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Laser-induced resonant transitions with Gaussian switching conditions

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Abstract. A simple closed-form formula is obtained for the transition probability of a *two-state atom* in a Gaussian laser pulse. The validity of the formula is tested by comparison with numerical results. The analytic formula is applicable over a wide range of the laser intensities, detunings and pulse durations. Particularly, it can be used for calculation of excitation probability when the Rabi frequency is of the order of or larger than a frequency associated with the pulse duration.

1. Introduction

The problem of calculation of transition probability in a system of two quantum states coupled by an oscillating time-dependent interaction, whose amplitude also varies in time in an arbitrary way, seems to have no general solution. An example of such a problem is the excitation of a two-level atom by a laser field whose amplitude is switched on and off as a time-dependent, bell-shaped function (in the reference frame of atom). While the numerical solution of this problem is relatively easy to carry out on modern computers, it is still important to derive reliable analytical results in which the transition probability dependence on the parameters of the problem is explicitly given.

A limited number of exact solutions exists for various pulse shapes (Bambini and Berman 1981, Carroll and Hioe 1986), but they apply unsatisfactorily in most real cases. Therefore, most theoretical work in this area has been devoted to the development of approximate solutions to the problem and to the study of their validity range (see, e.g., Rodgers and Swain (1987) and references therein).

The problem of switching on and off the laser field interaction is usually treated within either the sudden or adiabatic approximation. However, there are situations in practice where neither of these approximations is valid. In particular, the optical pumping of a two-level system by laser monochromatic radiation is related to the Rabi oscillations of the population of both the upper and lower level. In recent experiments (Kroon *et al* 1985, Lorent *et al* 1987) on fast atomic beams (2×10^3–$5\times10^5\ \mathrm{m\,s^{-1}}$), the transit time of atomic beam through the perpendicular laser beam and the time associated with the inverse of the Rabi frequency have been of the same order of magnitude. As a consequence, a first-order perturbational calculation cannot predict the pumping efficiency. It is, therefore, of great interest to obtain an accurate formula for the transition probability by using a non-perturbative approach. Indeed, in such intermediate cases, experimentalists can try to optimize the beam parameters to match a full inversion of the population.

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Let us consider an atom travelling with a velocity v and perpendicularly crossing the waist of a laser beam in the TEM_{00} mode of length L, as is schematically shown in figure 1. It is assumed that the spatial laser electric field distribution of the TEM_{00} mode inside the waist is described by a Gaussian function

$$E = E_0 \exp[-(l/L)^2]. \tag{1.1}$$

Since $l = vt$, the atom experiences a laser electric field of Gaussian time-dependent amplitude $E = E_0 \exp(-\gamma^2 t^2)$, where $\gamma = v/L$.

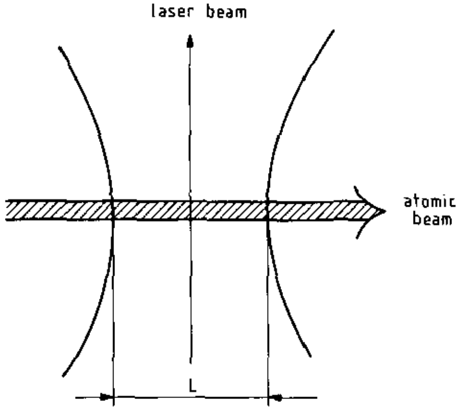

Figure 1. Geometry of an experiment with crossed atomic and laser beams.

A similar situation may be met when a two-level atom is overtaken by a Gaussian laser pulse of half-width $T = \gamma^{-1}(\ln(2))^2$, so that $E = E_0 \exp(-\gamma^2 t^2)$. With a constant laser field of frequency ω, the pulse parameters may significantly influence the transition probability, and the usual assumption of adiabatic switching on and off of the laser field seems to fail. We will focus our attention on the range of laser field parameters where the resonant Rabi frequency is larger than or of the same order as γ. This applies to both ps and ns lasers of intensity $I = 10^{10}\ \mathrm{W\ cm^{-2}}$ and larger.

The closed-form analytical expression which approximates the transition probability between two atomic states coupled by a Gaussian laser pulse is obtained in section 2. The method used in its derivation is the comparison equation method, which is well known in the field of slow atomic collision processes. The assumption of existence of strong coupling zones in both halves of the laser pulse, where the transitions are most probable, as well as 'the strong coupling' asymptotics of the resulting Bessel functions are used. As is demonstrated in section 3, by comparison of the analytical expression with the numerical solutions of the problem, this approach describes the process quite well over a wide range of the pulse parameters. Some applications of the obtained results as well as our concluding remarks are given in section 4.

2. Theory

Assume that an atom evolves in a laser field of frequency ω which near-resonantly couples two of its states, $|1\rangle$ and $|2\rangle$. The wavefunction of the atom may then be approximated by the following expansion:

$$\Psi(t)\rangle = \alpha(t)|1\rangle \exp(-\mathrm{i}\,W_a t) + \beta(t)|2\rangle \exp(-\mathrm{i}\,W_b t) \tag{2.1}$$

where $W_{a,b}$ are the eigenenergies of the unpertubed atomic Hamiltonian H_0. The full time-dependent Hamiltonian of the problem can be written as

$$H(t) = H_0 + V(t) \tag{2.2}$$

where $V(t)$ describes the laser-field–atom interaction, defined in the $\boldsymbol{p}\cdot\boldsymbol{A}$ gauge as

$$V(t) = \sum_{i=1,2} e(\boldsymbol{p}\cdot\boldsymbol{A}(t)/m) \tag{2.3}$$

and $\boldsymbol{A}(t)$ is the vector potential of the laser field, which is for a one-mode, linearly polarized laser, in the dipole approximation, given by

$$\boldsymbol{A}(t) = \boldsymbol{a}_0(t)\cos(\omega t). \tag{2.4}$$

The classical amplitude of the vector potential a_0 is a time-dependent function (slowly varying on the scale $1/\omega$), with the property $a_0(\pm\infty) = 0$, and a choice for $a_0(t)$ (as is discussed in Introduction) is

$$a_0(t) = \frac{\varepsilon_0(t)}{\omega} = \frac{E_0}{\omega}\exp(-\gamma^2 t^2). \tag{2.5}$$

Substitution of expansion (2.1) in the Schrödinger equation yields a system of first-order coupled differential equations for the amplitudes α and β. Assuming that the detuning $\delta = |\Delta W - \omega|$, where $\Delta W = W_2 - W_1$, is much smaller than ω, one can apply the rotating-wave approximation (RWA), which gives

$$\mathrm{i}\dot{\alpha} = \lambda \exp(-\gamma^2 t^2)\exp(-\mathrm{i}\delta t)\beta \tag{2.6a}$$

$$\mathrm{i}\dot{\beta} = \lambda \exp(-\gamma^2 t^2)\exp(\mathrm{i}\delta t)\beta. \tag{2.6b}$$

Here λ is the coupling strength, which is proportional to the maximum value E_0 of the laser electric field amplitude, as well as to the matrix element of the optical transition between $|1\rangle$ and $|2\rangle$. Defining $x = \gamma t$, $\mu = \Lambda/\gamma$, $\nu = \delta/\gamma$, the system (2.6) yields

$$\mathrm{i}\alpha' = \mu \exp(-x^2)\exp(-\mathrm{i}\nu x)\beta \tag{2.7a}$$

$$\mathrm{i}\beta' = \mu \exp(-x^2)\exp(\mathrm{i}\nu x)\alpha \tag{2.7b}$$

where α' denotes $(\partial/\partial x)\alpha$. Using the ansatze

$$\alpha = \exp(-\tfrac{1}{2}\mathrm{i}\nu x - \tfrac{1}{2}x^2)a \qquad \beta = \exp(\tfrac{1}{2}\mathrm{i}\nu x - \tfrac{1}{2}x^2)b \tag{2.8}$$

system (2.7) can be transformed into the two decoupled second-order differential equations

$$a'' + \mu^2 F(x)a = 0 \tag{2.9a}$$

$$b'' + \mu^2 F^*(x)b = 0 \tag{2.9b}$$

where

$$F(x) = -2\mathrm{i}(A/\mu)x + A^2 + \exp(-2x^2) - (1+x^2)/\mu^2 \tag{2.10}$$

and $A = \nu/2\mu = \delta/2\lambda$ is a detuning parameter, defined here as the ratio of the detuning and the Rabi frequency, and can, *in principle*, be *both small* (*at exact resonance* $A = 0$) and large. Without any loss of generality, it is assumed, for definiteness, that $A > 0$. Assuming that μ is the large parameter of the problem, an asymptotic expression is derived for both α and β, uniform in both A and x. In order to do so, the comparison equation method is used (see, e.g., Cherry 1950, Olver 1954, Nikitin and Umanskii

1984). The transition points of the equations (2.9) (roots of $F(x)=0$) in the x-complex plane are obtained up to the first power in small $1/\mu$ in the form

$$x_{1,3}=\tfrac{1}{2}\pi^{1/2}(\mp\exp(-\alpha/2)-\mathrm{i}\exp(\alpha/2))+\mathrm{i}/2\mu A+\mathrm{O}(1/\mu^2) \tag{2.11a}$$

$$x_{2,4}=\tfrac{1}{2}\pi^{1/2}(\mp\exp(-\alpha/2)+\mathrm{i}\exp(\alpha/2))+\mathrm{i}/2\mu A+\mathrm{O}(1/\mu^2) \tag{2.11b}$$

where

$$\sinh(\alpha)=\frac{2}{\pi}\ln(A). \tag{2.12}$$

$x_{1,2}$ lie in the $x<0$ complex half-plane, while $x_{3,4}$ lie in the $x>0$ half-plane, symmetrically located with regard to the $x_{1,2}$, with the imaginary axis as the axis of symmetry. It is convenient to write

$$x_1=x_{01}+\mathrm{i}/2A\mu \tag{2.13a}$$

$$x_2=x_{02}+\mathrm{i}/2A\mu \tag{2.13a}$$

where $x_{02}=x_{01}^*$. When $A\ll 1$ it follows $\alpha\ll 0$, i.e. $|\mathrm{Re}(x_i)|\gg 1$. Therefore, $x_{1,2}$ are far from the imaginary axis. But, since $\exp(\alpha/2)\ll 1$, x_1 and x_2 are close to each other and their effects should be simultaneously taken into account. Thus, two well separated and well localized zones of non-adiabatic transitions (the zones of avoided crossings of the adiabatic terms) appear during the development of the Gaussian pulse (at $x_{1,2}$ for $x<0$, and at $x_{3,4}$ for $x>0$). Outside of these zones, the atomic system in the laser field evolves adiabatically. On the other hand, when $A\simeq 1$ or even when $A\gg 1$, localization of the non-adiabatic regions is still present, as will be discussed later in this section. This enables one to extend the range of validity of the asymptotic ($\mu\gg 1$) theory to practically all values of A.

In the domain $x<0$, a choice for the etalon equation should be one of the equations of the special functions, which has two transition points as does the equation (2.10). A convenient choice is the equation of the Bessel functions

$$g''+\mu^2(\exp(2u)+C^2)g=0 \tag{2.14}$$

which has two independent solutions $J_{\pm\mathrm{i}\mu C}(\mu\,\mathrm{e}^u)$, in terms of the Bessel functions of the first kind, with both complex argument and index. C is the spectral parameter of the etalon equation and $u=u(x)$. Both C and $u(x)$ are found from the condition that the transition points in both (2.14) and (2.9a) are reached simultaneously. The solution of (2.9a) is assumed to be of the form

$$a=(u'(x))^{-1/2}g(u(x)) \tag{2.15}$$

which yields the equation for $u(x)$

$$(\exp(2u)+C^2)u'^2+\left[-2\mathrm{i}\frac{A}{\mu}x+A^2+\exp(-2x^2)-(1+x^2)/\mu^2\right]+\frac{1}{\mu^2}\left(\frac{3}{4}\left(\frac{u''}{u'}\right)^2-\frac{1}{2}\frac{u'''}{u'}\right). \tag{2.16}$$

Both u and C are assumed to be of the form of asymptotic series in powers of μ^{-1}, that is

$$u(x)=\sum_{k=0}^{\infty}u_k(x)\frac{1}{\mu^k}\qquad C=\sum_{k=0}^{\infty}C_k\frac{1}{\mu^k} \tag{2.17}$$

with initial conditions $u(x_{1,2})=0$. This yields (up to the small correction of the order of μ^{-2})

$$C=-\frac{\mathrm{i}}{\pi}\int_{x_{01}}^{x_{01}^*} f(x)\,\mathrm{d}x+\frac{\mathrm{i}}{2\mu}=C_0+\frac{\mathrm{i}}{2\mu} \tag{2.18a}$$

$$R+C_0[u_0-\ln(C_0+R)]+\frac{\mathrm{i}}{2\mu}[u-\ln(C_0+R)]$$
$$=\int_{x_{01}}^{x} f(x)\,\mathrm{d}x-\tfrac{1}{2}\mathrm{i}\pi C_0-\frac{\mathrm{i}}{2\pi}[\ln(A+f(x))+x^2] \tag{2.18b}$$

where

$$f(x)=[\exp(-2x^2)+A^2]^{1/2} \tag{2.19a}$$

is the Rabi frequency, divided by $\exp(x^2)$, and

$$R=[\exp(2u)+C_0^2]^{1/2}. \tag{2.19b}$$

From (2.18a) it follows that C_0 is a real number.

Similarly, by choosing equation (2.14) as an etalon equation for (2.9b), one obtains

$$C_b=C^* \qquad u_b=u^*. \tag{2.20}$$

Thus, using the relation

$$b=(u_b')^{-1/2}g \tag{2.21}$$

b is found in the form

$$b=\frac{(R^*)^{1/2}}{f(x)^{1/2}}\exp(-x^2/2)g \tag{2.22}$$

where

$$g=D_1J_z(\mu\exp(u^*))+D_2J_{-z}(\mu\exp(u^*)) \tag{2.23}$$

and D_1 and D_2 are the integration constants (to be found from the initial conditions of the problem), $z=\mathrm{i}\mu C^*$. Assuming that the system evolves from $t=-\infty$ to the transition zone adiabatically, with the initial conditions $\alpha(-\infty)=1$, $\beta(-\infty)=0$, and using (2.23) together with the form of Bessel functions of a small argument, it follows $D_2=0$. Finally, one gets

$$b=D_1p_0^{-1/2}\exp(-x^2/2)J_z(\mu\exp(u^*)) \tag{2.24a}$$

$$a=D_1[(C_0p_0^{1/2}-Ap_0^{-1/2})\exp(x^2/2)J_z(\mu\exp(u^*))$$
$$+ip_0^{1/2}\exp(u^*+x^2/2)J_z(\mu\exp(u^*))] \tag{2.24b}$$

where

$$p_0=f/R_0 \tag{2.25a}$$

R_0 is given by

$$R_0=(\exp(2u_0)+C_0^2)^{1/2} \tag{2.25b}$$

and

$$R_0-C_0[u_0-\ln(C_0+R_0)]=\mathrm{Re}\left(\int_{x_{01}^*}^{x} f(x)\,\mathrm{d}x\right). \tag{2.25c}$$

The evolution matrix **T**, which describes evolution of the system in time interval $(-\infty, 0)$ is defined by

$$T_{11}=\alpha(0) \qquad T_{21}=\beta(0) \qquad T_{12}=-T_{21}^{*} \qquad T_{22}=T_{11}^{*}. \tag{2.26}$$

Due to the symmetry of the transition points $x_{3,4}$ and $x_{1,2}$ with respect to the $\mathrm{Im}(x)$ axis, the evolution matrix **F** for the system in time interval $(0, +\infty)$ is found from **T** by using the relation (see, e.g., Krstić and Janev 1988)

$$\mathbf{F}=(\mathbf{T}^{*})^{-1} \tag{2.27}$$

which yields for the S-matrix of the system

$$\mathbf{S}=\mathbf{F}\cdot\mathbf{T}. \tag{2.28}$$

By straightforward calculations, the transition matrix element S_{21} is now found in the form

$$S_{21}=\frac{2\mathrm{i}\,\mathrm{Im}[b(0)a^{*}(0)]}{\mathrm{Det}} \tag{2.29a}$$

where

$$\mathrm{Det}=|a(0)|^{2}+|b(0)|^{2}. \tag{2.29b}$$

Equations (2.27) and (2.28) yield

$$S_{21}=-\frac{2i\,\mathrm{Re}[\exp(u_0)J_{z^{*}-1}(\mu\exp(u_0))J_z(\mu\exp(u_0^{*})]}{\mathrm{Det}\,1} \tag{2.30a}$$

with

$$\begin{aligned}\mathrm{Det}\,1=&[C_0^2p_0+(A^2+1)/p_0-2C_0A]|J_z(\mu\exp(u_0^{*}))|^2\\&+p_0\exp(2\,\mathrm{Re}(u_0))|J_{z-1}(\mu\exp(u_0^{*}))|^2\\&-2\,\mathrm{Im}[(C_0p_0-A)\exp(u_0)J_{z^{*}-1}(\mu\exp(u_0))J_z(\mu\exp(u_0^{*}))]\end{aligned} \tag{2.30b}$$

and in u_0 and p_0 it is assumed $x=0$.

Since both the arguments and the indices of the Bessel functions in (2.30) are proportional to the large parameter μ, the double asymptotics for the Bessel functions of both complex argument and index is used. As was discussed by Crothers (1972, 1975, 1976) on the example of parabolic cylinder functions, this kind of asymptotics, called 'strong coupling' asymptotics is suitable in calculation of the transition probability between two strongly coupled states, when the localization of the non-adiabatic regions is not pronounced, as in the case of transitions in the vicinity of the classical turning point in collisions of ions and atoms. From (2.25) it follows that, if $A\ll 1$

$$|\mu\exp(u)|\to\mu\pi^{1/2}/2 \qquad |z|\to\mu A\ln^{-1/2}(A) \tag{2.31a}$$

and therefore, the asymptotics of the Bessel functions for argument much larger than index would be suitable. This is in agreement with the discussion above that if $A\ll 1$, the localization of the non-adiabatic regions is pronounced. But when $A\gg 1$

$$|\mu\exp(u)|\to\mu A\ln^{-1/2}(A) \qquad |z|\to\tfrac{1}{8}\mu A\ln^{1/2}(A) \tag{2.31b}$$

and, in this case, the double asymptotics is needed. This kind of asymptotics of the Bessel functions, which is uniform in the index and therefore has the correct limit even in case $A \ll 1$, can be written in the form (Krstić and Janev 1988)

$$J_{i\nu+1/2}(\lambda) \to \frac{2}{1+i\nu} J_+ \qquad J_{i\nu-1/2}(\lambda) \to J_- \tag{2.32a}$$

where

$$J_\pm = \frac{2}{\Gamma(\frac{1}{2}+i\nu)} \left[\frac{\nu}{(\nu^2+\lambda^2)^{1/2}}\right]^{1/2} \exp[(i\nu \pm \tfrac{1}{2}) \ln(i\nu \pm \tfrac{1}{2}) \pm \tfrac{1}{2}]$$
$$\times \exp(i\nu - i\pi/4) \begin{matrix}\cos\\ \sin\end{matrix}\left\{(\nu^2+\lambda^2)^{1/2} - \nu \ln\left[\frac{\nu+(\nu^2+\lambda^2)^{1/2}}{\lambda}\right]\right.$$
$$\left. -\frac{i}{2}\left[\pi\nu \pm \frac{(\nu^2+\lambda^2)^{1/2}}{\nu} \mp \ln\left(\frac{\nu+(\nu^2+\lambda^2)^{1/2}}{\lambda}\right)\right]\right\}. \tag{2.32b}$$

This yields for the Bessel functions in (2.30)

$$J_z(\mu \exp(u^*)) \to G \sin(\vartheta_1) \tag{2.33a}$$

$$J_{z-1}(\mu \exp(u^*)) \to G \cos(\vartheta_2) \tag{2.33b}$$

where the complex quantities G, ϑ_1 and ϑ_2 are defined as

$$G = 2\mu^{-1/2}\Gamma^{-1}(z) R_0^{1/2} \exp[-i\mu C_0(1-\ln(\mu_0)) - \mu C_0\pi/2] \tag{2.34a}$$

$$\vartheta_1 = \mu \operatorname{Re} \int_{x_{02}} f(x)\,dx - i\mu C_0\pi/2 + \tfrac{1}{2}i \ln[A+(A^2+1)^{1/2}] \tag{2.34b}$$

$$\vartheta_2 = \vartheta_1 + i[u_0 - \ln(C_0+R_0)]. \tag{2.34c}$$

Using (2.34), expression (2.30) significantly simplifies, with the result

$$S_{21} = -\frac{i \sin(2\mu \operatorname{Re}\Gamma)}{\cosh(2\mu \operatorname{Im}\Gamma)} \tag{2.35a}$$

where

$$\Gamma = \int_0^{x_{02}} f(x)\,dx = \int_0^{x_{02}} [A^2 + \exp(-2x^2)]\,dx \tag{2.35b}$$

and x_{02} was defined in (2.11)-(2.13).

The probability of excitation of the state $|2\rangle$ by Gaussian laser pulse, starting from the state $|1\rangle$, is finally

$$P_{21} = |S_{21}|^2 = \frac{\sin^2(2\mu \operatorname{Re}\Gamma)}{\cosh^2(2\mu \operatorname{Im}\Gamma)}. \tag{2.36}$$

From the form of equations (2.9) it is obvious that the sign of detuning does not influence the probability, i.e.

$$P_{21}(-A) = P_{21}(A). \tag{2.37}$$

We note that the transition probability (2.36) exactly coincides with the one obtained for a more general case of both Gaussian interaction and detuning, in the limit of constant detuning (Janev and Krstić 1990), although the solution there was obtained in terms of confluent hypergeometric functions and the corresponding strong coupling asymptotics.

3. Validity of the result

The transition probability P_{21}, given by (2.36), is a simple function of two parameters, μ and Γ. The Γ is a function only of A and has to be calculated numerically. Expression (2.33b) can be transformed in a form convenient for numerical evaluation

$$\Gamma = x_{02} A \int_0^1 [1 + A^{2(y^2-1)} \exp(\mathrm{i}\pi y^2)]^{1/2}\, \mathrm{d}y. \tag{3.1}$$

Dimensionless μ is assumed a large number, while A is arbitrary. Their product $\mu A = \nu/2$ is a free parameter. It is convenient to have Γ tabulated in order to evaluate P_{21} in a simple and transparent way. Figures 2(a) and 2(b) represent Re Γ and Im Γ plotted against A, respectively, for A in the interval $(10^{-6}, 2)$.

In order to evaluate the bounds of validity of the result (2.36) as a function of μ and ν, a comparison with numerical solution of equations (2.7) is done and presented in figures 3, 4 and 5. As can be seen from figure 3, the agreement is satisfactory for all values of ν (the detuning), and it becomes better with increasing μ. Even for μ of

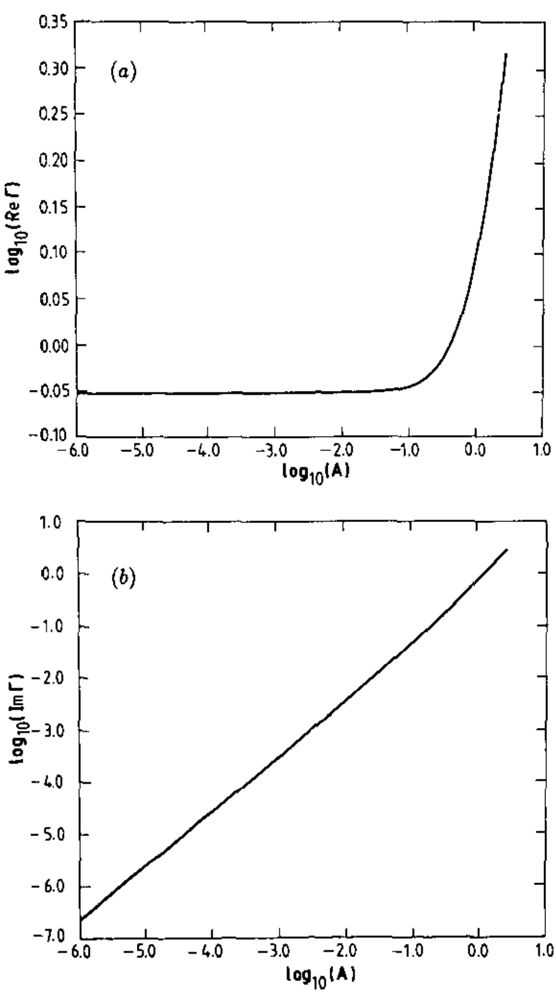

Figure 2. Numerically calculated: (a) Re(Γ), and (b) Im(Γ) plotted against $A = \nu/2\mu$.

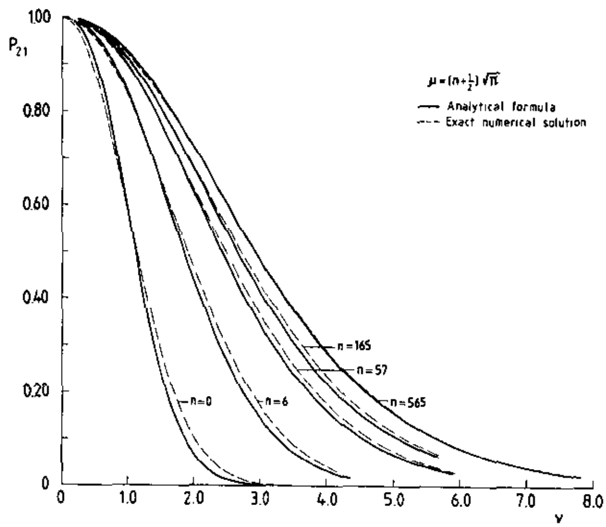

Figure 3. Comparison of the transition probability P_{21} calculated by formula in (2.36) and by numerical solution of (2.36). Results of Thomas (1983) and Carroll and Hioe (1986) are also presented.

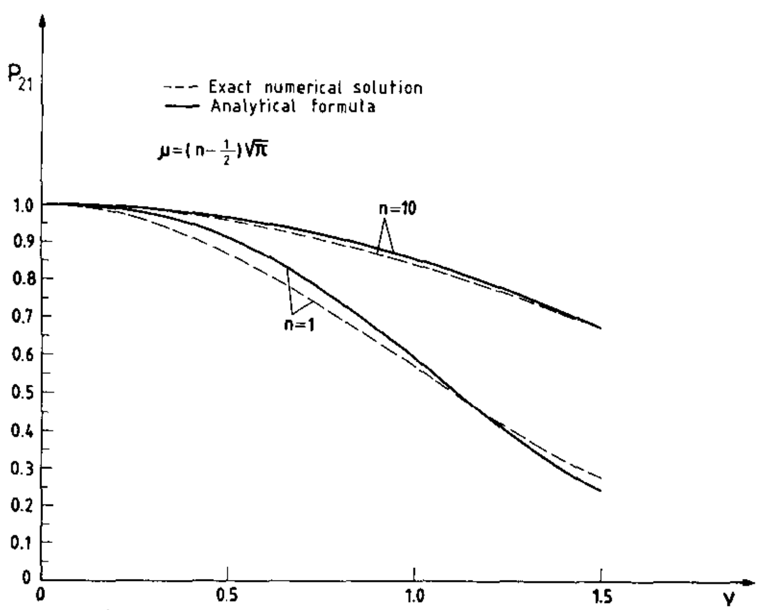

Figure 4. Transition probability plotted against ν for small values of μ.

the order of 1, the formula (2.34) reproduces basic properties of the numerical solution, as is shown in figures 4 and 5.

Calculation of Γ, and therefore of P_{21}, can be considerably simplified in the limit of small and large values of A. Γ can be expanded about $A=0$, with the result

$$|\mathrm{Re}(\Gamma)| \to \tfrac{1}{2}\pi^{1/2} \qquad |\mathrm{Im}(\Gamma)| \to \tfrac{1}{4}\pi A \ln^{-1/2}(A) \qquad A \ll 1. \tag{3.2}$$

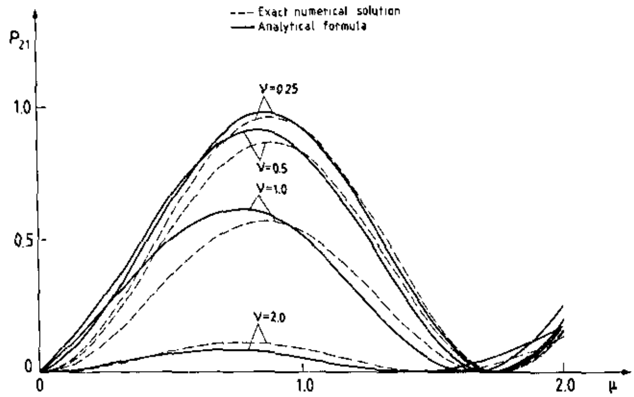

Figure 5. Transition probability plotted against μ, in the range of μ between 0 and 2.

From figure 2 it is obvious that approximations (3.2) may be applied up to $A=0.1$. Therefore, in the range $0<A<0.1$ the transition probability, equation (2.36), is correctly given by the formula

$$P_{21}=\frac{\sin^2(S)}{\cosh^2[\frac{1}{4}\pi\nu\ln^{-1/2}(\delta/2\lambda)]} \tag{3.3}$$

where

$$S=\mu\pi^{1/2} \tag{3.4}$$

is the pulse area, and δ, λ and $\nu=\delta/\gamma$ were defined in section 2. In the case of exact resonance ($A=0$), expression (3.3) goes to the exact result (Thomas 1983)

$$P_{21}=\sin^2(S). \tag{3.5}$$

In the limit $A\gg1$, Γ could be also simplified, with the result

$$|\mathrm{Re}(\Gamma)|\sim A\ln^{-1/2}(A) \qquad |\mathrm{Im}(\Gamma)|\sim\tfrac{1}{8}A\ln^{1/2}(A) \qquad A\gg1. \tag{3.6}$$

In the two limits, (3.2) and (3.6), the transition probability can be calculated in terms of algebraic functions. For A in between, Γ has to be calculated numerically. Although it could seem that in that case the direct numerical solution of the system of coupled differential equations (2.7) could be more acceptable, the computing time for calculation of Γ is about a hundred times shorter.

It is to be noted that, in the limit $\mu\to0$, from (2.36) it follows

$$P_{21}\sim\mu^2. \tag{3.7}$$

This is the correct behaviour of P_{21} when μ and ν are small, since from (2.7b) in the perturbation theory limit one gets

$$P_{21}^{\mathrm{pert}}=\pi\mu^2\exp(-\nu^2/2). \tag{3.8}$$

Carroll and Hioe (1986) developed a class of analytically solvable two-state problems. A special case is when the interaction matrix element and the detuning are of the form

$$H_{21}=\lambda\exp(-\gamma^2t^2) \qquad \Delta=\Delta_0\frac{\exp(-\gamma^2t^2)}{\cos(\pi/2\,\mathrm{erf}(\gamma t))}. \tag{3.9}$$

The resulting transition probability was obtained in the form

$$P_{21}=\frac{\sin^2(S)}{\cosh^2(\nu\pi^{1/2}/2)} \tag{3.10}$$

where ν and S are defined as below (3.3). Since $\Delta \simeq \Delta_0$ only when $\gamma t \ll 1$, one can expect that (3.10) can be acceptable for the model defined in section 2, if the important domain for transition (of avoided crossings of the terms) also satisfies the same condition ($\gamma t \ll 1$). This happens only in a very limited range of parameters μ and ν as can be seen in figures 3 and 4, where the comparison of expression (3.10) is given plotted against the exact numerical result. On the same figures, the result of Thomas (1983) is presented, which was obtained within the Magnus approximation, in the limit of small detunings ν, for the model of Gaussian interaction and constant detuning. This result can be written in the form

$$P_{21}=\exp(-\nu^2/2)\sin^2(S). \tag{3.11}$$

Obviously, both results, (3.10) and (3.11), apply well for $\mu \leqslant 1$ and for relatively small detunings. Although, the expression (2.36) is derived under condition $\mu \gg 1$, it also applies equally well in the region of validity of (3.10) and (3.11). Still, the results of Thomas and Carroll do not show correct values for larger μ, unless $\nu \ll 1$, when all P_{21} converges to a same value (the curves of (3.10) and (3.11) are universal at figures 3 and 4). We also note that the result of Thomas shows agreement with the Rosen–Zener conjecture. From the above discussion one can conclude that this conjecture is not valid for the considered model, unless $\mu \leqslant 1$ (very short pulses or a weak laser field). The extended discussion of the Rosen–Zener conjecture, given by Bambini and Berman (1981) applies to a class of asymmetric pulses (and to the symmetric Rosen–Zener itself), but not to the model of Gaussian interactions. Furthermore, in the derivation of the result (2.36), only the structure of the transition points is specific for the Gaussian pulses. Therefore, the validity of the expression (2.36) can be expanded beyond the Gaussian model, to any symmetric pulse which couples two states so that

$$H_{21}=\lambda f(t/\tau) \tag{3.12}$$

where τ is the pulse duration and the detuning δ is constant. Then, assuming there are only two (complex) transition points $t_{1,2}$ in domain $t<0$, and if the leading terms in expansion of $t_{1,2}$ in small $1/\lambda\tau$ are complex conjugated, then the transition probability in the RWA may be written in the form of (2.36), where $\mu=\lambda\tau$ and

$$\Gamma=\int_0^{t_1}(A^2+f^2(t/\tau))^{1/2}\,\mathrm{d}t/\tau \tag{3.13}$$

where $A=\delta/2\lambda$.

4. Discussion of the results

From the expression (2.36), and having in mind the behaviour of Γ in figures (2*a*) and (2*b*) one concludes that, for a fixed A, P_{21} oscillates with μ, and the oscillations are dumped with increasing of μ. The increase of μ, with A fixed, means an increase of the pulse half-width $1/\gamma$, and therefore, the short pulses are more efficient for the two-state excitation (as long as the maximum of the oscillation probability is concerned). In the limit $\mu\to\infty$ ($\gamma\to 0$, adiabatic limit), $P_{21}\to 0$. For a fixed μ, P_{21} decreases with an increase of the detuning, since Im(Γ) increases with A, while $\sin^2[\mathrm{Re}(\Gamma)]$ remains bounded. Finally, for a fixed detuning and γ, an increase of the laser intensity will cause the increase of μ and decrease of A.

There are some interesting consequences for the transition probability when the travelling Gaussian laser pulse resonantly couples the two states of an atom. Estimating the coupling matrix element λ as

$$\lambda \simeq eEa_0 = \mathrm{Ry}\left(\frac{I}{I_\mathrm{A}}\right)^{1/2} = \mathrm{Ry}\left(\frac{I_0}{I_\mathrm{A}}\right)^{1/2} \exp(-\gamma^2 t^2) \tag{4.1}$$

where I_A is the atomic unit for the radiation intensity, it follows

$$\mu = \frac{\mathrm{Ry}}{\gamma}\left(\frac{I_0}{I_\mathrm{A}}\right)^{1/2}. \tag{4.2}$$

A ns and ps laser yield $\gamma_\mathrm{n} \simeq 10^{-6}$ eV, and $\gamma_\mathrm{p} \simeq 10^{-3}$ eV, respectively. At the laser intensities of the order of $I \simeq 10^{10}$ W cm^{-2} it follows $\mu_\mathrm{n} \simeq 10^3$ and $\mu_\mathrm{p} \simeq 1$. Figure 6 represents the probability of the transition for a ns laser, at the exact resonance ($\nu = 0$) and at $\nu = 4$. As a function of the laser electric field amplitude, the probability is a rapidly oscillating function and the distance between two consecutive peaks is $\Delta\mu/\mu \simeq$ 0.5%. It is reasonable to assume that the instabilities in the laser intensity would average these oscillations to

$$P_{21} \simeq \frac{1}{2\cosh^2(2\mu\ \mathrm{Im}\,\Gamma)}. \tag{4.3}$$

The average excitation probability plotted against μ is presented in figure 7, for different values of the detuning ν. This is a slowly varying function of μ, but the averaging procedure is meaningful only when μ is very large, as was the case with the ns laser. When a ps laser ($\mu \simeq 1$) is used for the excitation, the transition probability is not a rapidly oscillating function of μ (figure 8), and the distance between the consequent

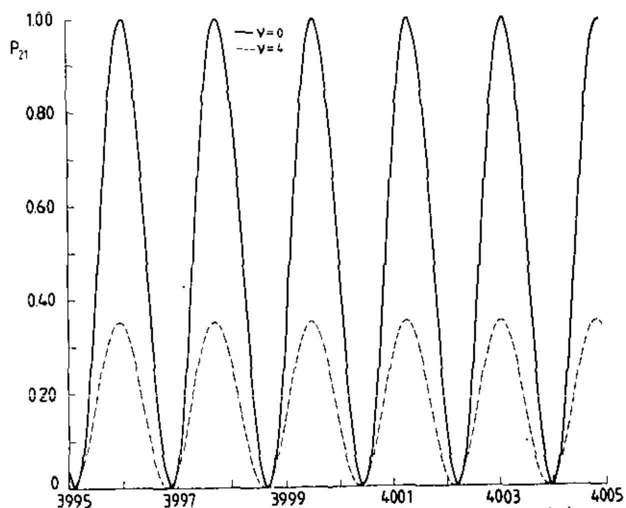

Figure 6. Transition probability plotted against μ for a ns laser of intensity of the order of 10^{10} W cm^{-2}.

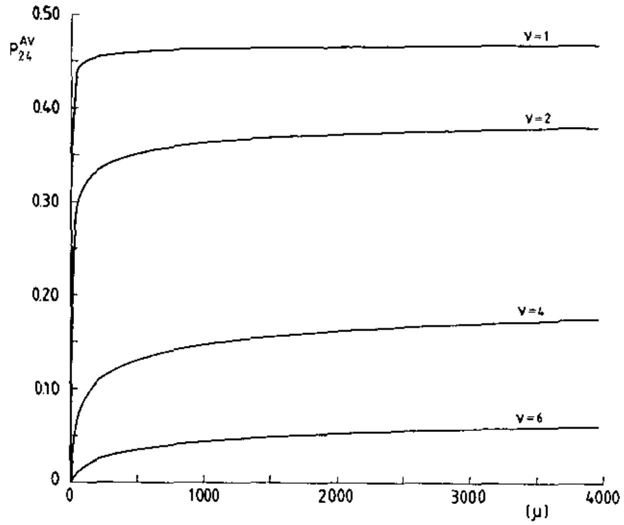

Figure 7. The averaged transition probability plotted against μ.

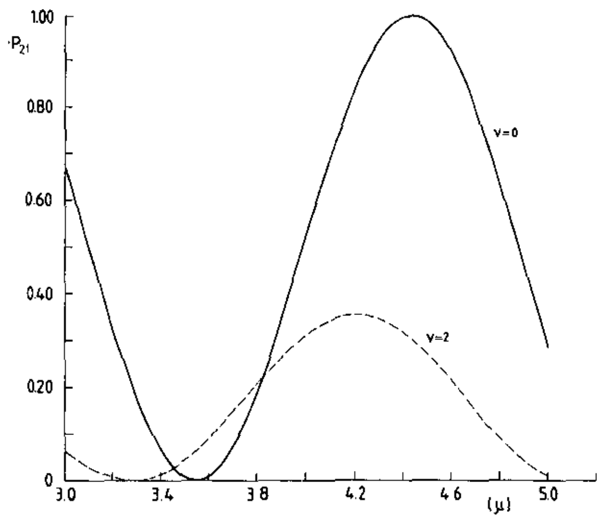

Figure 8. Transition probability plotted against μ for a ps laser of intensity of the order 10^{10} W cm^{-2}.

peaks is $\Delta\mu/\mu \simeq 25\%$. Therefore, in order to obtain maximum excitation efficiency, the laser intensity should be chosen carefully.

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