ W (this is a typical power for commercially available VUV lasers [19]) numerical estimations show that $n_{is}/n_{gr} \approx 1/3$, or $N_{is} \approx 2.5 \times 10^{15}$ during a period of time $t = (2-3)T^{is}_{1/2}$. Below we demonstrate that this quantity of isomeric nuclei in the sample is enough for the amplification of the spontaneous γ radiation.

The *effective* inverse population in the irradiated cooled sample is facilitated by the magnetic field. The Zeeman splitting of the nuclear levels is shown in Fig. 1. First we consider the case of a very low temperature, and a very strong magnetic field. Then only the lowest sublevels of the ground and isomeric states are populated. The selection rules for the $M1$ multipole give three transitions from the lowest sublevel $|J^{\pi}, m\rangle = |3/2^+, -3/2\rangle$ of the ground state to the upper sublevels $|5/2^+, -5/2\rangle$, $|5/2^+, -3/2\rangle$, and $|5/2^+, -1/2\rangle$ of the isomeric state. The sublevel $|3/2^+, -3/2\rangle$ is populated. The sublevels $|5/2^+, -5/2\rangle$, $|5/2^+, -3/2\rangle$, and $|5/2^+, -1/2\rangle$ are unoccupied. Therefore, we have the inverse population for these four Zeeman sublevels connected by the $M1$ transition. Since the radiative widths of considered transitions and the broadening of spectral line $\Delta\omega_{tot}$ by many orders of magnitude smaller than the typical distance between the Zeeman sublevels in the strong magnetic field (see below) the emitted photons can not excite the ^{229}Th nuclei from the lowest sublevel $|5/2^+, 5/2\rangle$ to the isomeric state, see Fig.1. Thus, in this case the necessary conditions for the amplification of the gamma radiation are satisfied.

Now we demonstrate that the *effective* inverse population and consequently the amplification of radiation are possible in a general case at the temperature $T = 0.01$ °K and in the static magnetic field H = 100 T. As we have already seen in the first case, the magnetic moments of the ground and isomeric states are very different. As a result, the ratio of the energy differences between the Zeeman sublevels of the ground and isomeric states is approximated by $|\mu_{gr}/\mu_{is}| \approx 6$. Therefore the population of the ground state sublevels falls down much faster, than the population of the isomeric sublevels. (Both populations are described by the Boltzmann distribution $\sim \exp(-\boldsymbol{\mu}_{gr(is)}\mathbf{H}/T)$.) The magnetic interaction energy μH is 1.4×10^{-6} eV for the ground state and 2.5×10^{-7} eV for the isomeric state. (Both values exceed considerably the radiative width $\Gamma_{rad}(M1; is \to gr)$ of the nuclear transition and the broadening of spectral line $\Delta\omega_{tot}$.) At the temperature $T = 0.01$ °K all sublevels of the isomeric state are populated (see Table 1). The population analysis of the ^{229}Th nuclei then gives 80% for the lowest sublevel $|5/2^+, 5/2\rangle$, 16% for $|5/2^+, 3/2\rangle$, and 3% for $|5/2^+, 1/2\rangle$. The population of the other sublevels is negligibly small.

Table I: The population of the Zeeman sublevels of the ^{229}Th nuclei.

Ground state	Population, cm^{-3}	Isomeric state	Population, cm^{-3}
$\|5/2^+, -5/2\rangle$	1.6×10^{14}	$\|3/2^+, 3/2\rangle$	4.0×10^{16}
$\|5/2^+, -3/2\rangle$	8.3×10^{14}	$\|3/2^+, 1/2\rangle$	5.3×10^{16}
$\|5/2^+, -1/2\rangle$	4.3×10^{15}	$\|3/2^+, -1/2\rangle$	7.1×10^{16}
$\|5/2^+, 1/2\rangle$	2.2×10^{16}	$\|3/2^+, -3/2\rangle$	9.5×10^{16}
$\|5/2^+, 3/2\rangle$	1.2×10^{17}		
$\|5/2^+, 5/2\rangle$	6.0×10^{17}		

Thus, the upper Zeeman sublevels of the ground state are unoccupied and transitions are possible to these states from the lower sublevels of the isomeric states. These transitions are shown by red arrows in Fig.1. Therefore, in the system of nuclear isomers an inverse population is effectively realized. In addition, it is worth noting that there is no resonant absorption for the photons emitted in these transitions. As one can see from Fig. 1 the energies of the "red" photos are too small to excite the ^{229}Th nuclei from the populated states $|5/2^+, 5/2\rangle$, $|5/2^+, 3/2\rangle$ and $|5/2^+, 1/2\rangle$ to the isomeric level.

The relative probability p of all these transitions taking into account the population of the states and the corresponding Clebsch-Gordan coefficients is 0.63. This value is then used for the calculation of the amplification factor, Eq. 1, where we put $n_{gr} = 0$ and pn_{is} instead of n_{is}. The result is $\chi \approx 20 - 25$ for the sample with $L = 1$ cm. (The attenuation coefficient $\mu \approx 3$ cm^{-1} was taken from [20].) It is clear that with mirrors χ is expected to increase considerably.

Next, we consider the following three transitions: $|3/2^+, -1/2\rangle \to |5/2^+, 1/2\rangle$, $|3/2^+, 1/2\rangle \to |5/2^+, 1/2\rangle$, and $|3/2^+, 3/2\rangle \to |5/2^+, 1/2\rangle$, shown by three left green arrows in Fig. 1. At the temperature and magnetic field specified above the population of the states involved in the transitions is also inversed, Table I. Therefore, these transitions also lead to the enhancement of the radiation, but their contribution to χ is very small (1%).

Now we turn to the transitions $|5/2^+, 1/2\rangle \rightarrow |3/2^+, 3/2\rangle$, $|5/2^+, 1/2\rangle \rightarrow |3/2^+, 1/2\rangle$, and $|5/2^+, 1/2\rangle \rightarrow |3/2^+, -1/2\rangle$, labeled by three right green arrows in Fig. 1. Since according to Table I the $|5/2^+, 1/2\rangle$ state is only weakly populated, here the resonant excitation of the isomeric level is also possible. Such an excitation is a result of the Mössbauer effect in the optical range. Emission of the gamma ray photons by the ^{229m}Th isomers and the resonant absorption of these photons by the ^{229}Th nuclei in a solid should occur without recoil. Indeed, the energy lost E_R due to the recoil is negligibly small: $E_R = \omega^2/2M \approx 1.4 \times 10^{-10}$ eV (here M is the ^{229}Th nucleus mass). The probability of the Mössbauer effect at low temperature estimated with the Debye-Waller factor is $f \approx \exp(-3E_R/2\theta_D)$ [21], where θ_D is the Debye temperature. It is evident that in our case $f = 1$ because $E_R/\theta_D \ll 1$.

The resonant absorption of the gamma radiation is more pronounced for the remaining transitions shown in Fig. 1 by three left and tree right blue arrows. There are no inverse populations there, and correspondingly there will be no amplification in the considered media.

It should be noted that the above mentioned estimations are made without considering the angular distribution of the gamma radiation. The intensities of the hyperfine components of the $M1$ radiation are proportional to $\sin^2(\vartheta)$ for the $\Delta m = 0$ transitions, and to $(1 + \cos^2(\vartheta))/2$ for the $\Delta m = \pm 1$ transitions (ϑ is the angle between the direction of the magnetic field and the photon momentum). For the transitions shown by red arrows in Fig. 1 in the $0.1 \times 0.1 \times 1$ cm sample we have found that the total amplification factor is almost independent on mutual orientation of the sample and the magnetic field. In other words, the angular distribution of the gamma radiation is not very important for the net result. However, it is conceivable that in certain situations the direction of magnetic field plays an important role. For example, in case the sample is oriented with its long side along the magnetic field one obtains mainly the circular polarization, because the $\Delta m = \pm 1$ transitions are amplified more than the others.

We close the consideration with a remark on the sign of the magnetic moment of the isomeric level. Its sign is also not critical for the amplification effect. The positive value of μ_{is} would simply half the general amplification factor. In such a case the radiation amplification is due to the three transitions to the upper sublevels of the ground state from the upper Zeeman isomeric sublevels, the population of which is smaller in comparison with the population of the lower sublevels, Table I.

In conclusion, in the present work we have suggested the conditions for obtaining of the coherent nuclear γ radiation in the optical range (with the energy 7.6 eV, or 3.5 eV) in the ensemble of the ^{229m}Th isomeric nuclei in a host dielectric crystal with a large band gap. According to our estimations this γ radiation has the linear amplification factor $\chi \geq 10$ cm^{-1} at $T = 0.01$ °K in the static magnetic field H = 100 T. The amplification is a result of the following factors: 1) the excitation of an appreciable number of the ^{229m}Th isomer ($n_{is} > 10^{17}$ cm^{-3} for the initial concentration $n_0 = 10^{18}$ cm^{-3}) by the laser radiation with the power of a few Watts; 2) the creation of the inverse population of the Zeeman sublevels of the isomeric and ground state of the ^{229}Th nuclei; 3) the recoilless emissions and absorption of the optical photons by thorium nuclei in the crystal (the Mössbauer effect in the optical range). Therefore, the required temperature and magnetic field are technically available and we suggest to carry out the necessary experiments to study the proposed setup for the gamma-ray laser.

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