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A. Turrin: CROSSING OF OPTICAL RESONANCES BY  
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ABSTRACT:

A model form is developed for an optical pulse having arbitrary envelope area and quasilinearly chirped frequency around the resonance crossing-point. It bridges the gap between the nearadiabatic and nonadiabatic limits in the dynamics of a two-level system.

INTRODUCTION

The basic and also the simplest formulation of a nonperturbative theory on population inversion produced in a two-level system by adiabatically chirping a laser field through the resonance frequency is embodied in the Landau and Zener (LZ) model<sup>1-5</sup>. In this model, a constant-intensity, linearly chirped radiation is assumed. The resulting probability that a two-level system which was in the ground state at time  $t = -\infty$  is in the excited state at  $t = +\infty$  is given by

$$(g g_{t=+\infty}^*)_{LZ} = 1 - \exp(-(\pi/2) \varrho_0), \quad (1a)$$

where

$$\varrho_0 = \omega_0^2 / \dot{A}_0. \quad (1b)$$

In eq. (1b),  $\omega_0 = p \mathcal{E} / \hbar$  is the (constant) Rabi flopping frequency, i. e.  $p$  is the dipole matrix element between the lower and the upper state and  $\mathcal{E}$  is the envelope of the oscillating, linearly polarized electric field.

$$\Delta_0(t) = \dot{\Delta}_0 t \quad (1c)$$

is the (linearly varying) detuning, i. e. the angular frequency offset between the laser frequency and the resonance frequency of the two-level system.

Remarkably, the LZ model leads to a formulation having only one independent parameter ( $\varrho_0$ ).

Conversely, a recent theoretical investigation<sup>6</sup> on the feasibility of nonadiabatic population inversion by the DeVoe and Brewer laser-frequency-switching technique<sup>7</sup> yields to a transition probability which is a function of two parameters, namely the pulse area

$$A = \int_{-\infty}^{+\infty} \omega(t) dt \quad (2a)$$

and the free-precession angle

$$P = 2 \int_0^{+\infty} \Delta(t) dt \quad (2b)$$

(as in refs. 1-5, the detuning  $\Delta(t)$  is assumed to be zero at  $t=0$ ).

Clearly the two models above represent physical situations that are basically different, the former being applicable in the near-adiabatic limit only; the latter describing, on the contrary, a typical nonadiabatic transition.

One would therefore like to have a more complete theory, viz., to add a new, intermediate model which neither assumes  $P$  to be finite, nor assumes  $\omega(t)$  to be constant, i. e. a model which depends upon the parameters

$$\varrho = \omega^2(0) / \dot{\Delta}(0) \quad (2c)$$

and  $A$ . Thus one could choose among three models, in any specific case. The present letter addresses itself to this task.

#### THE INTERMEDIATE MODEL.

The optical pulse we are going to consider may be modeled quite generally, by assuming  $\omega = \omega(t)$  to be an even, positive, arbitrary function of time, and such that  $\omega(\pm\infty)=0$ ; for  $\Delta$ , we write

$$\Delta = \Delta(t) = ((A/\pi)/\varrho) \omega \tan((\pi/A)\sigma), \quad (3a)$$

where  $\sigma = \int_0^t \omega dt \quad (3b)$

( $\omega(\pm\infty) = \pm A/2$ ). To give an example, the evolution of  $\Delta(t)$  as given by eqs. (3a, b) is shown explicitly in Fig. 1 for the special case where a gaussian profile for  $\omega(t)$  is adopted.

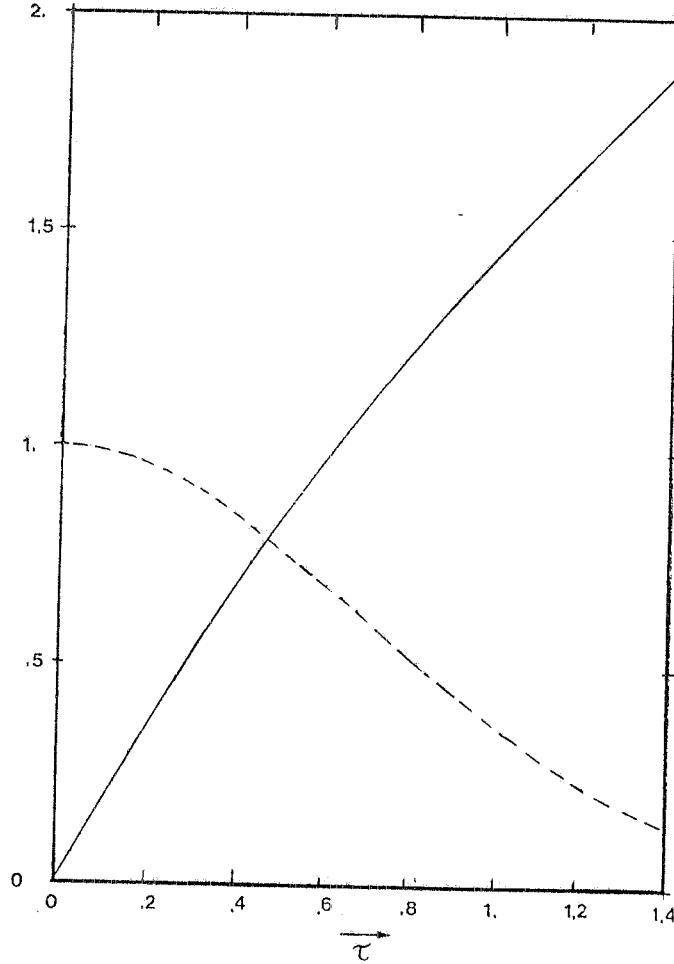


FIG. 1 - Dotted line:  $\exp(-\tau^2)$ , which represents the evolution of a gaussian optical pulse. Solid line:  $\exp(-\tau^2) \tan((\pi/2) \operatorname{erf}(\tau))$ , which, by eqs. (3a, b) of the text, exhibits the features of the (corresponding) sweeping program of the laser frequency through the resonance line.  $\tau = \sqrt{2 \ln 2} t/t_p$ ;  $t_p$  is the pulse width (FWHM).

ANALYSIS. In the rotating-wave approximation, the time-dependent

Schrödinger equation for the two-level atom interacting with the optical field leads to the pair of equations

$$i\dot{e} = (\omega/2)g \exp(-i \int_0^t \Delta dt) \quad (4a)$$

$$i\dot{g} = (\omega/2)e \exp(i \int_0^t \Delta dt), \quad (4b)$$

where  $g$  and  $e$  are the occupation numbers of the two states.

Decay terms are neglected. We set for the initial-values conditions

$$g=0, \quad e=1 \quad \text{at} \quad t=-\infty. \quad (5a, b)$$

The coupled differential equations (4a, b) can readily be separated by differentiation. Thus, we have the equation for g:

$$\ddot{g} - (i\Delta + \dot{\omega}/\omega)\dot{g} + (\omega/2)^2 g = 0. \quad (6)$$

Now, we insert the model  $\Delta(t)$  of eqs. (3a, b) in eq. (6) and introduce the new independent variable

$$x = (1/2) \left[ 1 + \sin((\pi/A)\sigma) \right]. \quad (7)$$

We get

$$x(1-x)g'' + \left[ (1+i\varepsilon/\varrho)/2 - (1+i\varepsilon/\varrho)x \right] g' + (\varepsilon/4)g = 0, \quad (8)$$

where  $\varepsilon = (A/\pi)^2$  and  $' \equiv d/dx$ ,  $'' \equiv d^2/dx^2$ . (9)

As time goes from  $-\infty$  to  $+\infty$ ,  $x$  spans the interval  $0 \leftarrow x \rightarrow 1$ . Eq. (8) is the hypergeometric differential equation<sup>8</sup>

$$x(1-x)g'' + \left[ c - (1+a+b)x \right] g' - abg = 0 \quad (10)$$

with the three constants  $a, b, c$ ,

$$2a = i\varepsilon/\varrho + \sqrt{\varepsilon - (\varepsilon/\varrho)^2} \quad (11a)$$

$$2b = i\varepsilon/\varrho - \sqrt{\varepsilon - (\varepsilon/\varrho)^2} \quad (11b)$$

$$2c = 1 + i\varepsilon/\varrho. \quad (11c)$$

Thus, the general solution of eq. (8) can be written in the form

$$g = C \cdot F(a, b; c; x) + Dx^{1-c} F(a-c+1, b-c+1; 2-c; x), \quad (12)$$

where  $C$  and  $D$  are integration constants and the  $F$ 's are hypergeometric functions (eqs. (15.5.3) and 15.5.4) of ref. 8).

In order that the boundary conditions expressed by Eqs. (5a, b) be satisfied we must set  $C = 0$ . To determine  $D$ , we reconsider eq. (4b) which, in terms of the new independent variable  $x$ , transforms into the equation

$$i2(\pi/A) \sqrt{x(1-x)} g' = e \exp(i \int_0^t \Delta dt). \quad (13)$$

This gives, in the limit  $t \rightarrow -\infty$ , i. e.  $x \rightarrow 0$ ,

$$g'g'^* = (1/4) \varepsilon/x. \quad (14)$$

Differentiating Eq. (12) (with  $C=0$ ) we have

$$g' = D(1-c)x^{-c} F(a-c+1, b-c+1; 1-c; x) \quad (15)$$

(use of the differentiation formula given by eq. (15.2.4) of ref. 8 has been made); combining eq. (14) and (15), we obtain, in the limit  $x \rightarrow 0$ ,

$$DD^* = \varepsilon / \left[ 1 + (\varepsilon/\varrho)^2 \right] . \quad (16)$$

Now, in the limit  $t \rightarrow +\infty$ , we find

$$gg_t^* \xrightarrow{t \rightarrow +\infty} DD^* \left| \frac{\Gamma(2-c)\Gamma(c-a-b)}{\{\Gamma(1-a)\Gamma(1-b)\}} \right|^2 \quad (17)$$

where the  $\Gamma$ 's are gamma functions.

In deriving this expression for the transition probability we have made use of eq. (15.1.20) of ref. 8.

Finally, we develop the squared modulus entering in eq. (17) and use eq. (16). We get for the desired transition probability

$$gg_{t=\infty}^* = 1 - \cos^2 \left[ (A/2) \sqrt{1 - ((A/\pi)/\varrho)^2} \right] \operatorname{sech}^2 \left[ (\pi/2)(A/\pi)^2/\varrho \right] \quad (18a)$$

for  $A/\pi \leq \varrho$  ;

$$gg_{t=\infty}^* = 1 - \cosh^2 \left[ (A/2) \sqrt{((A/\pi)/\varrho)^2 - 1} \right] \operatorname{sech}^2 \left[ (\pi/2)(A/\pi)^2/\varrho \right] \quad (18b)$$

for  $A/\pi \geq \varrho$  .

RESULTS. As mentioned in the introduction, the "natural" parameters which are used in the LZ, intermediate and nonadiabatic models are:

$$\begin{aligned} LZ^{(1-5)} & : \varrho \\ \text{intermediate} & : \varrho \text{ and } A \\ \text{nonadiabatic}^{(6)} & : A \text{ and } P. \end{aligned}$$

Each of these models has its own domain of validity.

Now, by looking first at eq. (18a), we see that it reduces precisely to the Rabi solution,  $\sin^2(A/2)$ , at  $\varrho = \infty$ . Likewise, the nonadiabatic model<sup>6</sup> tends towards the same special solution as  $P \rightarrow 0$  (eqs. (16, a, b) and (17) of ref. 6 with  $\alpha = 0$ ).

On eq. (18b) we note that it reduces precisely to eq. (1) for  $A$  large, whatever the pulse duration is (this can be very quickly derived by retaining only the zero-th and first order terms in the binomial expansion of the  $\sqrt{1-(\rho/(A/\pi))^2}$  expression entering in eq. (18b)). This is a general result, whose derivation does not involve the use of any particular model like the LZ model.

We have found it convenient to describe the response of the two-level system in a diagram (Fig. 2) which, e. g., for a given  $A/\pi$  value, gives the  $1/\rho$  values required to obtain inversion.

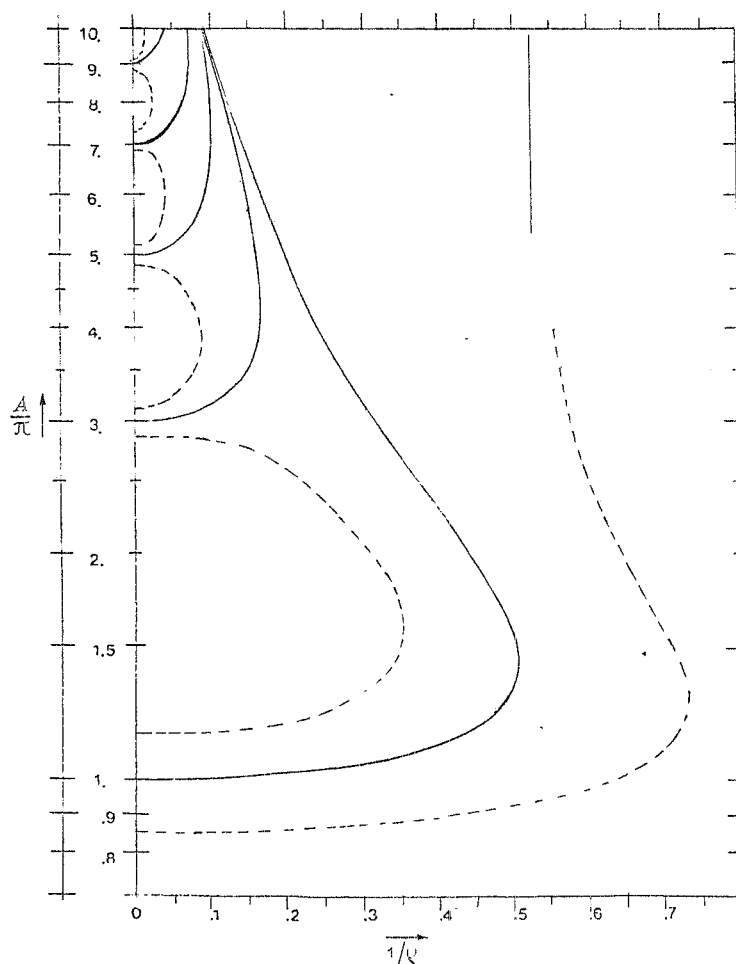


FIG. 2 - Diagram showing the response of the two-level system to a chirped optical pulse. The solid curves are the loci of complete population-inversion occurrence ( $gg_t^* \rightarrow +\infty = 1$ ). The loci where  $gg_t^* \rightarrow +\infty = 0.95$  are represented by the dotted lines. The vertical straight line at the top of the diagram represents the asymptotic value ( $(A/\pi) \rightarrow \infty$ ) for the  $gg_t^* \rightarrow +\infty = 0.95$  curve.

Inspection of Fig. 2 readily reveals that once the optical-pulse parameters are chosen properly, efficient population inversion can be achieved, and that no critical values of these parameters are involved anyway.

In conclusion, both the present and the nonadiabatic model<sup>6</sup> indicate that if the radiation is chirped, pulses with  $A/\pi > \sim 1$  have approximately the same potential to operate with high performance as do constant-intensity disturbances.

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