

Dynamics of elementary atomic-molecular processes in gas and plasma. (Ed. V. Ashcheglov). Nova Science Publishers, New York, 1996; p. 329-338

ON THE ELEMENTARY DECAY PROCESS OF THREE-LEVEL SYSTEMS

D.Yu. Kuznetsov

Introduction

The quantum-mechanical model of the interaction of atoms with the field has been studied in papers [1, 2]. These papers present some arguments in favour of the applicability of such models. Not analysing the applicability of the rotary wave approximation and the problem of making the effective Hamiltonian to be dimensionless, we shall assume that the velocity of light and Plank constant are equal to 1 and modes interacting efficiently with the atomic system have been selected in the field, and corresponding coupling constants have been calculated. For brevity, this atomic system will be called simply an "atom" although, of course, it can be an ion, molecule ~~and~~ more complex cluster.

or even

We shall assume that atom and field are described by the Hamiltonian

$$H_0 = \int a_k^+ a_k k dk - Eu^+u + Fv^+v, \quad (1)$$

and their interaction is described by the Hamiltonian

$$H_1 = \int \eta (a_k^+ w^+ u + a_k u^+ w) dk + \int \zeta (a_k^+ w^+ v + a_k v^+ w) dk + \int \xi (a_k^+ v^+ u + a_k u^+ v) dk, \quad (2)$$

where

$$a_k^+ a_p - a_p a_k^+ = \delta(k-p), \quad w^+ w + w w^+ = 1, \quad (3)$$

$$u^+ u + u u^+ = 1, \quad v^+ v + v v^+ = 1,$$

and all other commutators $a, a^+, u, u^+, v, v^+, w, w^+$ are equal to zero.

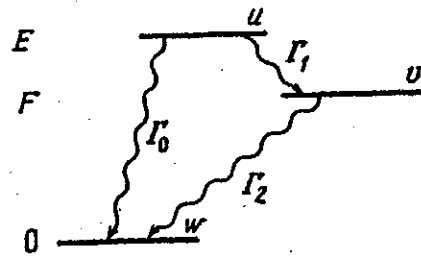


Fig 1. Diagram of atom levels.

Hamiltonian $H = H_0 + H_1$ describes a three-level atom interacting with a continuum of field modes: each of these modes can be treated as a harmonic oscillator. Fig. 1 presents the diagram of the atom levels. Parameters η , ξ , ζ in (2) are the coupling constants. In the approximation of narrow spectral lines they are considered to be independent on k . It will be clear below that $\Gamma = \Gamma_0 + \Gamma_1 = 2\pi\eta^2 + 2\pi\xi^2$, $\Gamma_2 = 2\pi\zeta^2$ are the decay rates of u and v levels.

In this paper we shall restrict our consideration to three specific cases when one of the constants η , ξ , ζ is zero. This means that the direct transition between one pair of levels is forbidden. In this case one can obtain the Schrodinger equation solution by the Weisskopf method [3]

$$i d\Psi/dt = H\Psi \quad (4)$$

in the visible analytic form.

1. Λ -TYPE TRANSITIONS (THE CASE OF $\zeta = 0$)

At $\zeta = 0$ the atom relaxes from u level to one of the stable levels w or v emitting one photon. It is convenient to search the equation (4) solution in the form

$$\Psi = h(t) e^{-iEt} u^+ |0\rangle + \int g_0(t) e^{-i(E-q)t} a_{q+E-F}^+ |0\rangle dq + \int f_q(t) e^{-i(E+q)t} \times a_{q+E}^+ |0\rangle dq, \quad (5)$$

where $|0\rangle$ is a vacuum, i.e. $a_k |0\rangle = u |0\rangle = v |0\rangle = w |0\rangle = 0$.

Substituting (5) into (4) we obtain the system

$$\begin{aligned} idh/dt &= \int (\eta f_q + \xi g_q) e^{-iqt} dq, \\ idg_q/dt &= \xi h e^{iqt}, \quad idf_q/dt = \eta h e^{iqt}. \end{aligned} \quad (6)$$

For the initial condition $h(0) = 1$, $g_q(0) = 0$, $f_q(0) = 0$ (which corresponds to the decay from the upper excited state) the system has a solution in the Weisskopf approximation

$$\begin{aligned} h &= \exp\{-(\Gamma/2)t\}, \\ g_q &= i\xi \frac{1 - \exp\{(iq - \Gamma/2)t\}}{iq - \Gamma/2}, \quad f_q = i\eta \frac{1 - \exp\{(iq - \Gamma/2)t\}}{iq - \Gamma/2}, \end{aligned} \quad (7)$$

where $\Gamma = \Gamma_0 + \Gamma_1$, $\Gamma_0 = 2\pi\eta^2$, $\Gamma_1 = 2\pi\xi^2$.

In the limit $t \rightarrow \infty$ the photon emitted at the transition from u level to one of the lower levels has a spectrum in the form of the Lorentzian bell-like profile localized near energy E in the case of transition to the lower level and near energy $E - F$ in the case of transition to the intermediate level. These bells have the same width which is determined by the total decay rate Γ . The relation between Γ_0 and Γ determines only relative areas of these bells, i.e. probabilities of atom transitions to levels w and v .

Note that $a_k^+ w^+ |0\rangle$ and $a_k^+ v^+ |0\rangle$ states are not identical and, hence, the interference between the two Lorentzian profiles does not occur even in the case when they overlap.

2. V-TYPE TRANSITION (THE CASE OF $\xi = 0$)

This case is interesting when $E \approx F$ and the upper levels overlap otherwise, the second level will in no way manifest itself at the transition from the upper level and the spectrum of the photon emitted will be determined by a single Lorentzian profile. Therefore, at $\xi = 0$ it would appear reasonable to seek the solution of (4) in the form

$$\Psi = h(t) e^{-iEt} u^+ |0\rangle + g(t) e^{-iFt} v^+ |0\rangle + \int f_q(t) e^{-i(G+q)t} w^+ |0\rangle, \quad (8)$$

where $G = (E + F)/2$. Substituting (8) in (4), we get the system

$$idh/dt = \int \eta f_q \exp\{-i(q + \varepsilon)t\} dq, \quad (9)$$

$$idg/dt = \int \zeta f_q \exp\{-i(q - \varepsilon)t\} dq,$$

$$idf_q/dt = \eta h \exp\{i(q - \varepsilon)t\} + \zeta g \exp\{i(q + \varepsilon)t\},$$

where $\varepsilon = (E - F)/2$. Let $f_q(0) = 0$. Then in the Weisskopf approximation the solution of system (9) has the form

$$h = \rho \cdot ((R_- + \gamma + i\varepsilon)h(0) + \sqrt{\gamma\beta}g(0))e^{(R_+ + i\varepsilon)t} - \quad (10a)$$

$$- \rho \cdot ((R_+ + \gamma + i\varepsilon)h(0) + \sqrt{\gamma\beta}g(0))e^{(R_- + i\varepsilon)t},$$

$$g = \rho \cdot (\sqrt{\gamma\beta}h(0) - (R_+ + \gamma + i\varepsilon)g(0))e^{(R_+ - i\varepsilon)t} -$$

$$- \rho \cdot (\sqrt{\gamma\beta}h(0) - (R_- + \gamma + i\varepsilon)g(0))e^{(R_- - i\varepsilon)t},$$

$$f_q = \frac{-\rho}{\sqrt{\pi}} \frac{(R_+ - i\varepsilon)h(0) - (R_+ + i\varepsilon)g(0)}{R_+ + iq} (1 - e^{(R_+ + iq)t}) +$$

$$+ \frac{\rho}{\sqrt{\pi}} \frac{(R_- - i\varepsilon)h(0) - (R_- + i\varepsilon)g(0)}{R_- + iq} (1 - e^{(R_- + iq)t}),$$

where

$$\rho = (R_- - R_+)^{-1}, \quad R_{\pm} = -(\gamma + \beta)/2 \pm [(\gamma + \beta)^2/4 - \varepsilon^2 + i\varepsilon(\gamma - \beta)]^{1/2},$$

(10b)

$$\gamma = \pi\eta^2 = \Gamma_0/2, \quad \beta = \pi\zeta^2 = \Gamma_2/2.$$

In the limit of interest to us $t \rightarrow \infty$

$$f_q = - \frac{\eta(q + \varepsilon)h(0) + \zeta(q - \varepsilon)g(0)}{(R_+ + iq)(R_- + iq)}. \quad (11)$$

At the transition from the upper excited state, the spectral curve has a hole (theoretically to zero) localized at the place of the second excited state. Fig.2 shows the characteristic form of such a curve. The curve is based on formulas (10b), (11) at $h(0) = 1$, $g(0) = 0$, $\gamma = 2\varepsilon$, $\beta = 0.3\varepsilon$. If the initial condition is given in the form of superposition of two excited states, one can obtain the superstable state when

$$h(0) = -g(0)\sqrt{\beta/\gamma}, \quad f_q = \sqrt{\frac{\gamma\beta}{\gamma + \beta}} \frac{2\varepsilon}{(R_+ + iq)(R_- + iq)}, \quad (12a)$$

and the superradiating state when

$$h(0) = g(0)\sqrt{\beta/\gamma}, \quad f_q = \sqrt{\frac{\gamma\beta}{\gamma + \beta}} \frac{2}{(R_+ + iq)(R_- + iq)}. \quad (12b)$$

In the case of $\beta \approx \gamma \gg \epsilon$ the emission spectrum width differs essentially from β, γ . Conversely, at $\epsilon \gg \beta, \gamma$ the spectrum takes the form of superposition of two nonoverlaped bells with half-widths β and γ .

Let us discuss the possibility of the experimental realization of such systems. For the Hamiltonian (2) to be applicable, it is important that both excited states would interact with the same modes of the field. The system of two similar ions in a Paul trap (see [4]) can be an appropriate simulation of such an atom in the case when the distance between ions exceeds substantially the atomic scale (on order of 10^{-8} cm^{-1}) but is considerably less than the emission wavelength (for example, on order of 10^{-4} cm^{-1}). Note that superradiating pairs of atoms similar to the (12b) case were described in [5]. Some problem consists in exciting only one of two close atoms (ions): the usual π pulse will excite efficiently both ions. The problem may be avoided using the difference between the coupling constants $\eta = \sqrt{\gamma/\pi}$ and $\zeta = \sqrt{\beta/\pi}$.

The same quasi-classical wave package may be $(2m + 1)\pi$ pulse for one level and $2n\pi$ pulse for another one.

If the splitting of the upper system levels is caused by the external (classical) field, one can prepare a system in the excited state in a strong field (when levels do not overlap) and then attenuate adiabatically the external field and observe the relaxation.

In this case the emission spectrum of such excited system will resemble Fig. 2.

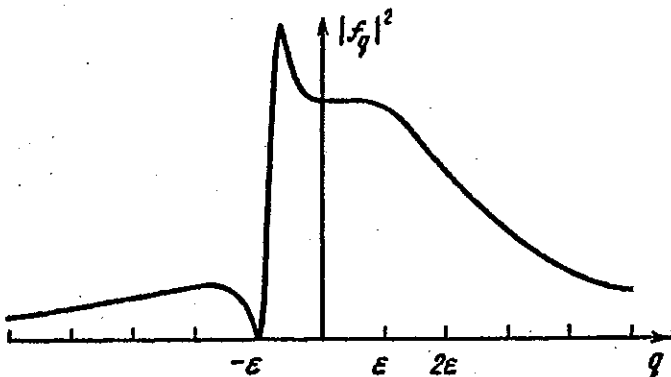


Fig.2. Form of the ^{expect} observed emission spectrum at the decay of the upper excited state by the v-type scheme constructed the formula (11) at $\gamma = 2\epsilon$, $\delta = 0.3\epsilon$, $g(0) = 0$, $h(0) = 1$

3. CASCADE TRANSITION AND FREQUENCY-TEMPORAL OSCILLATIONS (THE CASE OF $\eta = 0$)

At $\eta = 0$ only two-photon decay may occur. In this case it is convenient to seek the solution of Eq.(4) in the form

$$\Psi = h(t) e^{-iEt} u^+ |0\rangle + \int g_k(t) e^{-i(F+k)t} v^+ a_k^+ |0\rangle dk + \int \int f_{k,p}(t) e^{-i(k+p)t} a_k^+ a_p^+ w^+ |0\rangle dk dp. \quad (13)$$

The substitution of (13) to (4) gives

$$idh/dt = \int \xi g_k \exp\{i(E - F - k)t\} dk, \quad (14)$$

$$idg_k/dt = \int \zeta(f_{k,p}) + f_{p,k} \exp\{i(F - k)t\} dp + \xi h \exp\{-i(E - F - k)t\}, \quad (15)$$

$$idf_{k,p}/dt = \mu \xi g_k \exp\{-i(F - k)t\} + (1 - \mu) \zeta g_p \exp\{-i(F - p)t\}, \quad (16)$$

where μ is an arbitrary parameter. It is convenient to assume the equality $f_{k,p} = f_{p,k}$; to fit it to (16) one should assume $\mu = 1/2$.

Decay from the upper level corresponds to the initial condition

$$h(0) = 1, \quad g_k(0) = f_{k,p}(0) = 0. \quad (17)$$

The Weisskopf solution of the system (14) - (17) has the form

$$h(t) = \exp\{-\alpha t\}, \quad (18)$$

$$g_k(t) = -iR(\exp\{i(k - E + F)t - \alpha t\} - \exp\{-\beta t\}), \quad (19)$$

$$f_{k,p}(t) = 1/2 \tilde{f}_{k-E+F,p-F}(t) + 1/2 \tilde{f}_{p-E+F,k-F}(t),$$

$$\text{where } R = \xi/(ik + \alpha - \beta), \quad \alpha = \pi\xi^2 = \Gamma_1/2, \quad \beta = \pi\zeta^2 = \Gamma_2/2, \quad (20)$$

$$\tilde{f}_{q,r}(t) = R \frac{1 - \exp\{(iq + ir - \alpha)t\}}{iq + ir - \alpha} - R \frac{1 - \exp\{(ir - \beta)t\}}{ir - \beta}. \quad (21)$$

The distribution of emitted photons is determined by the function

$$\tilde{f}_{q,r} = \lim_{t \rightarrow \infty} \tilde{f}_{q,r}(t) = \frac{\sqrt{\alpha\beta}}{(iq + ir - \alpha)(ir - \beta)}. \quad (22)$$

One can measure $|\tilde{f}_{q,r}|^2$ using the spectral instrument. The Fourier transform module squared

$$f(x, y) = \frac{1}{2\pi} \int \exp\{-iqx - iry\} \tilde{f}_{q,r} dq dr = \sqrt{\alpha\beta} \theta(x) \theta(y - x) \exp\{-\alpha x - \beta(y - x)\} \quad (23)$$

can be measured by recording the time when emitted photons reach detectors. The meaning of formula (23) is clear: at first the upper level u decays with the rate of $\Gamma_1 = 2\alpha$ and then the intermediate level v decays with the rate of $\Gamma_2 = 2\beta$. One can check that the normalization has not been lost:

$$\int \int |\tilde{f}_{q,r}|^2 dq dr = \int \int |f(x, y)|^2 dx dy.$$

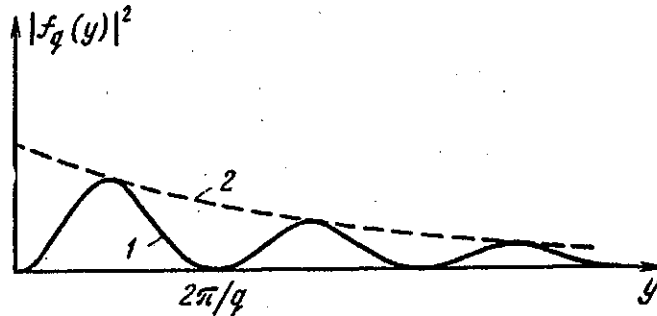


Fig. 3. The probability density of registration of the second photon at the instant of time y provided that the first photon has a frequency shift q (a curve 1) constructed for the case $\Gamma_1 = \Gamma_2 = \Gamma$ by formula (25); a curve 2 corresponds to the envelope $2 (\Gamma/q)^2 e^{-\Gamma y}$

It is of more interest is the case when the first photon of energy of $E - F$ order arrives at the spectral instrument, while the second photon (with the energy of F order) falls directly on the detector. After a sequence of tests such a scheme measures the function modulus squared

$$f_q(y) = \frac{1}{\sqrt{2\pi}} \int e^{-iqy} \tilde{f}_{q,r} dr.$$

Performing an integration one obtains

$$|f_q(y)|^2 = \frac{(2\pi)^{-1} \Gamma_1 \Gamma_2}{q^2 + (\alpha - \beta)^2} (e^{-2\alpha y} + e^{-2\beta y} - 2e^{-(\alpha+\beta)y} \cos(qy)). \quad (24)$$

At $\alpha = \beta$ ($\Gamma_1 = \Gamma_2 = \Gamma$) the formula (24) is simplified:

$$|f_q(y)|^2 = (2\pi)^{-1} (\Gamma/q)^2 e^{-\Gamma y} (1 - \cos(qy)). \quad (25)$$

This function is presented in Fig. 3. The oscillation period is determined by the first photon frequency shift $q = k$

- (E - F). One may say in some sense that there are beats occurring between the atom state and that of the field with slightly different energy.

The oscillating behaviour of the two-photon wave function may be explained on the basis of nearly classical arguments. Suppose the reduction of the two-photon wave package occurs at the second photon registration. In this case the first photon is emitted in a short time interval. The appropriate classical oscillator emits the wave package during $0 \leq t < y$ time interval; then the Fourier component amplitude with the frequency shift q is proportional to

$$c_q(y) = \int_0^y e^{iqt} dt = (1 - e^{iqy})/(iq). \quad (26)$$

Classical spectrum $|c_q(y)|^2$ determined by formula (26) involves the same oscillating factor $(1 - \cos(qy))$ as in the quantum theory (the formula (25)).

Correlations between photons emitted at the cascade transition may be considered as a realization of the Einstein-Podolskii-Rosen paradox (for noncascade transitions this paradox is described, for example, in [6]).

Note that distinction between F and E - F energies is not necessary if the photons involved can be distinguished by other characteristics, for example, by polarization. If the polarization is the same then photons have to be of "different colors", otherwise one can not say that one of the photons is "the first" and the other is "the second".

Few words can be said on the preparation of an atom in the excited state. This may be done either by two short π pulses at F and E - F frequencies (or by the equivalent pulse on the frequencies mixture) or by relaxation from the forth (higher than u) atom level with an obligatory registration of the photon emitted at the relaxation (the photon must turn the clock on to registrate the moment the last photon is emitted). Hence, the latter case deals with three-photon correlations.

CONCLUSION

The spectral distribution of photons emitted at the elementary decay of simple quantum-mechanical system was studied by an example of three-level atom. Even the rotary wave approximation allows to trace the influence of two close (neighbouring) levels on the emission spectrum. If there are, at least, two emitted quanta then a certain correlation between them occurs. Similar consideration may be performed in the case of many levels: if all cascades formed by the allowed transitions involve the same number of transitions then the solution may be obtained in the approximation of a certain number of quanta. The more general case when multi-quantum states are admixed (for example, at $\eta \neq 0$, $\xi \neq 0$, $\zeta \neq 0$) requires a special consideration. From the physical results of the paper one may set aside the following:

1. At the decay of one of the nearby levels connected with the ground state by the identical transitions, the hole appears in the spectral curve corresponding in frequency to the other level (see Fig. 2). The hole can shift at the decay of the superposition state (Eq.(11)). To study such dependences experimentally the special preparation of superpositional states is necessary.

2. At the cascade decay of three-level system two emitted photons are correlated in a certain way, the moment of the second photon registration is correlated specifically with the energy of the first photon. This relation is described by Eq. (25) and illustrated in Fig. 3. It can be detected with the coincidence circuit. It would be interesting to observe the oscillations, as at Fig.3, in the experiment with ions in a trap or atoms.

The author thanks A. V. Masalov, S.M. Kharchev, and V.A. Shcheglov for useful discussions and interest to this work.

ABSTRACT

The quantum-mechanical system (atom + field) is considered under assumption that one level is connected with two others by radiative transitions. The solutions of the Schrödinger equation are analyzed. The large-time limit of the solutions determines the distribution of the emitted photons. In some conditions this distribution has a nontrivial structure. Some ways to observe this structure in an experiment are discussed.

REFERENCES

- [1] S.V. Lavande and B.N. Jagtap, Cooperative effects in a system of strongly driven three-level atoms, Phys. Rev. A - Gen. Phys. 39, (1989) 694.
- [2] M.H. Mahran, A.-R. Samman, and A.-S.F. Obada, Quantum effects in 3-level atoms, J. Mod. Opt. 36, (1989) 53.
- [3] V. Weisskopf and E. Wigner, Berechnung der natürlichen Lichttheorie, Ztschr. Phys. 63, (1930) 54.
- [4] R. Blumel et al., Chaos and order of laser-cooled ions a Paul trap, Phys. Rev. A - Gen. Phys. 40, (1989) 808.
- [5] F.T. Arecchi and E. Courtens, Cooperative phenomena in resonant electromagnetic propagation, Ibid. 2, (1970) 1370.
- [6] D.N. Klyshko, The Einstein-Podolskii-Rosen paradox for the energy-time variables, Usp. Fiz. Nauk 158, (1989) 327.

also:

D. Kouznetsov. The decay of a three-level atom and time-frequency oscillations.

J. of Modern Optics 38, No. 1, 65-68 (1991)

H.J. Kimble, A. Mezzacappa.

Time dependence of photon correlations in a 3-level atomic cascade

Phys. Rev. A 31, No. 6, 3686-3697 (1985)
(1985)