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Natural line shape

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The observable line shape of the spontaneous emission depends on the procedure of atom's excitation. The spectrum of radiation emitted by a two-level atom excited from the ground state by a pi pulse of the resonant pump field is calculated for the case when the Rabi frequency is much larger than the relaxation rate. It is shown that the central part of the spectral distribution has a standard Lorentzian form, whereas for detunings from the resonance that are larger than the Rabi frequency the spectral density falls off faster. The shape of the wings of the spectral line is sensitive to the form of the pi pulse. The implications for the quantum Zeno effect theory and for the estimates of the duration of quantum jumps are discussed.

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We shall treat the problem of the natural line shape – the shape of the spectral line of the spontaneous emission – the radiation emission by a secluded atom accompanying the transition from the excited state (to some lower one) that originates from the interaction of the atom with the quantized electromagnetic field. In what follows we use the model of the two-level atom with the excited state $|2\rangle$ and the ground state $|1\rangle$ with the energies E_2 and E_1 respectively, which are connected by the electrical dipole transition. The theory originally developed by Weisskopf and Wigner [1] starts with the assumption of the exponential decay of the amplitude of the initial state,

$$B\left(t\right) = \mathrm{e}^{-\gamma t}. \tag{1}$$

It yields the Lorentzian line shape,

$$S\left(\omega\right) = \frac{1}{\pi} \cdot \frac{\gamma}{\left(\omega_0 - \omega\right)^2 + \gamma^2}, \tag{2}$$

where $\omega_0 = (E_2 - E_1)/\hbar$ is the transition frequency ($\hbar$ is the Planck's constant). The natural linewidth is $\gamma = \Gamma/2$, where the transition rate Γ is given by the equality

$$\Gamma = \frac{2\pi}{\hbar}\left|V_{nk}\right|^2 \rho\left(E_k\right), \tag{3}$$

that was originally derived by Dirac [2] and now is universally known as the Fermi golden rule. In Eq. (3) n and k denote the initial and final states of the system "atom + field", V_{nk} are the matrix elements of the interaction of atom with the quantized field, and $\rho(E_k)$ is the energy density of the final states. The summation over the quantum numbers others than energy is carried out.

The expression (3) includes the values of the matrix elements and of the density of states on the energy surface $E = E_n = E_k$. Since the relative variations of V_{nk} and $\rho(E_k)$ within the main peak of the spectral line (2) in the optical range have values about $\gamma/\omega_0 \sim 10^{-8}$, the Lorentzian line shape promises to be very accurate. To my knowledge, the deviations from (2) were never established experimentally.

However, from the theoretical point of view the Lorentzian form of spectrum is irritating. On one side, this expression can not be universally valid, since the negative frequencies of photons are physically meaningless. On the other side, Eq. (2) gives for the mean frequency of the radiation $\overline{\omega}$ the expression that diverges - literally logarithmically, but even faster, if the frequency dependence of the matrix elements, $V \propto \sqrt{\omega}$ (cf. Eq. (13)), and of the energy density of states, $\rho \propto \omega^2$, is taken into account [3].

The line shape Eq. (2) can be interpreted as the form of the energy distribution $W\left(E\right) = \hbar^{-1} S\left(E/\hbar\right)$ for the quasistationary initial state of the system $|\Psi\left(0\right)\rangle$. The law of decay of the initial state is given by the Fourier transform of the energy distribution,

$$\Phi\left(t\right) = \left|\langle\Psi\left(0\right) \mid \Psi\left(t\right)\rangle\right|^2 = \\ = \left|\int W\left(E\right)\exp\left(-i\frac{Et}{\hbar}\right)dE\right|^2. \tag{4}$$

The Lorentzian form of the energy distribution leads to the exactly exponential decay law $\Phi\left(t\right) = e^{-\Gamma t}$, that makes the Weisskopf and Wigner theory self-consistent. Thence the problem of the natural line shape can be reformulated as the problem of the law of the decay of the initial state: deviations from Eq. (2) will lead to the

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nonexponentiality of the decay – and vice versa. The additional incentive to study the detailed form of the initial stage of the decay law came from the concept of the quantum Zeno effect [4, 5]. If for small times the decay law is quadratic, $\Phi(t) \approx 1 - (t/\tau_Z)^2$ (the parameter τ_Z is known as the Zeno time), then frequent measurements of the energy of the system will prevent the decay of the initial excited state. This property permitted Schulman [6] to introduce the estimate for the duration of the quantum jump between the atomic states as the crossover time from the quadratic decay to the exponential (Fermi) decay:

$$\tau_J = \Gamma \tau_Z^2. \tag{5}$$

The logic behind this definition is lucid: if the measurement can influence the kinetics of the quantum jump, then it has not been completed yet.

Let's take the Hamiltonian of the system in the form $\hat{H} = \hat{H}_a + \hat{V}_q + \hat{H}_f$, where $\hat{H}_a$ and $\hat{H}_f$ are the Hamiltonians of the free atom and of the quantized field respectively, and $\hat{V}_q$ accounts for the atom's interaction with the quantized radiation field: $\hat{V}_q = \sum_\lambda (\hat{v}_\lambda + \hat{v}_\lambda^+)$,

$$\hat{v}_\lambda = i\sqrt{\frac{2\pi\hbar\omega_\lambda}{L^3}}\,\hat{\mathbf{d}}\mathbf{e}_\lambda \hat{a}_\lambda^+ \exp(-i\mathbf{k}_\lambda \mathbf{r}), \tag{6}$$

where the index λ numerates the modes of the quantized field. Here L^3 is the quantization volume, $\hat{\mathbf{d}}$ is the operator of the dipole moment of the atomic electron, ω_λ, $\mathbf{k}_\lambda$ and $\mathbf{e}_\lambda$ are the frequency of the photon, its wave vector and the polarization vector of the mode λ respectively, and $\hat{a}_\lambda^+$ and $\hat{a}_\lambda$ are the creation and annihilation operators for this mode.

If the atom is in the initial state $|\varphi\rangle$ and the field is in the vacuum state, then the dispersion of the energy,

$$\Delta E^2 = \frac{2\hbar}{3\pi c^3}\langle\varphi|\,\hat{\mathbf{d}}^2\,|\varphi\rangle \int \omega^3 d\omega, \tag{7}$$

is infinite, since the integral diverges at the upper limit, and the Zeno time $\tau_Z = 0$. In Eq. (7) c is the speed of light.

Several authors [7, 6, 8] have found finite values of τ_Z for the spontaneous emission of radiation by using the two-level model of the atom with the initial state $|\varphi\rangle$ and the final state $|\theta\rangle$. In this case

$$\Delta E^2 = \frac{\hbar c}{4\pi^2}\sum_{\mu=1}^{2}\int k\left|\langle\varphi|\,\hat{\mathbf{d}}\mathbf{e}_\mu e^{i\mathbf{k}\mathbf{r}}\,|\theta\rangle\right|^2 d\mathbf{k}. \tag{8}$$

The account of the momentum of the emitted photon suppresses the matrix elements in the high frequency domain and effectively cuts off the Lorentzian line shape at the frequencies around $\omega_+ = \alpha^{-1}\omega_a$, where $\alpha = e^2/\hbar c$ is the fine structure constant (e is the electric charge of the electron), and the atomic frequency unit $\omega_a = me^4\hbar^{-3}$ (m is the electron's mass). However, the results of these authors contradict their assumptions: if the hydrogen atom in the initial state $2p$ can indeed emit a photon with the energy about $E_+ = \hbar\omega_+ = 3.7\,\text{keV}$, then there is no reason to limit the channels of decay by the transition only to the state $1s$.

The influence of the truncation of the Lorentzian shape (2) in the domain $\omega < 0$ on the decay law was studied in Ref. [9]. This truncation diminishes the initial decay rate by half, but doesn't lead to the quadratic decay.

The physically unsatisfactory divergences of $\overline{\omega}$ and $\Delta\omega^2$ are rooted in the unphysical initial conditions. It is a commonplace of the theory of quasistationary states that their properties depend on the procedure of their preparation [10]. Therefore this procedure must be taken into account explicitly. The importance of this approach in the problem of the natural line shape was noted long time ago [11].

Let's assume that the atom initially was in the ground state $|1\rangle$ and then was excited by a pulse of the resonant pump field (of the frequency ω_0) that has the properties of the pi pulse [12], that conveys the two-level system (in the absence of relaxation) from one state into the other. We take the Hamiltonian of the system in the form $\hat{H} = \hat{H}_a + \hat{V}_q + \hat{V}_c + \hat{H}_f$, where $\hat{H}_a$ and $\hat{H}_f$ are the Hamiltonians of the (two-level) atom and of the quantized field respectively, $\hat{V}_q$ accounts for the atom's interaction with the quantized radiation field (in the dipole approximation) and $\hat{V}_c$ describes the atom's interaction with the classical pump field:

$$\hat{V}_c = -\hat{\mathbf{d}}\mathbf{E}(t)\cos\omega_0 t, \tag{9}$$

where $\mathbf{E}(t)$ is the envelope of the electric field of the pulse. For the sake of simplicity we assume that the pump carrier frequency equals the transition frequency ω_0.

The state vector of the system can be expanded as

$$|\Psi(t)\rangle = A\,|1,\text{V}\rangle\, e^{-i\omega_1 t} + B\,|2,\text{V}\rangle\, e^{-i\omega_2 t} + \\ + \sum_\lambda C_\lambda\,|1,\lambda\rangle\, e^{-i(\omega_1+\omega_\lambda)t}, \tag{10}$$

where $|j,\text{V}\rangle$ denote states of the system with the atom in the state $|j\rangle$ and the field in the vacuum state; in the state $|1,\lambda\rangle$ the atom is in the ground state $|1\rangle$, one photon is present in the mode λ, and there are no photons in other modes; $\omega_j = E_j/\hbar$.

The evolution of the system is governed by the system of equations

$$i\frac{dA}{dt} = B\Omega(t)\cos\omega_0 t e^{-i\omega_0 t}, \qquad (11)$$

$$i\frac{dB}{dt} = A\Omega(t)\cos\omega_0 t e^{i\omega_0 t} + \sum_\lambda u_\lambda C_\lambda e^{i\Delta_\lambda t}, \qquad (12)$$

$$i\frac{dC_\lambda}{dt} = u_\lambda B e^{-i\Delta_\lambda t}. \qquad (13)$$

Here $\Omega(t) = -\mathbf{d}_{12}\mathbf{E}(t)/\hbar$, where $\mathbf{d}_{12}$ is the matrix element of the dipole moment between the states $|1\rangle$ and $|2\rangle$,

$$u_\lambda = i\sqrt{\frac{2\pi\omega_\lambda}{L^3\hbar}}\,\mathbf{d}_{12}\mathbf{e}_\lambda, \qquad (14)$$

and $\Delta_\lambda = \omega_0 - \omega_\lambda$ is the frequency detuning between the atomic transition and the emitted photon.

Let's consider the pi pulse with the rectangular envelope,

$$\Omega(t) = \Omega \quad \left(-\frac{\pi}{\Omega} < t < 0\right), \qquad (15)$$
$$\Omega(t) = 0 \qquad \text{otherwise,}$$

and assume that the Rabi frequency is much larger than the relaxation rate, $\Omega \gg \gamma$. Then throughout the duration of the pulse we can neglect the spontaneous radiation, and take into account only the pump field. Thus from Eqs. (11) and (12) with the initial conditions $A(-\pi/\Omega) = 1$, $B(-\pi/\Omega) = 0$ in the rotating wave approximation we obtain

$$B(t) = -i\cos\frac{\Omega}{2}t \qquad \left(-\frac{\pi}{\Omega} < t < 0\right). \qquad (16)$$

To describe the second stage, that of the free emission, we can use the exponential decay of Eq. (1):

$$B(t) = -ie^{-\gamma t} \qquad (t > 0). \qquad (17)$$

The Eqs. (16) and (17) have the accuracy of the order $\gamma/\Omega \ll 1$, that is sufficient for our purposes.

By substitution of Eqs. (16) and (17) in Eq. (13) and integration we obtain for the limiting values of amplitudes C_λ:

$$C_\lambda(\infty) = -u_\lambda F(\omega_\lambda), \qquad (18)$$

where the spectral amplitude is given by the expression

$$F(\omega) = \frac{2\Omega\exp\left(i\frac{\pi\Delta}{\Omega}\right) - 4i\Delta}{\Omega^2 - 4\Delta^2} + \frac{1}{i\Delta + \gamma}. \qquad (19)$$

The spectral distribution of photons is $S(\omega)=N|F(\omega)|^2$, where N is the normalization constant; in our case $N \approx \gamma/\pi$. The explicit expression for $S(\omega)$ is too cumbersome; it is more convenient to work with the formula (19).

Firstly, it must be noted that the first term in the right-hand side (RHS) of (19) is regular at $\Delta = \pm\Omega/2$, since at these points both the numerator and the denominator have simple zeroes. Secondly, for small frequency detunings $|\Delta| \lesssim \Omega$ the second term in the RHS of (19) dominates, and the line shape is given just by the standard Lorentzian form (2). Thirdly, for large $|\Delta| \gg \Omega \gg \gamma$, after expanding both terms in negative powers of Δ, we find that two terms of the order Δ^{-1} cancel each other. The dominating contribution comes from the term

$$F(\omega) \approx -\frac{\Omega}{2}\exp\left(i\frac{\pi\Delta}{\Omega}\right)\Delta^{-2}, \qquad (20)$$

that defines the asymptotics of the spectral density

$$S(\omega) \approx \frac{\gamma}{4\pi}\frac{\Omega^2}{\Delta^4}. \qquad (21)$$

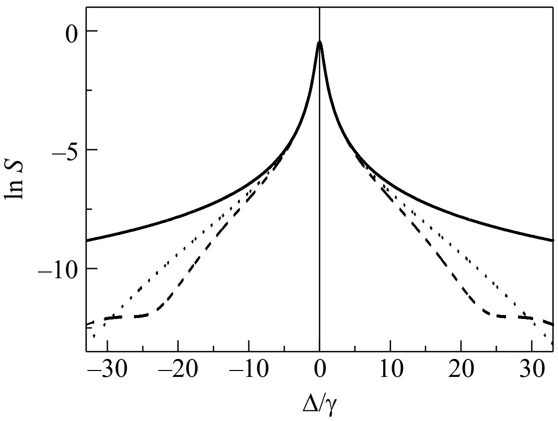
It decreases rapidly enough to provide a finite average value of the frequency of the emitted photons $\overline{\omega}$ (that in our approximation is indistinguishable from the transition frequency ω_0) and the finite value of the frequency dispersion $\Delta\omega^2 \approx 0.39\Omega\Gamma$.

It must be noted that the behavior of the wings of the spectral line depends on the form of the envelope of the pi pulse. We have calculated the spectral density $S(\omega)$ also for the pi pulse with the sine envelope:

$$\Omega(t) = -\frac{\pi}{2}\Omega\sin\Omega t\left(-\frac{\pi}{\Omega} < t < 0\right), \qquad (22)$$
$$\Omega(t) = 0 \quad \text{otherwise.}$$

The results are compared in Figure with the Lorentzian line shape and the spectral distribution for the rectangular envelope. It can be seen that for both types of pulses the crossovers between Lorentzian and asymptotic forms occur at $\Delta \sim \Omega$.

It has to be stressed that the process that we deal with should not be interpreted as a "filtering" of a pump pulse with the spectral width about Ω through the atomic transmission function of the Lorentzian shape. Firstly, in Figure one can see that the spectral density of the emitted radiation does not vanish for the frequency detuning $\Delta = 2\Omega = 20\gamma$, whereas there are no photons of this frequency in the spectrum of the incident pi pulse with the rectangular envelope, Eq. (15). Secondly, the pulse of the doubled duration, that of the form $\Omega(t) = \Omega$ for $-2\pi\Omega^{-1} < t < 0$ and $\Omega(t) = 0$ otherwise, will have



The dependence of the logarithm of the spectral density $\ln S$ on a scaled frequency detuning Δ/γ for the Rabi frequency $\Omega = 10\gamma$. Solid line – the Lorentzian form (2), dashed line corresponds to the rectangular envelope of the pi pulse, Eq. (15), dotted line – the same for the sine pulse envelope, Eq. (22)

the power spectrum that is twice as narrow and has the peak intensity four times as large as the power spectrum of the pulse Eq. (15). From the "filtering" reasoning one should expect similar changes in the spectrum of emitted radiation. However, in this case the radiation will be smaller than that from the pi pulse Eq. (15) by the factor about γ/Ω. Doubling the duration creates the 2 pi pulse that rapidly returns the system to its initial non-radiating state [12]: the atom can emit photons only throughout the short duration of the excitation pulse.

The process of radiation due to the excitation of an atom by a pi pulse is an essentially nonlinear process. It can be seen from the zeroth order expression for the probability amplitude

$$B(t) = -i \sin\left(\frac{1}{2}\int\limits_{t_0}^{t} \Omega\left(t'\right) dt'\right). \qquad (23)$$

Attempts to interpret the properties of this process by exploiting analogies with the one-photon scattering are doomed.

The asymptotic behavior of the radiation spectrum is determined by the degree of smoothness of the amplitude $B(t)$: the number of its first discontinuous derivative and the magnitude of the jump. It is possible to construct the "pi pulse" that will lead to the spectral wings of radiation that decay faster than Eq. (21), e.g. as Δ^{-6}.

The finite dispersion of the frequency defines the Zeno time of the system [6]; thus $\tau_Z = \left(\Delta\omega^2\right)^{-1/2} \sim \sim (\Omega\Gamma)^{-1/2}$. Then from Eq. (5) we obtain a somewhat trivial estimate for the duration of the quantum jump in our case, $\tau_J \sim \Omega^{-1}$. It must be noted that indefinite increase of the amplitude of the pi pulse will eventually violate the applicability of the two-level model. The transitions to other states of the system are necessarily important if the Rabi frequency is comparable to the transition frequency, $\Omega \sim \omega_0$. Thus our model assures that the duration of the quantum jump accompanying the spontaneous emission from a given transition will always be larger than the field period, $\tau_J \gtrsim \omega_0^{-1}$. This inequality is almost universally accepted by the community of physicists on the grounds of common sense and is reflected in the literature [13].

Our analysis is limited by the domain $\Omega \gg \gamma$. It is interesting to compare it with the opposite limiting case. For $\Omega \ll \gamma$ the rectangular "pi pulse" defined by Eq. (15) lasts much longer than the relaxation time γ^{-1}. Therefore during the most part of the pulse the initial state of the system is already ignorable, and the difference between the sequence of long "pi pulses" and the continuous pump field is unimportant. The spectrum of radiation of the two-level system under the influence of the continuous monochromatic field has been calculated by Mollow [14]. For $\Omega \ll \gamma$ and the pump frequency that equals that of the transition, ω_0, the power spectrum in our notation has the form

$$P\left(\omega\right) = \frac{\Omega^2}{2\gamma^2}\left[2\pi\delta\left(\omega-\omega_0\right) + \frac{2\Omega^2\gamma}{\left(\Delta^2+\gamma^2\right)^2}\right], \qquad (24)$$

where $\delta(z)$ is the Dirac's delta-function. The first term in the brackets describes the scattering of the pump radiation with unchanged frequency. Only the second, incoherent term can be interpreted as a spectrum of the "spontaneous" emission by the atom that is excited by a weak resonant field. The term "spontaneous" in this case may be too stretched, since the atom in the continuous monochromatic field can hardly be considered a secluded one. However, if one accepts this interpretation of the incoherent term, then it may be noted that for large $|\Delta|$ it follows the inverse quartic law, similar to our Eq. (21). Pushing the interpretation further, we may say that for the spectral distribution given by the incoherent term the Zeno time and the quantum jump time happen to be equal: $\tau_Z = \tau_J = \Gamma^{-1}$.

In conclusion, we have demonstrated that the line shape of the radiation, spontaneously emitted by an atom excited by a strong pi pulse is Lorentzian only in the domain of frequency detunings that do not exceed the Rabi frequency of the pulse, $|\Delta| < \Omega$. For larger values of $|\Delta|$ it falls off much faster. For the transition with the frequency $\omega_0 = 3.5 \cdot 10^{15}\,\mathrm{s}^{-1}$ and the matrix element of the dipole moment $d_{12} = 2.5 \cdot 10^{-18}$ CGS the spontaneous emission rate is $\Gamma = 1.3 \cdot 10^{7}\,\mathrm{s}^{-1}$. The condition $\Omega = 10\gamma$ corresponds to the intensity of the rectangular pi pulse $I = 92$ mW cm^{-2} and to its duration $\theta = \pi/\Omega = 47$ ns. In these conditions for the Lorentzian line shape approximately 6% of the emitted photons must have frequency detunings $|\Delta| > \Omega$. The observation of shortage of these photons seems to be accessible to the modern spectroscopic experiment.

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