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Two-level transition probabilities for asymmetric coupling pulses

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In a recent paper, Bambini and Berman [A. Bambini and P. R. Berman, Phys. Rev. A 23, 2496 (1981)] presented analytic solutions to a certain family of coherent-coupling pulses for a two-level system. They show, for nonresonant temporally asymmetric members of the class, that there are no solutions corresponding to vanishing transition probabilities. In this Comment, we examine the problem in greater generality and demonstrate that this property is the norm for asymmetric pulses, and that a vanishing transition probability is possible only if severely overdetermined conditions are satisfied.

The problem of a two-level system coupled by an external field has a long history in physics, dating back to the 1930's.[1,2] Originally motivated by investigations on atoms in magnetic fields, theories of such systems have more recently been applied to laser-related problems.[3]

Let a_1,a_2 be the amplitudes of the two states. We assume that the coupling potential connecting the two states is of variable amplitude and central frequency Ω, so that, in the rotating wave approximation, the time-dependent Schrödinger equation becomes a pair of coupled equations for a_1,a_2:

$$i\dot{a}_1=V(t)e^{i\Delta t}a_2 \,, \tag{1a}$$

$$i\dot{a}_2=V(t)e^{-i\Delta t}a_1 \,. \tag{1b}$$

Here Δ is the detuning of Ω from the atomic frequency. We work in a system of units where $\hbar=1$.

For the case where V is a constant in time, the solution for initial conditions $a_1=0$, $a_2=1$ at $t=0$ is

$$a_1=\frac{-iV}{(\Delta^2/4+V^2)^{1/2}}e^{i\Delta t/2}\sin[(\Delta^2/4+V^2)^{1/2}t] \,.$$

This is the Rabi problem. For this to be relevant, the approximation that the rise time of the field is much shorter than other characteristic times should be a good one. In their paper, Rosen and Zener[2] considered a case where this sudden approximation was not valid. They were motivated by a serious discrepancy between results of the sudden-approximation theory and experiment. They analyzed the effect of a smoothly varying pulse, choosing a hyperbolic secant because of the exactly solvable nature of the equations that result from such a time dependence. For the hyperbolic secant pulse, one may make a change of variable that transforms the equation of motion into the hypergeometric equation. Robiscoe[4] has shown how to generalize this to the case of decaying states.

Recently, Bambini and Berman[5] have gone beyond the Rosen-Zener problem. They show that there is an entire class of envelope functions that may be mapped into the hypergeometric equation, of which the hyperbolic secant pulse is merely one member. All $V(t)$ in the family, other than the hyperbolic secant, are asymmetric in time, i.e., $V(t)\neq V(-t)$. Bambini and Berman show that for these asymmetric pulses, there is no case, apart from exact resonance, where there is a nonvanishing transition probability, a striking and surprising result.

In the case of the Rabi problem, on the other hand, for any given detuning, there are always values of the pulse area for which the amplitude a_1 returns to zero. In the Rosen-Zener case, the amplitude $a_1(+\infty)$ goes like $(\sin A)/A$, where A is the pulse area, so that here too, once the hyperbolic secant envelope function is specified, one can find values of the area of the pulse for which $a_1(+\infty)$ vanishes. Similar remarks hold for other symmetric potentials, where solutions have been obtained with computers.[6,7] It is a most remarkable feature of the Bambini-Berman problem that it admits no asymmetric envelopes for $\Delta \neq 0$ with a nonvanishing transition probability. That is, it asserts that for asymmetric pulses of the form studied, if the amplitudes $a_2 = 1$ and $a_1 = 0$ at $t = -\infty$, then at time $t = +\infty$, the probability for finding the system in state 1 is nonvanishing, i.e., there will always be some population in state 1 for this class of off-resonant asymmetric pulse. No previous prediction of this kind of behavior seems extant in the literature. It should be understood that only envelopes of a single algebraic sign are being considered, so that, for example, pulses that are completely antisymmetric in time are excluded from this discussion.

Bambini and Berman[5] reach their conclusion by obtaining a complete analytic solution to their problem. Since most pulse shapes do not admit of closed form solutions, it is of interest to inquire whether the nonvanishing of transtion probabilities holds for other smooth, asymmetric pulses and whether this property can be demonstrated in a general way, i.e., through the structure of the equations of motion. It is to this question that we address the present work.

Equations (1) may be put in the form of uncoupled second-order equations

$$\ddot{a}_1 - (\dot{V}/V + i\Delta)\dot{a}_1 + V^2 a_1 = 0 , \quad (2a)$$

$$\ddot{a}_2 - (\dot{V}/V - i\Delta)\dot{a}_2 + V^2 a_2 = 0 . \quad (2b)$$

Defining $z = \int_{-\infty}^{t} f(t')dt' - \frac{1}{2}$, with $A = \int_{-\infty}^{\infty} V\,dt$ and $f = V/A$, Eqs. (2) become, in the z plane,

$$a_1'' - i\frac{\Delta}{f}a_1' + A^2 a_1 = 0 , \quad (3a)$$

$$a_2'' + i\frac{\Delta}{f}a_2' + A^2 a_2 = 0 . \quad (3b)$$

We assume, with Bambini and Berman,[5] that $f(t)$ does not change sign, so that the transformation, which differs from theirs, is single valued. If one transforms Eq. (3a) via the substitution

$$b = a_1 \exp\left[(-i\Delta/2)\int_0^z dz'/f(z')\right] = a_1 e^{-i\Delta t/2} ,$$

into an equation with the first derivative missing, we have

$$b'' + \left[\frac{\Delta^2}{4f^2} + \frac{i\Delta f'}{2f^2} + A^2\right] b = 0 , \quad (3a')$$

or

$$-b'' + \left[\frac{-\Delta^2}{4f^2} - \frac{i\Delta f'}{2f^2}\right] b = A^2 b . \quad (3b')$$

Equation (3b′) resembles a one-dimensional, time-independent Schrödinger equation for a particle of mass $\frac{1}{2}$ moving in the complex "potential"

$$V = -\left[\frac{\Delta^2}{4} + \frac{i\Delta f'}{2}\right]\frac{1}{f^2} ,$$

where $\hbar$ has been set $=1$.

This equation is to be solved subject to the initial conditions that $b = 0$ at $z = -\frac{1}{2}$. If the dynamics of the problem permit a transition probability of zero for certain pulse areas, this means $b(z = +\frac{1}{2})$ also vanishes for those values of A. In short, we must solve an eigenvalue problem and find those values of A^2 for which the solutions of Eq. (3b′) vanish at $z = \pm\frac{1}{2}$. Now, for physical pulses, only real envelopes exist. For these, A^2 is real and positive. If none of the eigenvalues A^2 meet this criterion, A will have an imaginary part for all the eigenfunctions of Eq. (3b′), and none will correspond to a system driven by an actual pulse, i.e., there will be no physically meaningful pulse areas for which the system undergoes a transition probability of zero. In the following, we shall assume a nonvanishing detuning. Note that the case of exact resonance is entirely equivalent to the elementary quantum mechanical problem of a particle in a box, whose eigenvalues A^2 are $n^2\pi^2$. In this way, we confirm the simple result that the transition probability vanishes for pulse areas that are integral multiples of π, if $\Delta = 0$.

We should comment that if one constructs an asymmetric potential from two temporally distinct symmetric pulses, one can, by making each of the component pulses produce a net transition amplitude of zero, cause the overall probability to vanish. To force the components to be exactly nonoverlapping in time requires that they be sharply cut off. Thus, these pulses do not conform to the smoothness criterion of Bambini and Berman.[5]

We consider now pulses where the imaginary term is present. We examine first the case of symmetric pulses. Let A^2 be a typical eigenvalue. If we replace the imaginary term by its negative, then the resulting equation will have A^{2*} for its eigenvalue. Now, since $f(z)$ is symmetric in z, $f'(z)$ will be antisymmetric. Therefore, the transformation $z \to -z$ reverses the sign of the imaginary term on the left-hand side of Eq. (3b′), but leaves the eigenvalue unchanged. Immediately, $A^2 = A^{2*}$, i.e., all the eigenvalues are real, although not necessarily larger than zero. For asymmetric pulses, the transformation $z \to -z$ does not reproduce the complex-conjugate equation, and A^2 will not, in general, be the same as A^{2*}. This does not absolutely rule out the possibility that for particular $f(t)$ and detuning, one might have one or more real and positive eigenvalues, but demonstrates that it could occur only by accident. We shall show in the following that the conditions that must necessarily be fulfilled for A^2 to be real for asymmetric pulses are severely overdetermined.

To proceed, we will analyze the problem from a perturbative viewpoint, and assume that the entire perturbation expansion can be summed. We do not restrict ourselves to the first few terms, but study the parity-related properties of the full series. We take the zero-order problem to be

$$-b_0'' - \left[\frac{\Delta^2}{4f^2}\right] b_0 = A_0^2 b_0 \ . \tag{4}$$

This is Hermitian and identical to a time-independent Schrödinger equation, which has only real eigenvalues. The imaginary term $-i\Delta f'/2f^2$ is to be considered as a perturbation.

We wish to contrast the case of symmetric and asymmetric pulse envelopes. Assume $f(t)$ to be symmetric—$f(z)$ is also symmetric. [If $f(t)$ were not symmetric about $t=0$, $f(z)$ would lack symmetry about its origin.] For this case, the unperturbed eigenfunction b_0 has definite parity, and the perturbation $-i\Delta f'/f^2$ is odd under reflection. It follows directly that if one writes a perturbation series for A^2 as an expansion in the usual way, contributions from odd powers of the "strength" of the "interaction" will be absent. Since only the even orders survive, and the strength parameter is purely imaginary, the resulting eigenvalues will be real.

If the potential $V(t)$ is not symmetric neither $1/f^2$ nor f'/f^2 will be operators of definite parity, nor will unperturbed solutions b_0 possess well-defined inversion properties. Hence, both even and odd terms in the perturbation expansion will be present, and the eigenvalues A^2 will all be complex, unless there is a case where, for a specific detuning, the odd powers of the expansion sum to zero.

The latter is an extremely unlikely circumstance. Equation (3b′) is of the form

$$-b'' - \left[\frac{\mu}{f^2} + i\frac{\lambda f'}{f^2}\right] b = A^2 b \ . \tag{5}$$

We require not only that the odd powers sum to zero, but that they do so for a value of λ that is exactly the square root of μ. We cannot quite exclude this possibility, but it is evidently highly overdetermined.

To summarize, we have shown that the result obtained for particular asymmetric pulses by Bambini and Berman,[5] namely that there are no nonresonant cases for which the transition probability vanishes, is the normal consequence of the general structure of the equations of motion, and applies, apart from some remotely possible accidental cases, to all smoothly varying, asymmetric pulses which possess envelopes of a single algebraic sign.

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[2]N. Rosen and C. Zener, Phys. Rev. 40, 502 (1932).
[3]An extensive compilation of references is given by L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).
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[5]A. Bambini and P. R. Berman, Phys. Rev. A 23, 2496 (1981).
[6]S. Yeh and P. R. Berman (private communication).
[7]E. J. Robinson (unpublished).