Validity of the Rosen-Zener conjecture for Gaussian-modulated pulses

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Using the Magnus approximation we show that the Rosen-Zener conjecture holds for a two-level system in near-resonant interaction with a Gaussian-modulated pulse of sufficiently short duration.

Within the rotating-wave approximation[1] (RWA), the equation of motion for the interaction-picture state amplitudes of a two-level system dipole interacting with an oscillating field $E^0 f(t)\cos\omega t$, arbitrarily directed along the z axis, is

$$i\frac{d}{dt}\begin{pmatrix} b_1(t) \\ b_2(t)\end{pmatrix} = -\tfrac{1}{2}\mu E^0 f(t)\begin{pmatrix} 0 & e^{-i\nu t} \\ e^{i\nu t} & 0\end{pmatrix}\begin{pmatrix} b_1(t) \\ b_2(t)\end{pmatrix} , \quad (1)$$

where $\mu = \langle 1|\mu_z|2\rangle = \langle 2|\mu_z|1\rangle$ is the electric dipole transition matrix element coupling the states $|1\rangle$ and $|2\rangle$, which are of opposite parity and have a level separation ω_{21}. In (1), $\nu=\omega_{21}-\omega$ is the detuning frequency, where ω is the carrier frequency of the oscillating field; the field, which is of amplitude E^0, is modulated by a time-varying pulse envelope $f(t)$.

Uncoupling (1) gives the second-order differential equation

$$\ddot{b}_1 + [-\dot{f}(t)/f(t) + i\nu]\dot{b}_1 + (\tfrac{1}{2}\mu E^0)^2 f^2(t) b_1(t) = 0 , \quad (2)$$

with a similar equation for $b_2(t)$. Under the transformation $z(t) = \frac{1}{2}\mu E^0 \int_{-\infty}^{t} dt' f(t')$, (2) becomes

$$(b_1'' + b_1)(\tfrac{1}{2}\mu E^0) f(t) + i\nu b_1' = 0 . \quad (3)$$

On resonance $\nu = 0$ and, in this case, the general solution of (3) is

$$b_1(t) = C\cos[z(t)] + D\sin[z(t)] , \quad (4)$$

where C and D are arbitrary constants to be fixed by the initial conditions.

Equation (4) is the solution to (2) for any pulse envelope $f(t)$ when $\nu=0$. If $b_1(-\infty)=1$ and $\dot{b}_1(-\infty)=0$, then it follows from (4) that the induced transition probability for excitation from $|1\rangle$ to $|2\rangle$, i.e., $P_2(t,-\infty) = 1-|b_1(t)|^2$, is given by

$$P_2(t,-\infty) = \sin^2\left(\tfrac{1}{2}\mu E^0 \int_{-\infty}^{t} dt' f(t')\right) \quad (5)$$

for any modulating envelope $f(t)$. Clearly, $P_2(t,-\infty)$ is dependent upon the nature of $f(t)$. The steady-state transition probability is

$$P_2(+\infty,-\infty) = \sin^2 A , \quad (6)$$

where A is the so-called pulse "area" defined by

$$A = \tfrac{1}{2}\mu E^0 \int_{-\infty}^{+\infty} dt' f(t') , \quad (7)$$

assuming $f(t)$ to be an even function of t. Thus, irrespective of the nature of $f(t)$, the steady-state value of the induced transition probability is the same for all pulses of equal area interacting on-resonance with a two-level system within the RWA.

It is more difficult to solve (2) when $\nu \neq 0$. For a rectangular pulse with a modulating envelope given by

$$f(t) = \begin{cases} 1 & \text{for } |t| \le \frac{1}{2}\tau \\ 0 & \text{for } |t| > \frac{1}{2}\tau \end{cases} \quad (8)$$

where τ is the pulse duration, (2) is a differential equation with constant coefficients whose general solution is

$$b_1(t) = e^{-i\nu t}(C\cos\{\tfrac{1}{2}t[\nu^2+(\mu E^0)^2]^{1/2}\} + D\sin\{\tfrac{1}{2}t[\nu^2+(\mu E^0)^2]^{1/2}\}) , \quad (9)$$

where C and D are arbitrary constants to be fixed by the initial conditions. If $b_1(-\frac{1}{2}\tau)=1$ and $\dot{b}_1(-\frac{1}{2}\tau)=0$, then it follows from (9) that the induced transition probability $P_2(t,-\frac{1}{2}\tau)$ is given by

$$P_2(t,-\tfrac{1}{2}\tau) = \frac{(\mu E^0)^2}{\nu^2+(\mu E^0)^2} \times \sin^2\{\tfrac{1}{2}(t+\tfrac{1}{2}\tau)[\nu^2+(\mu E^0)^2]^{1/2}\} , \quad (10)$$

the so-called Rabi formula.[1,2] The steady-state transition probability is $P_2(\frac{1}{2}\tau,-\frac{1}{2}\tau)$. If τ is arbitrarily large, so that the oscillating field executes many optical cycles throughout the duration of the pulse, then the steady-state transition probability is given by

$$P_2(+\infty,-\infty) = \frac{(\mu E^0)^2}{\nu^2+(\mu E^0)^2}\lim_{\tau\to\infty}\frac{1}{\tau}\int_0^{\tau} d\tau' \sin^2\{\tfrac{1}{2}\tau'[\nu^2+(\mu E^0)^2]^{1/2}\} = \frac{1}{2}\,\frac{(\mu E^0)^2}{\nu^2+(\mu E^0)^2} . \quad (11)$$

The rectangular pulse is an idealization since a physically realizable pulse cannot be switched on and off instantaneously as suggested by (8). A laboratory pulse will have an envelope varying smoothly with time and is switched on $[f(-\infty)=0]$ and off $[f(+\infty)=0]$ so that the pulse has an epochal existence $[f(t)\to 1$ as $|t|\to 0]$ of finite duration. The modulating envelope $f(t)=\text{sech}(\pi t/\tau)$, $|t|<\infty$ is "bell shaped," and like the rectangular pulse in (8), has an area $A=\frac{1}{2}\mu E^0\tau$, where τ is a constant characteristic of the pulse duration. Rosen and Zener[3] have shown that for this pulse (2) is exactly soluble. For the initial conditions $b_1(-\infty)=1$ and $\dot{b}_1(-\infty)=0$, they report the asymptotic value of P_2 as

$$P_2(+\infty,-\infty)=\frac{\sin^2 A}{A^2}|A\,\text{sech}(\tfrac{1}{2}\nu\tau)|^2\,, \tag{12}$$

and recognizing that

$$A\,\text{sech}(\tfrac{1}{2}\nu\tau)=A\mathfrak{F}\left[\frac{1}{\tau}\,\text{sech}\left(\frac{\pi t}{\tau}\right)\right],$$

where

$$\mathfrak{F}[f(t)]=\int_{-\infty}^{\infty}dt'e^{-i\nu t'}f(t') \tag{13}$$

is the Fourier transform of $f(t)$, Rosen and Zener further conjectured that for an arbitrary envelope $f(t)$, of area $A=\frac{1}{2}\mu E^0\tau$,

$$P_2(+\infty,-\infty)=\frac{\sin^2 A}{A^2}\left|A\mathfrak{F}\left(\frac{1}{\tau}f(t)\right)\right|^2 \tag{14}$$

is approximately true for a pulse which is near resonant with the two-level system. The basis for this suggestion is twofold: firstly, if $\nu=0$, then using (7) and (13) in (14) gives (6); and secondly, if A is small, so that $\sin A\simeq A$, then (14) reduces to

$$P_2(+\infty,-\infty)=\left|A\mathfrak{F}\left(\frac{1}{\tau}f(t)\right)\right|^2, \tag{15}$$

precisely the result given in first-order perturbation theory using the RWA.

With increasing use of pulsed light sources in physical applications it is important to establish the validity of the Rosen-Zener formula (14) for arbitrary pulse envelopes. To do this it is necessary to adopt a different approach to the solution of (2) than that employed by these authors since their approach is applicable (albeit exactly) only to the hyperbolic secant shaped pulse and to a class of envelopes asymmetric in time.[4]

The Magnus[5] unitary approximate solution to (1) is given by

$$\underline{b}(t)=\exp[\underline{M}(t,-\infty)]\underline{b}(-\infty)\,, \tag{16}$$

where $\underline{M}(t,-\infty)=-\underline{M}^{\dagger}(t,-\infty)$ and $\underline{b}^T(t)=[b_1(t)b_2(t)]$. With

$$\underline{M}(t,-\infty)=\sum_{k=1}^{\infty}\underline{M}^{(k)}(t,-\infty)\,,$$

$$\underline{M}^{(1)}(t,-\infty)=-i\int_{-\infty}^{t}dt'\,\underline{H}(t')\,, \tag{17a}$$

$$\underline{M}^{(2)}(t,-\infty)=\frac{1}{2}\int_{-\infty}^{t}dt'\int_{-\infty}^{t'}dt''[\underline{H}(t'),\underline{H}(t'')]\,, \tag{17b}$$

and $\underline{M}^{(k)}(t,-\infty)$ is generally a sum of k-fold integrals of k-fold–nested commutators of $\underline{H}(t)$, the 2×2 coefficient matrix in (1). For the Gaussian envelope

$$f(t)=\exp(-\pi t^2/\tau^2),\quad |t|<\infty \tag{18}$$

having the same area $A=\frac{1}{2}\mu E^0\tau$ as the rectangular and hyperbolic secant pulses,

$$\underline{M}^{(1)}(+\infty,-\infty)=iA\exp\left(\frac{-\nu^2\tau^2}{4\pi}\right)\begin{bmatrix}0&1\\1&0\end{bmatrix} \tag{19}$$

and

$$\underline{M}^{(2)}(+\infty,-\infty)=-\frac{1}{2}A^2\exp\left(\frac{-\nu^2\tau^2}{2\pi}\right)\text{erf}\left(\frac{i\nu\tau}{(2\pi)^{1/2}}\right)\begin{bmatrix}1&0\\0&-1\end{bmatrix}, \tag{20}$$

respectively. Thus the second-order Magnus approximation gives

$$\underline{b}(+\infty)=\exp\begin{bmatrix}\alpha&i\beta\\i\beta&-\alpha\end{bmatrix}\underline{b}(-\infty)\,, \tag{21}$$

where $\alpha=-\frac{1}{2}\beta^2\text{erf}[i\nu\tau/(2\pi)^{1/2}]$ and $\beta=A\times\exp(-\nu^2\tau^2/4\pi)$, as the asymptotic solution to (1). For the initial conditions $b_1(-\infty)=1$ and $b_2(-\infty)=0$, (21) gives, for $P_2(+\infty,-\infty)=|b_2(+\infty)|^2$,

$$P_2(+\infty,-\infty)=A^2\exp\left(\frac{-\nu^2\tau^2}{2\pi}\right)\left|\sum_{l=0}^{\infty}\frac{(\alpha^2-\beta^2)^l}{(2l+1)!}\right|^2. \tag{22}$$

If $\nu\simeq 0$ and τ is sufficiently small, say, $\nu^2\tau^2\ll 2\pi$, $\alpha^2-\beta^2\simeq -A^2$ which, when used in (22), gives

$$P_2(+\infty,-\infty)=\exp(-\nu^2\tau^2/2\pi)\sin^2 A\,. \tag{23}$$

This is in agreement with the Rosen-Zener conjecture (14), since

$$\mathfrak{F}\left(\frac{1}{\tau}f(t)\right)=\exp\left(\frac{-\nu^2\tau^2}{4\pi}\right)$$

for $f(t)$ defined in (18). Both theoretical[6] and experimental[7] evidence suggest that, ideally, the field outputted from a perfectly mode- and phase-locked pulsed laser consists of a train of Gaussian-amplitude sinusoids. For a two-level system, (23) represents the asymptotic value of the induced transition probability for excitation to level $|2\rangle$ under the assumption that the Gaussian-modulated pulse, of sufficiently short duration τ, is near resonant with the system. Skinner[8] has numerically confirmed the validity of (23) for sufficiently small ν.

In comparing (12) and (23) with (11) we note that for the cw case the two-level system saturates at on-resonance frequencies or as the coupling strength μE^0 becomes sufficiently large. In contrast, for amplitude-modulated fields complete population inversion may occur, such as for a resonant $\pi/2$ pulse, since the two-level system undergoes induced absorption only over the epochal duration of the pulse.

It is 50 years since Rosen and Zener presented their analysis in this journal within the context of a two-level atom interacting with a magnetic field. The essence of their treatment is the transformation of (2) into the hypergeometric equation. More recently, Robiscoe[9]—recognizing the relevance of their results to the problem of a two-level atom interacting with an oscillating field within the RWA—has shown how to generalize the Rosen-Zener approach to the case of decaying states and has given a comprehensive comparison between the rectangular and hyperbolic secant pulses. In a further generalization of the Rosen-Zener problem, Bambini and Berman[4] have shown that there is an entire class of modulating envelope functions that may be mapped into the hypergeometric equation; of this class only the hyperbolic secant is symmetric in time. In contrast to the rectangular and hyperbolic secant pulses, this class of asymmetric envelopes is such that at off-resonance frequencies one cannot define a value of the pulse area for which the transition probability vanishes. Direct numerical integration[10] for the transition probabilities, without invoking the RWA, indicates an oscillatory behavior in P_2 as a function of A at both on- and off-resonance frequencies for Gaussian-modulated pulses, and only in the case of ultrashort pulses does P_2 fail to vanish throughout the pulse duration. Robinson,[11] in an investigation of the findings of Bambini and Berman, introduced a transformation of (2) which Robiscoe[12] used in conjunction with the Jeffreys-Wentzel-Kramers-Brillouin approximation to estimate the transition amplitudes in the asymptotic region for an arbitrary smoothly varying pulse envelope, although the important question of establishing the validity of the Rosen-Zener formula (14) for Gaussian pulses was not addressed. Using the Magnus approach we find the conditions of validity are that the field be near resonant with the two-level system and that it be of sufficiently short duration.

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