DIRECT TWO-PHOTON EXCITATION OF ISOMERIC TRANSITION IN THORIUM-229 NUCLEUS

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A possibility of the two-photon excitation of an isomeric state in a nucleus of thorium-229 has been discussed. The fluorescence intensity of the excitation is demonstrated to be identical for the irradiation of nuclei with either monochromatic light or polychromatic radiation consisting of a sequence of short light pulses of the same intensity. The two-photon excitation of Th^{3+} ion in an electromagnetic trap with a focused laser beam with a wavelength of about 320 nm and power of 100 mW can lead to the absorption saturation, at which the fluorescence emission with the frequency of the transition in a nucleus is maximal. In crystals doped with Th^{4+} to a concentration of about 10^{18} cm^{-3} and irradiated with a laser radiation 10 W in power, the emission of several photons per second with a wavelength of about 160 nm becomes possible.

1. Introduction

A permanent interest to works dealing with the creation of quantum-mechanical frequency standards (atomic clocks) is stimulated by both the development of fundamental science and engineering demands. Modern methods used for the determination of time unit, a second, are associated with the cesium atomic frequency standard. In particular, one second is defined as the duration of 9 192 631 770 periods of the radiation corresponding to the transition between the two hyperfine levels of the ground state of a ^{133}Cs atom [1]. The accuracy of the primary cesium frequency standard based on the cesium fountain clock was 4×10^{-16} [2]. A possibility for atoms and ions to be cooled down to ultralow temperatures allowed a stability of the order of 10^{-17} to be obtained using cold Al^+ ions [3]. Atomic standards are of interest for navigating systems, such as GPS, GLONASS, and GALILEO, as well as for telecommunication networks.

A promising candidate for the role of reference quantum-mechanical frequency standard is the isomeric transition in a ^{229}Th nucleus. This isotope has an extremely low energy of the isomeric state. According to the most recent data, it equals 7.8 ± 0.6 eV [4], which corresponds to a radiation wavelength of about 160 nm. Progress in the development of the generation of high laser-radiation harmonics allows the nuclear standard to be regarded as a real possibility. For instance, in work [5], radiation with a wavelength of 205 nm was obtained with the use of the generation of the fourth harmonic of radiation emitted by a titanium-sapphire laser, which is close to that required for the implementation of a nuclear standard. The development of a nuclear standard is also stimulated by the capability of its application for studying the evolution of the ratio between fundamental constants [6, 7]. It is so because the ^{229}Th-based frequency standard is supposed to be by several orders of magnitude more sensitive to a possible variation of the hyperfine structure constant than the frequency standards based on the transitions in the electron shell of atoms or ions. Analogously to their atomic counterparts, nuclear

clocks, if being realized, could be applied to metrology, spectroscopy, global navigating systems, and so forth.

It should be noted that, till now, the energy of the isomeric transition is known only from the analysis of γ-spectra with energies that considerably exceed the nuclear isomeric transition energy. Therefore, it cannot be considered as ultimately known, until direct measurements of the level have not been carried out. For this purpose, rather a large number of thorium nuclei must be excited simultaneously. At the present time, either thorium ions in traps [8–11] or thorium-doped crystals transparent in the ultra-violet range [12, 13] are proposed to be used. The advantage of traps consists in the possibility of a better control over the fields that act on thorium ions. At the same time, crystals allow a much larger number of nuclei to be engaged simultaneously than those in traps.

Instead of using the radiation with a frequency close to that of the transition in a nucleus, the latter can be excited with the use of two- or multiphoton processes. The choice of that or another excitation way depends on the parameters of available laser radiation sources, such as the intensity, spectral width, and generation mode (pulsed or continuous). Owing to the short wavelength of radiation needed for the excitation of a nucleus, this radiation can be obtained by generating the radiation harmonics of pulsed lasers operating in the visible spectral range. At the repetition frequency of an order of 100 MHz and under single-photon excitation conditions, only one of the spectral components, which is close to the frequency of the transition in a nucleus, can expectedly stimulate a transition between nuclear states. This means that, if the radiation emitted by picosecond and femtosecond lasers and characterized by a considerable number of spectral components is used as the pumping one, only a small fraction of the emitted intensity can be used for the nuclear excitation. At the same time, in the case of two-photon excitation, almost all spectral components (grouped in pairs consisting of the frequencies locating above and below the transition frequency and at the same distance from it) stimulate the nuclear excitation. On the other hand, the probability of a two-photon transition is much lower than that of a single-photon one, and a possibility to develop a frequency standard on the basis of a two-photon transition is not so evident.

It should be noted that the two-photon excitation, in contrast to the single-photon one, is insensitive, to an accuracy of the squared Doppler effect, to the atomic velocity, if counter-propagating waves are used, i.e. the excitation by a standing wave is executed [14]. Really, if an atom that moves with the velocity v along the z-axis is excited by a standing wave with the frequency ω, it is subjected to the action of two monochromatic waves with the frequencies $\omega \pm kv$, where $k = 2\pi\omega/c$, in the accompanying reference frame. Absorption of two photons from the counter-propagating waves results in an increase of the atomic energy by the quantity $\hbar(\omega + kv) + \hbar(\omega - kv) = 2\hbar\omega$, which is independent of the atomic velocity.

An interesting method of isomeric transition excitation with the use of two photons was proposed in work [9]. The cited authors suggested to populate an intermediate level in the electron shell of Th^+ ion, the energy of which is well-known, with the help of a laser with narrow spectral width. The frequency of the second laser is scanned near the difference between the predicted frequency of the isomeric transition and the frequency of the first laser. If this procedure gives rise to the excitation of a level in the ionic electron shell with the energy close to that of the isomeric transition, this energy can be transferred to the nucleus. In this case, owing to the variation in the nucleus state, the position of the ionic intermediate level also changes, so that the first laser field ceases to interact with it, and the fluorescence at the first-laser frequency diminishes. At the same time, the issue concerning the lifetime of the isomeric state in the single-charged ion is not clear in detail [15]. Note that the resonance width for such an excitation is determined by the lifetime of the intermediate level. Therefore, the method of two-photon nuclear excitation proposed in work [9] does not give any advantages while developing a frequency standard, in which a long-term isomeric state of the nucleus is excited through the excitation of the electron shell, in comparison with that, where the electron transition in an atom or ion is excited. However, the method of excitation of a nucleus into the isomeric state, which was proposed in work [9], can undoubtedly facilitate the registration of the optical nuclear transition.

This work is devoted to the analysis of the "direct" two-photon nuclear transition in a thorium nucleus making no allowance for any intermediate levels. Such an excitation can be carried out, generally speaking, in any thorium ion. We have considered the excitation of nuclei with either the monochromatic or polychromatic field created by a light wave or counter-propagating light waves. In Section 2, the major spectroscopic characteristics of thorium are described, which are relevant to the transition from the ground nuclear state into the excited one. Section 3 contains basic equations. In Section 4, we will derive the effective Hamiltonian, which forms a basis for the description of the time evolution of slowly

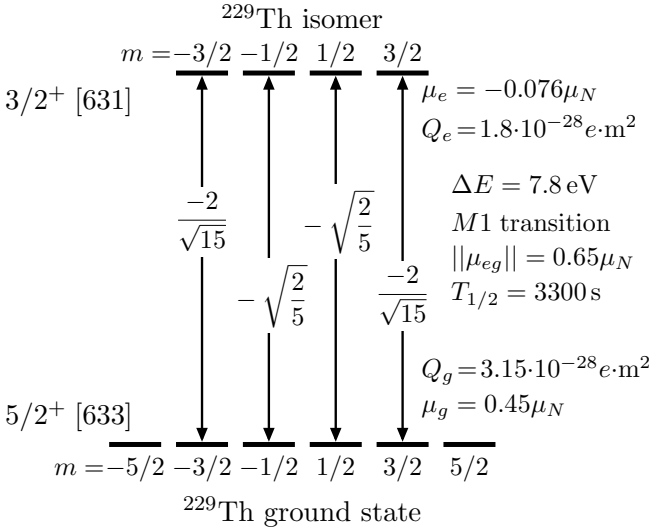
changing, in comparison with the laser radiation period, components of the probability amplitudes for a state of the nucleus. The two-photon absorption in a monochromatic field is discussed in Section 5, and that in the field created by a sequence of propagating light pulses in Section 6. In Section 7, the two-photon excitation in the field of sequences of counter-propagating light pulses is considered. In Section 8, the efficiency of the two-photon excitation is evaluated and discussed. In Conclusions, the results obtained in this work are summarized.

2. ^{229}Th Isomer

^{229}Th isotope is α-active with a half-life period of 7340 year [16]. In Figure, the available spectroscopic information concerning the magnetic dipole ($M1$) transition between the first excited level $I_e = 3/2$ and the ground state level $I_g = 5/2$ in a ^{229}Th nucleus is summarized.

Each of two levels with the total momentum I_i ($I_g = 5/2$ and $I_e = 3/2$) splits into $2I_i + 1$ sublevels described by the magnetic quantum number $m = -I_i, -I_i + 1, \ldots I_i - 1, I_i$. If the nucleus is subjected to the action of a constant magnetic field, those levels have different energies. In a solid, the nucleus undergoes the influence of the electric field created by atoms of the environment. As a result, there arises a quadrupole shift of the nucleus energy, which depends on m. Let the nucleus be subjected to the action of linearly polarized laser radiation, which can depend on the time; in particular, it can be a sequence of pulses with period T. Provided this assumption, it is sufficient to analyze the two-level model of interaction between the nucleus and the field to evaluate the efficiency of two-photon excitation.

Among the whole set of transitions between the atomic or ion states and the nucleus in the ground or excited state, we consider the transition between the states with the same orbital moment and spin of electrons in the ground and excited states. The frequency of this transition, to an accuracy of corrections associated with the hyperfine level structure, coincides with that of the transition between the ground and excited state of a nucleus, i.e. the reference frequency of optical nuclear frequency standard. Although the state of electrons remains invariable, their presence induces the variation of the total moment for the system "electron shell + nucleus", as well as the g-factor responsible for the Stark shift of energy levels in the magnetic field. A fourfold-charged Th^{4+} ion has a closed electron shell of radon. Figure illustrates the interaction of this ion with the field. For Th^{3+} ion, the ground state is $5F_{5/2}$ [11]. As a result,



Ground and first excited states of ^{229}Th nucleus. The levels are classified according to the Nilsson model [17]. The radiation lifetime of a free nucleus $T_{1/2} = 55$ min (the rate of spontaneous radiation emission $\gamma = 0.00021\ \mathrm{s}^{-1}$) and the reduced matrix element of the transition $\|\mu_{eg}\| = \sqrt{\frac{4\pi}{3}B(M1)(2I_e+1)} = 0.65\mu_N$, where $B(M1)$ is the reduced probability of decay, were calculated on the basis of data taken from work [18]. The magnetic moments of levels in terms of nuclear magneton units and the quadrupole moment in the ground state, $Q_g = 3.15 \times 10^{-28}\ e \cdot \mathrm{m}^2$, were taken from works [19, 20]. The estimate $Q_e = 1.8 \times 10^{-28}\ e \cdot \mathrm{m}^2$ for the quadrupole momentum in the excited state was taken from work [21]. The arrows indicate probable transitions between magnetic sublevels in the field of linearly polarized laser radiation. The Clebsch–Gordan coefficients, which are proportional to the matrix elements of the magnetic dipole moments of corresponding transitions, are also indicated

there emerges a hyperfine structure of the ground state with the total moment $F = 0, 1, 2, 3, 4, 5$ and the excited state with $F = 1, 2, 3, 4$. In a magnetic field, each level of hyperfine structure splits into $2F + 1$ sublevels. Owing to the Zeeman effect, those sublevels are shifted with respect to their position in the zero field by

$$\Delta E = \mu_{\mathrm{B}} g_F m_F B, \tag{1}$$

where B is the magnetic field induction, and μ_{B} the Bohr magneton. The multiplier g_F in the case of zero orbital moment (which is of interest for us) equals

$$g_F = g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)} +$$

$$+ g_I \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)}, \tag{2}$$

where I is the nuclear moment, J the angular momentum of a valence electron, $g_I = -\mu_i/(\mu_{\rm B} I)$ is the nuclear g-factor, $(i = e, g)$,

$$g_J = 1 + \frac{J(J+1) - L(L+1) + S(S+1)}{2J(J+1)}(g_e - 1) \quad (3)$$

is the electron g-factor, L the orbital moment of electrons, S the spin, and $g_e = 2.0023$ is the g-factor of a free electron (in the further calculations, we adopt $g_e = 2$).

For Th^{4+} ion, $J = 0$ and $g_F = g_I$. For Th^{3+} ion in the ground state, $g_F = \frac{3}{7} + \frac{1}{2} g_I$ for every hyperfine sublevel. For Th^{3+} ion with the excited nucleus ($I = \frac{3}{2}$), similar calculations give, e.g., $g_F = \frac{15}{28} + \frac{3}{8} g_I$ for the hyperfine sublevel with $F = 4$. Since $g_I \sim \mu_N/\mu_{\rm B} \ll 1$, the last term can be neglected. Hence, the g-factor for Th^{3+} is by three orders of magnitude larger than that for Th^{4+}. As one can see from what follows, this circumstance brings about a substantial increase in the efficiency of two-photon excitation of the thorium nucleus in Th^{3+} ion in comparison with Th^{4+} one.

3. Model of Nucleus Interaction with the Field

In order to estimate the possibility of the direct two-photon excitation of a thorium nucleus, let us consider transitions between thorium states characterized by a definite quantum number m related to the ground and excited nucleus states. If ions are in a solid matrix, their energies corresponding to different m-values are different owing to the interaction between the ions and the environment. However, if ions or atoms are in the trap, we consider that a magnetic field eliminating the degeneration with respect to m is applied to them. Therefore, to analyze the excitation of a nucleus, we may use the two-level model.

Let the ground state be designated as $|g\rangle$, and the excited one as $|e\rangle$. The magnetic dipole transition between the states of an atom in the ground and excited states has the constant dipole moments $\mu_{gg} = -\mu_{\rm B} g_g m$ and $\mu_{ee} = -\mu_{\rm B} g_e m$, where g_g and g_e are the g-factors for the ground and excited, respectively, states. The magnetic dipole moment of the transition between the states $|g\rangle$ and $|e\rangle$ equals μ_{ge}. Let the atom be subjected to the action of a field—monochromatic or created by a sequence of light pulses—with a carrier frequency ω such that the value of 2ω is close to the frequency ω_0 of the transition between the states $|g\rangle$ and $|e\rangle$. Moreover, let the transition in the nucleus leave the state of atomic electrons intact. In addition, we suppose that only the terms responsible for the change in the nucleus state are essential in the Hamiltonian describing the interaction between the atom and the field.

The induction of the magnetic field of laser radiation with the carrier frequency ω, which affects the atom, is described by the expression

$$B(t) = \tilde{B}(t) e^{-i\omega t} + \tilde{B}(t)^* e^{i\omega t}. \quad (4)$$

The probability amplitudes C_g and C_e to find the nucleus in the state $|g\rangle$ or $|e\rangle$, respectively, vary according to the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \boldsymbol{C} = H \boldsymbol{C}, \quad (5)$$

where $\boldsymbol{C}$ is the column vector with the components C_g and C_e, and the Hamiltonian H looks like

$$H = \begin{pmatrix} -\mu_{gg} B(t) & -\mu_{ge} B(t) \\ -\mu_{eg} B(t) & \hbar\omega_0 - \mu_{ee} B(t) \end{pmatrix}, \quad (6)$$

where $\hbar\omega_0$ is the energy difference between the states $|g\rangle$ and $|e\rangle$.

In the general case of a polychromatic field with a narrow, in comparison with ω, spectral width, expression (4) can be interpreted as rapid oscillations with the optical frequency ω and a slowly varying amplitude. For a monochromatic field, $\tilde{B}(t)$ does not depend on the time. For instance, for a propagating monochromatic wave,

$$\tilde{B}(t) = \tfrac{1}{2} B_0 e^{ikz}, \quad (7)$$

and, for a monochromatic field of two counter-propagating waves (standing wave),

$$\tilde{B}(t) = B_0 \cos kz, \quad (8)$$

where $k = \omega/c$, z is the z-coordinate of the atom, and B_0 is the wave amplitude.

If the atom is subjected to the action of the field created by a sequence of light pulses with the pulse-repetition period T, the magnetic field induction at the atom location point can be written down in the form

$$B(t) = B_0 \sum_{n=-\infty}^{\infty} a_n \cos\left[(\omega + n\Delta)\, t - k_n z + \varphi_n\right], \quad (9)$$

where $k_n = (\omega + n\Delta)/c$ and $\Delta = 2\pi/T$. The relative amplitudes of spectral components, a_n, are normalized so that the component with $n = 0$ has the maximum amplitude $a_0 = 1$.

Comparing Eqs. (4) and (9), one can see that, for the field of a sequence of propagating light pulses,

$$\tilde{B} = \tfrac{1}{2} B_0 \sum_{n=-\infty}^{\infty} a_n \exp\left(-in\Delta t + ik_n z - i\varphi_n\right). \quad (10)$$

For the field of the counter-propagating sequences of pulses

$$B(t) = B_0 \sum_{n=-\infty}^{\infty} a_n \cos\left[(\omega + n\Delta)\, t - k_n z + \varphi_n\right] +$$

$$+B_0 \sum_{n=-\infty}^{\infty} a_n \cos\left[(\omega + n\Delta)\, t + k_n z + \varphi_n\right], \qquad (11)$$

we have

$$\tilde{B} = B_0 \sum_{n=-\infty}^{\infty} a_n \cos k_n z \exp\left(-in\Delta t - i\varphi_n\right). \qquad (12)$$

Here, the reference point for the z-coordinate is chosen so that, at the point $z = 0$, the field of a wave propagating in the negative direction of the z-axis reproduces the field of the wave propagating in the positive direction of the z-axis.

4. Effective Hamiltonian

Let us make the substitutions

$$C_g(t) = c_g(t) \exp\left(\frac{i\mu_{gg}}{\hbar} \int_0^t B(t')\, dt'\right),$$

$$C_e(t) = c_e(t) \exp\left(-Ni\omega t + \frac{i\mu_{ee}}{\hbar} \int_0^t B(t')\, dt'\right). \qquad (13)$$

in the Schrödinger equation (5). The variation of the column vector $\boldsymbol{c}$ with the components c_g and c_e in time is described by the Schrödinger equation with the Hamiltonian

$$\mathcal{H} = \begin{pmatrix} 0 & -\mu_{ge} B(t) e^{i\Phi(t)} \\ -\mu_{eg} B(t) e^{-i\Phi(t)} & \hbar\omega_0 - 2\hbar\omega \end{pmatrix}, \qquad (14)$$

where

$$\Phi(t) = -2\omega t + \frac{\mu_{ee} - \mu_{gg}}{\hbar} \int B(t')\, dt'. \qquad (15)$$

The lower limit of integration is not indicated, because the proper choice of the time reference point makes the value of the primitive at this limit equal to zero.

We consider that the characteristic time τ of field amplitude variations (e.g., the pulse duration) considerably exceeds the reciprocal carrier frequency of the field, so that the inequality

$$\omega\tau \gg 1 \qquad (16)$$

is valid. Then

$$\Phi(t) = -2\omega t + \frac{\mu_{ee} - \mu_{gg}}{i\hbar\omega}\left[\tilde{B}(t)^* e^{i\omega t} - \tilde{B}(t) e^{-i\omega t}\right]. \qquad (17)$$

In particular, for the field of a monochromatic propagating wave ($\tau = \infty$), we obtain

$$\Phi(t) = -2\omega t + B_0 \frac{\mu_{ee} - \mu_{gg}}{\hbar\omega} \sin(\omega t - kz), \qquad (18)$$

whereas, for the monochromatic field of counter-propagating waves,

$$\Phi(t) = -2\omega t + 2B_0 \frac{\mu_{ee} - \mu_{gg}}{\hbar\omega} \cos kz \sin\omega t. \qquad (19)$$

Let us introduce the following functions slowly varying in time:

$$\beta(t) = \frac{\tilde{B}(t)\mu_{ge}}{\hbar}, \qquad \alpha(t) = \frac{\tilde{B}(t)\left(\mu_{ee} - \mu_{gg}\right)}{\hbar\omega}. \qquad (20)$$

Since $|\alpha| \ll 1$, the further calculations will be carried out to an accuracy of the terms linear in α. Substituting Eqs. (17) and (20) into Hamiltonian (14), we obtain

$$\mathcal{H} = \frac{\hbar}{2} \begin{pmatrix} 0 & \Omega(t) + \tilde{\Omega}(t) \\ \Omega(t)^* + \tilde{\Omega}(t)^* & 2\delta \end{pmatrix}. \qquad (21)$$

Here, in the non-diagonal elements, we separated the terms $\tilde{\Omega}(t)$ rapidly varying in time with the frequency ω and the slowly varying terms $\Omega(t)$ and introduced the two-photon detuning $\delta = \omega_0 - 2\omega$. The fast and slow components of the Rabi frequency equal

$$\Omega = -2\alpha^*\beta^*,$$

$$\tilde{\Omega} = -2\beta^* e^{-i\omega t} - 2\beta e^{-3i\omega t}. \qquad (22)$$

Hereafter, in order to simplify the notation and if it cannot lead to misunderstanding, we do not indicate the time dependence of quantities. In expression (22), we neglected terms with higher orders of smallness with respect to $\alpha \ll 1$. At the same time, it should be noticed that only the nonzero value of α makes the component of the Rabi frequency that slowly varies in time different from zero for the two-photon process considered here.

The solution of the Schrödinger equation is sought as a sum of two terms, slowly and rapidly varying in time

with a characteristic time of the order of $2\pi/\omega$, for each of the probability amplitudes,

$$c_n(t) = \sigma_n(t) + \tilde{\sigma}_n(t), \quad n = g, e, \tag{23}$$

where the rapidly varying terms are marked by the tilde sign. We adopt that the average value of rapidly varying term over the period $2\pi/\omega$ vanishes. From the Schrödinger equation, it follows that

$$i\dot{\sigma}_g + \underline{i\dot{\tilde{\sigma}}_g} = \tfrac{1}{2}\Omega\sigma_e + \underline{\underline{\tfrac{1}{2}\tilde{\Omega}\tilde{\sigma}_e}} + \tfrac{1}{2}\tilde{\Omega}\sigma_e + \underline{\tfrac{1}{2}\Omega\tilde{\sigma}_e},$$

$$i\dot{\sigma}_e + \underline{i\dot{\tilde{\sigma}}_e} = \tfrac{1}{2}\Omega^*\sigma_g + \underline{\underline{\tfrac{1}{2}\tilde{\Omega}^*\tilde{\sigma}_g}} + \tfrac{1}{2}\tilde{\Omega}^*\sigma_g + \underline{\tfrac{1}{2}\Omega^*\tilde{\sigma}_g} +$$

$$+\delta\sigma_e + \underline{\delta\tilde{\sigma}_e}. \tag{24}$$

The underlined terms rapidly oscillate and vanish after having been averaged over the oscillation period $2\pi/\omega$. The twice underlined terms consist of products of oscillating multipliers. They can be presented as a sum of a component that slowly varies in time and an oscillating component with the averaged value equal to zero. To exclude the rapid motion, let us collect together, into two groups, rapidly and slowly oscillating terms in Eqs. (24). The oscillating terms in Eqs. (24) give

$$i\dot{\tilde{\sigma}}_g = \tfrac{1}{2}\tilde{\Omega}\sigma_e,$$

$$i\dot{\tilde{\sigma}}_e = \tfrac{1}{2}\tilde{\Omega}^*\sigma_g. \tag{25}$$

Here, we took into account that $\left|\dot{\tilde{\sigma}}_n(t)\right| \sim \omega\,|\tilde{\sigma}_n(t)|$, $\omega \gg (\Omega, \left|\tilde{\Omega}(t)\right|, |\delta|)$, and $|\tilde{\sigma}_n(t)| \ll 1$. The solution of Eqs. (24) reads

$$\tilde{\sigma}_g = -\left(\frac{\beta^*}{\omega}e^{-i\omega t} + \frac{\beta}{3\omega}e^{-3i\omega t}\right)\sigma_e,$$

$$\tilde{\sigma}_e = \left(\frac{\beta}{\omega}e^{i\omega t} + \frac{\beta^*}{3\omega}e^{3i\omega t}\right)\sigma_g. \tag{26}$$

The terms in Eqs. (24) that slowly vary in time bring about

$$i\dot{\sigma}_g = \tfrac{1}{2}\Omega\sigma_e + \left\langle \tfrac{1}{2}\tilde{\Omega}\tilde{\sigma}_e \right\rangle,$$

$$i\dot{\sigma}_e = \tfrac{1}{2}\Omega^*\sigma_g + \left\langle \tfrac{1}{2}\tilde{\Omega}^*\tilde{\sigma}_g \right\rangle + \delta\sigma_e, \tag{27}$$

where the notation $\langle\cdots\rangle$ means the time-averaging over the interval $2\pi/\omega$. After carrying out this averaging, we obtain

$$i\dot{\sigma}_g = \tfrac{1}{2}\Omega\sigma_e - \frac{4|\beta|^2}{3\omega}\sigma_g,$$

$$i\dot{\sigma}_e = \tfrac{1}{2}\Omega^*\sigma_g + \frac{4|\beta|^2}{3\omega}\sigma_e + \delta\sigma_e. \tag{28}$$

As a result, we find that the variation of the slow components of the probability amplitudes is described by the Schrödinger equation with the effective Hamiltonian

$$\mathcal{H}_{\text{eff}} = \frac{\hbar}{2}\begin{pmatrix} -S & \Omega \\ \Omega^* & 2\delta + S \end{pmatrix}, \tag{29}$$

where

$$S = \frac{8|\beta|^2}{3\omega} = \frac{8|\tilde{B}\mu_{ge}|^2}{3\hbar^2\omega},$$

$$\Omega = -2\alpha^*\beta^* = -2\frac{\tilde{B}^{*2}}{\hbar^2\omega}\mu_{ge}(\mu_{ee} - \mu_{gg}). \tag{30}$$

As one can see from Eq. (30), the light shift is proportional to the laser radiation intensity. In effect, this relation was obtained because we have gone beyond the rotating wave approximation [23]. The ratio $|S/\Omega| \sim |\mu_{ge}/(\mu_{ee} - \mu_{gg})|$ is of the order of 10^{-3} in the case where the g-factors of the ground and excited atomic states are determined by the atomic electron structure, e.g., for a triple-charged thorium ion in the trap.

5. Two-Photon Interaction of a Nucleus with a Monochromatic Field

To estimate the rate of nucleus fluorescence under the influence of monochromatic laser radiation, it is convenient to write down expressions for the Rabi frequency and the light shift in the case where the nucleus is excited by the propagating monochromatic wave. From the expression for the radiation intensity in the SI system,

$$I = \frac{c}{2\mu_0}B_0^2, \tag{31}$$

and Eqs. (7) and (30), we obtain

$$S = \frac{4|\mu_{ge}|^2}{3\hbar^2\omega c}\mu_0 I,$$

$$\Omega = -\frac{\mu_0 I}{\hbar^2 \omega c}\mu_{ge}(\mu_{ee} - \mu_{gg})e^{-2ikz}. \tag{32}$$

In the case of monochromatic standing wave, from expressions (8), (30), and (31), we obtain

$$S = \frac{16|\mu_{ge}|^2}{3\hbar^2\omega c}\mu_0 I \cos^2 kz,$$

$$\Omega = -\frac{4\mu_0 I}{\hbar^2 \omega c}\mu_{ge}(\mu_{ee} - \mu_{gg})\cos^2 kz, \tag{33}$$

where I is the intensity of either of the counter-propagating waves.

For an atom or ion moving in the trap, in the case of a propagating monochromatic wave, the proportionality of the Rabi frequency to the quantity e^{-2ikz}, in which z linearly depends on the time, brings about a Doppler shift of the resonance by the magnitude of $2kv$, where v is the atom velocity. This means that, for narrow resonances to be obtained, it is necessary to cool atoms down to ultralow temperatures. For two counter-propagating waves (standing light wave), the multiplier

$$\cos^2 kz = \frac{1}{2} + \frac{1}{4}e^{2ikz} + \frac{1}{4}e^{-2ikz} \tag{34}$$

in expression (33) for the Rabi frequency testifies that there emerge three resonances, when the atom interacts with the field; namely, two of them are shifted with respect to the transition frequency by $\pm 2kv$, and one resonance is located at the transition frequency. Since all groups of atoms make contribution to the latter resonance, the dependence of the fluorescence intensity on the detuning δ looks like a wide peak associated with the Doppler line broadening and a narrow high peak at its center. This phenomenon serves as a basis for two-photon spectroscopy [14, 24]. Therefore, there is no need to use ultracold atoms or their localization in a small volume (the Dicke effect) at the two-photon excitation in order to reduce a negative influence of the Doppler effect on the frequency standard functioning.

6. Interaction of a Nucleus with the Field of a Sequence of Propagating Light Pulses

Let the atom undergo the action of the field of a sequence of light pulses with the pulse period T. The magnetic field induction at the point, where the atom is located, is described by expression (9). The spectrum of this field is equidistant, with the difference between the frequencies of spectral components $\Delta = 2\pi/T$. On the basis of expressions (10) and (30), it is evident that the light shift and the Rabi frequency can be expanded in the Fourier series

$$S = \sum_{n=-\infty}^{\infty} S_n e^{in\Delta t} \tag{35}$$

and

$$\Omega = \sum_{n=-\infty}^{\infty} \Omega_n e^{in\Delta t}, \tag{36}$$

where

$$S_n = \frac{2|B_0\mu_{ge}|^2}{3\hbar^2\omega} \sum_{j=-\infty}^{\infty} a_{j-n}a_j \times$$

$$\times \exp\left(i\varphi_j - i\varphi_{j-n} + ik_{j-n}z - ik_j z\right) \tag{37}$$

and

$$\Omega_n = -\frac{{B_0}^2}{2\hbar^2\omega}\mu_{ge}(\mu_{ee} - \mu_{gg}) \sum_{j=-\infty}^{\infty} a_{n-j}a_j \times$$

$$\times \exp\left(i\varphi_j + i\varphi_{n-j} - ik_{n-j}z - ik_j z\right). \tag{38}$$

The light shift S and the Rabi frequency Ω include time-independent terms and terms oscillating with the period $T = 2\pi/\Delta$,

$$S = S_0 + \tilde{S}, \qquad \Omega = \Omega_0 + \tilde{\tilde{\Omega}}. \tag{39}$$

We consider the low intensities of laser radiation, for which $|S| \ll \Delta$ and $|\Omega| \ll \Delta$. Then, by applying the procedure of averaging over the rapid motion (in this case, with the frequency Δ), which was described in Section 4, it is possible to determine the effective Hamiltonian describing the variation of the slow components of the probability amplitudes σ_g and σ_e in time. Similarly to Eq. (23), we write down the quantities σ_g and σ_e as sums of slowly varying in time and oscillating (marked by the tilde sign) terms,

$$\sigma_j(t) = b_j(t) + \tilde{b}_j(t), \quad j = g, e. \tag{40}$$

From the Schrödinger equation with Hamiltonian (29), it follows that

$$i\dot{b}_g + \underline{i\dot{\tilde{b}}_g} = \tfrac{1}{2}\Omega_0 b_e + \underline{\tfrac{1}{2}\tilde{\tilde{\Omega}}\tilde{b}_e} + \tfrac{1}{2}\tilde{\tilde{\Omega}}b_e + \tfrac{1}{2}\Omega_0\tilde{b}_e -$$

$$-\tfrac{1}{2}S_0 b_g - \underline{\tfrac{1}{2}\tilde{S}\tilde{b}_g} - \tfrac{1}{2}\tilde{S}b_g - \tfrac{1}{2}S_0\tilde{b}_g,$$

$$i\dot{b}_e + \underline{i\dot{\tilde{b}}_e} = \tfrac{1}{2}\Omega_0^* b_g + \underline{\tfrac{1}{2}\tilde{\tilde{\Omega}}^*\tilde{b}_g} + \tfrac{1}{2}\tilde{\tilde{\Omega}}^* b_g + \tfrac{1}{2}\Omega_0^*\tilde{b}_g +$$

$$+\tfrac{1}{2}S_0 b_e + \underline{\tfrac{1}{2}\tilde{S}\tilde{b}_e} + \tfrac{1}{2}\tilde{S}b_e + \tfrac{1}{2}S_0\tilde{b}_e + \delta b_e + \underline{\delta\tilde{b}_e}. \quad (41)$$

Here, analogously to the notation in Eq. (24), the underlined terms oscillate and vanish after being averaged over the oscillation period, whereas the twice underlined terms consist of products of oscillating multipliers and can be presented as sums of slow and rapid components. Equating the rapid terms independently, we obtain

$$i\dot{\tilde{b}}_g = \tfrac{1}{2}\tilde{\tilde{\Omega}}b_e - \tfrac{1}{2}\tilde{S}b_g,$$

$$i\dot{\tilde{b}}_e = \tfrac{1}{2}\tilde{\tilde{\Omega}}^* b_g + \tfrac{1}{2}\tilde{S}b_e. \quad (42)$$

Here, we took into account that $\left|\dot{\tilde{b}}_n(t)\right| \sim \Delta\left|\tilde{b}_n(t)\right|$, $\Delta \gg (\Omega_0, |\tilde{\tilde{\Omega}}|, |\delta|)$, and $\left|\tilde{b}_n(t)\right| \ll 1$. The solution of Eqs. (42) looks like

$$\tilde{b}_g = \frac{1}{2\Delta}\sum_{\substack{n=-\infty\\ n\neq 0}}^{\infty}\left(S_n b_g - \Omega_n b_e\right)\frac{e^{i\Delta nt}}{n},$$

$$\tilde{b}_e = -\frac{1}{2\Delta}\sum_{\substack{n=-\infty\\ n\neq 0}}^{\infty}\left(\Omega_{-n}^* b_g + S_n b_e\right)\frac{e^{i\Delta nt}}{n}. \quad (43)$$

The terms in Eq. (41) that slowly change in time give

$$i\dot{b}_g = \tfrac{1}{2}\Omega_0 b_e - \tfrac{1}{2}S_0 b_g + \left\langle\tfrac{1}{2}\tilde{\tilde{\Omega}}\tilde{b}_e\right\rangle - \left\langle\tfrac{1}{2}\tilde{S}\tilde{b}_g\right\rangle,$$

$$i\dot{b}_e = \tfrac{1}{2}\Omega_0^* b_g + \tfrac{1}{2}S_0 b_e + \left\langle\tfrac{1}{2}\tilde{\tilde{\Omega}}^*\tilde{b}_g\right\rangle + \left\langle\tfrac{1}{2}\tilde{S}\tilde{b}_e\right\rangle + \delta b_e, \quad (44)$$

where the notation $\langle\cdots\rangle$ means the averaging over the time interval $2\pi/\Delta$. From expressions (37), (38), (43), and (44), one can see that, in the case of two-photon interaction, the averaged terms are quadratic in the intensity, and they can be neglected. Then the equations for the slowly varying components of the probability amplitudes are as follows:

$$i\dot{b}_g = \tfrac{1}{2}\Omega_0 b_e - \tfrac{1}{2}\overline{S}b_g,$$

$$i\dot{b}_e = \tfrac{1}{2}\Omega_0^* b_g + \tfrac{1}{2}\overline{S}b_e + \delta b_e, \quad (45)$$

where

$$\overline{S} = S_0. \quad (46)$$

Hence, the variation in time of the slow components of the probability amplitudes in the case where the nucleus is excited by a sequence of propagating pulses is described by the Schrödinger equation with the effective Hamiltonian

$$\mathcal{H}_{\text{eff}} = \frac{\hbar}{2}\begin{pmatrix} -\overline{S} & \Omega_0 \\ \Omega_0^* & 2\delta + \overline{S}\end{pmatrix}, \quad (47)$$

where $\overline{S}$ and Ω_0 are defined by expressions (46) and (38) at $n = 0$, respectively.

For the further calculations, we must select a model for the field. We assume that the phases φ_n of all spectral components of the field equal zero, and the amplitudes are described by the Gaussian distribution

$$a_n = \exp\left(-\frac{n^2}{n_0^2}\right). \quad (48)$$

Let $n_0 \gg 1$, so that the summation in the expressions given above can be replaced by the integration if the time t satisfies the condition $n\Delta t \ll 1$. This inequality means that we do not deal with time intervals far from light pulses, where the field strength is very low. The calculation of $\tilde{B}$ gives

$$\tilde{B} = B_0\frac{n_0\sqrt{\pi}}{2}\exp\left[ik_0 z - \frac{n_0^2\Delta^2}{4}\left(t - \frac{z}{c}\right)^2\right]. \quad (49)$$

This expression describes the time dependence of the field created by one of the pulses that follow one another with period T. As one can see from Eq. (49), the field created by pulses is described by the Gaussian function $\exp\left(-t^2/\tau_p^2\right)$, where

$$\tau_p = \frac{2}{\Delta n_0} = \frac{T}{\pi n_0}. \quad (50)$$

The time-averaged intensity of laser radiation is equal to

$$I = \frac{2c\langle|\tilde{B}|^2\rangle}{\mu_0} = \frac{n_0 c\sqrt{2\pi^3}}{4\mu_0}B_0^2. \quad (51)$$

The light shift is determined by the expression

$$\overline{S} = \frac{4\mu_{ge}^2\mu_0}{3\hbar^2\omega c}I. \quad (52)$$

The calculation of the Rabi frequency Ω_0 gives

$$\Omega_0 = -\frac{\mu_{ge}(\mu_{ee} - \mu_{gg})}{\hbar^2\omega c}\mu_0 I e^{-2ik_0 z} \quad (53)$$

Expression (52) for the light shift and expression (53) for the Rabi frequency coincide with the corresponding formulas (32) obtained for the field of a propagating monochromatic wave.

The coordinate dependence of the Rabi frequency (53) for a moving atom testifies, as was indicated in Section 5 in the case of propagating monochromatic waves, to the Doppler shift of the resonance frequency of the interaction of the nucleus with the field by $2kv$. Therefore, in the case of free atoms or ions, it is necessary to analyze the interaction of the atom with the field of counter-propagating pulse sequences, for which the formation of a narrow resonance is possible, at least under the interaction with a monochromatic field.

7. Two-Photon Interaction of a Nucleus with the Field of Counter-Propagating Sequences of Light Pulses

From expressions (30) for the light shift and the Rabi frequency at the two-photon interaction between the nucleus and the field of counter-propagating sequences of light pulses (see Eqs. (11) and (12)), we obtain

$$S = \sum_{n=-\infty}^{\infty} S_n e^{in\Delta t} \tag{54}$$

and

$$\Omega = \sum_{n=-\infty}^{\infty} \Omega_n e^{in\Delta t}, \tag{55}$$

where

$$S_n = \frac{8|B_0\mu_{ge}|^2}{3\hbar^2\omega}\sum_{j=-\infty}^{\infty} a_{j-n}a_j \cos k_{j-n}z \times$$

$$\times \cos k_j z \exp\left(i\varphi_j - i\varphi_{j-n}\right) \tag{56}$$

and

$$\Omega_n = -2\frac{{B_0}^2}{\hbar^2\omega}\mu_{ge}(\mu_{ee}-\mu_{gg})\sum_{j=-\infty}^{\infty} a_{n-j}a_j \times$$

$$\times \cos k_{n-j}z \cos k_j z \exp\left(i\varphi_j + i\varphi_{n-j}\right). \tag{57}$$

As was done in Section 6, we normalize the relative amplitudes of spectral components a_n in such a way that the component with $n = 0$ has the maximum amplitude $a_0 = 1$. Carrying out the calculations similar to those made in Section 6, we determine the effective Hamiltonian (47) describing the two-photon interaction between the atom and the field, where $\overline{S} = S_0$.

Let us choose the same model of field as that in the previous section,

$$a_n = \exp\left(-\frac{n^2}{n_0^2}\right). \tag{58}$$

For pico- and femtosecond-pulses, which we are interested in, $n_0 \gg 1$, and the summation in the expressions given above can be replaced by the integration at $n\Delta t \ll 1$. The calculation of $\tilde{B}$ gives

$$\tilde{B} = B_0\frac{n_0\sqrt{\pi}}{2}\exp\left[ik_0 z - \frac{n_0^2\Delta^2}{4}\left(t-\frac{z}{c}\right)^2\right] +$$

$$+B_0\frac{n_0\sqrt{\pi}}{2}\exp\left[-ik_0 z - \frac{n_0^2\Delta^2}{4}\left(t+\frac{z}{c}\right)^2\right] \tag{59}$$

for two counter-propagating pulses. The whole sequence of pulses is obtained from Eq. (59) by repeating it with period T. As is evident from Eq. (59), the field of pulses is described by the Gaussian function $\exp\left(-t^2/\tau_p^2\right)$, where τ_p is determined by expression (50).

The light shift is calculated from Eq. (56) taken at $n = 0$ with regard for Eq. (51) as

$$S_0 = \frac{8\mu_{ge}^2\mu_0 I}{3\hbar^2\omega c}\left(1 + e^{-2z^2/l_p^2}\cos 2kz\right), \tag{60}$$

where $l_p = c\tau_p$, and I is the intensity of either of the counter-propagating waves. Expression (60) is valid if the value of $\Delta z/(2\pi c)$ is close to an integer. Hence, the light shift consists of two components; one of them is constant in space, and the other is modulated with the spatial period $\lambda = 2\pi/k_0$ and has the envelope in the form of a set of Gaussian curves located in a vicinity of $z = cTn$, where n is an integer; here, the counter-propagating pulses "collide".

Similar calculations for the Rabi frequency Ω_0 in Eq. (57) give rise to

$$\Omega_0 = -\frac{2\mu_0 I}{\hbar^2\omega c}\mu_{ge}(\mu_{ee}-\mu_{gg})\left(\cos 2kz + e^{-2z^2/l_p^2}\right). \tag{61}$$

The exponential function in Eq. (61) corresponds to the spatial overlapping of pulses. The component of Ω_0 proportional to $\cos 2kz$ is responsible for the formation of resonances shifted by $\pm 2kv$ owing to the Doppler effect in the case of moving atoms or ions. The resonance, which is independent of the atom velocity, arises owing to the term with the Gaussian coordinate dependence. It is connected with a simultaneous absorption of photons from two counter-propagating pulses. The S_0- and Ω_0-quantities averaged over the wavelength equal

$$\langle S_0\rangle = \frac{8\mu_{ge}^2\mu_0 I}{3\hbar^2\omega c} \tag{62}$$

and

$$\langle\Omega_0\rangle = -\frac{2\mu_0 I}{\hbar^2\omega c}\mu_{ge}(\mu_{ee}-\mu_{gg})e^{-2z^2/l_p^2}. \quad (63)$$

It is this value of Rabi frequency averaged over the wavelength that is responsible for the formation of a narrow resonance at the frequency equal to half a frequency of the transition in the nucleus.

One can see that the magnitude of Rabi frequency averaged over the wavelength is described by a Gaussian with the maximum located at the point where the pulses "collide". Whence, it follows that, for the atoms to effectively interact with the field, they must be localized in a volume with the linear dimension along the direction of pulse propagation of an order of l_p. For instance, it can be a cell filled with a thorium-containing gas, or atoms can be localized in an optical trap. Two-photon excitation of nuclei with a sequence of laser pulses is effective as much as that using a monochromatic field, because all spectral field components are engaged at that. For example, in the resonance case $2\omega = \omega_0$, the pairs $(n, -n)$ of spectral components participate in the formation of the two-photon transition.

8. Discussion of Results

Let us estimate the expected fluorescence of a specimen in the framework of the two-level model of interaction between the nucleus and the laser radiation field. Provided that the rate of relaxation of level populations is equal to γ (we assume that the relaxation occurs owing to the spontaneous radiation emission from the excited state), and their coherence equals γ', the stationary population of the excited state amounts to

$$\rho_{22} = \frac{1}{2}\frac{|\Omega_0|^2\gamma'}{|\Omega_0|^2\gamma' + \gamma\,(\gamma'^2 + (\delta+S)^2)}, \quad (64)$$

where δ is the detuning from the two-photon resonance. For a free atom or ion in the field of laser radiation, $\gamma' = \frac{1}{2}(\gamma+\gamma_L)$, where γ_L is the diffusion coefficient for the laser radiation phase [26], i.e. the width of the laser spectrum in the case of monochromatic radiation. From Eq. (64), taking into account that $\gamma \ll \gamma_L$, one can see that, for the absorption to saturate at the resonance, $\delta + S = 0$, the condition $|\Omega_0| > \sqrt{\gamma\gamma_L} \sim 0.03\ \mathrm{s}^{-1}$ must be satisfied. For instance, if $\gamma_L/2\pi \approx 1$ Hz, it is obeyed at $|\Omega_0| \approx 0.03\ \mathrm{s}^{-1}$, and, if $\gamma_L/2\pi \approx 100$ Hz, at $|\Omega_0| > 0.3\ \mathrm{s}^{-1}$. The number of photons emitted at the absorption saturation reaches the maximum value of $\gamma/2 \sim 0.0001\ \mathrm{s}^{-1}$ per each nucleus on the average.

Since the nucleus spin changes from 5/2 to 3/2 at the excitation of the nucleus, the hyperfine structure of energy levels of an atom or ion with the nucleus in the isomeric state also changes. As a result, there emerges a possibility of detecting the excited nucleus with the help of optical laser radiation with the frequency tuned to one of the transition frequencies between hyperfine levels. Since the linewidths Γ's for the corresponding electron transitions are of the order of $10^7\ \mathrm{s}^{-1}$, the excitation of each nucleus stimulates the emission of about 10^7 photons during a second. The implementation of this registration setup requires that atoms or ions should be excited simultaneously from all sublevels of the hyperfine structure of the ground electron state of an atom or ion with the isomeric nucleus in order to prohibit atoms or ions to be accumulated on one of them when the transition from the excited state into the ground one takes place in the course of spontaneous radiation emission.

Let us evaluate the Rabi frequency at the two-photon excitation of Th^{3+} ion. Its ground state has six hyperfine components with the total moment F ranging from 0 to 5, and the ground state of the electron shell of an ion with the excited nucleus has four hyperfine components with the F-values ranging from 1 to 4. From expressions (32), (53), and (63), one can see that the Rabi frequency is maximal, if the product $(\mu_{ee}-\mu_{gg})\mu_{eg}$ is maximal. This condition is satisfied for the transition between the levels $|g\rangle = |F=1, m=0, I=5/2\rangle$ and $|e\rangle = |F=1, m=1, I=3/2\rangle$. In this case,

$$\mu_{ee} - \mu_{gg} = \frac{3}{2}\mu_{\mathrm{B}}, \quad (65)$$

$$\mu_{eg} = (-1)^{F_g+I_e+J-1}\|\mu_{eg}\|\sqrt{2F_g+1}\times$$

$$\times C^{F_e m_e}_{F_g m_g 1q}\left\{\begin{array}{ccc} I_g & J & F_g \\ F_e & 1 & I_e \end{array}\right\} = -\sqrt{\frac{7}{30}}\frac{\|\mu_{eg}\|}{2}. \quad (66)$$

For a laser radiation power of 100 mW and provided that radiation is focused into a spot 1 μm in diameter, we obtain $I = 10^7\ \mathrm{W/cm^2}$, with the Rabi frequency of the transition, according to Eq. (63), being equal to $\Omega_0 \approx 0.07\ \mathrm{s}^{-1}$ and the light shift, according to Eq. (62), being equal to about $5.3\times10^{-6}\ \mathrm{s}^{-1}$. So a small value of light shift testifies that the very procedure of recording the signal from a thorium nucleus inserts, in essence, no additional error into the results of transition frequency measurements. The actual accuracy of optical clocks is governed by other factors, which are not associated with the thorium excitation; in particular, these are the

frequency stability of a radiation source and the field in the trap. The indicated evaluation of the Rabi frequency testifies to a capability of achieving the absorption saturation at the two-photon transition in a thorium nucleus by applying moderate-power laser radiation.

In order to enhance the fluorescence signal, it is necessary to increase the number of irradiated thorium nuclei. A high concentration of thorium nuclei can be achieved in crystals. In this case, on the one hand, the Rabi frequency becomes lower by three orders of magnitude, because thorium is tetravalent in known chemical compounds. On the other hand, however, the number of thorium ions can be increased by many orders of magnitude. Let us evaluate the number of emitted photons in this case. In the case of the transition in Th^{4+}, the expression for the transition dipole moment looks like

$$\mu_{eg} = \frac{\|\mu_{eg}\|}{\sqrt{2I_e+1}} C^{I_e m_e}_{I_g m_g 1 q}, \quad q = m_e - m_g. \tag{67}$$

For $|g\rangle = |I = 5/2, m = 3/2\rangle$ and $|e\rangle = |I = 3/2, m = 3/2\rangle$, we obtain $\mu_{eg} = \frac{1}{\sqrt{15}} \|\mu_{eg}\|$. For a laser radiation power of 100 mW and if radiation is focused into a spot 1 μm in diameter, we obtain $I = 10^7$ W/cm^2. Such a radiation intensity cannot guarantee the absorption saturation. The Rabi frequency Ω_0 of the transition, according to Eq. (63), is of the order of 1×10^{-5} s^{-1}, and the term containing Ω_0 in the denominator of expression (64) for ρ_{22} can be neglected.

Let the crystal be illuminated with a laser beam characterized by the spatial intensity distribution

$$I = \frac{2P}{\pi w^2} \exp\left(-\frac{2r^2}{w^2}\right), \tag{68}$$

where P is the laser radiation power,

$$w = w_0 \sqrt{1 + \frac{z^2}{b^2}}, \tag{69}$$

r is the distance from the point to the beam axis, w_0 the minimum beam radius (the minimum distance from the beam axis, at which the field decreases by a factor of e), z the coordinate along the beam, $b = \pi w_0^2/\lambda$ is the confocal parameter, and λ the wavelength of laser radiation. Let us determine the number of photons emitted per unit time from the irradiated crystal volume at the two-photon excitation. Let the concentration of thorium nuclei in the specimen be n. Then $\gamma n \rho_{22}$ photons are emitted per unit volume per unit time. Taking the inequality $\Omega_0 \ll \gamma\gamma'$ into account and integrating over the whole specimen volume, we obtain the total number of emitted photons per unit time,

$$F = \int_0^\infty dr\, 2\pi r \int_{-\infty}^\infty dz\, n\mathcal{N} = \frac{n\pi P^2}{2\lambda\gamma'} \left[\frac{\mu_{ge}(\mu_{ee} - \mu_{gg})}{\hbar^2 \omega c} \mu_0\right]^2. \tag{70}$$

In work [21], while estimating the possibility of the creation of an optical γ-laser on a $LiCaAlF_6$ crystal with a thorium impurity, the concentration of the latter was taken to equal 10^{18} cm^{-3}, so that Th^{4+} ions did not change the crystal structure substantially. For our estimation, we adopt, as was done in the case of free ion, that $\gamma' = \gamma_L$. Then, according to Eq. (70), the specimen emits 3.6 photons per second if the radiation intensity is 10 W and $\gamma_L/2\pi = 1$ Hz.

Now, let us evaluate the volume of the active region in the crystal. For $w_0 = 10$ μm and $\lambda = 320$ nm, we obtain $b = 1$ mm. Therefore, the crystal volume can be smaller than 0.1 mm^3. Provided that the concentration equals $n = 10^{18}$ cm^{-3}, such a volume contains $n = 10^{14}$ thorium atoms, 400 of which decay every second. As a result, additional radiation may be generated, by depending on the crystal used. For example, in a CaF_2 crystal 25 mm^3 in dimension, every α-decay of ^{241}Am invoked scintillations (about 40 photons) during 10^{-5} s in the spectral range of 220–400 nm far from the length of the excited ^{229}Th fluorescence signal [27]. This radiation can be reduced considerably by applying a thin specimen in order to allow α-particles to leave it quickly after the decay. In particular, a crystal 1 μm in thickness on a metal substrate can be used, in which the fluorescence can be excited with the help of a surface electromagnetic wave.

9. Conclusions

Our analysis of two-photon optical transitions in a ^{229}Th nucleus showed that the averaged intensities of a propagating monochromatic wave and the field of a sequence of short light pulses, which are required for the identical excitation of nuclei, are equal to each other. The light shifts in the polychromatic and monochromatic fields are also identical. Since radiation for the excitation of nuclei (with a wavelength of about 320 nm) is generated in multiphoton processes, it is easier to obtain the required intensity of laser radiation from a pulse sequence rather than from monochromatic radiation. Moreover, the application of two-photon transitions allows the background scattering signal to be reduced substantially, because nuclei are excited at a fre-

quency that is half as high as that for the fluorescence signal. Another advantage of the pulse excitation consists in a possibility of measuring the frequency of the nuclear transition, provided that the excitation is carried out with the help of a broadband frequency comb. While irradiating Th^{3+} thorium ions in an electromagnetic trap and at a pumping power of 100 mW, the absorption saturation at the two-photon transition can be attained. With regard for the long lifetime of excited nuclei at their detection, an auxiliary radiation can be used, which is in resonance with one of the transitions between the components of the hyperfine structure of a thorium ion with the isomeric nucleus. This circumstance allows the fluorescence signal to be enhanced by several orders of magnitude. If a 10-W laser radiation is used to excite Th^{4+} ions in a solid, several photons per second can expectedly be emitted with a frequency equal to that of the optical transition in a thorium-229 nucleus. Thus, the method proposed here for the creation of excited isomeric state of thorium-229 on the basis of the two-photon absorption can be used in both solid-state optical nuclear clocks and clocks on the basis of ions in an electromagnetic trap.

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ПРЯМЕ ДВОФОТОННЕ ЗБУДЖЕННЯ ІЗОМЕРНОГО ПЕРЕХОДУ В ЯДРІ ТОРІЮ-229

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Розглядається можливість двофотонного збудження ізомерного стану в ядрі торію-229. Показано, що інтенсивність флуоресценції однакова при збудженні ядер монохроматичним випромінюванням або поліхроматичним випромінюванням послідовності коротких світлових імпульсів тієї ж інтенсивності. При двофотонному збудженні іона Th^{3+} в електромагнітній пастці сфокусованим випромінюванням лазера з довжиною хвилі ~ 320 нм і потужністю близько 100 мВт можна досягти насичення поглинання, за якого випромінювання флуоресценції з частотою переходу в ядрі максимальне. В кристалах, допованих Th^{4+} з концентрацією близько 10^{18} см$^{-3}$, у полі лазерного випромінювання потужністю 10 Вт можливе випромінювання кількох фотонів за секунду з довжиною хвилі ~ 160 нм.