Inducing complete population oscillations in systems with externally induced dipole moments

Duje Bonacci
Physical Chemistry Department, R. Bošković Institute, Bijenička 54, 10000 Zagreb, Croatia
(Dated: April 1, 2022)

A two level system is considered which has no static dipole moment, e.g. molecule $H_2$ in its ground electronic state. If strong enough external field is applied, it will dynamically distort such a system and supply it with time (and field) dependent dipole moment. Although it is impossible to do so in the undistorted system which has no coupling to the dipole component of the external field, having induced in it a dipole moment, the rotational and vibrational dynamics of such system can be manipulated using lasers. In this work, a system is considered in which the external perturbation dynamically induces the transition dipole moment between only two distinct levels. The aim of the work is to show how the driving pulse of the following form:

$$F[t] = F_0 m[t] \cos [\omega [t] \ t]$$  \hspace{1cm} (1)

can be analytically designed, that will produce Rabi-like complete population oscillations between the two levels.

PACS numbers: 3.65.Sq

*Electronic address: dbonacci@irb.hr
I. CALCULATION

A. Dynamical equation

Consider a two level system for which in unperturbed state no spectroscopically allowed dipole transition exists between its two levels (e.g. molecule $H_2$). Such system cannot be directly manipulated using the standard methods of laser control.

However, if such a system is exposed to sufficiently intensive external perturbation, time-dependent dipole transition moment between its two states can be dynamically induced. This dipole transition moment can then be used to manipulate the system’s internal (rotational, vibrational, ... ) dynamics.

It can be demonstrated [1] that the dynamics of such two-level system subjected to strong coherent external radiation is governed by the following equation:

$$\frac{d}{dt} \left( \begin{array}{c} c_\alpha[t] \\ c_\beta[t] \end{array} \right) = \frac{dF[t]}{dt} \left( \begin{array}{cc} \mu_{\alpha\alpha}[t] & \omega_{\alpha\beta}[t] \\ \omega_{\alpha\beta}[t] & \mu_{\beta\beta}[t] \end{array} \right) \left( \begin{array}{c} e^{i\omega_{\alpha\beta}[t]t} - \frac{m[t]}{\omega_{\alpha\beta}[t]} \cos \omega_{\alpha\beta}[t]t \\ i \frac{m[t]}{\omega_{\alpha\beta}[t]} \sin \omega_{\alpha\beta}[t]t \end{array} \right) \left( \begin{array}{c} c_\alpha[t] \\ c_\beta[t] \end{array} \right)$$

(2)

In this equation: $c_{\alpha,\beta}[t]$ are the wave-function expansion coefficients; $F[t]$ is the external field, $\mu_{ij}[t] \equiv \langle i|\hat{\mu}|j \rangle$ is the field-induced dipole moment of state or dipole transition moment ($\hat{\mu}$ is the dipole moment operator); $\omega_{\alpha\beta} \equiv |E_\alpha - E_\beta|$ is the absolute value of the instantaneous transition frequency between the two states of the system; and $s_{\alpha\beta} \equiv \text{Sign}[E_\alpha - E_\beta]$ is the sign of that transition frequency.

B. The external field

The external field is constructed so as to fit the following analytic form:

$$F[t] = F_0 \ m[t] \ \cos [\omega[t] \ t]$$

(3)

Here, $F_0$ is the maximum amplitude of the field achieved throughout the pulse, $m[t]$ is the pulse envelope ($0 \leq m[t] \leq 1$) and $\omega[t]$ is the time-dependent frequency of the external perturbation. All of these three parameters are freely tunable.

It should be noted that such ‘analytic’ form needs not be fully analytic - pulse envelope and pulse chirp can be purely numerical functions. Also, such seemingly rigid form of the pulse puts no serious restriction on the possible variety of actual pulse shape. Indeed, by varying the three pulse parameters, any reasonable pulse shape can be constructed. The only important caveat is that pulse must be such that at any time the field varies much more rapidly than the envelope. If this is granted, it straightforwardly follows that:

$$\frac{dF[t]}{dt} \approx -F_0 \ m[t] \ \omega[t] \ \sin [\omega[t] \ t]$$

$$= i \ F_0 \ m[t] \ \omega[t] \ \frac{e^{i\omega[t]t} - e^{-i\omega[t]t}}{2}$$

(4)

C. Transforming the time variable

Next, for further analytic calculation convenience, the following transformations and shorthands are introduced:

$$d\tau \equiv F_0 \ m[t] \ \omega[t] \ \frac{\mu_{\alpha\beta}[t]}{\omega_{\alpha\beta}[t]} \ dt$$

$$\Delta_{\pm}[\tau] \equiv \omega[t(\tau)] \pm \omega_{\alpha\beta}[t(\tau)]$$

$$f_i[\tau] \equiv 2 \frac{\mu_{ii}[t]}{\mu_{\alpha\beta}[t]} \ \omega_{\alpha\beta}[t(\tau)] \ \sin[\omega[t(\tau)] \ t(\tau)] \ : \ (i = \alpha, \beta)$$

(7)

(8)

With Eq. (4) included, in terms of these quantities the dynamical equation Eq. (2) becomes:
Introducing transformation (11) into (10), the following equation is obtained:

\[
\frac{d}{dt} \begin{pmatrix} a_\alpha[\tau] \\ a_\beta[\tau] \end{pmatrix} = -i \begin{pmatrix} -f_\alpha[\tau] & s_{\alpha\beta}[t[\tau]](e^{i s_{\alpha\beta}[t]\Delta_-[t]t} - e^{-i s_{\alpha\beta}[t]\Delta_+[t]t}) \\ s_{\alpha\beta}[t[\tau]](e^{-i s_{\alpha\beta}[t]\Delta_-[t]t} - e^{i s_{\alpha\beta}[t]\Delta_+[t]t}) & -f_\beta[\tau] \end{pmatrix} \begin{pmatrix} a_\alpha[\tau] \\ a_\beta[\tau] \end{pmatrix}
\]  

(9)

Now the final unitary transformation is sought:

\[
\begin{pmatrix} b_\alpha(\tau) \\ b_\beta(\tau) \end{pmatrix} = e^{-i\hat{A}(\tau)} \begin{pmatrix} a_\alpha(\tau) \\ a_\beta(\tau) \end{pmatrix}
\]  

(11)

with:

\[
\hat{A}[\tau] = \begin{pmatrix} \rho_1[\tau] & 0 \\ 0 & \rho_2[\tau] \end{pmatrix}
\]  

(12)

where \(\rho_1[\tau]\) and \(\rho_2[\tau]\) are freely adjustable functions, such that the final transformed system vector satisfies:

\[
\frac{d}{d\tau} \begin{pmatrix} b_\alpha[\tau] \\ b_\beta[\tau] \end{pmatrix} = \pm i \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} b_\alpha[\tau] \\ b_\beta[\tau] \end{pmatrix}
\]  

(13)

As Eq. (13) is identical to the standard form of Rabi oscillations equation in a two-level system (for details, see e.g. [3]), the corresponding solution would represent complete population transfer oscillations between the two levels. Introducing transformation (11) into (10), the following equation is obtained:

\[
\frac{d}{dt} \begin{pmatrix} b_\alpha[\tau] \\ b_\beta[\tau] \end{pmatrix} = -i \begin{pmatrix} -f_\alpha[\tau] + \frac{d}{d\tau}\rho_1[\tau] & s_{\alpha\beta}[t[\tau]] e^{-i(s_{\alpha\beta}[t]\Delta_-[\tau]t + (\rho_1[\tau] - \rho_2[\tau])) t[\tau]} \\ s_{\alpha\beta}[t[\tau]] e^{i(s_{\alpha\beta}[t]\Delta_-[\tau]t - (\rho_1[\tau] - \rho_2[\tau])) t[\tau]} & -f_\beta[\tau] + \frac{d}{d\tau}\rho_2[\tau] \end{pmatrix} \begin{pmatrix} b_\alpha[\tau] \\ b_\beta[\tau] \end{pmatrix}
\]  

(14)

If this is to be fitted to form (13), the following conditions must be fulfilled:

\[
\frac{d}{d\tau}\rho_1(\tau) = f_\alpha[\tau],
\]

(15)

\[
\frac{d}{d\tau}\rho_2(\tau) = f_\beta[\tau],
\]

(16)

\[
s_{\alpha\beta}[t]\Delta_-[\tau]t[\tau] + (\rho_1[\tau] - \rho_2[\tau]) = 0,
\]

(17)

which can be compactly written as:

\[
\frac{d}{d\tau}(\Delta_-[\tau]t[\tau]) = -s_{\alpha\beta}[t](f_\alpha[\tau] - f_\beta[\tau])
\]  

(18)

D. Introduction of RWA and Rabi oscillations condition

Under certain conditions (see e.g. [2]) whose validity can be checked retrospectively, rotating wave approximation can be introduced to simplify the obtained full dynamical equation of the system, Eq. (9). If RWA is valid, then the dynamical significance of the rapidly rotating elements in the dynamical matrix (the ones containing \(\Delta_+[\tau]\)) is negligible, and these elements can be dropped from all further calculations. Hence, approximate dynamical equation is obtained:

\[
\frac{d}{dt} \begin{pmatrix} a_\alpha[\tau] \\ a_\beta[\tau] \end{pmatrix} = -i \begin{pmatrix} -f_\alpha[\tau] & s_{\alpha\beta}[t[\tau]](e^{i s_{\alpha\beta}[t]\Delta_-[t]t} - e^{-i s_{\alpha\beta}[t]\Delta_+[t]t}) \\ s_{\alpha\beta}[t[\tau]](e^{-i s_{\alpha\beta}[t]\Delta_-[t]t} - e^{i s_{\alpha\beta}[t]\Delta_+[t]t}) & -f_\beta[\tau] \end{pmatrix} \begin{pmatrix} a_\alpha[\tau] \\ a_\beta[\tau] \end{pmatrix}
\]  

(10)
E. Analyticaly optimized frequency chirp

Integrating Eq. (18) and reverting to the original time coordinate $t$ yields the recurrent formal solution for the optimized driving frequency $\omega[t]$:

$$\omega[t] = \omega_{\alpha\beta}[t] - s_{\alpha\beta}[t] \frac{E_0}{t} \int_{t_0}^{t} (\mu_{\alpha\alpha}[t_1] - \mu_{\beta\beta}[t_1]) \ m[t_1] \ \omega[t_1] \ t_1 \ \sin[\omega[t_1] \ t_1] \ dt_1$$  \hspace{1cm} (19)

As this is a recurrent equation, it does not provide directly the optimized frequency. However, with numerical computational power nowadays available it should be a fairly simple and quick task to obtain the solution using some a rather simple computer iteration scheme.

Hence, using this solution, fully controlled and complete population oscillations can be induced in the strongly perturbed system.

Acknowledgment

I am very grateful to Prof. Gabriel Balint-Kurti for insightful discussion and sharing of some ideas from his own work. These provided both indispensable sparks and firm guidelines in the development of the results presented herein.

[1] G.G. Balint-Kurti, private communication (June 2004).
[2] D. Bonacci, Rabi spectra - a simple tool for analyzing the limitations of RWA in modelling of the selective population transfer in many-level quantum systems, quant-ph/0309126.
[3] W. Demtroeder, Laser spectroscopy: basic concepts and instrumentation, 1st Edition, Springer-Verlag, Berlin, 1988.