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Spontaneous emission in a cavity

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Contents

Abstract. A brief review is given of theoretical and experimental investigations of the spontaneous emission from an atom in a resonator cavity. A theory of spontaneous emission from a two-level atom in free space and in a cavity is presented in a methodical unified manner. In a single-mode resonator the structure of the spectrum of the emitted photons is a singlet or a doublet, depending on the ratio of the cavity damping to the constant of the interaction of an atom with a field. The emission spectrum of an atom in a cavity with two modes with similar frequencies may have a triplet structure.

1. Brief historical review

The problem of spontaneous emission is one of the classical topics in quantum electrodynamics. Much work has been done on this problem, beginning with the well-known paper of Weisskopf and Wigner [1]. The theory of spontaneous emission has been dealt with in many books and review papers. Only some of them are mentioned below [2 – 13], but the list is not exhaustive.

In the course of the last few years the problem of spontaneous emission has been attracting attention in connection with the study of the process of spontaneous emission from an atom or a molecule inside a resonator cavity. Although this problem postdates the spontaneous emission in free space, it also now has a long history. The interest in it has recently increased because opportunities have become available for an experimental verification of the theoretical predictions.

The probability of spontaneous emission in a cavity was first pointed out by Purcell [14]. He stated that the probability w_s^c of spontaneous emission in a single-mode cavity can be deduced from the probability w_s of spontaneous emission of an atom in free space if the latter is divided by the density of field oscillators in free space $\rho(\omega) = \omega^2/3\pi^2c^3$ and multiplied by a quantity $Q/\pi\omega_c V$ which represents nominally the density of field oscillators in a cavity mode (Q is the Q factor of a cavity, V is its volume, $\lambda = 2\pi c/\omega$, and c is the velocity of light):

$$w_s^c = w_s \frac{3\lambda^3 Q}{8\pi^2 V}. \tag{1}$$

Eqn (1) predicts a considerable increase in the probability of spontaneous decay if free space is replaced by a high-Q cavity in the microwave range of wavelengths when the wavelength is comparable with the linear dimensions of the cavity. For example, the probability is $w_s^c \approx 4 \times 10^2 w_s$ for $\lambda^3 \propto V$ and $Q \approx 10^4$.

A more consistent theory shows that Purcell and several other authors [15, 16] are absolutely right, but only when the cavity frequency w_c is tuned exactly to the atomic frequency transition w_a. The expression (1) for the probability of spontaneous emission in a cavity does not include in any way the detuning (offset) between the eigenfrequency (natural frequency) of the cavity and the resonance frequency of the atomic transition. A later calculation of Bunkin and Oraevskii [17] has shown that

$$w_s^c = \frac{4\pi|\boldsymbol{\mu}|^2}{\hbar V} \frac{\omega_c^2/Q}{(\omega_c - \omega_a)^2 + (\omega_c^2/Q^2)}, \tag{2}$$

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where $\boldsymbol{\mu}$ is a matrix element of the dipole moment corresponding to a light-emitting transition. If $\omega_a = \omega_c$ is substituted in Eqn (2), the Purcell expression follows directly. However, it also follows from Eqn (2) that when the difference between ω_a and ω_c is sufficiently large, the process of spontaneous emission in a cavity is strongly inhibited. For example, if $\omega_a - \omega_c$ lies between the fundamental and first mode of a cavity, so that $\omega_a - \omega_c \sim \omega_c$, the probability decreases by a factor Q^2 compared with the resonance case. This result naturally suggests that the placing of an excited atom in a waveguide with a critical frequency higher than the resonance frequency of the light-emitting atom will inhibit spontaneous emission [18, 19].

In the late fifties and early sixties a series of experiments has been carried out on a maser with a beam of ammonia molecules in which two (or even three) consecutive cavities have been used [3, 20–23]. The usual generation of coherent microwave radiation ($\lambda = 1.25$ cm) takes place in the first cavity. The second cavity is used to observe collective (coherent) spontaneous emission. Although in these experiments the authors' aim has not been to demonstrate specifically an increase in the probability of a spontaneous transition of a molecule in a cavity, these experiments have provided a clear evidence of this increase. In the second cavity, whose Q factor is several thousand, a strong collective spontaneous emission has been observed so that the molecules emerging from the second cavity have been found to be de-excited. On the other hand, the passage of a beam of molecules in free space between the cavities has not produced any significant de-excitation of molecules.

The first direct observation of an increase in the probability of spontaneous emission from an atom inside a cavity was reported in 1983 [24]. The Rydberg state of sodium atoms, corresponding to the principal quantum number of 23, was used in this experiment. The atoms were placed in a niobium superconducting cavity with its eigenfrequency close to 340 GHz. Tuning of this frequency to a resonance with an atomic transition shortened the lifetime of the excited atoms.

In the optical range a similar experiment was carried out by Heinzen and Feld [25]. They were able to detect an increase in the probability of spontaneous emission in a multimode optical cavity resonator.

The inhibition of spontaneous emission has also been studied experimentally. The first experiment known to the present author was reported by Drexhage in 1974 [26]. An investigation was made of the fluorescence emitted by a thin dye film placed near a mirror at a node of an electromagnetic field formed as a result of interference between the waves incident on the mirror and reflected from it. Drexhage reported a 25% reduction in the probability of spontaneous decay. Similar experiments were also described by DeMartini et al. [27]. Gabrielse and Dehmelt carried out experiments involving the inhibition of spontaneous transitions in a cavity [28]. They observed a tenfold increase in the lifetime of a single electron on an excited cyclotron orbit in a Penning trap. The electrodes of this trap served as the cavity.

An increase in the lifetime of the Rydberg states of atoms inside the cavity have also been reported by others [29, 30].

The idea of inhibition of spontaneous radiative transitions has recently been developed on the basis of periodic optical structures. When electromagnetic waves propagate inside such structures, there are forbidden ranges of frequencies (and wave vectors) of these waves. If the reson-ance frequency of an atom falls inside such a forbidden range, the spontaneous decay of an atom is strongly inhibited [9, 31–33]

Eqn (2) is derived on the assumption of a relatively strong cavity damping when the inequality $\omega/Q \gg (|\boldsymbol{\mu}|^2\langle E^2\rangle)^{1/2}/\hbar$ is satisfied; here, $\langle E^2\rangle$ is the average value of vacuum fluctua-tions of the electric field intensity. In the microwave range a dipole transition with the usual value $|\boldsymbol{\mu}| \approx 10^{-18}$ cgs units and a cavity with $Q = 10^{13}-10^{4}$ such an inequality is readily satisfied. The frequency distribution of the spontaneously emitted photons is then of monoresonance nature. However, in the case of transitions between high Rydberg atomic states the dipole moment is $|\boldsymbol{\mu}| \approx 10^{-16}$ cgs units and the Q factor of a superconducting cavity can reach 10^9 [34]. In this case the inequality $\omega/Q \ll (|\boldsymbol{\mu}|^2\langle E^2\rangle)^{1/2}/\hbar$ is obeyed and the spectral distribution of the photons emitted in the cavity is a doublet [35–37].

It is well known that the cavity modes can be degenerate: one and the same eigenfrequency can correspond to several different field configurations. If the modes are assumed to be rigorously orthogonal, the transition probability should in this case be multiplied by the number of degenerate modes. However, such modes are frequently not exactly orthogonal. It is more consistent to regard them as mutually coupled oscillators. The problem of spontaneous emission from a particle in a cavity with two coupled modes has been investigated [38]. The spontaneous emission into coupled degenerate (or close on the frequency scale) modes has been found to occur at a lower rate than the emission of independent (orthogonal) modes.

Very recently many papers have been published on the problem of spontaneous emission in a cavity with specific applications to various tasks in quantum electronics [39–47]. In particular, consideration has been given to special features of open plane–plane cavity resonators [39, 40] and cavities with a different geometry [40], the noise in optical amplifiers [39], and the spontaneous emission in semiconductor lasers [41]. The reported investigations are interesting also from the methodological point of view.

A brief historical review will be followed by an ordered presentation of the theory of spontaneous emission and this theory will be used to prove all the results mentioned above.

2. Spontaneous emission in free space

Although the main purpose of this review is to provide a systematic account of the problem of spontaneous emission in a single-mode cavity, it is logical to begin with the theory of spontaneous emission in free space.

Let us begin with the classical model of Weisskopf and Wigner: a two-level atom interacts with a continuum of oscillators of a free-space field. The Hamiltonian of the atom–field system is

$$\hat{H} = \hat{H}_a + \hat{H}_f + \hat{V}, \tag{3}$$

where $\hat{H}_a$ is the operator of the energy of the atom with two eigenvalues: $W_1 = 0$ and $W_2 = \hbar\omega_a$; $\hat{H}_f$ is the operator of the energy of the electromagnetic field and its eigenvalues $n_{k,e}\hbar\omega_k$ describe the energy of a field

oscillator corre-sponding to a plane wave with the wave vector $\boldsymbol{k}$ and a polarisation $\boldsymbol{e}$; $\hat{V}$ is the operator of the interaction of the atom with the electromagnetic field.

The combined atom–field quantum states will be described by two indices $|a, \alpha\rangle$. The first index describes the state of the atom and the second is the combined index of the field oscillator $\alpha \equiv (\boldsymbol{k}, \boldsymbol{e})$.

When the two-level atom interacts with the field, the system may be in one of the following states:

$|1, 0\rangle$ when the atom is excited and all the field oscillators are in the ground state;

$|0, 1_\alpha\rangle$ when the atom is de-excited and it has emitted one photon with the combined index α.

The state $|0, 1_\alpha\rangle$ is in fact a continuous set of states because there is a definite probability of the excitation of any one of the field oscillators.

The operators $\hat{H}_\mathrm{a}$ and $\hat{H}_\mathrm{f}$ are diagonal in the representation of the states $|1, 0\rangle$ and $|0, 1_\alpha\rangle$:

$$\hat{H}_\mathrm{a}|1, 0\rangle = W_2|1, 0\rangle, \quad \hat{H}_\mathrm{a}|0, 1_\alpha\rangle = W_1|0, 1_\alpha\rangle, \tag{4}$$

$$\hat{H}_\mathrm{f}|1, 0\rangle = 0, \quad \hat{H}_\mathrm{f}|0, 1_\alpha\rangle = \hbar\omega_\alpha|0, 1_\alpha\rangle. \tag{5}$$

We shall assume that the operator $\hat{V}$ is off-diagonal in the selected representation:

$$\hat{V}|1, 0\rangle = V_\alpha|0, 1_\alpha\rangle, \quad \hat{V}|0, 1_\alpha\rangle = V_\alpha^*|1, 0\rangle. \tag{6}$$

We shall seek the wave function of the system in the form of a superposition of these states:

$$\Psi(t) = A(t)\exp(-\mathrm{i}\omega_\mathrm{a}t)|1, 0\rangle + \sum_\alpha B_\alpha(t)\exp(-\mathrm{i}\omega_\alpha t)|0, 1_\alpha\rangle. \tag{7}$$

The Schrödinger equation together with Eqns (4), (5), and (6) yields the following equations for the coefficients $A(t)$ and $B(t)$ [2]:

$$\mathrm{i}\hbar\frac{\mathrm{d}A}{\mathrm{d}t} = \sum_\alpha V_\alpha \exp[\mathrm{i}(\omega_\mathrm{a} - \omega_\alpha)t]B_\alpha + \mathrm{i}\hbar\delta(t), \tag{8a}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}B}{\mathrm{d}t} = V_\alpha^* \exp[-\mathrm{i}(\omega_\mathrm{a} - \omega_\alpha)t]A. \tag{8b}$$

In the above equation the δ function describes the initial state: it represents the absence of the atom up to the moment $t = 0$ and its appearance at the moment $t = 0$ in an excited state.

We shall seek the solution of Eqns (8a) and (8b) by the Laplace transform method. We shall introduce the Laplace transforms

$$A_p \leftrightarrow A(t), \quad B_{\alpha p} \leftrightarrow B_\alpha(t)\exp[-\mathrm{i}(\omega_\alpha - \omega_\mathrm{a})t], \tag{9}$$

which, in accordance with Eqns (8a) and (8b), obey the following equations:

$$A_p = \sum_\alpha \frac{V_\alpha}{\mathrm{i}\hbar}B_{\alpha p} + 1, \tag{10a}$$

$$[p - \mathrm{i}(\omega_\mathrm{a} - \omega_\alpha)]B_{\alpha p} = \frac{V_\alpha^*}{\mathrm{i}\hbar}A_p. \tag{10b}$$

If $B_{\alpha p}$ is expressed in terms of Eqn (10b) and is substituted in Eqn (10a), the result is

$$(p + \Gamma_p)A_p = 1, \tag{11}$$

where

$$\Gamma_p = \sum_\alpha \left(|V_\alpha|\hbar^{-1}\right)^2 \left[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})\right]^{-1}. \tag{12}$$

Eqn (11) has a trivial solution if the sum (12) can be calculated. This can be done by summation over two polarisations for each value of the vector $\boldsymbol{k}$ and integration with respect to the directions and magnitudes of this vector, because $\boldsymbol{k}$ is a continuously variable parameter. The replacement of the summation with the integral sign requires introduction of a density of states $\rho(\omega_\alpha)$ which depends only on the frequency (modulus) of the wave vector because space is regarded as isotropic. Consequently Eqn (12) becomes

$$\Gamma_p = \sum_\nu \int \left(|V_\nu(\omega_\alpha)|\hbar^{-1}\right)^2 \times \rho(\omega_\alpha)\left[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})\right]^{-1} \mathrm{d}\Omega\,\mathrm{d}\omega_\alpha, \tag{13}$$

where the index ν denotes the direction of the polarisation and $\mathrm{d}\Omega$ is the differential of the solid angle. Summation over the polarisations and integration with respect to the angular variables (directions of $\boldsymbol{k}$) is carried out in the Appendix 1 on the assumption that the interaction of the atom with the field is of the dipole nature:

$$\hat{V} = -\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{E}}$$

The result is

$$\Gamma_p = 2(3\pi\hbar c^3)^{-1}\int_0^\infty |\boldsymbol{\mu}|^2\omega^3\left[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})\right]^{-1}\mathrm{d}\omega_\alpha. \tag{14}$$

It now remains to integrate with respect to the frequency. It is usual to proceed as follows (see, for example, Ref. [2]).

The value of the required function $A(t)$ is determined by the poles of its Laplace transform A_p. Later calculations will show that the value of p at a pole is much less than ω_α or ω_a. Therefore, $|\boldsymbol{\mu}|^2\omega_\alpha^3$ is a slowly varying function compared with $[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})]^{-1}$. In the limit of very small values of p, compared with ω_α and ω_a, the results are

$$[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})]^{-1} = \pi\delta(\xi) - \mathrm{i}P\xi^{-1}, \quad \xi = \omega_\alpha - \omega_\mathrm{a}, \tag{15}$$

$$\Gamma_p = 2(3\pi\hbar c^3)^{-1}\int_{-\omega_\mathrm{a}}^\infty |\boldsymbol{\mu}|(\omega_\mathrm{a} + \xi)|^2(\omega_\mathrm{a} + \xi)^3 \times [\pi\delta(\xi) - \mathrm{i}P\xi^{-1}]\,\mathrm{d}\xi. \tag{16}$$

Since $\omega_\mathrm{a} \gg p$, the lower limit of integration in Eqn (16) can be assumed to be $-\infty$. The imaginary part of Eqn (16) is the principal value of the integral. It determines the radiative correction to the atomic transition frequency. The problem of radiative corrections to the energy levels is of interest for its own sake and it represents an important chapter of quantum electrodynamics. However, the radiative correction to the frequency does not play a significant role in the theory of decay of atomic levels and of the natural width of spectral lines. Therefore, in the

majority of investigations of spontaneous emission, the discussion of the radiative correction usually ends at this stage. We shall do the same here.

A decisive role in the theory of the natural width of a line is played by $\mathrm{Re}\,\Gamma_p$. It follows from Eqn (16) that

$$\mathrm{Re}\Gamma_p = \gamma = 2|\boldsymbol{\mu}|^2\omega_\mathrm{a}^3(3\hbar c^3)^{-1}. \tag{17}$$

This quantity is independent of p because A_p has one pole $p = -\gamma$. Therefore,

$$A(t) = \exp(-\gamma t), \tag{18a}$$

$$B_\alpha(t) = V_\alpha^*(\mathrm{i}\hbar)^{-1}\,\frac{1-\exp\{-[\gamma+\mathrm{i}(\omega_\mathrm{a}-\omega_\alpha)]t\}}{\gamma+\mathrm{i}(\omega_\mathrm{a}-\omega_\alpha)} \tag{18b}$$

The probability w_s of spontaneous transitions per unit time is given by

$$w_\mathrm{s} = -\frac{1}{|A|^2}\,\frac{\mathrm{d}|A|^2}{\mathrm{d}t} = 2\gamma = 4|\boldsymbol{\mu}|^2\omega_\mathrm{a}^3(3\hbar c^3)^{-1}. \tag{19}$$

The frequency distribution of the emitted photons is

$$\int |B_\alpha(\infty)|^2\rho(\omega_\alpha)\,\mathrm{d}\Omega = \gamma\pi^{-1}[\gamma^2+(\omega_\mathrm{a}-\omega_\alpha)^2]^{-1}. \tag{20}$$

In writing down Eqn (20) it is assumed that $\rho(\omega_\alpha) = \rho(\omega_\mathrm{c})$, which is permissible since $\rho(\omega_\alpha)$ is a slowly varying function compared with the Lorentzian $[\gamma^2+(\omega_\mathrm{a}-\omega_\alpha)^2]^{-1}$.

All seems to be well: the integral (14) is calculated subject to quite obvious assumptions and the value of the integral is of general nonmodel nature. Unfortunately, in the majority of the well-known books the discussion of the calculation of this integral ends here. However, the situation is more dramatic than it might seem at first sight. The point is this: the integral

$$\mathrm{Re}\,\Gamma_p = 2(3\pi\hbar c^3)^{-1}\int_0^\infty |\boldsymbol{\mu}|^2\omega_\alpha^3 p\left[p^2+(\omega_\alpha-\omega_\mathrm{a})^2\right]^{-1}\mathrm{d}\omega_\alpha \tag{21}$$

converges only subject to more stringent assumptions about the function $|\boldsymbol{\mu}|^2\omega_\alpha^3$ than simply its slow variation compared with the function $[p^2+(\omega_\alpha-\omega_\mathrm{a})^2]^{-1}$. If, for example, it is assumed that $|\boldsymbol{\mu}|^2$ decreases at higher values of ω_α in accord-ance with the power law $|\boldsymbol{\mu}|^2 \propto \Omega^n/(\Omega^n+\omega_\alpha^n)$, then the integral (21) converges if $n > 2$.

The need to assume a reduction in the dipole moment on increase in the frequency ω_α is not a serious problem. At high frequencies, when the wavelength becomes less than the dimensions of an atom, the field changes its phase many times within these dimensions. In this case the dipole approximation is, strictly speaking, invalid and one should speak of a matrix element of the interaction operator $\hat{V}$. Since [2]

$$V_\alpha \approx \iiint \exp(i\boldsymbol{k}\cdot\boldsymbol{r})\Psi_2^*(\boldsymbol{r})\boldsymbol{p}\Psi_1(\boldsymbol{r})\,\mathrm{d}^3r, \tag{22}$$

where $\boldsymbol{p}$ is the momentum of an electron in an atom and $\Psi_j(\boldsymbol{r})$ are the wave functions of the atomic states between which a transition takes place, it follows that for $|\boldsymbol{k}|a \gg 1$ (a is the effective size of an atom) the value of $|V_\alpha|^2$ decreases with increase in the frequency faster than ω_α^{-2}. This can be readily demonstrated by assuming that, for example, $\Psi_j(\boldsymbol{r}) \propto \mathrm{e}^{-r/a}$.

However, strictly speaking, the two-level approximation for the description of an atom is invalid at high frequencies because the levels with higher energies can then be excited right up to the state of ionisation of an atom. If $\hbar\omega_\alpha > mc^2$, pair creation begins, and so on. Therefore, we can see that at high frequencies our calculation model becomes meaningless.

How can this problem be solved? The solution is given by the following procedure. The δ-function approximation used in the calculation of the integral (14) can be regarded as the first approximation in the expansion of the integral as a series in p. We have seen that this approximation is independent of p and it is not based on any specific model of the process at high values of ω_α. Calculation of the results in higher approximations requires however either modelling of the process at high frequencies or allowance for the whole complex range of phenomena (ionisation, pair creation, etc.) which are involved in the process in the limit $\omega_\alpha \to \infty$, which is hardly practical. However, if the terms of higher orders in p are small compared with the first approximation, they can be simply ignored.

The question is: are these terms small? We shall answer this by specific calculations of the integral (14) and by modelling the frequency dependence of the dipole moment. For example, let us assume that $|\boldsymbol{\mu}|^2 = |\boldsymbol{\mu}_0|^2\,\Omega^4/(\Omega^2+\omega_\alpha^2)^2$. Then the ratio of the terms which are ignored to the main term is of the order of $p\Omega^2/\omega_\mathrm{a}^3 = \gamma\Omega^2/\omega_\mathrm{a}^3$. If we assume that Ω is governed by the atomic dimensions ($\Omega \approx c/a$) and that $\boldsymbol{\mu}_0 \approx 10^{-17}-10^{-18}$ cgs units, it follows that $\gamma\Omega^2/\omega_\mathrm{a}^3 \ll 1$ (see also the Appendix 7.2).

It therefore follows that at sufficiently low values of the probability of a spontaneous transition, compared with its frequency, the use of the δ function in the calculation of (14) is justified. The result obtained in this way should be regarded as the first term of the expansion of the probability as a series in terms of the small parameter γ/ω_a. Calculation of higher-order corrections is strictly speaking impossible within the framework of the two-level model and the dipole approximation. We can only say at sufficiently low values of γ/ω_a the first-order term predominates, so that the higher-order terms of the expansion can be ignored.

We shall now give numerical estimates of the probability of spontaneous transitions. We shall assume that the wavelength of the spontaneously emitted radiation lies in the visible part of the spectrum and, therefore, we shall postulate that $\lambda = 0.5$ μm. Let the dipole moment be $|\boldsymbol{\mu}| = 5 \times 10^{-18}$ cgs units; this value is typical of atomic transitions in the visible part of the spectrum. It then follows from Eqn (19) that $w_\mathrm{s} = 6 \times 10^6\ \mathrm{s}^{-1}$. This value agrees well with the experimentally determined lifetimes ($\sim 10^{-8}$ s) of excited atomic states. In the microwave range when $\lambda = 1$ cm the transition probability is $(2 \times 10^4)^3$ times smaller: $w_\mathrm{s} \approx 10^{-5}\ \mathrm{s}^{-1}$. This spontaneous emission probability corresponds to a lifetime of the order of days.

3. Spontaneous emission in a cavity

3.1. Formulation of the problem and principal equations

We shall begin with the model proposed earlier [17]: in this model a two-level atom is assumed to interact with an oscillator which in turn interacts with the system characterised by a continuous absorption spectrum. The oscillator describes a cavity mode closest in its frequency to

the two-level atom. The interaction of the oscillator with the system that has a continuous spectrum (walls of the resonator cavity) allows for the cavity damping.

The Hamiltonian of such a complex system is

$$\hat{H} = \hat{H}_{\mathrm{a}} + \hat{H}_{\mathrm{c}} + \hat{H}_{\mathrm{w}} + \hat{U} + \hat{V}, \tag{23}$$

where $\hat{H}_{\mathrm{a}}$ is the eigenenergy of the atom, $\hat{H}_{\mathrm{c}}$ is the energy of the oscillator, $\hat{H}_{\mathrm{w}}$ is the energy of the system with a continuous spectrum (cavity walls), $\hat{U}$ is the interaction of the atom with the oscillator, and $\hat{V}$ is the interaction of the oscillator with the walls.

The states of the combined atom–oscillator–walls system will be described by three indices $|\mathrm{a}, \mathrm{c}, \mathrm{w}\rangle$. The first index indicates the state of the atom, the second that of the oscillator, and the third represents the walls.

When the atom interacts with the cavity, the system may be in the following states;

$|1,0,0\rangle$ when the atom is excited, the oscillator is in the ground state, and the walls are not excited; the energy of the state W_{a} is equal to the energy of the excited atomic level (the lower state of the atom is assumed to be the ground) and we then find that $W_{\mathrm{a}} = \hbar\omega_{\mathrm{a}}$, where ω_{a} is the Bohr frequency of the atomic transition;

$|0,1,0\rangle$ when the atom is in the lower state, the oscillator is in the first excited state, and the walls are not excited; the energy of the state is $W_{\mathrm{c}} = \hbar\omega_{\mathrm{c}}$, where ω_{c} is the eigenfrequency of the cavity;

$|0,0,1_{\omega}\rangle$ when the atom is in the lower state, the oscillator is in the ground state, and the walls have absorbed a photon of frequency ω; the energy of the state is $W_{\omega} = \hbar\omega$.

Since the walls with a continuous absorption spectrum can absorb a photon of any frequency, the third of the above states represents in fact a set of states which can be described by a continuous parameter ω.

We shall seek the wave function $\Psi(t)$ of the system in the form of a superposition of the above states, so that

$$\begin{aligned}\Psi(t) &= A(t)\exp(-\mathrm{i}\omega_{\mathrm{a}}t)|1,0,0\rangle \\ &\quad + B(t)\exp(-\mathrm{i}\omega_{\mathrm{c}}t)|0,1,0\rangle \\ &\quad + \sum_{\omega} C\omega(t)\exp(-\mathrm{i}\omega t)|0,0,1_{\omega}\rangle.\end{aligned} \tag{24}$$

It follows from the Schrödinger equation that

$$\mathrm{i}\hbar\frac{\mathrm{d}A}{\mathrm{d}t} = U\exp\Big[\mathrm{i}(\omega_{\mathrm{a}} - \omega_{\mathrm{c}})t\Big]B + \mathrm{i}\hbar\delta(t), \tag{25a}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}B}{\mathrm{d}t} = U^{*}\exp\Big[-\mathrm{i}(\omega_{\mathrm{a}} - \omega_{\mathrm{c}})t\Big]A + \textstyle\sum_{\omega} V_{\omega}\exp\Big[\mathrm{i}(\omega_{\mathrm{c}} - \omega)t\Big]C_{\omega}, \tag{25b}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}C_{\omega}}{\mathrm{d}t} = V_{\omega}^{*}\exp\Big[-\mathrm{i}(\omega_{\mathrm{c}} - \omega)t\Big]B. \tag{25c}$$

In the above equations the quantities U and V_{ω} are the matrix elements of the following interaction operators:

$$U = \langle 0,1,0|\hat{U}|1,0,0\rangle, \quad V_{\omega} = \langle 0,0,1_{\omega}|\hat{V}|0,1,0\rangle; \tag{26}$$

$\delta(t)$ is the function on the right-hand side of Eqn (25a), which allows for the initial state of the system when the atom in an excited state appears at the moment $t = 0$.

As in the preceding section, we shall solve the system of equations (25) by the Laplace transformation. We shall introduce the Laplace transforms of the required quantities $A(t)$, $B(t)$, and $C_{\omega}(t)$:

$$\begin{aligned}A_p &= A(t)\exp\Big[-\mathrm{i}(\omega_{\mathrm{a}} - \omega_{\mathrm{c}})t\Big], \\ B_p &= B(t), \\ C_{\omega p} &= C_{\omega}(t)\exp\Big[\mathrm{i}(\omega_{\mathrm{c}} - \omega)t\Big].\end{aligned} \tag{27}$$

It then follows from Eqns (25a)–(25c) that

$$\Big[p + \mathrm{i}(\omega_{\mathrm{a}} - \omega_{\mathrm{c}})\Big]A_p = \frac{U}{\mathrm{i}\hbar}B_p + 1\ , \tag{28a}$$

$$pB_p = \frac{U}{\mathrm{i}\hbar}A_p + \sum_{\omega}\frac{V_{\omega}}{\mathrm{i}\hbar}C_{\omega p}\ , \tag{28b}$$

$$\Big[p + \mathrm{i}(\omega - \omega_{\mathrm{c}})\Big]C_{\omega p} = \frac{V_{\omega}^{*}}{\mathrm{i}\hbar}B_p\ . \tag{28c}$$

3.2. Cavity damping

Before we analyse the system of equations (28), let us consider the intermediate problem of the energy decay in a cavity in the absence of an atom. We shall assume that initially the cavity is in the first excited state and the 'walls' are in the ground energy state. We can easily see that in this case the wave function is described by Eqn (24) with $A(t) = 0$, where $B(t)$ and $C_{\omega}(t)$ obey

$$pB_p = \sum_{\omega}\frac{V_{\omega}}{\mathrm{i}\hbar}C_{\omega p} + 1\ , \tag{29a}$$

$$\Big[p + \mathrm{i}(\omega - \omega_{\mathrm{c}})\Big]C_{\omega p} = \frac{V_{\omega}^{*}}{\mathrm{i}\hbar}B_p\ . \tag{29b}$$

The structure of these equations is fully analogous to the system (10) which describes the process of spontaneous emission in free space. Elimination of $C_{\omega p}$ from the above system gives

$$(p + \Gamma_{\mathrm{c}p})B_p = 1\ , \tag{30}$$

where

$$\Gamma_{\mathrm{c}p} = \sum_{\omega}\frac{|V_{\omega}|^2}{\hbar^2}\frac{1}{p + \mathrm{i}(\omega - \omega_{\mathrm{c}})}\ . \tag{31}$$

We now have to calculate the sum in Eqn (31). Since in the formulation of the problem the absorption spectrum of the walls is assumed to be continuous, the sum in Eqn (31) can be reduced to an integral by introducing the density of states $\rho(\omega)$ for the walls:

$$\Gamma_{\mathrm{c}p} = \int_0^{\infty}\frac{|V_{\omega}|^2}{\hbar^2}\rho(\omega)\frac{1}{p + \mathrm{i}(\omega - \omega_{\mathrm{c}})}\,\mathrm{d}\omega\ . \tag{32}$$

Its calculation requires assumptions about the frequency dependence of $|V_{\omega}|^2\rho(\omega)$. Formally we are dealing with the same problem as in the calculation of a similar sum in the

process of spontaneous emission in free space. However, in this case the situation is less dramatic and there is more freedom in the choice of the model to describe the absorption of radiation by the walls.

We shall not need to analyse the dependence of Γ_{cp} on the microscopic parameters of the medium. It will be sufficient to know that

$$\Gamma_{cp} = \gamma_c - i\delta_c \tag{33}$$

is a complex quantity dependent on p. We can say this on the basis of the results of the preceding section if γ_c and δ_c are small compared with the eigenfrequency of the cavity ω_c. This can be demonstrated by a direct calculation of the integral (32) and a selection of the profile of the absorption band of the walls in, for example, the same form as adopted in the Appendix 7.2. In this case the quantity Ω is the spectral width of the absorption coefficient of the walls. If Ω is sufficiently large, then

$$\gamma_c = \pi |V_{\omega_c}|^2 \rho(\omega_c)\hbar^{-2}. \tag{34}$$

We can now easily calculate that $p = -\gamma_c + i\delta_c$ is a pole of B_p and

$$B(t)\exp(-i\omega_c t) = \exp\left[-(\gamma_c - i\delta_c)t\right]. \tag{35}$$

We can see that γ_c describes the cavity damping. It is usually found at $\gamma_c \ll \omega_c$ and this has been used here to calculate Γ_{cp} and γ_c. The probability of the state $|B(t)|^2$ decays at a rate characterised by the constant $2\gamma_c$. The quantity δ_c is the radiative correction to the cavity frequency; it only corrects this frequency, but does not lead to any qualitatively new effects.

3.3. Spontaneous decay rate and structure of the emission spectrum of an atom in a cavity

We shall now return to the task of solving the system of equations (28). Elimination of $C_{\omega p}$ from Eqn (28b) by means of Eqn (28c) gives

$$\left[p + \sum_{\omega} \frac{|V_\omega|^2}{\hbar^2} \frac{1}{p + i(\omega - \omega_c)}\right] B_p = \frac{U^*}{i\hbar} A_p. \tag{36}$$

The sum in the brackets in the above equation is known from the preceding section. Therefore,

$$B_p = \frac{U^*/i\hbar}{p + \gamma_c - i\delta_c} A_p, \tag{37}$$

which in turn makes it possible to calculate A_p:

$$A_p = \frac{p + \gamma_c - i\delta_c}{(p + i\Delta)(p + \gamma_c - i\delta_c) + |u|^2}, \tag{38}$$

where

$$\Delta = \omega_a - \omega_c, \quad u = U/i\hbar,$$

and to obtain the final expressions for B_p and $C_{\omega p}$:

$$B_p = \frac{-u^*}{(p + i\Delta)(p + \gamma_c - i\delta_c) + |u|^2}, \tag{39}$$

$$C_{\omega p} = \frac{u^* v_\omega^*}{[(p + i\Delta)(p + \gamma_c - i\delta_c) + |u|^2](p + i\Delta_\omega)}, \tag{40}$$

where

$$\Delta_\omega = \omega - \omega_c, \text{ and } v_\omega = V_\omega/i\hbar.$$

The inverse Laplace transformation, applied to Eqns (39) and (40), gives

$$A(t) = \frac{p_1 + \gamma_c - i\delta_c}{p_1 - p_2}\exp(p_1 t) - \frac{p_2 + \gamma_c - i\delta_c}{p_1 - p_2}\exp(p_2 t), \tag{41}$$

$$C_\omega(t) = u^* v_\omega^* \left(\frac{\exp(p_1 t)}{(p_1 - p_2)(p_1 - p_3)} + \frac{\exp(p_2 t)}{(p_2 - p_1)(p_2 - p_3)} + \frac{\exp(p_3 t)}{(p_3 - p_1)(p_3 - p_2)}\right), \tag{42}$$

where $p_{1,2}$ are general poles of the functions A_p and $C_{\omega p}$,

$$p_{1,2} = -\frac{1}{2}\left(\gamma_c - i\delta_c + i\Delta\right) \pm \left[\frac{1}{2}\left(\gamma_c - i\delta_c - i\Delta\right)^2 - 4|u|^2\right]^{1/2}, \tag{43}$$

are the roots of the equation

$$(p + i\Delta)(p + \gamma_c - i\delta_c) + |u|^2 = 0, \tag{44}$$

and

$$p_3 = -i\Delta_\omega \tag{45}$$

is the third pole of the function $C_{\omega p}$. The spectral distribution of the photons is given by

$$|C_\omega(\infty)|^2 \rho(\omega) = \frac{|u|^2 |v_\omega|^2}{|p_3 - p_1|^2 |p_3 - p_2|^2} \rho(\omega). \tag{46}$$

The frequency dependence of the denominator of Eqn (46) is decisive: $|v_\omega|^2 \rho(\omega)$ is a continuous function of the frequency and in fact can be replaced by a constant quantity $|v_{\omega_c}|^2 \rho(\omega_c) = \gamma_c/\pi$.

Let us now estimate typical parameters governing the process of spontaneous emission in a cavity. The dominant form of the interaction of atoms or molecules with an electromagnetic field is the dipole mechanism, so that

$$\hat{U} = -\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{E}}$$

Consequently, $|u| = (\boldsymbol{\mu}/\hbar)\langle 0,1,0|\hat{\boldsymbol{E}}|1,0,0\rangle$. A matrix element of the field is $\langle 0,1,0|\hat{\boldsymbol{E}}|1,0,0\rangle = \boldsymbol{e}(2\pi\hbar\omega_c)^{1/2}$ [2]; see also Eqn (7.1.3) in the Appendix 7.1.

In the classical microwave spectroscopy experiments a typical object of an investigation is the rotational spectrum of a molecule [48]. Such spectra correspond to dipole moments $|\boldsymbol{\mu}| \approx 10^{-18}$ cgs units. Since in this range we have $\omega_c \approx 10^{11}$ Hz, it follows that $|u| \approx 10^3\ \mathrm{s}^{-1}$. These experiments are relevant to the first work in the field of quantum electronics, which were studies of masers with molecular beams of ammonia, formaldehyde etc. [3]. Metal cavity resonators used in spectroscopic and maser experiments have Q factors $Q \approx 10^3 - 10^4$. Such values of the Q factor in the centimetre range of wavelengths

correspond to $\gamma_s \approx 10^7\ s^{-1}$. We can see that in these experiments the inequality $\gamma_c \gg |u|$ is obeyed. This is the reason why an approximation based on this inequality was used by Bunkin and Oraevski [17].

A different class of recent experiments is based on the use of transitions in the spectra of highly excited atoms (Rydberg transitions) [34]. The wavelengths corresponding to these transitions also lie in the microwave range. The correspond-ing dipole moment is $|\boldsymbol{\mu}| \approx 10^{-16}$ cgs units. Super-conducting cavities with $Q \approx 10^9$ are used in such experiments. Therefore, these experiments correspond to the inequality $\gamma_c \ll |u|$.

We shall now analyse these two cases.

In the first case ($\gamma_c \gg |u|$) not only γ_c, but also the detuning Δ can be large, so that we can calculate the values of the poles of Eqn (43) on the assumption that $\frac{1}{2}(\gamma_c - i\delta_c - i\Delta) \gg |u|$. We then find that

$$p_1 = -\gamma_c + i\delta_c, \quad p_2 = -\gamma + i\delta - i\Delta, \tag{47}$$

where

$$\gamma = |u|^2 \frac{\gamma_c}{\gamma_c^2 + (\delta_c + \Delta)^2}, \quad \delta = |u|^2 \frac{\delta_c + \Delta}{\gamma_c^2 + (\delta_c + \Delta)^2}. \tag{48}$$

The function A_p has thus in fact only one pole p_2 and

$$A(t) = \exp[-(\gamma - i\delta)t]. \tag{49}$$

We can see that δ is the radiative shift of the transition frequency of an atom in a cavity resonator. It disappears when $\delta_c + \Delta = 0$, i.e. when the cavity frequency (after allowance for the radiative shift) coincides with the frequency of the atomic transition.

It follows from Eqn (47) that

$$\frac{d|A|^2}{dt} = -2\gamma|A|^2. \tag{50}$$

The quantity $w_s^c = 2\gamma$ determines the rate of decay of the upper atomic level. If it is assumed that the interaction is of the dipole nature, $\hat{U} = -\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{E}}$, then w_s^c is described by Eqn (2).

Eqn (46) for the frequency distribution of the emitted photons reduces in this case to

$$|C_\omega(\infty)|^2 \rho(\omega) = \frac{\gamma}{\pi}[\gamma^2 + (\Delta_\omega - \delta)^2]^{-1}. \tag{51}$$

We can see that the emission line is a Lorentzian monoresonance with a half-width γ.

In the second case when $|u| \gg \gamma_c$:

$$p_{1,2} = -\frac{1}{2}\gamma_c \pm i|u|. \tag{52}$$

We shall now calculate the frequency distribution of the emitted photons given by Eqn (46). In this case we have

$$|C_\omega(\infty)|^2 \rho(\omega) = \frac{|u|^2\gamma_c/\pi}{[(\gamma_c/2)^2 + (\Delta_\omega + |u|)^2][(\gamma_c/2)^2 + (\Delta_\omega - |u|)^2]}\rho(\omega). \tag{53}$$

Eqn (53) describes a spectral distribution composed of two peaks (Fig. 1). A physical interpretation of this distribution is as follows. If $|u| \gg \gamma_c$, a photon emitted by an atom is reabsorbed by the atom with a much higher probability than the absorption by a wall. In this case the atom and the cavity can be regarded as one coupled system. The energy of this coupled system has two values. At $\omega_c = \omega_a$ these two values are $\omega_a + |u|$ and $\omega_a - |u|$. This is the reason for the appearance of the doublet. Such an interpretation is supported by direct calculation of the atom–cavity system

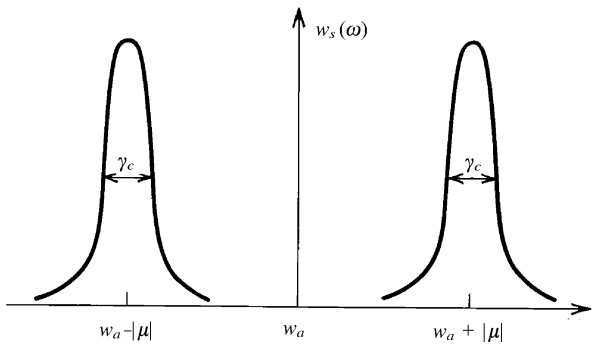

Figure 1. Doublet structure of the spectrum of spontaneous emission of an atom in a single-mode cavity. Here, $w_s(\omega)$ is a spectral density of the probability. The scale of the figure is arbitrary.

$$\Psi(t) = A(t)\exp(-i\omega_a t)|1,0,0\rangle + B(t)\exp(-i\omega_c t)|0,1,0\rangle \tag{54}$$

in the $|u| \gg \gamma_c$ approximation. For simplicity, we shall assume that $\omega_a = \omega_c = \omega_0$. In this approximation the poles of the functions A_p and B_p coincide and are given by Eqn (52). Therefore,

$$A(t)\exp(-i\omega_0 t) = \frac{1}{2}\exp\left[-\left(\frac{\gamma_c}{2}\right)t\right]\left\{\exp\left[-i(\omega_0 + |u|)t\right] + \exp\left[-i(\omega_0 - |u|)t\right]\right\}, \tag{55a}$$

$$B(t)\exp(-i\omega_0 t) = \frac{i}{2}\exp\left[-\left(\frac{\gamma_c}{2}\right)t - i\phi\right] \times\left\{\exp\left[-i(\omega_0 + |u|)t\right] - \exp\left[-i(\omega_0 - |u|)t\right]\right\}. \tag{55b}$$

In Eqns (55a) and (55b) the terms $\gamma_c/2$ are retained in the arguments of the exponential functions, but they are omitted from the pre-exponential factors. They do not play a significant role in these factors; they alter only slightly the values of the coefficient. The retention of the terms $\gamma_c/2$ in the arguments allows for a qualitatively important effect which is the decay of an excited state of the atom–cavity system. It is interesting to note that the decay of an excited state of this system is characterised by an exponential function with an argument half that describing the decay of energy in a cavity without an atom.

If allowance is made for Eqns (55a) and (55b), the wave function can be represented in the form

$$\Psi(t) = \exp\left[-\left(\frac{\gamma_c}{2}\right)t\right]\left\{\exp\left[-i(\omega_0 + |u|)t|+\rangle\right] + \exp\left[-i(\omega_0 - |u|)t|-\rangle\right]\right\}, \tag{56}$$

where

$$|\pm\rangle = \frac{1}{2}|[|1,0,0\rangle \pm \mathrm{i}\exp(-\mathrm{i}\phi)|0,1,0\rangle]. \tag{57}$$

The wave function (56) represents a superposition of two slowly decaying (quasistationary) states with energies $\hbar(\omega_0 \pm |u|)$. In accordance with these two values of the energy, the spontaneous emission spectrum has two maxima.

However, if we follow solely the fate of the states of an atom, we find from Eqns (55a) and (55b) that the probability of finding it in the upper or lower state oscillates at a frequency $2|u|$ [49].

We shall now find the ratio of the parameters $|u|$ and γ_c for which a monoresonance becomes a doublet. We shall do this by calculating $|C_\omega(\infty)|^2$ without invoking the inequality $|u| \gg \gamma_c$, but simply assuming that $|u| > \gamma_c/2$. We shall postulate that the cavity frequency is tuned exactly to the frequency of a spectral line: $\delta_c + \Delta = 0$. We then have $p_{1,2} = -(\gamma_c/2) + \mathrm{i}(\delta_c \pm \beta)$, $\beta = (|u|^2 - -\gamma_c^2/4)^{1/2}$, and

$$|C_\omega(\infty)|^2\rho(\omega) = \frac{|u|^2(\gamma_c/\pi)\rho(\omega)}{[(\gamma_c/2)^2 + (\Delta_\omega + \delta_c + \beta)^2][(\gamma_c/2)^2 + (\Delta_\omega + \delta_c - \beta)^2]}\rho(\omega). \tag{58}$$

It is obvious that a doublet in the spectral distribution of the emitted photons corresponds to two maxima with a minimum between them. The positions of the extrema of the function (58) on the frequency axis are given by the roots of the equation

$$(\Delta_\omega + \delta_c)\left[(\Delta_\omega + \delta_c)^2 + |u|^2 - \frac{1}{2}\gamma_c^2\right] = 0. \tag{59}$$

When considered as a function of the frequency, there are three real roots if

$$|u| > \frac{\gamma_c}{\sqrt{2}}\,. \tag{60}$$

It therefore follows that when an atom emits spontaneously in a single-mode cavity resonator, a doublet may appear in the emission spectrum if the cavity has a sufficiently large Q factor. It has been stated [50, 51] that in a certain range of values of the ratio $|u|/\gamma_c$ a triplet may appear in the spectrum of spontaneous emission from a two-level atom in a *single-mode* cavity. However, this is in conflict with the results of other investigations [35–37] and is not supported by the above theory.

4. Experimental verification of the doublet structure of the spectrum

The splitting of the energy states in a sufficiently strong external radiation field has been known from the time of the experiments of Autler and Townes [48]. However, observation of the splitting in the field of the intrinsic radiation of an atom the energy of which is equal to one photon, is difficult for fundamental reasons. It is necessary to study the emission from a single atom in a cavity because the situation becomes much more complex in the presence of many atoms: in the final analysis, the atoms may be in the field of many photons emitted by different atoms and even the number of atomic collective states involved in the process may become large [52].

The spectral distribution of the photons emitted spontaneously in a cavity can be recorded provided a sensitive detector capable of reacting to single photons is available. The construction of such a detector for microwave photons is a difficult experimental task. It is much simpler to detect the state of an atom which escapes from a cavity. It is therefore preferable to use an alternative manifestation of the spectral doublet effect which is a periodic time dependence of the probability of an atomic state. The probability of finding an atom in the upper or lower state at the exit from a cavity, depending on the time τ of its flight across the cavity, oscillates at a frequency $2|u|$. In fact, it follows from Eqns (55a) and (55b) that

$$|A(\tau)|^2 = \tfrac{1}{2}\exp(-\gamma_c\tau)[1 + \cos(2|u|\tau)]\,, \tag{61a}$$

$$|B(\tau)|^2 = \tfrac{1}{2}\exp(-\gamma_c\tau)[1 - \cos(2|u|\tau)]\,, \tag{61b}$$

which can indeed be observed experimentally.

The recent technological progress has made it possible to study the process of emission from single atoms in a cavity with a very high Q factor [34, 53]. The state of an atom emerging from a cavity can then be recorded.

The experiment involving observation of the probability of oscillations described by Eqns (61a) and (61b) is difficult to carry out in its pure form. First, it is necessary to ensure that there is no thermal radiation inside the cavity. In the microwave range this requires cooling to temperatures well below 1 K. Second, the experimental conditions must be such that the radiation from the atoms crossing consecutively the cavity does not accumulate inside it because otherwise the effect is not purely spontaneous. This means that at each moment in time there should be only one atom in the cavity and the time interval between the consecutive atoms should be much greater than the photon lifetime in the cavity $\tau_c = (2\gamma_c)^{-1} = Q_c/\omega_c$. If $Q \approx 10^9$ and $\omega_c \approx 10^{11}$ Hz, then $\tau_c = 10^{-2}$ s, so that the intensity of the atomic beam should not exceed 100 atoms per second. Moreover, a high degree of the selection of the velocities of the atoms is required.

Eqns (61a) and (61b) can be readily generalised by allowing for the presence of thermal radiation inside the cavity. In this case the probability $P_A(\tau)$ of finding an atom in an excited state is given by [53]

$$P_A(\tau) = \tfrac{1}{2}\exp(-\gamma_c\tau)\left[1 - \exp\left(-\frac{\hbar\omega_c}{kT}\right)\right] \times \sum_n \exp\left(-\frac{n\hbar\omega_c}{kT}\right)\left\{1 + \cos\left[2|u|(n+1)^{1/2}\tau\right]\right\}. \tag{62}$$

It is this expression that was checked experimentally at $T = 3$ K [53]. In this experiment use was made of a velocity-selected beam of ^{85}Rb atoms and a niobium resonator cavity with $Q_c = 8 \times 10^8$. The atoms were injected into the cavity in a highly excited (Rydberg) state $63p_{3/2}$. The resonator was tuned to the frequency $\omega_c = 2\pi \times 21.5$ GHz, corresponding to the $63p_{3/2} \to 61d_{3/2}$ transition in an atom. Time was measured from the moment of injection of an atom into the cavity. Measurements were carried out

over an interval of 30–140 μs. The authors found that Eqn (62) agreed excellently with the experimental results (Fig. 2).

We shall once again draw attention to the fact that the oscillations of the probability of finding an atom in a specific state at the exit from the cavity are related uniquely, in accordance with Eqns (56), (61a), and (61b), to the splitting of the energy levels of the atom–cavity mode system when these levels interact. Therefore, the experiments of H Walther and his colleagues, the results of which are plotted in Fig. 2, demonstrate unambiguously the splitting of the energy levels of the atom–cavity mode system, leading to the doublet structure of the spectrum.

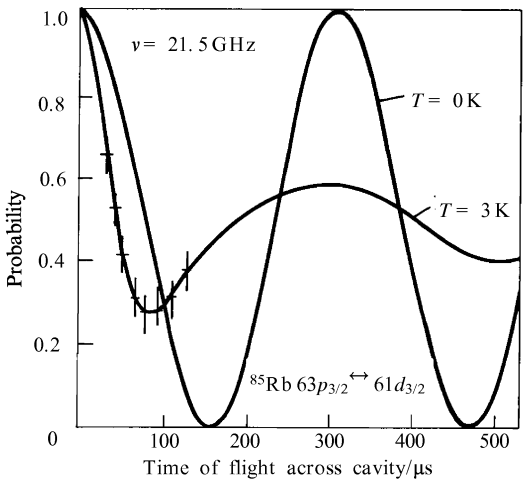

Figure 2. Oscillations of the probability of finding an atom in the upper energy state: theory and experiments [53].

Quite recently H J Kimble and his colleagues (California Institute of Technology) have succeeded in observing what is known as the *vacuum* Rabi splitting of the energy level of an atom into two closely spaced sublevels by carrying out experiments in the optical range [54]. This is in fact a direct observation of a spontaneous doublet.

5. Possibility of a triplet structure in the spectrum

A triplet structure of the emission spectrum may appear due to the interaction of an atom with two cavity modes which have similar frequencies or are even degenerate. It readily follows from the above discussion that the system of equations describing the dynamics of the process is

$$\mathrm{i}\hbar\frac{\mathrm{d}A}{\mathrm{d}t} = U_1 \exp\left[\mathrm{i}(\omega_\mathrm{a}-\omega_1)t\right]B_1 + U_2 \exp\left[\mathrm{i}(\omega_\mathrm{a}-\omega_2)t\right]B_2 + \mathrm{i}\hbar\delta(t)\,, \tag{63a}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}B_1}{\mathrm{d}t} = U_1^* \exp\left[-\mathrm{i}(\omega_\mathrm{a}-\omega_1)t\right]A + \sum_\omega V_{1\omega} \exp\left[\mathrm{i}(\omega_1-\omega)t\right]C_\omega\,, \tag{63b}$$

$$\mathrm{i}\hbar\frac{\mathrm{d}B_2}{\mathrm{d}t} = U_2 \exp\left[-\mathrm{i}(\omega_\mathrm{a}-\omega_2)t\right]A + \sum_\omega V_{2\omega} \exp\left[\mathrm{i}(\omega_2-\omega)t\right]C_\omega\,, \tag{63c}$$

and

$$\mathrm{i}\hbar\frac{\mathrm{d}C_\omega}{\mathrm{d}t} = V_{1\omega}^* \exp\left[-\mathrm{i}(\omega_1-\omega)t\right]B_1 + V_{2\omega}^* \exp\left[-\mathrm{i}(\omega_2-\omega)t\right]B_2\,. \tag{63d}$$

The notation is as follows: U_j is the operator of the interaction of an atom with the jth mode; V_j is the operator of the interaction of the jth mode with the cavity walls; ω_j is the eigenfrequency of the cavity; B_j is the amplitude of the state of the jth mode ($j = 1, 2$). The resut of the notation is the same as before.

We shall assume that the line emitted by such an atom is half-way between the frequencies of two normal modes of the cavity: $\omega_1 = \omega_\mathrm{a} - \Delta$, $\omega_2 = \omega_\mathrm{a} + \Delta$. Then, introduction of the Laplace transforms $A_p = A(t)$, $B_{1p} = B_1(t)\exp(\mathrm{i}\Delta t)$, $B_{2p} = B_2(t) \times \exp(-\mathrm{i}\Delta t)$, $C_{\omega p} = C_\omega(t)\exp(-\mathrm{i}\Delta_\omega t)$, where $\Delta_\omega = \omega - \omega_\mathrm{a}$, leads to the following equations:

$$pA_p = \frac{U_1}{\mathrm{i}\hbar}B_{1p} + \frac{U_2}{\mathrm{i}\hbar}B_{2p} + 1, \tag{64a}$$

$$(p-\mathrm{i}\Delta)B_{1p} = \frac{U_1^*}{\mathrm{i}\hbar}A_p + \sum_\omega \frac{V_{1\omega}}{\mathrm{i}\hbar}C_{\omega p}, \tag{64b}$$

$$(p+\mathrm{i}\Delta)B_{2p} = \frac{U_2^*}{\mathrm{i}\hbar}A_p + \sum_\omega \frac{V_{2\omega}}{\mathrm{i}\hbar}C_{\omega p}, \tag{64c}$$

$$(p+\mathrm{i}\Delta_\omega)C_{\omega p} = \frac{V_{1\omega}^*}{\mathrm{i}\hbar}B_{1p} + \frac{V_{2\omega}^*}{\mathrm{i}\hbar}B_{2p}. \tag{64d}$$

Elimination of $C_{\omega p}$ from these equations yields the following sums:

$$\Gamma_j = \sum_\omega \frac{|V_{j\omega}|^2}{\hbar^2}\frac{1}{p+\mathrm{i}\Delta_\omega}, \qquad K_{mn} = \sum_\omega \frac{V_{m\omega}V_{n\omega}^*}{\hbar^2}\frac{1}{p+\mathrm{i}\Delta_\omega}. \tag{65}$$

The complex quantities $\Gamma_j = \gamma_{\mathrm{c}j} + \mathrm{i}\delta_{\mathrm{c}j}$ describe, as before, the damping and the radiative shift of the eigenfrequency of the jth cavity mode. The quantities K_{mn} are the intermode coupling coefficients.

In order to concentrate our attention on the main effect, we shall simplify the calculations by postulating that $\delta_\mathrm{c} = 0$, $K_{mn} = 0$, $|V_{1\omega}| = |V_{2\omega}|$, $|U_1| = |U_2| = \hbar|u|$. This implies equality of the damping coefficients of the modes ($\gamma_1 = \gamma_2 = \gamma_\mathrm{c}$) and the absence of any intermode coupling.

At first sight the coupling coefficients are linked 'rigidly' to the damping coefficients, so that by selecting the values of $\gamma_{1,2}$ we are limited in the range of K_{mn}. There is naturally a linkage: in the selected model the values of $|K_{mn}|$ cannot exceed the largest of the coeffcients $\gamma_{1,2}$. However, within the limits of this constraint the coefficients K_{mn} can have any values since they depend not only on the moduli of $V_{1\omega}$ and $V_{2\omega}$, but also on the difference between their phases. In the case of a random phase distribution we have $K_{12} = K_{21} = 0$.

In order to answer the question of the spectral distribution of the emitted photons, we have to calculate — as before — the value of $|C_\omega(\infty)|$. Standard transformations give

$$|C_\omega(\infty)|^2 = 4|uv_\omega|^2 \frac{\gamma_c^2 + \Delta_\omega^2}{|(p_1 + i\Delta_\omega)(p_2 + i\Delta_\omega)(p_3 + i\Delta_\omega)|^2}, \quad (66)$$

where p_j are the roots of the following cubic equation with real coefficients:

$$p(p + \gamma_c)^2 + \Delta^2 p + 2|u|^2(p + \gamma_c) = 0. \quad (67)$$

If one of the roots of Eqns (61a) and (61b) is real (for example p_1), and the other two are complex conjugates ($p_{2,3} = \alpha \pm i\beta$), then

$$|C_\omega(\infty)|^2 \propto \frac{\gamma_c^2 + \Delta_\omega^2}{(p_1^2 + \Delta_\omega^2)[\alpha^2 + (\Delta_\omega - \beta)^2][\alpha^2 + (\Delta_\omega + \beta)^2]}. \quad (68)$$

In this case three spectral peaks appear if $\Delta^2 > 0$ and β is sufficiently large. If Δ^2 is small, then

$$|C_\omega(\infty)|^2 \propto \frac{1}{[\alpha^2 + (\Delta_\omega - \beta)^2][\alpha^2 + (\Delta_\omega + \beta)^2]} \times \left[1 + \frac{\Delta^2}{|u|^2} \frac{\gamma_c^2}{\gamma_c^2 + \Delta_\omega^2}\right]. \quad (69)$$

We can see that the appearance of a triplet is not subject to a threshold of the detuning Δ. If $|u| > \gamma_c/2$, the third peak appears as soon as $\Delta^2 > 0$. However, if Δ^2 is small, the amplitude of this peak is small because it is proportional to Δ^2. This is illustrated in Fig. 3.

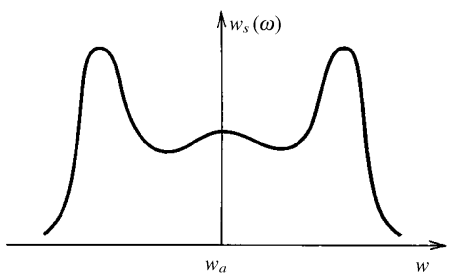

Figure 3. Triplet structure of the spectrum of spontaneous emission of an atom in a two-mode cavity. Here, $w_s(\omega)$ is a spectral density of the probability. The scale of the figure is arbitrary.

If $\Delta = 0$, it follows that $p_1 = -\gamma_c$, $\alpha = -\gamma_c/2$, and $\beta = [2|u|^2 - (\gamma_c^2/4)]^{1/2}$. We then have

$$|C_\omega(\infty)|^2 \propto \frac{1}{[\alpha^2 + (\Delta_\omega - \beta)^2][\alpha^2 + (\Delta_\omega + \beta)^2]}. \quad (70)$$

Let us now return to the doublet. The difference between a two-mode doublet when $\Delta = 0$ and a single-mode doublet is only this: there are two identical modes, the strength of the interaction of an atom with the cavity field is doubled, so that the condition for the appearance of a doublet is modified somewhat to

$$|u| > \gamma_c/2. \quad (71)$$

Can a triplet appear when $\Delta = 0$? It may appear if there is an intermode coupling. It follows from the classical theory of oscillations that two linearly coupled oscillators with absolutely identical frequencies ω_c can be represented by two independent normal oscillations of frequencies $\omega_+ = \omega_c(1 + \kappa)$ and $\omega_- = \omega_c(1 - \kappa)$, where κ is the frequency-normalised intermode coupling coefficient. However, this situation is practically equivalent to the case of two uncoupled modes with a detuning $2\Delta = 2\omega_c\kappa$ considered above. On the same basis we can predict a triplet in the spectrum of spontaneous emission of radiation by a moving atom: for a certain configuration of the cavity field a standing wave is 'seen' by an atom as two modes with frequencies displaced symmetrically (because of the Doppler effect) relative to the resonance frequency of the atom. These ideas are supported qualitatively by later calculations [55], but they represent a special case of a more complex pattern. It has been shown [55] that the motion of an atom may lead to the appearance of a multiplet structure. A complex structure of the spontaneous emission spectrum of a moving atom is associated with the fact that the motion of the atom modulates periodically the phase of the atom — cavity mode system. Periodic phase modulation is known to correspond to an infinite Fourier series with an equidistant set of frequencies. Interference between a large number of states corresponding to the Fourier components is in fact responsible for the multiplet structure of the spectrum. For certain velocities v of an atom (the important parameter is $\omega v/c|u|$) the structure of the spectrum may look like a doublet and a triplet. A very fast motion of an atom 'smears out' the structure of the spectrum and it is converted to a singlet even when the condition (71) is obeyed.

6. Conclusions

1. The process of spontaneous emission is frequently said to be due to quantum fluctuations. Quantum fluctuations of both the electromagnetic vacuum and of the dipole moment are important in this interpretation [11]. Since quantum fluctuations disappear in the classical approximation, it is sometimes stated that spontaneous emission is a pure quantum effect. However, this is incorrect, because spontaneous emission also occurs within the classical theoretical framework, as has rightly been pointed out by Ginsburg [12]. Nevertheless, the role of fluctuations in the quantum theory is important. The point is this: that in the classical theory a radiator is usually represented by a model of an excited dipole oscillator. However, if the dipole oscillator is excited, this means that either the dipole moment or its derivative or both are 'automatically' different from zero. A system of this kind unavoidably begins to emit radiation as soon as it can oscillate freely. The oscillations die out, as is known, because of the reaction of the radiation field on the oscillator.

The situation is different in the quantum theory. The excitation of a quantum system does not imply that the system must have an average dipole moment. Quite the reverse: this moment is usually zero and the system requires an initial impulse to start the process of emission. Quantum fluctuations provide such an impulse. In this connection it is important to stress that initiating fluctuations are needed also in some classical models. An example of such a model

is provided by the Bloch equations describing the dynamics of the magnetisation vector of a material. These equations have been derived by Bloch from purely classical considerations, but they have an exact quantum analogue and have been used widely in the description of the dynamics of masers and lasers beginning from the work of Fain [56] and Oraevskii [57]. Within the framework of these equations the energy stored by a radiator is governed by the z-component of the magnetisation and it cannot be zero if the system is excited. However, the radiation is generated because of the x- or y-components of the magnetisation, and initially these components may be zero. The system does not begin to emit of its own until the initial values of the x- or y-components of the magnetisation or of the emitted field are induced by fluctuations or in some other way.

2. It is well known that in addition to spontaneous emission, there is also stimulated emission which is the basis of the operation of generators of coherent radiation, in particular, of lasers. There is a close relationship between these two processes, as described by Einstein [58]

$$A_{21} = \hbar\omega^3(\pi^2c^3)^{-1}B_{21}. \tag{72}$$

The standard notation is used in the above relationship: A_{21} is the probability of a spontaneous transition from a level 2 to a level 1; B_{21} is known as the stimulated transition coefficient. A consequence of this fundamental relationship is that the probability of spontaneous emission can be calculated if we know the linear response of the investigated system (for example, an atom–cavity system) to an external monochromatic field [8]. A linear response of the system may be known in connection with investigations of other problems. The problem of spontaneous emission is then solved almost automatically. However, if the problem is formulated for the first time, the question is which is simpler: calculation of the linear response of the system or direct solution of the problem of spontaneous emission?

3. The reader can see that this review deals with the problem of spontaneous emission from a *single* atom. It is the interaction of a single atom with an unexcited field oscillator (free space or a cavity) that makes it possible to detect and understand fine fundamental effects of the interaction between matter and electromagnetic radiation (including the electromagnetic vacuum).

This review has not dealt with the problems of simultaneous interaction of many atoms or molecules with electromagnetic radiation. In the simultaneous interaction of many atoms with an electromagnetic field we are faced unavoidably with the problem of cooperative effects in spontaneous emission [52]. The cooperative effects represent such a very large problem that it cannot be fitted in the Procrustean bed of a single paper. I shall therefore confine ourselves to references to fairly recent reviews dealing with the cooperative effects [59, 60]. The cooperative effects represent a topic in which it is difficult to draw the line between the processes of spontaneous and stimulated emission. This leads to the hazard of sinking in the boundless sea of the results relating to the physics of masers, lasers, and even radiophysics and optics in general. For the same reason this paper does not deal with the problem of spontaneous emission in the presence of a strong monochromatic field. The reader is referred to the treatment of this problem elsewhere [61, 62].

Even the problem of the emission of radiation by a single atom can hardly be dealt with fully and permanently by a single paper. It is continuing to attract the attention of investigators to this day.

7. Appendices

7.1. Calculation of Eqn (14)

It follows from the canons of quantum electrodynamics that the electric vector of an electromagnetic field can be represented by a series

$$\hat{\boldsymbol{E}}(\boldsymbol{r}, t) = \sum_\alpha \hat{q}_\alpha^+ \boldsymbol{F}_\alpha^*(\boldsymbol{r}) + \hat{q}_\alpha \boldsymbol{F}_\alpha(\boldsymbol{r}), \tag{7.1.1}$$

where $\boldsymbol{F}_\alpha(\boldsymbol{r})$ are the eigenfunctions of the electrodynamic boundary-value problem corresponding to the problem in hand: $\hat{q}_\alpha^+$ and $\hat{q}_\alpha$ are the photon creation and annihilation operators. The electromagnetic field in free space can be described by eigenfunctions selected usually in the form of plane waves

$$\boldsymbol{F}_\alpha(\boldsymbol{r}) = \exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}),$$

so that

$$\boldsymbol{E}(\boldsymbol{r}, t) = \sum_\alpha \exp\left[\hat{q}_\alpha^+ \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}) + \hat{q}_\alpha \exp(-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r})\right]; \tag{7.1.2}$$

$\boldsymbol{e}$ is a unit vector in the direction of polarisation of the field; $\boldsymbol{e}\,\boldsymbol{k} = 0$, α is the combined $(\boldsymbol{k},\boldsymbol{e})$ index. Each wave vector $\boldsymbol{k}$ corresponds to two field oscillators in accordance with two linearly independent directions of the polarisation of an electromagnetic wave.

In the dipole interaction case we have

$$\hat{V} = -\hat{\boldsymbol{\mu}}\cdot\hat{\boldsymbol{E}}$$

and the calculation of V_{ω_α} involves calculation of the matrix elements of the operators $(\hat{\boldsymbol{\mu}}\cdot\mathbf{e})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\hat{q}_\alpha^+$ and $(\hat{\boldsymbol{\mu}}\cdot\boldsymbol{e})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\hat{q}_\alpha^+$. The matrix elements of the operators $\hat{q}_\alpha^+$ and $\hat{q}_\alpha$ are given by the following relationship which applies to the selected normalisation of the eigenfunctions:

$$\langle 0, 1|\hat{q}_\alpha^+|1, 0\rangle = \langle 1, 0|\hat{q}_\alpha|0, 1\rangle = (2\pi\hbar\omega)^{1/2}. \tag{7.1.3}$$

Let ϑ be the angle between the vectors $\boldsymbol{\mu}$ and $\boldsymbol{e}$. We then have $(\hat{\boldsymbol{\mu}}\cdot\boldsymbol{e})\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} = \hat{\boldsymbol{\mu}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}\cos\vartheta$. Calculation of the matrix element $\langle 0, 1_{\omega_\alpha}|\boldsymbol{\mu}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}|1, 0\rangle$ requires generally the knowledge of the specific wave functions of an atom. However, the value of $\boldsymbol{\mu}$ considered as a function of the coordinates differs from zero within the limits of the dimensions of an atom. If we then confine ourselves to spontaneous emission at wave-lengths not shorter than in the visible range, then within the limits of the dimensions of an atom we have $\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}} \approx 1$. Therefore,

$$\langle 0, 1_{\omega_\alpha}|\boldsymbol{\mu}\,\mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}}|1, 0\rangle = \langle 0, 1_{\omega_\alpha}|\boldsymbol{\mu}|1, 0\rangle = \mu(\omega_\alpha), \tag{7.1.4}$$

where $\boldsymbol{\mu}(\omega_\alpha)$ is a purely atomic characteristic found as usual from the experimental results.

It remains to calculate $\cos\vartheta$ and to sum in Eqn (13) over the polarisations and to integrate with respect to the directions of the emitted photons. The position and orien-

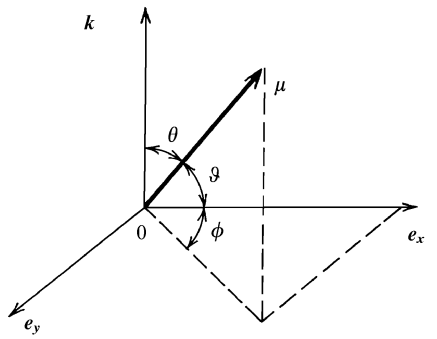

Figure 4. Orientation of the dipole moment of an emitting atom in a coordinate system linked to the unit vectors of a photon, i.e. with the wave vector $\boldsymbol{k}$ and the directions of polarisaitons $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$.

tation of an atom in space are assumed to be given. In the integration process the variable is the orientation of the vector $\boldsymbol{k}$. However, in calculations it is more convenient to link the coordinate system to the wave vector $\boldsymbol{k}$ by selecting its direction as the z axis; the other two axes can then be linked conveniently to two directions of linearly independent polarisations (Fig. 4). Then, integration over the directions of $\boldsymbol{k}$ in Eqn (13) is equivalent to integration with respect to the angles of the orientation of the vector $\boldsymbol{\mu}$ in the selected coordinate system. It is clear from Fig. 4 that $\cos^2\vartheta = \sin^2\theta\cos^2\phi$, if the emitted photon is polarised along the x axis and that $\cos^2\vartheta = \sin^2\theta\sin^2\phi$, if this photon is polarised along the y axis. Summation over these two polarisations has the effect that the square of the interaction matrix element is proportional to $\sin^2\theta$. Since $\mathrm{d}\Omega = \sin\theta\,\mathrm{d}\theta\,\mathrm{d}\phi$, integration over the directions of the vector $\boldsymbol{k}$ reduces to calculation of the following simple integral:

$$\int_0^{\pi}\mathrm{d}\theta\int_0^{2\pi}\mathrm{d}\pi\,\sin^3\theta = \frac{8}{3}\pi. \tag{7.1.5}$$

Substitution of Eqns (7.1.3)–(7.1.5) in Eqn (13) gives Eqn (14).

7.2. Calculation of the frequency integral

The reasoning given in Section 2 can be illustrated by calculating

$$\Gamma_p = 2(3\pi\hbar c^3)^{-1}\int_0^{\infty}|\boldsymbol{\mu}|^2\omega^3[p + \mathrm{i}(\omega_\alpha - \omega_\mathrm{a})]^{-1}\,\mathrm{d}\omega_\alpha \tag{7.2.1}$$

on the assumption that

$$|\boldsymbol{\mu}(\omega_\alpha)|^2\omega_\alpha^3 = \frac{|\boldsymbol{\mu}_0|^2}{\pi}\frac{\Omega^2}{\Omega^2 + (\omega_\alpha - \omega_\mathrm{a})^2}, \tag{7.2.2}$$

where $\boldsymbol{\mu}_0$ and Ω are constant quantities. This form of the dependence on ω_α in the case of a sufficiently high value of Ω satisfies the assumptions about the interaction matrix element made in the derivation of Eqn (17). However, this dependence is of purely model nature and it differs from the dependence $\mu(\omega_\alpha)$ for which estimates are given in the main text above. The selection made here simplifies maximally the calculation aspect of the problem without altering the fundamentals.

Calculation of the integral (7.2.1) subject to Eqn (7.2.2) gives the relationship

$$\Gamma_p = \gamma\frac{\Omega^2}{\Omega^2 + p^2}\left[\left(1 - \frac{1}{\pi}\arctan\frac{p}{\omega_\mathrm{a}} - \frac{p}{2\Omega} - \frac{p}{\pi\Omega}\arctan\frac{\omega_0}{\Omega}\right)\right.$$
$$\left. - \frac{\mathrm{i}}{2\pi}\ln\frac{\omega_\mathrm{a}^2}{\Omega^2 + \omega^2}\right], \tag{7.2.3}$$

where $\gamma = 2|\boldsymbol{\mu}_0|^2\omega_\mathrm{a}^3/3\hbar c^3$.

The poles of the function A_p are given, in accordance with Eqn (11), by

$$p = -\Gamma_p. \tag{7.2.4}$$

If we assume that $\gamma/\omega_\mathrm{a} \ll 1$ and $\gamma/\Omega \ll 1$, we can find the solution of Eqn (7.2.4) by the method of successive approximations, where $p = p_1 + p_2 + \ldots$ and

$$p_1 = -\gamma\left(1 - \frac{\mathrm{i}}{2\pi}\ln\frac{\omega_\mathrm{a}^2}{\Omega^2 + \omega^2}\right). \tag{7.2.5}$$

We can see that in this approximation the real part of a pole governing the decay of the upper atomic state is independent of the model parameter Ω. The imaginary part, governing the radiative frequency shift, depends on this model parameter. In the limit $\Omega \to \infty$ the imaginary part rises without limit and this is closely related to the general problem of divergences in quantum electrodynamics [38].

The corrections obtained in the next approximation are of the order of γ/ω_a and γ/Ω. These corrections depend on the model parameter Ω. It is obvious that their actual nature will change with the model.

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