The dynamics of spontaneous decay on short and long time scales

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A theory is developed that describes the spontaneous decay of a hydrogenlike atom on time scales that are small or large compared to the reciprocal photon emission frequency. On short time scales the spontaneous decay rate is found to tend in an oscillatory manner to the exponential law of spontaneous decay. The Lamb shift of the energy levels also experiences similar temporal oscillations at the initial stage. Finally, corrections to the exponential decay at late times are shown to depend on the choice of the atom–electromagnetic-field interaction Hamiltonian.
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1. INTRODUCTION

Processes of spontaneous decay of time-dependent quantum systems and the formation of a photon as a wave packet are of great interest to quantum optics.[1–13] The latest investigations in these fields have produced a deeper understanding of the main principles of quantum mechanics of open systems. Another aspect of these studies has been stimulated by the ambiguity in the description of the Lamb shift of the energy levels of an atom and other system parameters by different atom–electromagnetic-field interaction Hamiltonians:[12,13]

$$H_i = -\frac{e}{c\mu}(\mathbf{A}(\mathbf{r},t)\mathbf{p}) + \frac{e^2}{2\mu c^2}\mathbf{A}^2(\mathbf{r},t),$$

$$H_i' = e\int_0^r (d\mathbf{r}'\mathbf{E}(\mathbf{r}',t)).$$

Here $\mathbf{e}(\mathbf{r},t)$ and $\mathbf{A}(\mathbf{r},t)$ are the strength and vector potential of the electromagnetic field, and e, μ,$\mathbf{r}$, and $\mathbf{p}$ are, respectively, the charge, reduced mass, radius vector, and momentum of the electron with respect to the center of mass of the hydrogenlike atom.

The first report on nonexponential spontaneous decay of quasistationary states can be found in the work of Khalfin,[1,2] who demonstrated, that the corrections to the exponential law of spontaneous decay behave like $1/t$, with t the time, basing his reasoning on the fact that the energy distribution density is semifinite. Khalfin's papers did not remain unnoticed, and simultaneously with the development of model atom-field interaction Hamiltonians more exact expressions for the corrections to the exponential spontaneous decay law appeared in the literature.[4,5] The papers of Wilkiewicz and Eberly[7] and Atkins and Woolloy[9] should also be mentioned in this connection, since the researchers point to the need to allow for the time lag within the atom when the corrections to the exponential spontaneous decay law are taken into account. More rigorous calculations show that in time intervals large compared to the reciprocal photon emission frequency ω^{-1} the correction to the exponential spontaneous decay law behaves like $1/t$ and constitutes a small addition to the exponential.[3–8] Also of interest is the effect of the antiresonant terms in the interaction Hamiltonian on the Lamb shift of the energy levels in the process of spontaneous decay. An interesting mathematical study of this problem is given in Refs. 7 and 8.

In contrast to the papers mentioned above, in this paper great attention is paid to the initial stage in the interaction of an excited atom with the vacuum fluctuations of an electromagnetic field and to the transition to the quasiexponential decay over times long compared to ω^{-1}. Note that during the collapse of an excited atom into the ground state the vacuum fluctuations of the electromagnetic field are of the order of the wavelength of the emitted photon. Naturally, the length of the photon's wave packet considerably exceeds the quantum size of the atom proper (we are speaking of the radius of the first Bohr orbit). Hence because of the finiteness of the group velocity the wave packet has not enough time to leave the atom instantaneously, and in small time intervals there is an exchange of energy between an excited nonstationary state of the atom and a state of the electromagnetic field. Such an exchange manifests itself over times of order ω^{-1}.

A new method is also suggested for allowing for the time lag within the atom. The idea is based on integrating the right-hand sides of the kinetic equations over the frequencies of the electromagnetic field and the coordinates of the atom. Allowance for the antiresonance terms in the equation for inversion and for the time lag shows that at early times the emerging wave packet interacts with the excited and ground states [see Eq. (15a)]. For times t of order ω^{-1} the probability of stimulated action on the ground and excited states of the atom is of the same order of magnitude as the reciprocal spontaneous decay time of the atom, and only at late times does the probability tend to the small corrections to the exponential law of spontaneous decay, which are well-known from the literature.

Note that the paper also studies the dependence of spontaneous decay over times that are small or large compared to ω^{-1} on the choice of the interaction Hamiltonian.[9,10] Section 2 focuses on studying spontaneous decay by employing the common interaction Hamiltonian $e(\mathbf{A}(\mathbf{r},t)\cdot\mathbf{p})/c\mu$. There it is shown that over long times $t \gg \omega^{-1}$ the expressions for the Lamb shift and corrections to the exponential spontaneous decay law coincide with the ordinary expressions for these quantities (see Ref. 8). Also the

oscillatory nature of the decay of the system in time intervals $t \leqslant \omega^{-1}$ is extensively discussed.

Section 3 studies the same problem with a new gauge imposed on the vector and scalar potentials of the electromagnetic field. There it is shown that the corrections to the exponential spontaneous decay law obtained with a multiple interaction Hamiltonian[9-11] are much smaller than those obtained by the common interaction Hamiltonian for a time interval $t > \omega^{-1}$. The oscillatory behavior of the inversion of the atom and the rate of spontaneous decay at early times is also obtained in that section.

Finally, Sec. 4 discusses the various assumptions made in this paper that lead to discrepancies between the results obtained with the two forms of the interaction Hamiltonian.

2. SPONTANEOUS DECAY WITH ALLOWANCE FOR THE INTERACTION HAMILTONIAN $e(\mathbf{A}(\mathbf{r},t)\mathbf{p})/c\mu$

The process of studying the interaction of a hydrogenlike atom and a variable electromagnetic field is simplified by shifting the origin to the center of mass of the electron and nucleus. Ignoring the motion of the nucleus with respect to the center of mass, we can describe the motion of the electron in the central field of the nucleus and the variable electromagnetic field by the Hamiltonian

$$H(t)=\frac{1}{2\mu}\left\{\mathbf{p}-\frac{e}{c}\mathbf{A}^{g}(\mathbf{r},t)\right\}^{2}-\frac{ze^{2}}{r}+e\Phi^{g}(\mathbf{r},t). \tag{1}$$

Here $\mathbf{r}$ and $\mathbf{p}=i\hbar\nabla$ are the electron position and momentum vectors with respect to the center of mass, and z is the charge of the nucleus. The potentials $\mathbf{A}^{g}(\mathbf{r},t)$ and $\Phi^{g}(\mathbf{r},t)$ can be transformed into a new pair of potentials by a gauge transformation (see, e.g., Ref. 7)

$$\begin{aligned}&\mathbf{A}(\mathbf{r},t)=\mathbf{A}^{g}(\mathbf{r},t)-\nabla\chi(\mathbf{r},t),\\&\Phi(\mathbf{r},t)=\Phi^{g}(\mathbf{r},t)+\partial\chi(\mathbf{r},t)/\partial t,\end{aligned} \tag{2}$$

where $\chi(\mathbf{r},t)$ is a transformation function dependent on the coordinate $\mathbf{r}$ and time t. If we use (2) to select the function $\chi(\mathbf{r},t)$ is such a way that the scalar potential satisfies $\Phi(\mathbf{r},t)=0$, the Hamiltonian of the system consisting of the atom and the radiation field can be written as[7,9,10]

$$H=H_{0}+H_{i}, \tag{3a}$$

$$H_{0}=\frac{\mathbf{p}^{2}}{2\mu}-\frac{ze^{2}}{r}+\frac{1}{2\pi}\int dV(\mathbf{B}^{2}+\mathbf{E}^{2}), \tag{3b}$$

$$H_{i}=-\frac{e}{c\mu}(\mathbf{A}(\mathbf{r},t)\mathbf{p})+\frac{e^{2}}{2\mu c^{2}}\mathbf{A}^{2}(\mathbf{r},t). \tag{3c}$$

Here $\mathbf{E}$ and $\mathbf{B}$ are the vectors of the electric and magnetic components of the transverse electromagnetic field.

The aim of this section is to allow for the retardation of the emitted electromagnetic field in the process of collapse of the excited electron to the ground state. Hence in the interaction Hamiltonian (3c) in the nonrelativistic approximation we ignore the term proportional to the square of the vector potential, which is common practice. If we assume that when the interaction between the hydrogenlike atom and the electromagnetic-field vacuum is switched on the electron is in the quantum state $|n=2,l=1,m=0\rangle$ (here n, l, and m are, respectively, the principal, orbital, and magnetic quantum numbers), the spontaneous transition between the states $|2,1,0\rangle$ and $|1,0,0\rangle$ can be described by the Hamiltonian equation

$$\begin{aligned}H=\hbar\omega_{0}R_{3}+\sum_{k}\hbar\omega_{k}(a_{k}^{+}a_{k}+1/2)+\sum_{k}g_{k}\{(\varphi_{12}(k)a_{k}^{+}\\+\varphi_{12}(-k)a_{k})R^{-}+(\varphi_{21}(k)a_{k}+\varphi_{21}(-k)a_{k}^{+})R^{+}\}.\end{aligned} \tag{4}$$

Here

$$\begin{aligned}&\varphi_{12}(k)=\frac{ie\hbar}{c\mu}\int dV\psi_{100}(r)(\mathbf{e}_{\lambda},\nabla)\exp\{-(\mathbf{kr})\}\psi_{210}(r),\\&\varphi_{21}(k)=\varphi_{12}^{+}(k),\quad g_{k}=\sqrt{2\pi c^{2}\hbar/V\omega_{k}},\end{aligned} \tag{5}$$

R^{+}, R^{-}, R_{3}, a_{k}^{+}, and a_{k} are the operators of the atomic subsystem and the electromagnetic field satisfying the commutation relations

$$\begin{aligned}&[R^{+},R^{-}]=2R_{3},\quad [R^{\pm},R_{3}]=\mp R^{\pm},\\&R^{+}R^{-}+R^{-}R^{+}=1,\\&[a_{k},a_{k}^{+}]=\delta_{kk'},\quad [a_{k},a_{k'}]=[a_{k}^{+},a_{k'}^{+}]=0,\end{aligned} \tag{5a}$$

V is the quantization volume, $\mathbf{e}_{\lambda}$ is the photon polarization vector $(\lambda=1,2)$, $k=(\mathbf{k},\lambda)$, $\psi_{100}=\exp(-\omega)/\sqrt{\pi}$ and $\psi_{210}=i\omega\cos(\vartheta)/4\sqrt{2\pi}$ are the wave functions of the ground ($|1s\rangle$) and the excited ($|2p\rangle$) states of the hydrogenlike atom, $\rho=r/a$, $a=\hbar/\mu e^{2}z$ is the first Bohr radius, and $\hbar\omega_{0}$ is the energy separation of the ground $|1s\rangle$ and excited $|2p\rangle$ states. In deriving the Hamiltonian (4) from (3) we allowed for the antiresonant terms in the interaction with the electromagnetic field. Note that to ensure the convergence of integrals when calculating the Lamb shift and the corrections to the spontaneous decay we did not employ the common dipole approximation $\varphi\simeq i\omega_{0}(\mathbf{e}_{\lambda}\cdot\mathbf{d})\hbar/c$ in (4) and (5) ($\mathbf{d}$ is the dipole moment of the transition).

The Heisenberg equation for the population difference assumes the form

$$\begin{aligned}\frac{dR_{3}(t)}{dt}=\frac{i}{\hbar}\sum_{k}g_{k}\{(\varphi_{12}(k)a_{k}^{+}(t)+\varphi_{12}(-k)a_{k}(t))R^{-}(t)\\-(\varphi_{21}(k)a_{k}(t)+\varphi_{21}(-k)a_{k}^{+}(t))R^{+}(t)\}.\end{aligned} \tag{6}$$

The solution of the Heisenberg equation for the electromagnetic-field operators $a_{k}^{+}(t)$ and $a_{k}(t)$ is

$$\begin{aligned}&a_{k}^{+}(t)=a_{k}^{+}(0)\exp(i\omega_{k}t)+i\frac{g_{k}}{\hbar}\int_{0}^{t}d\tau\{\varphi_{12}(-k)R^{-}(t-\tau)\\&\qquad+\varphi_{21}(k)R^{+}(t-\tau)\}\exp[i\omega_{k}\tau],\\&a_{k}(t)=[a_{k}^{+}(t)]^{+}.\end{aligned} \tag{7}$$

After substituting (7) into (6) and averaging over the initial state $\psi(t=0)=|f\rangle|A\rangle$ of the system (here $|f\rangle$ and $|A\rangle$ are the states of the electromagnetic-field vacuum and the atom at $t=0$) we exclude the free solution of Eq. (7), $a_{k}(0)|f\rangle=0$ and $\langle f|a_{k}^{+}(0)=0$. Thus, Eq. (6) assumes the form

$$\frac{d\langle R_3(t)\rangle}{dt}=-\sum_k \frac{g_k^2}{\hbar^2}\int_0^t d\tau\{\langle(\varphi_{21}(k)R^+(t-\tau)+\varphi_{12}(-k)\times R^-(t-\tau))(R^-(t)\varphi_{12}(k)-\varphi_{21}(-k)\times R^+(t))\rangle\}\exp[i\omega_k\tau]+\text{ h.c.} \tag{8a}$$

An equation for $\langle R^+(t)|$ can be obtained in the same way:

$$\frac{d\langle R^+(t)\rangle}{dt}=i\omega_0\langle R^+(t)\rangle+2\sum_k \frac{g_k^2}{\hbar^2}\int_0^t d\tau\{|\varphi_{12}(k)|^2\times[\langle R_3(t)R^+(t-\tau)\rangle\exp(-i\omega_k\tau)-\langle R^+(t-\tau)R_3(t)\rangle\exp(i\omega_k\tau)]-\varphi_{12}(k)\varphi_{12}(-k)\times[\exp(i\omega_k\tau)\langle R^-(t-\tau)R_3(t)\rangle-\exp(-i\omega_k\tau)\langle R_3(t)R^-(t-\tau)\rangle]\}. \tag{8b}$$

The right-hand sides of Eqs. (8a) and (8b) contain expressions of the form

$$J(\tau)=\sum_k \varphi_{21}(k)\varphi_{12}(k)\exp(i\omega_k\tau)g_k^2/\hbar^2, \tag{9}$$

which are integrated over neither the spatial variables of the atom nor $\mathbf{k}$. Since the emerging wave packet has the size of approximately the radiation wavelength and is larger than the first Bohr orbits by a factor of several thousands, exact integration of Eq. (9) would make it possible to study the behavior of the spontaneous decay rate in the initial stage of development, when, due to its finite size, the photon cannot leave, the atom immediately because of the finiteness of the group velocity. The sum over the polarizations λ and the integral over all the directions of $\mathbf{k}$ can easily be calculated if in the integration over the angles we direct the polar axis along the retardation vector $\mathbf{r}_1-\mathbf{r}_2$ in (9):

$$\sum_{\lambda=1}^{2}\int d\Omega_k \exp[i\mathbf{k}\cdot(\mathbf{r}_1-\mathbf{r}_2)](\mathbf{e}_\lambda\cdot\mathbf{p}_1)(\mathbf{e}_\lambda\cdot\mathbf{p}_2)=2\pi\left\{\frac{\sin(kr_{12})}{kr_{12}}[(\mathbf{p}_1\cdot\mathbf{p}_2)+(\mathbf{n}_{12}\cdot\mathbf{p}_1)(\mathbf{n}_{12}\cdot\mathbf{p}_2)]-\frac{1}{\mathbf{r}_{12}^2}\frac{\partial^2}{\partial k^2}\frac{\sin(kr_{12})}{kr_{12}}[(\mathbf{p}_1\cdot\mathbf{p}_2)-3(\mathbf{n}_{12}\cdot\mathbf{p}_1)(\mathbf{n}_{12}\cdot\mathbf{p}_2)]\right\}, \tag{10}$$

where

$$\mathbf{n}_{12}=(\mathbf{r}_1-\mathbf{r}_2)/r_{12},\quad r_{12}=|\mathbf{r}_1-\mathbf{r}_2|,\quad \mathbf{p}_1=\nabla_1,\quad \mathbf{p}_2=\nabla_2$$

(here $\nabla_i=\text{grad}|_{\mathbf{r}_i}$). Thus, the integral with respect to the absolute value of the wave vector $\mathbf{k}$ and the electron coordinate assumes the form

$$J(\tau)=\frac{e\hbar^2}{12\pi\mu^2a^5}\int_0^\infty \frac{\exp(i\omega\tau)}{\omega^2}d\omega\int_0^\infty \rho_2^3d\rho_2\int_0^\infty \rho_1^3d\rho_1\times\int_{-1}^{1}dx\exp\left(-\frac{3}{2}(\rho_1+\rho_2)\right)\left[(2\rho_1-x\rho_1\rho_2)\times(1-x^2)\frac{1}{u}\frac{\partial}{\partial u}\frac{1}{u}\frac{\partial}{\partial u}+2(2\rho_1-u^2)\frac{\partial}{\partial u}\right]\times\frac{\sin(\omega au/c)}{u}, \tag{11}$$

where $u=\rho_1^2+\rho_2^2-2x\rho_1\rho_2$.

Simultaneous integration over the frequencies ω and the electron coordinate in (11) is reduced to the following dependence on the time lag τ:

$$J(\tau)=\frac{2^2e^2\hbar}{3^6a^4\mu^2}\left\{\frac{\partial}{\partial b}-\frac{3}{2}\frac{\partial^2}{\partial b^2}+\frac{3}{4}\frac{\partial^3}{\partial b^3}\right\}\left\{i\pi\times\exp\left(-\frac{cb\tau}{a}\right)-\exp\left(-\frac{cb\tau}{a}\right)\text{Ei}\left(\frac{cb\tau}{a}\right)-\exp\left(\frac{cb\tau}{a}\right)\text{Ei}\left(-\frac{cb\tau}{a}\right)\right\}\Bigg|_{b=3/2}. \tag{12}$$

Here $\text{Ei}(x)$ is the exponential integral. Simple transformations show that

$$K(\tau)=\sum \varphi_{12}^2(k)\exp(i\omega_k\tau)g_k^2/\hbar^2 \tag{13}$$

is reduced to the integral (12): $K(\tau)=-J(\tau)$.

For exact integration over the time lag τ we must know the time-dependence of the correlators $\langle f(t-\tau)f(t)\rangle$, where $f(t)=R^+(t)$, $R^-(t)$, or R_3 on the right-hand sides of Eqs. (8a) and (8b). Since $J(\tau)$, $K(\tau)$, and similar Hermitian conjugate functions rapidly decrease as functions of τ in time intervals that are large compared to a/c, it is convenient to separate the rapidly oscillating part (with respect to τ) in the operators $R^\pm(t-\tau)$:

$$R^\pm(t-\tau)=\tilde{R}^\pm(t-\tau)\exp[\pm i\omega_0(t-\tau)].$$

Here the functions $\tilde{R}^\pm(t-\tau)$ are assumed to vary smoothly compared to $\exp[i\omega_0(t-\tau)]$ in the sense that $|d\tilde{R}^\pm(t-\tau)/d(t-\tau)|\ll|\tilde{R}^\pm(t-\tau)|\omega_0$. This can easily be verified by observing that the main contribution to the integrals with respect to τ on the right-hand sides of Eqs. (8a) and (8b) is provided only by the power expansion of $\tilde{R}^\pm(\tau-\tau)$ in τ. As Eq. (12) implies, τ is of order a/c, i.e., the expansion corresponds to the series expansion in the small parameter ω_0a/c. In the above approximation the correlators on the right-hand sides of Eqs. (8a) and (8b) can be approximated by the following expressions both for early and late times (compared to ω_0^{-1}):

$$\begin{aligned}
&\langle R^+(t-\tau)R^+(t)\rangle\simeq\exp(-i\omega_0\tau)\langle R^+(t)R^+(t)\rangle=0,\\
&\langle R^+(t-\tau)R^-(t)\rangle\simeq\exp(-i\omega_0\tau)[\langle R_3(t)\rangle+0.5],\\
&\langle R^-(t-\tau)R^+(t)\rangle\simeq\exp(i\omega_0\tau)[0.5-\langle R_3(t)\rangle],\\
&\langle R^+(t-\tau)R_3(t)\rangle\simeq-\exp(-i\omega_0\tau)\langle R^+(t)\rangle/2,\\
&\langle R^-(t-\tau)R_3(t)\rangle\simeq\exp(i\omega_0\tau)\langle R^-(t)R^+(t)\rangle/2.
\end{aligned} \tag{14}$$

The other correlators in (8a) and (8b) are the Hermitian conjugates of (14). Thus, with allowance for the correlators (12)–(14) Eqs. (8a) and (8b) assume the form

$$\frac{d\langle R_3(t)\rangle}{dt}=-[A_\parallel(t)+B(t)][\langle R_3(t)\rangle+0.5]+B(t)[0.5-\langle R_3(t)\rangle], \tag{15a}$$

$$\frac{d\langle R^+(t)\rangle}{dt}=i(\omega_0+\Delta_0+iA_\perp)\langle R^+(t)\rangle+(\Delta_0-iA_\perp)\times\langle R^-(t)\rangle. \tag{15b}$$

Here

$$A_\parallel(t)=-\frac{2^7z^2\alpha^3}{3^8a}\hat{E}(u)L(u,t)\Big|_{u=1},$$

$$B(t)=-\frac{2^6z^2\alpha^3c}{3^8a}\hat{E}(u)M(u,t)\Big|_{u=1}, \tag{16}$$

$$\Delta_0=\frac{2^5z^2\alpha^3c}{3^8\pi a}\hat{E}(u)\chi(u,t)\Big|_{u=1},$$

$$A_\perp=\frac{2^5z^2\alpha^3c}{3^8\pi a}\hat{E}(u)\sigma(u,t)\Big|_{u=1}, \tag{17}$$

$$L(t,u)=\frac{1}{u^2+\delta^2}\{\delta-\exp(-\nu ut)[u\sin(\omega_0t)+\delta\cos(\omega_0t)]\}, \tag{16a}$$

$$M(u,t)=\frac{1}{u^2+\delta^2}\left\{\left[\frac{u}{\pi}[\exp(-\nu ut)\times\mathrm{Ei}(\nu ut)-\exp(\nu ut)\mathrm{Ei}(-\nu ut)]+\delta\times\exp(-\nu ut)\right]\cos(\omega_0t)+\left[u\exp(-\nu ut)-\frac{\delta}{\pi}[\exp(-\nu ut)\mathrm{Ei}(\nu ut)+\exp(\nu ut)\mathrm{Ei}(-\nu ut)]\right]\sin(\omega_0t)+\delta\frac{2}{\pi}\times\left[\mathrm{Si}(\omega_0t)-\frac{\pi}{2}\right]\right\}, \tag{16b}$$

$$\chi(u,t)=\frac{1}{u^2+\delta^2}\{\delta\cos(\omega_0t)[\exp(-\nu ut)\mathrm{Ei}(\nu ut)+\exp(\nu ut)\mathrm{Ei}(-\nu ut)]+u\sin(\omega_0t)\times[\exp(-\nu ut)\mathrm{Ei}(\nu ut)-\exp(\nu ut)\mathrm{Ei}(-\nu ut)]-2\delta[\mathrm{Ci}(\omega_0t)-\ln(\nu u)]\}, \tag{17a}$$

$$\sigma(u,t)=-\frac{\pi}{2(u^2+\delta^2)}\{\delta\cos(\omega_0t)\exp(-\nu ut)+u\sin(\omega_0t)\exp(-\nu ut)-\delta\}+\frac{\pi}{2}M(u,t), \tag{17b}$$

$\nu=3c/2a$, $\delta=3\omega_0a/2c$, $\mathrm{Si}(x)$ and $\mathrm{Ci}(x)$ are the sine and cosine integrals,[14] and

$$\hat{E}(u)=\frac{\partial}{\partial u}-\frac{\partial^2}{\partial u^2}+\frac{1}{3}\frac{\partial^3}{\partial u^3}. \tag{18}$$

Equation (15a) implies that the emerging wave packet (photon) in the time interval $t\sim1/\omega_0$ has a strong effect not only on the excited state of the atom, $\langle R_3(t)\rangle+0.5$, but also on the ground state $-\langle R_3(t)\rangle+0.5$. It can easily be shown that the coefficient of the stimulating action of the emerging photon field, $B(t)$, is of the same order of magnitude as the spontaneous transition coefficient $A_\parallel(t)$ only for $a/c\sim t\leqslant1/\omega_0$. Since the atom is several thousand times smaller than the radiation wavelength, $a\ll\lambda_0=2\pi c/\omega_0$, the expressions for $A_\parallel(t)$, $B(t)$, Δ_0 and $A_\perp$ in (16) and (17) simplify considerably when $t\gg a/c$. Indeed, expanding the function $1/(u^2+\delta^2)$ in a power series in the small parameter δ,

$$\frac{1}{u^2+\delta^2}=\frac{1}{u^2}\left[1-\left(\frac{\delta}{u}\right)^2+\left(\frac{\delta}{u}\right)^2-\cdots\right], \tag{19}$$

and allowing for the asymptotic behavior of the exponential integral at large values of the argument ($t\gg2a/3c$; see Refs. 14 and 15),

$$\mathrm{Ei}(\nu ut)\simeq\frac{\exp(\nu ut)}{\nu ut}\left[1+\frac{1}{\nu ut}+\frac{2!}{(\nu ut)^2}+\cdots\right], \tag{19b}$$

we arrive at the following expressions for the above parameters to lowest order in the smallness parameter $(\nu ut)^{-1}$:

$$A_\parallel=\frac{1}{\tau_0},\quad B(t)\simeq\frac{1}{\pi\tau_0}\left\{\mathrm{Si}(\omega_0t)-\frac{\pi}{2}+\frac{\cos(\omega_0t)}{\omega_0t}\right\},$$
$$A_\perp=\frac{1}{\pi\tau_0}\left[\mathrm{Si}(\omega_0t)+\frac{\cos(\omega_0t)}{\omega_0t}\right], \tag{20}$$

$$\Delta_0=\frac{1}{\pi\tau_0}\left\{\ln\left(\frac{\nu}{\omega_0}\right)-\frac{11}{12}+\mathrm{Ci}(\omega_0t)-\frac{\sin(\omega_0t)}{\omega_0t}\right\}, \tag{21}$$

$$\tau_0=3^{11}\nu^2\hbar c/2^{18}e^2\omega_0^3.$$

Here we used only the first term in each expansion (18) and (19).

In the asymptotic limit in expressions (16) and (20) the coefficients $A_\parallel$ and $B(t)$ in the time interval $a/c\ll t\sim\omega_0$ acquire a simpler form. Note that at late times $t>\omega_0^{-1}$ the coefficient of the stimulating action of the emitted field on the ground and excited states of the atom, $B(t)$, decays in an oscillatory manner. In this situation we can use the asymptotic expansion of the sine integral $\mathrm{Si}(\omega_0t)$ (see Ref. 14):

$$\mathrm{Si}(\omega_0t)\simeq\frac{\pi}{2}-f(\omega_0t)\cos(\omega_0t)-g(\omega_0t)\sin(\omega_0t), \tag{22}$$

$$f(\omega_0t)\simeq\frac{1}{\omega_0t}\left[1-\frac{2!}{(\omega_0t)^2}+\frac{4!}{(\omega_0t)^4}-\cdots\right],$$

$$g(\omega_0t)\simeq\frac{1}{(\omega_0t)^2}\left[1-\frac{3!}{(\omega_0t)^2}+\frac{5!}{(\omega_0t)^4}-\cdots\right].$$

If we allow for the expansion (22), we arrive at the following expression for the coefficient $B(t)$:

$$B(t)\approx -\frac{1}{\pi\tau_0}\frac{\sin(\omega_0 t)}{(\omega_0 t)^2} \quad \text{for } \omega_0 t\gg 1, \tag{23}$$

which coincides with the correction to the exponential spontaneous decay law calculated in Refs. 4–8. However, the equations and expressions obtained in this section for the spontaneous decay rate allow for the corrections to the exponential spontaneous decay law not only in late times ($t\gg\omega_0^{-1}$) but also at early and intermediate times ($t\leqslant\omega_0^{-1}$; see Eqs. (16)–(20)).

The parameters Δ_0 and $A_\perp(t)$ in Eq. (15b) take into account the correction to the oscillation frequency of the quantum dipole (i.e., the Lamb shift of the energy levels) and the damping of the transverse polarization of this dipole at a given time t in the evolution of the system. As Eq. (21) implies, the expression for these parameters simplifies considerably for $t\gg a/c$. From Eqs. (17) and (21) it also follows that for $t\sim\omega_0^{-1}$ the quantities Δ_0 and $A_\perp(t)$ grow in an oscillatory manner, reaching a certain limit when $t\gg\omega_0^{-1}$. If we employ the asymptotic expansion of the cosine integral for large values of the argument,

$$\mathrm{Ci}(\omega_0 t)=f(\omega_0 t)\sin(\omega_0 t)-g(\omega_0 t)\cos(\omega_0 t) \tag{24}$$

(the functions $f(\omega_0 t)$ and $g(\omega_0 t)$ are defined in (22a)), in the lowest order of the power expansion in the smallness parameter $1/\omega_0 t$ we arrive at the following expressions for $\Delta_0(t)$ and $A_\perp(t)$:

$$\Delta_0(t)\approx -\frac{1}{\pi\tau_0}\left\{\ln\left(\frac{\nu}{\omega_0}\right)-\frac{11}{12}-\frac{\cos(\omega_0 t)}{(\omega_0 t)^2}\right\}, \tag{25a}$$

$$A_\perp(t)\approx \frac{1}{\pi\tau_0}\left\{\frac{\pi}{2}-\frac{\sin(\omega_0 t)}{(\omega_0 t)^2}\right\}. \tag{25b}$$

The expression (25a) for $\Delta_0(t)$ makes it possible to compare the suggested method of studying spontaneous decay with the results obtained via the Laplace transformation method.[3–8] Allowing for the accuracy of the calculations of the Lamb shift of energy levels done by Seke and Herfort[8] by the Laplace transformation method, we can compare the above expression for Δ_0 with their results. This is a meaningful approach if only because Seke and Herfort[8] demonstrated the need to allow for antiresonant terms in calculating the Lamb shift of energy levels. In our notation the expression for Δ_0 given in Ref. 8 is

$$\Delta_0^{\mathrm{SH}}=\frac{1}{\pi\tau_0}\left[\ln\left(\frac{\nu}{\omega_0}\right)-\frac{11}{12}\right]. \tag{26}$$

As Eqs. (25a) and (26) imply, the constant part of the expression for Δ_0 is contained in (26). In other words, our expression for the energy-level shift tends to the one obtained earlier for this parameter Δ_0^{SH} in the limit $\omega_0 t\gg 1$. Note that the energy-level shift (21) is a function of the spontaneous decay process, which means it is impossible to introduce an average value for the shift that would encompass both short and long times compared to ω_0^{-1}.

To conclude this section, here is the solution of Eq. (15a) with the initial data $\langle R_3(0)\rangle=0.5$:

$$\langle R_3(t)\rangle=-0.5+\exp\left\{-\int_0^t d\tau[A_\parallel(\tau)+2B(\tau)]\right\} + \int_0^t d\tau\, B(\tau)\exp\left\{-\int_\tau^t d\tau'[A_\parallel(\tau')+2B(\tau')]\right\}. \tag{27}$$

For the time interval $a/c\ll\tau_0$ the solution (27) can be written as

$$\langle R_3(t)\rangle=-0.5+\exp\{-t[1+2\chi(t)]/\tau_0\}+t\chi(t)/\tau_0, \tag{27a}$$

where

$$\chi(t)=\mathrm{Si}(t)-\frac{\pi}{2}+[\cos(\omega_0 t)-1+\mathrm{Ci}(\omega_0 t) + \ln(\nu/\omega_0)-11/12]/\omega_0 t.$$

Equation (27a) implies that the rate of variation of the population difference between the atomic levels tends in an oscillatory manner to the exponential spontaneous decay law as $\omega_0 t$ increases.

3. SPONTANEOUS DECAY WITH A MULTIPOLE HAMILTONIAN

If we use the operator

$$\Lambda=\exp\left\{-\frac{i}{\hbar}\frac{e}{c}\int_\zeta^{\mathbf{r}}(d\mathbf{r}'\cdot\mathbf{A}(\mathbf{r}',t))\right\}, \tag{28}$$

then, according to the unitary transformation procedure suggested in Refs. 9 and 10, $\tilde{H}=\Lambda H\Lambda^{-1}$, and we can obtain a new form [compared to (3)] of the atom–electromagnetic-field interaction Hamiltonian:

$$\tilde{H}=\tilde{H}_0+\tilde{H}_i, \tag{29a}$$

where

$$H_0=\frac{p^2}{2\mu}-\frac{e^2}{r}+\frac{1}{8\pi}\int dV'[\mathbf{E}_\perp^2(\mathbf{r}',t)+\mathbf{B}^2(\mathbf{r}',t)], \tag{29b}$$

$$H_i=-\int dV'\{(\mathbf{P}(\mathbf{r}')\cdot\mathbf{E}_\perp(\mathbf{r}'))+(\mathbf{M}(\mathbf{r}')\cdot\mathbf{B}(\mathbf{r}'))\} + 2\pi\int dV'\mathbf{P}_\perp^2(\mathbf{r}')+\frac{1}{2\mu}\mathbf{Q}^2(\mathbf{r}). \tag{29c}$$

Here, following Ref. 10, we have employed the following notation: $\mathbf{P}(\mathbf{r}')=\mathbf{P}_\perp(\mathbf{r}')+\mathbf{P}_\parallel(\mathbf{r}')$ is the atomic polarization,

$$\mathbf{P}(\mathbf{r}')=\int_\zeta^{\mathbf{r}}d\mathbf{r}''\delta(\mathbf{r}''-\mathbf{r}'),$$

$\mathbf{P}_\perp(\mathbf{r}')$ and $\mathbf{P}_\parallel(\mathbf{r}')$ are the transverse and longitudinal atomic polarizations,

$$\mathbf{P}_\parallel(\mathbf{r}')=\mathrm{grad}|_{\mathbf{r}'}\left\{\frac{1}{|\mathbf{r}-\mathbf{r}'|}-\frac{1}{r'}\right\}\frac{e}{4\pi},$$

$$\mathbf{Q}(\mathbf{r})=\int_\xi^1 v\,dv[\mathbf{r}\cdot\mathbf{B}(v\mathbf{r})]\frac{e}{c},$$

$$\mathbf{M}(\mathbf{r}')=\frac{e}{\mu}\sum_{n=1}^{\infty}\frac{(-1)^{n+1}n}{(n+1)!}(\mathbf{r}\cdot\nabla_{\mathbf{r}'})^{n-1}\mathbf{r}\times\mathbf{p}\delta(\mathbf{r}'),$$

ζ is a small parameter, the distance to the point occupied by the electron when the interaction with the electromagnetic field is switched on, and $\xi=\zeta/r$. After substituting the expressions for the polarization and $\mathbf{M}(\mathbf{r})$ into (29c) we arrive at the following expression for the first term in that formula:

$$\int dV'\{(\mathbf{P}(\mathbf{r}')\mathbf{E}_{\perp}(\mathbf{r}')+\mathbf{M}(\mathbf{r}')\mathbf{B}(\mathbf{r}'))\}$$
$$=e\int_{\zeta}^{\mathbf{r}}(d\mathbf{r}'\mathbf{E}_{\perp}(\mathbf{r}'))+\frac{e}{\mu c}\int_{\xi}^{1}v\,dv(\mathbf{r}\times\mathbf{p}\cdot\mathbf{B}(v\mathbf{r})). \tag{29d}$$

In classical terms Eq. (29d) describes the energy needed for moving an electron from the initial point ζ to a point at a distance r from the nucleus in an external electromagnetic field.

Since an unperturbed atom is assumed to be in the $|s\rangle$ state, it convenient to let the parameter ζ go to zero. This, however, will be done only after integrating over the electromagnetic-field frequencies. The form of the suggested Hamiltonian is convenient because the interaction with the electromagnetic field can be interpreted as the interaction of the field and electric and magnetic multipoles.[9,10] In the interaction Hamiltonian (29b) the term $2\int dV'\mathbf{P}_{\perp}^2(r')$ acts only on the electron states of the atom and renormalizes these states thanks to the presence of an electromagnetic field. Hence this term in the Hamiltonian (29a) can be ignored since it contributes very little to the energy states of the atoms in comparison to $\tilde{H}_0$. The term which is nonlinear in the electromagnetic-field operators, $\mathbf{Q}^2/2\mu$, can also be dropped from (29c) because in the nonrelativistic approximation ($v/c\ll 1$) its contribution is small compared to the linear term.

Bearing all this in mind, we consider the spontaneous decay as occurring only between the $|2p\rangle$ and $|1s\rangle$ states of the hydrogenlike atom. Then, as in Sec. 2, we arrive at the following Hamiltonian in the second-quantization representation:

$$\tilde{H}=\hbar\omega_0 R_3+\sum_k\hbar\omega_k(b_k^+b_k+1/2)+i\sum_k q_k\{[(\Phi_{12}(k)-\mu_{12}(k))R^-+(\Phi_{21}(-k)+\mu_{21}(-k))R^+]b_k^+ - b_k[(\Phi_{12}(-k)+\mu_{12}(-k))R^+ +(\Phi_{21}(k)-\mu_{21}(k))R^-]\}, \tag{30}$$

where

$$\Phi_{12}(k)=e\int dV\psi_{100}(r)\int_{\zeta}^{\mathbf{r}}(\mathbf{e}_{\lambda}\cdot d\mathbf{r}')\times\exp\{-(\mathbf{k}\cdot\mathbf{r})\}\psi_{210}(r),$$
$$\mu_{12}(k)=\frac{\hbar\omega_k e}{2\mu c^2}\int dV\psi_{100}(r)(\mathbf{e}_{\lambda}\cdot\mathbf{r})\int_{\xi}^{1}v^2dv\times\exp\{-(\mathbf{k}\cdot\mathbf{r})v\}\psi_{210}(r), \tag{31}$$

$$\Phi_{21}=\Phi_{12}^+(k),\quad \mu_{21}(k)=\mu_{12}^+(k),\quad q_k=\sqrt{2\pi\hbar\omega_k/V}.$$

The remaining notation is similar to that used in Sec. 2: b_k^+, b_k, R^+, R^-, and R_3 are the operators of the electromagnetic field and the atom; they satisfy the same commutation relations (5a).

Eliminating the electromagnetic-field boson operators from the Heisenberg equations for $R_3(t)$ and $R^+(f)$ as we did in Sec. 2, we arrive at the following equations for the averages of these operators:

$$\frac{d\langle R_3(t)\rangle}{dt}=-\sum_k\frac{q_k^2}{\hbar^2}\int_0^t d\tau\exp(i\omega_k\tau)\{\langle[(\Phi_{21}(k)-\mu_{21}(k))\times R^+(t-\tau)+(\Phi_{12}(-k)+\mu_{12}(-k))\times R^-(t-\tau)][(\Phi_{12}(k)-\mu_{12}(k))R^-(t) -(\Phi_{21}(-k)-\mu_{21}(-k))R^+(t)]\rangle\}+\text{h.c.}, \tag{32a}$$

$$\frac{d\langle R^+(t)\rangle}{dt}=i\omega_0\langle R^+(t)\rangle+2\sum_k\frac{q_k^2}{\hbar^2}\int_0^t d\tau\{\exp(i\omega_k\tau)\times\langle[(\Phi_{21}(k)-\mu_{21}(k))R^+(t-\tau)+(\Phi_{12}(-k)+\mu_{12}(-k))R^-(t-\tau)]R_3(t)(\Phi_{21}(-k)+\mu_{21}(-k))\rangle-\exp(-i\omega_k\tau)\langle(\Phi_{21}(k)-\mu_{21}(k))R_3(t)[(\Phi_{21}(-k)+\mu_{21}\times(-k))R^+(t-\tau)+(\Phi_{12}(k)-\mu_{12}(k))R^-(t-\tau)]\rangle\}. \tag{32b}$$

Let us now study the sum over k in expressions like

$$I(\tau)=\sum_k(\Phi_{21}(k)-\mu_{21}(k))(\Phi_{12}(k)-\mu_{12}(k))\exp(i\omega_k\tau)q_k^2/\hbar^2. \tag{33}$$

Here the integral over the solid angle and the sum over photon polarizations in the transition from summation with respect to k to integration can be reduced to an expression of type (10) if we assume that the electron moves from point ζ to point $\mathbf{r}$ along the straight line connecting these points:

$$\sum_k\int d\Omega\int_{\zeta}^{r_1}dr_1'\int_{\zeta}^{r_2}dr_2'\exp[i(\mathbf{k}\cdot(\mathbf{r}_1'-\mathbf{r}_2'))](\mathbf{e}_{\lambda}\cdot\mathbf{n}_1')\times(\mathbf{e}_{\lambda}\cdot\mathbf{n}_2')$$
$$=4\pi\int_{\zeta}^{r_1}dr_1'\int_{\zeta}^{r_2}dr_2'\left\{\frac{\sin(kr_{12}')}{kr_{12}'}[(\mathbf{n}_1'\cdot\mathbf{n}_2')+(\mathbf{n}_{12}'\cdot\mathbf{n}_1')\times(\mathbf{n}_{12}'\cdot\mathbf{n}_2')]-[(\mathbf{n}_1'\cdot\mathbf{n}_2')-3(\mathbf{n}_{12}'\cdot\mathbf{n}_1')\times(\mathbf{n}_{12}'\cdot\mathbf{n}_2')]\frac{\partial^2}{\partial(kr_{12}')^2}\frac{\sin(kr_{12}')}{kr_{12}'}\right\}, \tag{34}$$

where $\mathbf{n}_{12}=(\mathbf{r}_1-\mathbf{r}_2)/r_{12}'$, $r_{12}'=|\mathbf{r}_1'-\mathbf{r}_2'|$, and $\mathbf{n}_i=\mathbf{r}_i'/r_i'$, with $i=1,2$.

Substitution of (34) into (33) yields the following expression for $I(\tau)$:

$$I(\tau)=\frac{e^2}{6\pi\hbar a}\int_0^\infty \exp(i\omega\tau)d\omega\int_0^\infty \rho_2^3 d\rho_2\int_0^\infty \rho_1^3 d\rho_1$$
$$\times\exp\left(-\frac{3}{2}(\rho_1+\rho_2)\right)\int_\xi^1 \frac{ds}{s}\left(1-\frac{\hbar\omega}{2\mu c^2}s^2\right)$$
$$\times\int_\xi^1 \frac{dq}{q}\left(1-\frac{\hbar\omega}{2\mu c^2}q^2\right)$$
$$\times\int_{|\rho_1 s-\rho_2 q|}^{|\rho_1 s+q\rho_2|} d\kappa(s^2\rho_1^2+q^2\rho_2^2-\kappa^2)$$
$$\times\left\{\left[1-\frac{(q^2\rho_1^2+s^2\rho_2^2-\kappa^2)^2}{4(qs\rho_1\rho_2)^2}\right]\frac{\partial}{\partial\kappa}\frac{1}{\kappa}\frac{\partial}{\partial\kappa}\right.$$
$$\left.+\frac{q^2\rho_1^2+s^2\rho_2^2-\kappa^2}{(qs\rho_1\rho_2)^2}\frac{\partial}{\partial\kappa}\right\}\frac{\sin(\omega a\kappa/c)}{\kappa}. \qquad (35)$$

Since exact integration of the right-hand side of (35) is impossible, we give only the expression for $I(\tau)$ integrated over the variables of the atomic subsystem:

$$I(\tau)=\lim_{p\to\infty}\frac{2^8e^2c}{3^7\hbar a^2}\int_0^\infty dz\, z\exp\left(i\frac{3}{2}\frac{cz\tau}{a}\right)\left\{\frac{2z}{(z^2+1)^2}\right.$$
$$+\frac{3z}{z^2+1}+3\arctan z-\frac{2pz}{((pz)^2+1)^2}-\frac{3pz}{(pz)^2+1}$$
$$\left.-3\arctan(pz)\right\},\quad p=1/\xi. \qquad (35a)$$

In deriving Eqs. (35) we ignored $\hbar\omega s^2/2\mu_c^2$ in comparison to unity. To express the integral in Eq. (35a) in terms of analytic functions we must write $\arctan z$ in the form of a series:[14,15]

$$\arctan z=\frac{z}{1+z^2}\sum_{n=0}^\infty\frac{(2n)!!}{(2n+1)!!}\frac{z^{2n}}{(1+z^2)^n}. \qquad (36)$$

After substituting (36) into (35a) and letting the parameter p go to infinity we arrive at the following expression for $I(\tau)$:

$$I(\tau)=\left(\frac{2}{3}\right)^7\frac{e^2c^2}{p\hbar c^2}\hat{D}(u)\{i\pi\exp(-vu\tau)$$
$$-\exp(-vu\tau)\mathrm{Ei}(vu\tau)-\exp(vu\tau)\mathrm{Ei}(-vu\tau)\}, \qquad (37)$$

where

$$\hat{D}(u)=3^2-\frac{3}{2}\frac{1}{u}\frac{\partial}{\partial u}-\left(\frac{1}{u}\frac{\partial}{\partial u}\right)^2+\frac{1}{3\times4}\left(\frac{1}{u}\frac{\partial}{\partial u}\right)^3$$
$$+\sum_{n=0}^\infty\frac{(2n)!!}{(2n+1)!!}\frac{1}{n!}\left[2\times3^2+\frac{3}{n+1}\frac{1}{u}\frac{\partial}{\partial u}\right.$$
$$\left.-\frac{3}{(n+1)(n+2)}\left(\frac{1}{u}\frac{\partial}{\partial u}\right)^2\right]u^{2n}+3^2$$
$$\times\sum_{n=0}^\infty\sum_{m=n}^\infty\frac{(2n)!!(2(m-n))!!}{(2(m-n)+1)!!(2n+1)!!}$$
$$\times\frac{1}{(m+1)!}\left(\frac{1}{2u}\frac{\partial}{\partial u}\right)^{m+1}u^{2(m+1)}. \qquad (38)$$

Using the same approximations (14) in Eqs. (32), we obtain an analytical equation for the population difference of the atomic levels:

$$\frac{d\langle R_3(t)\rangle}{dt}=-(\mathscr{A}_\parallel(t)+\mathscr{B}(t))N_2(t)+\mathscr{B}(t)N_1(t), \qquad (39)$$

where $N_2(t)=\langle R_3(t)\rangle+0.5$ is the population of the upper level, $N_1(t)=0.5-\langle R_3(t)\rangle$ is the population of the ground state, and

$$\mathscr{A}_\parallel(t)=2\left(\frac{2}{3}\right)^8\frac{e^2}{\hbar a}\hat{D}(u)L(u,t),$$
$$\mathscr{B}(t)=2\left(\frac{2}{3}\right)^8\frac{e^2}{\hbar\pi a}\hat{D}(u)M(u,t), \qquad (40)$$

with $L(u,t)$ and $M(u,t)$ defined, respectively, in (16a) and (16b).

Note that although the expressions for $\mathscr{A}_\parallel(t)$ and $\mathscr{B}$ are identical in form with Eqs. (16), the fact that the operator $\hat{D}(u)$ acts on $M(u,t)$ and $L(u,t)$ drastically alters the dependence. This becomes especially evident in studying the decay of the atom in the time interval $a/c\ll t<\omega_0/c$. In this case we can use the expansions (18) and (19) in the right-hand sides of Eqs. (16a) and (16b). Here the result of the action of $\hat{D}(u)$ on the right-hand sides of $M(u,t)$ and $L(u,t)$ simplifies considerably if one recalls that

$$\left(\frac{1}{2u}\frac{\partial}{\partial u}\right)^n u^{2m}=0\quad\text{for } n>m.$$

Now one can easily show that the result of the action of $\hat{D}(u)$ on the first term in the expansion (18) is zero and that only the second term in (18) provides a finite contribution:

$$\hat{D}(u)\frac{1}{u^2}=0,\quad \hat{D}(u)\frac{1}{u^4}=2^6.$$

Note that in Sec. 2 we left only the first term in the expansion (18), since $\hat{E}(u)(1/u^2)\neq0$. Thus, allowing for the various terms proportional to $1/u^4$, we arrive at the following expressions for $\mathscr{A}_\parallel$ and $\mathscr{B}(t)$ when $t\gg a/c$:

$$\mathscr{B}(t)=\frac{1}{\pi\tau_0}\left\{\mathrm{Si}(\omega_0 t)-\frac{\pi}{2}+\frac{\cos(\omega_0 t)}{\omega_0 t}+\frac{\sin(\omega_0 t)}{(\omega_0 t)^2}\right.$$
$$\left.-\frac{2\cos(\omega_0 t)}{(\omega_0 t)^3}\right\},\quad \mathscr{A}_\parallel=\frac{1}{\tau_0}. \qquad (41)$$

Equations (41) and (23) show that $\mathscr{B}(t)$ and $B(t)$ depend differently on time. Figure 1 clearly illustrates the difference. Although these functions have different values at each moment of time t, their common feature is that they oscillate with periods of order ω_0^{-1}. Such behavior of $\mathscr{B}(t)$ and $B(t)$ is apparently due to the complex nature of the creation of a wave packet (photon), whose spatial size is several thousand times greater than the quantum size of the atom. Over time $t\gg1/\omega_0$ both $\mathscr{B}(t)$ and $B(t)$ become much smaller than τ_0^{-1}. However, the functions $B(t)$ and $\mathscr{B}(t)$ differ for such

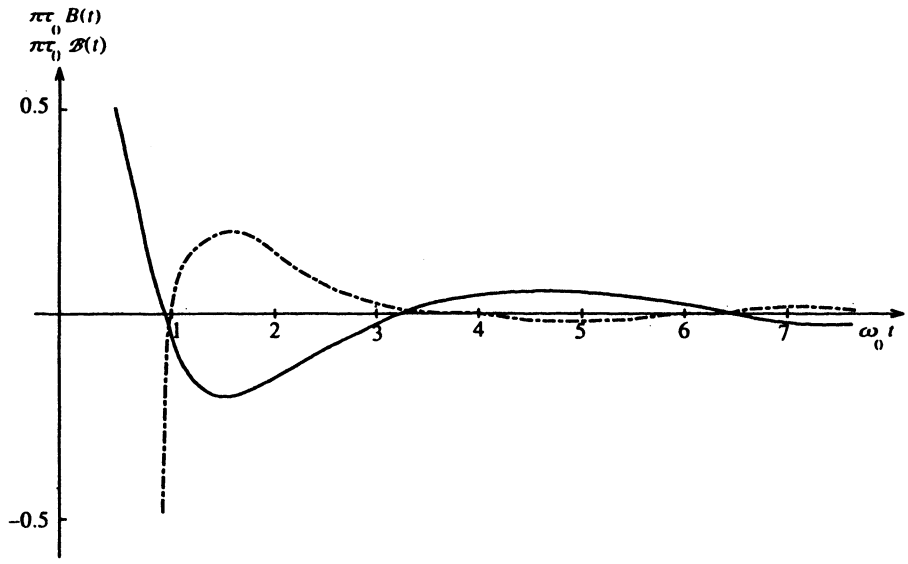

FIG. 1. The time dependence of the stimulated transition coefficients $B(t)$ (the solid curve) and $\mathscr{B}(t)$ (the dot-dash curve) at the initial stage in spontaneous decay.

times. Indeed, if we use the asymptotic expansion of the sine integral $\mathrm{Si}(\omega_0 t)$ [Eq. (22)], we arrive at the following expression for $\mathscr{B}(t)$:

$$\mathscr{B}(t) \simeq \frac{12\sin(\omega_0 t)}{\pi\tau_0(\omega_0 t)^4} \quad \text{for } \omega_0 t \gg 1. \tag{42}$$

This is the correction to the exponential decay law for an atom with $t \gg \omega_0^{-1}$ obtained via the multipole Hamiltonian (30). Clearly, the correction behaves like $1/(\omega_0 t)^4$ and, naturally, is much smaller than the correction obtained via the common interaction Hamiltonian $e(\mathbf{p}\cdot\mathbf{A}(\mathbf{r},t))/c\mu$ [Refs. 4–8; see also Eq. (23)].

If we use the approximation suggested for deriving Eq. (39), we get the same equation for $\langle R^+(t)\rangle$ as in Sec. 2 [see Eq. (15b)]. But in the given situation the expressions for $\Delta_0(t)$, $\mathscr{A}_\perp$, and $\mathscr{A}_\parallel$ differ from (16) and (17). Note that if the parameter ξ goes to zero and then $\Delta_0(t)$ is integrated over the frequencies ω, as done in (37), we get a divergent expression for the energy-level shift $\tilde{\Delta}_0$. For this reason, integrating the expression for $\tilde{\Delta}_0$ over the time lag τ and allowing for Eq. (35a), we get

$$\tilde{\Delta}_0 = \frac{2^9 e^2}{2\times 3^8 \pi\hbar a}\int_0^\infty z\,dz\left\{\frac{2}{\delta^2-z^2} - \frac{\cos[(\delta+z)\nu t]}{\delta+z} - \frac{\cos[(\delta-z)\nu t]}{\delta-z}\right\}\left\{\frac{3z}{z^2+1} + \frac{2z}{(z^2+1)^2} + 3\arctan z - \frac{3z\xi}{(\xi z)^2+1} - \frac{2z\xi}{[(\xi z)^2+1]^2} - 3\arctan(\xi z)\right\}^2. \tag{43}$$

If we let the parameter ξ go to zero and then integrate (43) with respect to z, we see that

$$2\delta\int_0^\infty \frac{z\,dz}{\delta^2-z^2}\arctan z = \infty,$$

i.e., the integral is divergent. We can make the Lamb shift converge if we first integrate (43) with respect to z and only then let the parameter ξ go to zero:

$$J = \lim_{\xi\to 0}\lim_{C\to\infty} 2\delta\int_0^C \frac{z\,dz}{\delta^2-z^2}[\arctan z - \arctan(\xi z)]^2. \tag{44}$$

As (44) implies, $\lim\limits_{\xi\to 0}\lim\limits_{C\to 0} \neq \lim\limits_{C\to 0}\lim\limits_{\xi\to 0}$. Hence introducing the variable $u = \xi z$ and integrating (44) by parts, we get

$$J = -\lim_{\xi\to 0}\delta\ln(|(\xi\delta)^2-u^2|)\left[\arctan\frac{u}{\xi} - \arctan u\right]\Bigg|_0^\infty + 2\delta\lim_{\xi\to 0}\int_0^\infty du\,\ln|u^2-(\xi\delta)^2|\left[\arctan\frac{u}{\xi} - \arctan u\right]\times\left\{\frac{\xi}{\xi^2+u^2} - \frac{1}{u^2+1}\right\}. \tag{45}$$

Clearly, the first term in (45) is equal to zero at the upper and lower limits of integration. The second term is finite for any value of the parameter ξ and is equal to

$$J = 4\delta\int_0^\infty \ln u\left[\frac{\pi}{2} - \arctan u\right]\frac{1}{u^2+1} = -\frac{7}{2}\delta\zeta(3), \tag{45a}$$

where $\zeta(3) = 1.20206$ is the Riemann zeta function.

Thus, if the expressions (35a) and (45a) are taken into account, for $\tilde{\Delta}(t)$ and $\mathscr{A}_\perp(t)$ we have

$$\tilde{\Delta}_0(t)=\frac{3^4}{2^{13}\tau_0\delta^2}\left\{\frac{3^2\times 7}{2}\zeta(3)+6\pi^2+\frac{31}{3}\right\}$$
$$+\frac{1}{2\pi\tau_0}\left\{\ln\frac{\nu}{\omega_0}-\frac{137}{120}+\mathrm{Ci}(\omega_0 t)-\frac{\sin(\omega_0 t)}{\omega_0 t}\right.$$
$$\left.+\frac{\cos(\omega_0 t)}{(\omega_0 t)^2}+\frac{2\sin(\omega_0 t)}{(\omega_0 t)^3}\right\}, \tag{46}$$

$$\mathscr{B}_\perp(t)=\frac{1}{\pi\tau_0}\left[\mathrm{Si}(\omega_0 t)+\frac{\cos(\omega_0 t)}{\omega_0 t}+\frac{\sin(\omega_0 t)}{(\omega_0 t)^2}\right.$$
$$\left.-\frac{2\cos(\omega_0 t)}{(\omega_0 t)^3}\right]. \tag{47}$$

The expression (46) for the Lamb shift derived with the Hamiltonian (29) is much larger than that obtained in Sec. 2 [see Eq. (21)]. Note that for long times the temporal corrections to $1/\tau_0$ in (47) are smaller than the respective corrections (20).

4. CONCLUSION

The above results suggest that the inverse effect of the radiation field on the spontaneous decay rate is important in small time intervals $t\sim\omega_0^{-1}$. Both approaches remove the divergences in the Lamb shift of the energy levels. From Eqs. (20) and (41) it follows that with the passage of time (i.e., $\omega_0 t\gg 1$) the corrections to the exponential spontaneous decay law tend to zero in an oscillatory manner.

Notwithstanding the general characteristics of restoration of the exponential spontaneous decay law, the expressions for the observables, which allow for the multipole interaction Hamiltonian and the Hamiltonian $e(\mathbf{A}(\mathbf{r},t)\mathbf{p})/c\mu$, still diverge. The following pattern can be observed. As we go over to the new form of the interaction Hamiltonian (29b) the increase in the Lamb shift of the energy levels [see Eq. (46)] is accompanied by a decrease in the corrections to the exponential spontaneous decay law (42) in comparison to the similar correction (23).

The study has shown that the evolution of the spontaneous decay process depends not only on the choice of the initial conditions imposed on the atom and field but also on the boundary conditions at the moment of switch-on of the atom–electromagnetic-field interaction. This last factor is known to affect not only the shape of the atom–field interaction Hamiltonian but also the phase of the system wave function. In selecting the state of the system in Sec. 3 we assumed that the electron and the nucleus were separated by a distance ξ when the interaction with the electromagnetic field was switched on. In this paper the parameter ζ was set equal to zero ($\zeta\to 0$) with allowance for the fact that in the dipole approximation the system Hamiltonian acquires the well-known form $(\mathbf{d}\cdot\mathbf{E}(\mathbf{r},t))$, where $\mathbf{d}=e\mathbf{r}$. For instance, Mandelstam[13] selected ζ equal to infinity, which corresponds to moving the electron from infinity to a distance $\mathbf{r}$ from the nucleus. This would imply that Eqs. (35a) and (43) should be integrated under the assumption that the parameter ξ tends to infinity. Clearly, both $\mathscr{B}(t)$ and $\tilde{\Delta}(t)$ acquire new values in the process. Without going into the details of such calculations, we note that apparently an optimal choice of this parameter and the initial conditions of the state of the electromagnetic field and the atom can remove the discrepancies between the above forms of the interaction Hamiltonian.

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