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A. Turrin: NONADIABATIC POPULATION INVERSION BY LASER FREQUENCY SWITCHING.

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ABSTRACT. -

Efficient nonadiabatic population inversion in two-level systems is predicted by theory in the case where a pulsed laser driving field of width ~ 85 psec (FWHM) is frequency shifted ~10 GHz through the resonance line with a rise time ~ 65 psec.

Chirped laser pulses are at present an indispensible part of the facilities for obtaining population inversion in two-level systems in order to observe coherent transient phenomena in optically excited samples.

While the Stark-switching method [1] is still used [2] for adiabatic population inversion purposes, it seems safe to assume that the future belongs to the laser-frequency-switching technique [3].

Very recently, considerable progress has been made in this field, leading to the extension of these techniques [3] to a 100-psec scale [4].

This opens up, as we will see, new possibilities for achieving almost complete inversion in an irradiated sample.

In the present letter we study in detail the problem of optimising the characteristic pulse parameters in order to achieve the maximum population inversion in a two-level system, initially in the ground state, subjected to a very short and rapidly chirped optical pulse.

DeVoe and Brewer reported [4] that they are able to shift the frequency of their laser light in the range 0 to 10 GHz with a rise time $T \sim 100$ psec. This means that the extent of region of time where a near-resonant pulse can be effective for population inversion equals T, approximately. On the other hand, it is well known that in order for the inversion process to be adiabatic, the chirped pulse must satisfy the requirements [5]

$$\omega(t) << \Delta(t) \qquad \text{at} \quad |t| > \sim T/2 \tag{1a}$$

and

$$\left[\omega^2(t) + \Delta^2(t)\right]^{1/2} T/2 \gg 2\pi \quad \text{at} \quad -T/2 < t < +T/2 \tag{1b}$$

(from here downwards the assumption is made that the resonance crossing ($\Delta=0$) occurs at $t=0$).

Here, $\omega(t)=p\,\mathcal{E}(t)/\hbar$ is the Rabi flopping frequency (p is the dipole matrix element between the lower and the upper state; $\mathcal{E}(t)$ is the envelope of the oscillating, linearly polarized electric field) and $\Delta(t)$ is the detuning, i. e. the angular frequency offset between the laser frequency and the resonance frequency of the two-level system.

Condition (1b) requires that at the resonance crossing $\omega(0) >> 120$ GHz. One is then dealing with a very high input intensity which has a temporal profile centered at $t=0$ and which must drop $\sim$ one order of magnitude in ~ 50 psec ($\sim$FWHM). Thus it is necessary to abandon the adiabatic conditions (1, a, b) and focus our attention on the response to a near-resonant radiation which induces a small precession frequency throughout the pulse. A sketch of the Bloch-vector motion in the case where such a nonadiabatic population inversion occurs is given in Fig. 4 of a recent communication by Tsukada [6].

As a model that exhibits rapid sweeping of the pulse carrier frequency through the atomic resonance frequency, let $\omega = \omega(t)$ to be an even (i. e. $\omega(t) \equiv \omega(-t)$), positive, arbitrary function of time, going to zero at $t \rightarrow \pm\infty$, and

$$\Delta = \Delta(t) = \alpha\,\omega\,\tanh\varphi \quad , \quad \text{where} \quad 2\varphi = \int_0^t \omega\, dt \quad . \tag{2a,b}$$

Here, α is assumed to be a constant parameter.

Integration of Eq. (2a) over all positive time leads to

$$P/4 \;=\; \alpha \ln \cosh (A/4), \tag{3a}$$

where

$$P \equiv 2 \int_0^{\infty} \Delta\, dt \qquad \text{and} \qquad A \equiv \int_{-\infty}^{+\infty} \omega\, dt \tag{3b,c}$$

are recognized to be the "free-precession" angle and the pulse envelope "area" [7]. Graphs of $\Delta(t)$ ($\equiv -\Delta(-t)$) are shown in Fig. 1 for two values of α in the case where a gaussian temporal profile for ω is adopted. These graphs will be examined later to assess "a posteriori" the adequacy of our model.

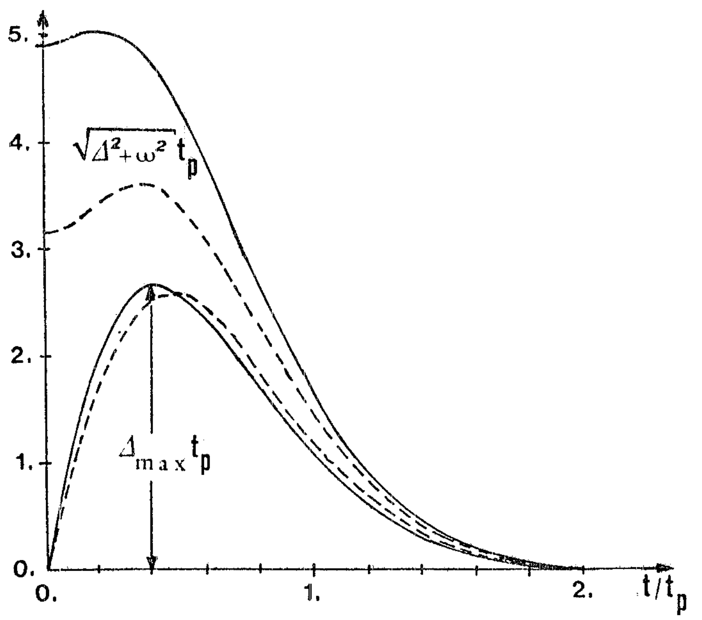

FIG. 1 - Plots of $\Delta(t)$ and $(\Delta^2(t) + \omega^2(t))^{1/2}$ for the case where a gaussian pulse of width t_p (FWHM) is assumed. Solid and dotted lines correspond respectively to the points indicated by the symbols ● and x of Fig. 2.

Summarizing so far, by the model above the manner in which the near-resonant driving pulse interacts with the two-level system can be completely characterized by P and A. Thus we have a very general description of the pulse with a minimal set of assumptions. Lastly, we note that by eq. (3a) only one parameter has been introduced.

Once the model above is adopted, the object is to solve the conventional coupled-equations of motion

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

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

(which can be derived by considering the time-dependent Schrödinger equation for the two-level system interacting with the optical field) written, as done above, in rotating-wave approximation. Decay terms are neglected.

In the following, the corresponding transition probability $gg^*_{t\to+\infty}$ in the long-time limit for the two-level system initially in the ground state

$$g_{t\to-\infty}=0 \quad , \quad e_{t\to-\infty}=1 \tag{5a,b}$$

will be calculated by inserting eqs. (2a, b) in (4a, b) and solving these equations.

The first step consists of decoupling the motion equations. We have, e. g., for g,

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

Eq. (6) is easily transformed into the equation

$$x^2(1-x)g'' + x\left[(1+i\alpha)-(1-i\alpha)x\right] g' + (1-x)/4\, g = 0 \tag{7}$$

by the transformation

$$x = -\exp(2\varphi) \qquad (' \equiv d/dx\, ;'' \equiv d^2/dx^2) \quad . \tag{8}$$

On introduction of the new function y by the substitution [8]

$$g = x^\lambda\, y, \tag{9}$$

Eq. (7) becomes [8]

$$x(1-x)y'' + \left[c-(a+b+1)x\right] y' - aby = 0\,. \tag{10}$$

The constants λ, c, a and b are:

$$2\lambda_{\pm} = -i\alpha \pm ir\ , \quad c_{\pm} = 1 \pm ir, \quad r = (1+\alpha^2)^{1/2}\ , \tag{11a,b,c}$$

$$a_+ = -i\alpha + ir\ , \quad b_+ = -i\alpha\ , \tag{11d,e}$$

$$a_- = -i\alpha\ , \quad b_- = -i\alpha - ir\ . \tag{11f,g}$$

Eq. (10) is the well known hypergeometric differential equation [9]. Thus, the general solution of eq. (7) in the vicinity of x=-0 can be written [10] as follows:

$$g = C(-x)^{-i\alpha/2+ir/2} F(-i\alpha+ir, -i\alpha\ ; 1+ir;\ x) + \\ + D(-x)^{-i\alpha/2-ir/2} F(-i\alpha-ir, -i\alpha\ ;\ 1-ir;\ x). \tag{12}$$

Here, C and D are integration constants and the F's are hypergeometric functions [9] (Note that $F(a,b;c;x) \equiv F(b,a;c;x)$).

We are interested in C and D in the limit $t \longrightarrow -\infty$, which corresponds to $x = -\exp(-A/2)$. Thus, for $A \gtrsim 3\pi/2$, we may replace in eq. (12) the F functions by unity. One gets in the limit $t \longrightarrow -\infty$

$$g_{t \longrightarrow -\infty} \approxeq C \exp(-irA/4) + D \exp(irA/4) = 0. \tag{13}$$

Here, the boundary condition expressed by Eq. (5a) has been imposed

To derive a second relation between C and D it is necessary to reconsider Eq. (4b), which, in terms of the new independent variable x, transforms into the equation

$$2xg' = -ie \exp\left(i \int_0^t \Delta\, dt\right). \tag{14}$$

For $A \gtrsim 3\pi/2$, and in the limit $t \longrightarrow -\infty$ where the boundary condition (5b) must be satisfied, Eq. (14) becomes

$$(-\alpha + r)\, C \exp(-irA/4) - (\alpha + r)\, D \exp(irA/4) \cong 1\ . \tag{15}$$

In deriving eq. (15) use has been made of the differentiation formula [11] for the F functions which gives, in our approximation ($A \gtrsim 3\pi/2$), $xF'(a,b;c;x) \approx 0(x)$. In addition, we have dropped in Eq. (15) a common phase-factor $\exp(i\psi)$ which comes in, since the transition probability gg^* will be independent of factors like these .

It is now straightforward to obtain C and D from Eqs. (13) and (15) and insert their resulting expressions in eq. (12). Once this substitution has been made, the asymptotic form for g as $t \rightarrow +\infty$, i. e. $x=-\exp(A/2)$ (with $A \gtrsim 3\pi/2$), can be found by making use of the appropriate linear transformation formula [12)] for the $F(a,b\ ;c;\ x \rightarrow -\infty)$ functions appearing in Eq. (12). Replacing in the resulting linearly-transformed [12)] form of $g_{t \rightarrow +\infty}$ the $F(a,b;c;1/x)$ functions by unity and performing a fair amount of elementary and Γ-algebra, one finds

$$r\, g_{t \rightarrow +\infty} \cong -S + \alpha\, r\, \mathrm{Re}\,(G) - ir^2\, \mathrm{Im}\,(G), \qquad (16)$$

where

$$S = \sinh(\alpha\pi)/\sinh(r\pi) \qquad (16a)$$

and

$$G = \exp(irA/2)\,\Gamma^2(ir)\left[\Gamma(i\alpha+ir)\ \Gamma(-i\alpha+ir)\right]^{-1}. \qquad (16b)$$

Here, the Γ's are gamma functions, and a further incoming common phase-factor has been suppressed in Eq.(16).

Developing the squared modulus $gg^*_{t \rightarrow +\infty}$, we can derive from it an equation for α in the form $\alpha = f(\alpha)$

$$\alpha = (r/2)\left[-gg^*_{t \rightarrow +\infty} + (S/r)^2 + \alpha^2\, \mathrm{Re}^2(G) + r^2\, \mathrm{Im}^2(G)\right]\left[\mathrm{Re}(G)S\right]^{-1}, \qquad (17)$$

where $gg^*_{t \rightarrow +\infty}$ plays the role of a parameter.

The problem of finding the roots of Eq. (17) has been solved by computer using a zero finding routine and a complex gamma function routine. The results are shown in Fig.2, where we have plotted the "pulse-performance" curves for various values of $gg^*_{t \rightarrow +\infty}$.

Fig. 2 indicates that the laser-frequency-switching technique makes available region in the P, A plane (i. e. $1.5 \lesssim A/\pi \lessapprox 2.6$ and $1.2 \lesssim P/\pi \lessapprox 1.6$) in which almost complete population inversion can be obtained. This means

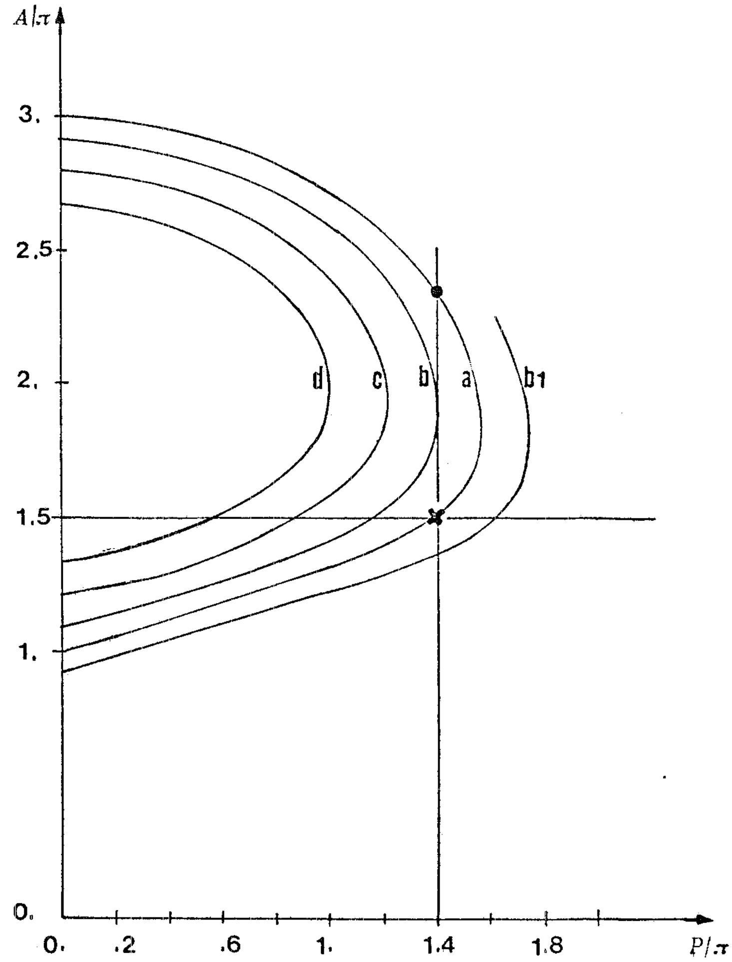

Fig. 2 - Diagram showing the A and P values required to obtain almost complete population inversion. Curves labeled by a; b; c; d; b1 correspond to $gg^{*}_{t \to +\infty}$ = 1.; 0.98; 0.90; 0.75; 0.98 respectively. A possible operating line at P/π = 1.4 is indicated. For $A/\pi < 1.5$ our approximation fails.

that with P fixed (consider, e.g., the operating vertical line at $P = 1.4\ \pi$) inversion remains still insensitive even to wide variations of the light intensity due to the bell-shaped transverse spatial profile of the excitation pulse.

Specializing now to the case of a gaussian temporal profile of the excitation optical pulse of width t_p ($\equiv$ FWHM)

$$\omega = \omega_o \exp\left[-(2\ln 2)(t/t_p)^2\right], \tag{18}$$

we will find that $\Delta(t)$, given by Eqs. (2a, b), is a very weakly dependent function of α when P is fixed and A is varied (as supposed above) throughout the range of interest. From Eqs. (18), (2a, b) and (3c) we obtain

$$\omega t_p = (2\pi \ln 2)^{1/2} (A/\pi) \exp\left[-(2\ln 2)(t/t_p)^2\right] \tag{19a}$$

and

$$\Delta(t) t_p = \alpha \omega t_p \tanh\left[(A/4)\,\mathrm{erf}((2\ln 2)^{1/2}(t/t_p))\right]. \tag{19b}$$

Graphs of $\Delta(t)t_p$ for two extreme values of α at $P=1.4\pi$ (points marked by the • and × signs in Fig. (2)) are shown in Fig. 1, which also includes the $(\omega^2+\Delta^2)^{1/2} t_p$ curves corresponding to these • and × points. In calculating the $\Delta(t)t_p$ function throughout the range bounded by the • and × points, profiles very nearly equal to those of Fig. 1 have resulted. This demonstrates the suitability of our model and constitutes therefore the central result of our letter.

In order to convey a feeling for the characteristics of an optimal pulse, we choose conditions that are representative of those actually occurring in the laser-frequency-switching applications [4]. Thus the frequency shift is chosen as $2\,\Delta_{max}/(2\pi) = 10$ GHz. From Fig. 1 ($P=1.4\,\pi$) one gets $\Delta_{max} t_p = 2.6$ and, for the rise-time T, $(T/2)/t_p = 0.4$, which correspond to $t_p = 83$ psec and $T = 66$ psec. By inspection of the graphs of $(\omega^2+\Delta^2)^{1/2} t_p$ and with the parameters above we obtain, in the whole range bounded by the • and × points of Fig. 2, 17 GHz $< (\omega^2 + \Delta^2)^{1/2} <$ 60 GHz for $-t_p < t < t_p$, a figure comfortably greater than, e. g., the width $1/T_2^* \approx 10$ GHz of the inhomogeneously broadened spectrum of gaseous absorbers. Thus, no complications due to T_2^* effects are predicted, even for atoms that are excited less strongly by nonuniform radial portions of the laser beam.

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