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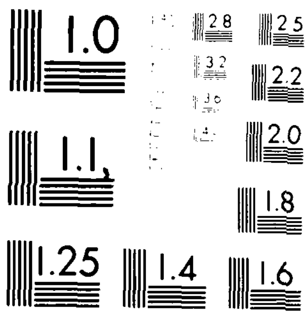

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New Results for Transition Probabilities in Two-Level Systems:

The Large Detuning Regime

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-- SEVERAL NEW EXPERIMENTAL LASER SPECTROSCOPIC TECHNIQUES AND MAY ALSO
-- LEAD TO THE PREDICTION OF NEW OPTICAL EFFECTS.
-- 24 - APPROACH: (U) THE TECHNICAL APPROACH INVOLVES THE STUDY OF TWO BASIC
-- PROBLEM AREAS. FIRST, OPTICAL COLLISIONS AND RADIATIVE COLLISIONS WILL
-- BE STUDIED IN THREE-LEVEL ATOMIC SYSTEMS. NEITHER THE PHYSICAL PROCESSES
-- NOR THE ROLE OF INTERFERENCE EFFECTS IS UNDERSTOOD IN THESE SYSTEMS. THE
-- SECOND PROBLEM AREA INVOLVES THE STUDY OF COLLISIONS IN SATURATION
-- SPECTROSCOPY. IN PARTICULAR, IT IS BELEIVED THAT SATURATION SPECTROSCOPY
-- HAS CONSIDERABLE POTENTIAL FOR DETERMINING EXCITED STATE CROSS SECTIONS
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-- ARTICLE ON LASER-ASSISTED COLLISIONAL PROCESSED HAS BEEN WRITTEN AND
-- SUBMITTED FOR PUBLICATION. THE WORK ON THE EFFECTS OF COLLISIONS ON

-- ATOMIC STATE COHERENCES HAS BEEN USED TO ANALYZE RECENT EXPERIMENTAL
-- WORK AND PROVIDES THE FIRST CLEAR PICTURE OF THESE PROCESSES. THE
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Abstract

The problem of calculating transition probabilities in two-level systems is studied in the limit where the detuning is large compared to the inverse duration of the interaction. Coupling potentials whose Fourier transforms $\tilde{V}(\omega)$ are of the form $f(\omega)e^{-(|\tau\omega|)}$ for large frequencies give rise to solutions which may be classified into families according to the form of ~~$f(\omega)$~~. Within each family, transition probabilities may be calculated from formulae that differ only in the numerical value of a scaling parameter. In cases where the coupling function has a pole in the complex time plane, the families are identified with the order of this singularity. In particular, for poles of first order, a connection with the Rosen-Zener solution can be made.

The analysis is performed via high-order perturbation expansions, which are shown to always converge for two-level systems driven by coupling potentials of finite pulse area.

1. Introduction

In many areas of physics, one encounters problems involving two states of a quantum-mechanical system coupled by a time-dependent potential.[1-10] In the interaction representation, the equations of motion for a_1 and a_2, the probability amplitudes of levels 1 and 2, are of the form

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

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

where ω is the frequency separation of the states and $V(t)$ is the coupling potential. Decay effects are neglected in Eqs. (1) (and throughout this paper) and we work in a system of units in which $\hbar = 1$.

Equations of this type arise in many semiclassical problems. A problem of current interest to which they apply is the coupling of two levels of an atom by a laser pulse that has a temporal width which is small compared to the natural lifetimes of the levels. The pulse, $V(t)$ is of the form

$$V(t) = 2A(t)\cos\Omega t, \tag{2}$$

where Ω is the central frequency of the pulse, and $2A(t)$ is the envelope function of its amplitude. Assuming that $\frac{|\Omega-\omega|}{\Omega+\omega} << 1$, one can recast Eqs. (1) in terms of Δ, the detuning of the pulse from resonance (rotating wave approximation) as

$$i\dot{a}_1 = A(t)\,e^{i\Delta t}\,a_2, \qquad (3a)$$

$$i\dot{a}_2 = A(t)\,e^{-i\Delta t}\,a_1. \qquad (3b)$$

Eqs. (3) or (1) are deceptively simple in form, and one might, at first glance, believe that the system must be completely understood, so that nothing remains to be investigated about the equations or their solution. Actually, there is very little known about the overall qualitative nature of the solutions to Eqs. (3) for arbitrary A(t). Apart from any intrinsic interest one might have in the dynamics of two-level systems, such information could be useful, for example, in applications where one wishes to choose the pulse shape to maximize the excitation probability for a given detuning Δ.

To appreciate that our assertion concerning the lack of knowledge about the behavior of systems described by Eqs. ([illegible]) is valid, one need only recognize that the answer to the following question is not known in general. "Starting with initial conditions $a_1(-\infty) = 1$, $a_2(-\infty) = 0$, how does the probability amplitude $a_2(t)$ depend qualitatively on the pulse area S, defined by

$$S = \int_{-\infty}^{\infty} A(t)\,dt,$$

on the detuning, and on the shape of the envelope function A(t)?"

A response to this query can be made for a limited number of cases. Analytic solutions are available if A(t) belongs to a class of functions[1], (including the hyperbolic secant of Rosen and Zener[2,3]) mappable into the hypergeometric equation, or if $A(t) = (\text{constant}) \exp(-\alpha|t|)$[9,10] or if $A(t)$ is a step function (Rabi problem), or if the detuning is zero.* In addition, there are approximate solutions available in adiabatic[4] or perturbative limits. Yet, there remains a wide range of parameters and pulse shapes for which an answer to the basic question cannot be provided.

In this paper, we shall examine the solutions to Eqs. (3) in the limit where the product of the detuning $|\Delta|$ and the characteristic pulse duration τ has a magnitude greatly in excess of unity. In other words, we are assuming that the pulse does not possess the appropriate Fourier components to significantly compensate for the detuning. In consequence, the transition probability $|a_2(\infty)|^2$ will always be very small (but still great enough to be experimentally measurable in atomic vapors of densities $\sim 10^{12}$ atoms/cm^3). We note that numerical solutions of Eq. (3) in this detuning range may be possible but are very costly in computer time and plagued with technical difficulties.

For the case $|\Delta\tau| \gg 1$, we shall establish the following results: (1) low-order perturbative approximations for $a_2(\infty)$ are not valid for arbitrary pulse area θ, despite the fact that $|a_2(t)|^2 \ll 1$ for all time. (2) an iterative solution to Eqs. (1) always converges for well-behaved envelope functions. (3) Asymptotic solutions for $a_2(t)$, t finite, may be easily found, but expressions for $a_2(\infty)$ are difficult to obtain. (4) Asymptotic solutions for $a_2(\infty)$ can be obtained for a limited class of

*Kaplan[7] has also considered cases where the detuning varies as prescribed functions of the amplitude, and obtained closed-form expressions.

pulse envelope functions using contour integration techniques. This is a broader set than that for which exact solutions are known. (5) The asymptotic dependence of $a_2(\infty)$ depends critically on the nature of the singularities of the pulse envelope function $A(t)$, analytically continued into the complex plane. (6) If two pulse functions have the same Fourier transforms in the limit of large frequencies and if the dominant dependence of the transform is an exponential decay in the frequency, then the asymptotic forms of the solutions $a_2(\infty)$ for these functions in the limit of large $|\Delta|$ are simply related. In this paper, we address points (1), (2), (3), and (6); methods for actually obtaining asymptotic solutions (points (4) and (5)) will be discussed in a future article.

II. Asymptotic solutions.

As we have indicated, the Rosen-Zener[2,3] (hyperbolic secant coupling pulse) problem is one of the few for which exact solutions are known. In this case, a simple expression gives the transition amplitude as a function of detuning and area for all values of these parameters. Naturally, since this formula

$$a_2(\infty) = -i\sqrt{2\pi}\,\tilde{V}(\Delta)\,\frac{\sin S}{S}, \tag{4}$$

where $\tilde{V}$ is the Fourier transform of $A(t)$, is exact, it is valid in the special case of the asymptotic limit.

We shall show that there is an entire class of pulses for which the asymptotic transition amplitude, as a function of S and Δ, may be written down by inspection, once the Rosen-Zener problem has been solved. We shall also demonstrate that there are other classes of pulses whose solutions to these are unrelated to Rosen-Zener, but are connected to each other in the sense that once one has been solved, the solutions for the entire class may be obtained by inspection.

The existence of these related solutions will be established via term-by-term comparison of n^{th} order perturbation expansions which, under very general conditions, are convergent in two-level problems (see Appendix). With suitable scaling of the coupling strengths, the series for different members of particular classes will be seen to be identical, in the limit of large detunings.

The particular potentials analyzed in this paper are $V(t)$ whose Fourier transforms for large ω assume the form $g(\omega)\exp(-|b\omega|)$, where g is a slowly varying function of ω, and b a constant. It is convenient to make a variable change, such that $\nu = |b|\omega$ and $x = t/|b|$. Consequently, the exponential decay factor in the Fourier transform becomes $\exp(-|\nu|)$ and the equations of motion transform to

$$i\dot{a}_1 = \beta f(x) e^{i\alpha x} a_2, \qquad (3a')$$

$$i\dot{a}_2 = \beta f(x) e^{-i\alpha x} a_1, \qquad (3b')$$

where $\alpha = |b\Delta|$ and where the dot now signifies differentiation with respect to x. β, previously designated as θ, is the pulse area. The reduced potential function $f(x)$ is defined such that $\int_{-\infty}^{\infty} f(x)dx = 1$. The pulse area is invariant under the indicated change of variable. One may also write Eqs. ([illegible]) as a pair of uncoupled second-order equations

$$\ddot{a}_1 - \left(\frac{\dot{f}}{f} + i\alpha\right)\dot{a}_1 + \beta^2 f^2 a_1 = 0, \tag{5a}$$

$$\ddot{a}_2 - \left(\frac{\dot{f}}{f} - i\alpha\right)\dot{a}_2 + \beta^2 f^2 a_2 = 0. \tag{5b}$$

There are two aspects to the solution of Eqs. ([illegible]) or ([illegible]). These are the calculation of the amplitudes at finite and infinite times, respectively. The former are of interest if the transients are to be used as input to other problems, such as multiphoton ionization[illegible], while the latter, with which we are mainly concerned here, are the transition amplitudes, $a_2(\infty)$. The two temporal regimes differ greatly in the methods that must be used to perform accurate calculations.

Apart from the trivial problem, the problem which has attracted the most study is that of Rosen and Zener[illegible], $f(t) = (\mathrm{sech}\, \pi t/T)/T$, for which the solutions are

$$a_1 = {}_2F_1(a, b, c, z), \tag{6a}$$

$$a_2 = -iKz^{1-c^*}\,{}_2F_1(a-c^*+1,\ b-c^*+1,\ 2-c^*,\ z), \tag{6b}$$

or

$$a_2 = -iKz^{1-c^*}(1-z)^{c^*-a-b}\,{}_2F_1(1-a, 1-b, 2-c^*, z), \tag{6b'}$$

where $a = -b = \dfrac{\beta}{\pi}, \quad c = \dfrac{1}{2} - \dfrac{i\alpha}{4\pi},$

$$z = \frac{-\tanh\frac{\pi x}{2} + 1}{2}, \quad K = \frac{\beta}{4\pi\left(\frac{1}{2} - \frac{i\alpha}{4\pi}\right)},$$

and ${}_2F_1$ designates the hypergeometric function. The form of a_2 given by Eq. ([illegible]) is valid for all x, while that given by Eq. ([illegible]') holds only for finite x, unless [illegible] corresponds to [illegible] a pulse area for which $a_2(+\infty)$ vanishes. We recall that [illegible] the transition amplitude for the Rosen-Zener problem is given by Eq. ([illegible]).

One may write the solution to Eq. ([illegible]) in perturbation series in the usual fashion, noting that only [illegible] terms contribute to the expression for a_1, while only odd [illegible] in the [illegible] for a_2. The expression for $a_2(+\infty)$ is

$$a_2 = -i \sum_{k=0}^{\infty} a_2^{(2k+1)} \beta^{2k+1} (-1)^k, \quad \text{where}$$

$$a_2^{(2k+1)} = \int_{-\infty}^{\infty} A(x_1) e^{-i\alpha x_1} dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} A(x_j) e^{i(-1)^j \alpha x_j} dx_j .$$

In the Appendix, it is shown that this series converges for all finite pulse areas.

For the remainder of the paper we will restrict ourselves to the case of pulses that are symmetric in time and where $|\alpha| \gg 1$ --

the adiabatic or asymptotic limit. The [illegible] order term will be symmetric in x. We shall [illegible] the finite and infinite time solutions of the [illegible] problem, which exemplify relevant properties of [illegible].

We may obtain the finite [illegible] the Γ_1 function of Eq. ([illegible])

$$a_2 = -\frac{i\beta}{4\pi\left(\frac{1}{\alpha}-\frac{i\alpha}{4\pi}\right)}\, e^{-i\alpha x} \operatorname{sech}\frac{\pi x}{2}\Big[\,1 + \frac{\left(1+\frac{\beta}{\pi}\right)\left(1-\frac{\beta}{\pi}\right)}{3-\frac{i\alpha}{2\pi}}\left(-\tanh\frac{\pi x}{2}+1\right)+\cdots\Big].$$

For large α, it is sufficient to retain [illegible]

$$a_2 \simeq \frac{\beta}{\alpha}\, e^{-i\alpha x} \operatorname{sech}\frac{\pi x}{2}.$$

This is equivalent to first-order perturbation theory in the adiabatic limit

$$a_2^{(1)} = -i\int_{-\infty}^{x} V(x')\, e^{-i\alpha x'}\, dx' \simeq \frac{V(x)}{\alpha}\, e^{-i\alpha x},$$

where subsequent parts integrations are neglected, since they are $O(\frac{1}{\alpha^n})$, $\alpha > 1$. We immediately see that this sequence of parts integrations is unsuitable for calculating $a_2(\infty)$, since each term separately vanishes when $x \to \infty$. Even including the third- and higher-order terms in the perturbation [illegible] via analogous sequences of parts integral [illegible] not [illegible] are [illegible] [illegible]. [illegible] quently, other [illegible] are necessary [illegible] for $a_2(t)$.

It [illegible] from [illegible] paragraph that for [illegible] enough α, first-order perturbation theory is a sufficiently accurate approximation for [illegible], provided α is finite. For infinite [illegible], not only [illegible] [illegible] [illegible] [illegible] correction [illegible] $_2$ [illegible] integral may [illegible]. This is [illegible], [illegible] [illegible] Eq. ([illegible]), in which [illegible] first-order [illegible] the first-order theory [illegible] [illegible] [illegible] present [illegible] [illegible] coupling [illegible] [illegible] resonance. Simple first-order theories [illegible] for a rectangular pulse of the same strength. As we shall see, other [illegible] pulses also possess this "saturation memory". In fact, in some cases, a higher-order theory is necessary off resonance even for a case where a first-order theory would suffice at resonance. This is exemplified by the formulae of Eqs. ([illegible]) below.

Since each coupling function $f(t)$ is different, one might be led to believe that separate calculations must be performed for each individual case. Fortunately, as we have stated earlier, there prove to the classes of pulses where, if one knows the functional dependence of the asymptotic transition amplitude on ϵ and λ for one member of the class, one knows it for all members of the class, although the actual time dependence of the potentials may be drastically different. What is significant is that their Fourier transforms approach the same form as $\epsilon \to \infty$.

When Rosen and Zener derived Eq. (4), they suggested that similar formulae might hold for other pulse shapes.[illegible] This conjecture proved not to hold in general. It is manifestly false for asymmetric pulses, nor is it even valid for all symmetric pulses.[illegible] What we shall show is that a kind of Rosen-Zener conjecture does apply at large detunings for pulses in which $f(x)$ has simple poles at $x = i$. This law does not apply to pulses which have higher order poles at this point, although scaling laws for these do exist, different for each order.

The following theorem will be established. Let two coupling pulses $f(x)$ and $f_0(x)$ have Fourier transforms $\tilde{V}(\nu)$ and $\tilde{V}_0(\nu)$. The Fourier transforms of both approach, for large values of the argument, the same asymptotic form $\tilde{V}_a(\nu)$. If $\tilde{V}_a$ is of the form $g(\nu)e^{-|\nu|}$ where $g(\nu)$ is a slowly varying function of ν, then the asymptotic transition amplitudes generated by the two pulses will be the same, provided that the pulse areas are both finite. A sufficient condition for the indicated asymptotic behavior of the Fourier transforms is that they be equal, for large ν, to a contour integration whose value is given by the product of the residue at $x = i$ and the usual Cauchy factor $2\pi i$. If two such pulses are to have the same $\phi(v)$, they must possess poles of the same order at $x = i$.

The contribution of order $(2k+1)$ to the transition amplitude may be rewritten slightly

$$a_2^{(2k+1)} = \int_{-\infty}^{\infty} A(x_1) e^{-i\alpha x_1} dx_1 \prod_{j=2}^{2k+1} \lim_{\lambda_j \to 0} \int_{-\infty}^{x_{j-1}} A(x_j) e^{\{i(-1)^j \alpha + \lambda_j\} x_j} dx_j .$$

The factors $e^{\lambda_j x_j}$ do not affect the integrals. They are used to remove ambiguities at $x_j \to -\infty$ in the argument below, where we express the amplitude in terms of integrals in the frequency domain. The limits $\lambda_j \to 0$ are to be taken before the x_1 integration is performed. Expressing each $A(x_j)$, $j \geq 2$, in terms of its Fourier transform, we have

$$a_2^{(2k+1)} = \frac{1}{(2\pi)^k} \int_{-\infty}^{\infty} A(x_1) e^{-i\alpha x_1} dx_1 \prod_{j=2}^{2k+1} \lim_{\lambda_j \to 0} \int_{-\infty}^{x_{j-1}} dx_j \int_{-\infty}^{\infty} \tilde{V}(\nu_j) e^{i(\nu_j + (-1)^j \alpha - i\lambda_j) x_j} d\nu_j .$$

By working in the frequency domain, we shall be able to examine the structure of the integrals for $a_2^{(2k+1)}$ and establish that the contribution from regions where the asymptotic form of $\tilde{V}$ is not valid is lower by $O(\frac{1}{\alpha})$ than the contributions from regions where it is valid.

The integrals over the x_i are trivial to perform. We obtain

$$a_2^{(2k+1)} = \lim_{\lambda_\ell \to 0} \frac{1}{(2\pi)^{k-\frac{1}{2}}} \int_{-\infty}^{\infty} d\nu_2 \cdots d\nu_{2k+1} \, \tilde{V}\Big(\sum_{j=2}^{2k+1} \nu_j - \alpha\Big)$$

$$\prod_{j=2}^{2k+1} \frac{\tilde{V}(\nu_j)}{\sum_{\ell=2k+3-j}^{2k+1} (\nu_\ell + (-1)^\ell - i\lambda_\ell)} \, .$$

We now proceed to determine the asymptotic form of these amplitudes. The analysis is easiest to follow for the third-order contribution $a_2^{(3)}$, but exactly the same reasoning and conclusions will apply for the higher order terms. (The theorem is true by inspection for first-order, since that contribution is, apart from a constant multiplier, just the [illegible] transform itself.) Thus, if the coupling [illegible] of the same asymptotic form, their [illegible] the same way with [illegible]. The leading [illegible] $a_2^{(3)}$,

$$a_2^{(3)} = \lim_{\lambda \to 0} \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\tilde{V}(\nu_1)\,\tilde{V}(\nu_2)\,\tilde{V}(\nu_1+\nu_2-\alpha)\, d\nu_1\, d\nu_2}{(\nu_1 - \alpha - i\lambda)(\nu_2 + \nu_1 - i\lambda)} \, .$$

It is convenient to make the change of variable $\nu_i = y_i \alpha$.

$$a_2^{(3)} = \lim_{\lambda \to 0} \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \frac{\tilde{V}(\alpha y_1)\,\tilde{V}(\alpha y_2)\,\tilde{V}(\alpha[y_1 + y_2 - 1])\, dy_1\, dy_2}{(y_1 - 1 - i\lambda)(y_1 + y_2 - i\lambda)} =$$

$$\frac{1}{\sqrt{2\pi}}\left\{ P\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\tilde{V}(\alpha y_1)\,\tilde{V}(\alpha y_2)\,\tilde{V}(\alpha[y_1+y_2-1])\,dy_1\,dy_2}{(y_1-1)\,(y_1+y_2)} + i\pi \lim_{\lambda\to 0}\int_{-\infty}^{\infty} dy_2\left[\frac{\tilde{V}(\alpha)\left(\tilde{V}(\alpha y_2)\right)^2}{1+y_2-i\lambda} + \frac{\left(\tilde{V}(\alpha y_2)\right)^2\tilde{V}(\alpha)}{-y_2-1-i\lambda}\right]\right\},$$

where P indicates that the integrand excludes infinitesimal regions near $y_1 = -y_2$ and $y_1 = 1$. We may formally integrate the last two terms. If, (-1) is factored from the second of the two integrals, they combine to become

$$i\pi \lim_{\lambda\to 0}\int_{-\infty}^{\infty} dy_2\,\tilde{V}(\alpha)\left(\tilde{V}(\alpha y_2)\right)^2\left\{\frac{1}{1+y_2-i\lambda} - \frac{1}{1+y_2+i\lambda}\right\}.$$

It is immediately obvious that if these are partitioned according to the rule

$$\lim_{\epsilon\to 0}\int\frac{\phi(x)\,dx}{x-x_0-i\epsilon} = P\int\frac{\phi(x)\,dx}{x-x_0} + i\pi\,\phi(x_0),$$

the principal value contributions exactly cancel, while the $i\pi$ terms are proportional to $e^{-2\alpha}$, and exponentially small compared to $a_2^{(1)}$, which decays only like $e^{-\alpha}$. Terms proportional to exponentials which decay more rapidly than $e^{-\alpha}$ do not contribute to the asymptotic form.

We now proceed to examine the remaining contributions to $a_2^{(2)}$, where it is again understood that the small regions in the neighborhood of $y_1 = -y_2$ and $y_1 = 1$ are excluded from the integrals. For all regions

except where $|y| < \left|\frac{a}{\alpha}\right|$, where a is a number of order unity, $\tilde{V}(\alpha y) \to \tilde{V}_a(\alpha y)$.
Thus, for the entire y_1-y_2 plane, except where $y_1 \sim 0$, $y_2 \sim 0$ (but not both simultaneously) and $y_1 + y_2 \approx 1$, the numerator of the integrand is well represented by its asymptotic form. Furthermore, since at most one of the three Fourier transform factors departs from its asymptotic form in any given region of space, the area in the $y_1 - y_2$ plane over which one of the $\tilde{V}$ both departs from its asymptotic form and decays no more rapidly than $e^{-\alpha}$ is $O(1/\alpha)$. It is, of course implicitly assumed that the exact and asymptotic forms of the Fourier transforms remain bounded as their arguments $\to 0$. For the former, this is equivalent to the requirement, which we have already stated, that β be finite.

Now consider that portion of the y_1-y_2 plane where all factors in the numerator are well-approximated by their asymptotic forms. Examine in particular the exponential decay factors

$$e^{-\alpha|y_1|}\, e^{-\alpha|y_2|}\, e^{-\alpha|y_1+y_2-1|}\,.$$

The only portion of the plane where the combined effect of the exponential factors leads to an overall decay that is not faster than $e^{-\alpha}$ is the range $0 < y_1 < 1$, $0 < y_2 < 1-y_1$. The integrand does not change sign in this portion of y_1-y_2 space, which encompasses an area $\sim 1/2$, compared to the area $1/\alpha$, which is the corresponding extent in which the nonasymptotic integrand decays no more rapidly than $e^{-\alpha}$. Note that there is no portion of the plane in which the integrand decays more slowly than $e^{-\alpha}$. Thus the nonasymptotic integrand contribution is $O(\frac{1}{\alpha})$ compared to that of the asymptotic integrand.

Similar considerations enable one to deduce that one may also replace the Fourier transforms in the higher-order integrals by their asymptotic forms.

We thus conclude that if the time-dependences of two coupling functions are such that the asymptotic forms of their Fourier transforms are identical and of the indicated form, the large detuning transition amplitudes are the same.

As we have indicated, a sufficient condition that two pulses have the same $a_2(\omega)$ for large α is that both asymptotic Fourier transforms be equal to contour integrations given by [illegible] (Res(x=i)). We compare the hyperbolic secant of Rosen and Zener, $f = \frac{1}{[illegible]}$ sech [illegible] with the Lorentzian $f = \frac{1}{\pi}(1+x^2)^{-1}$. The corresponding $A(x)$ are

$$A_L(x) = \frac{\beta}{\pi}(1+x^2)^{-1},$$

$$A_H(x) = \frac{\beta}{2}\,\mathrm{sech}\,\frac{\pi x}{2}.$$

The transforms for both may be calculated via contour integrations. The Lorentzian case is trivial and applies to all ν, not just large frequencies. We choose a contour that runs along the real axis from $-R$ to $+R$ and is closed by a semicircle in the upper half plane. The contribution to the contour integral from the arc vanishes as $R \to \infty$, so that the Fourier transform is identical to the contour integral, whose value is determined by the residue at the simple pole at $x = i$.

The result is

$$\tilde{V}_L = \frac{\beta}{\sqrt{2\pi}} e^{-|\nu|} \quad . \tag{7a}$$

For the hyperbolic secant we choose a rectangular contour which runs from $-R$ to $+R$ along the real axis, that is continued by rectangular segments parallel to the imaginary from the points $(\pm R, 0)$ to the points $(\pm R, \pi i)$, and is closed by a line parallel to the real axis which runs from $(R, \pi i)$ to $(-R, \pi i)$. The two vertical segments give vanishing contributions as $R \to \infty$, and the horizontal segment off the real axis goes exponentially to zero compared to the segment along the real axis as $\nu \to \infty$. Thus, for the hyperbolic secant, the Fourier transform is identical to that of the Lorentzian in the asymptotic region. For large ν it is given by

$$\tilde{V}_H \simeq \frac{2\beta}{\sqrt{2\pi}} e^{-|\nu|} \quad . \tag{7b}$$

Since the Rosen-Zener solution gives the transition amplitude for all detunings, according to Eq. (4), as $-i\sqrt{2\pi}\, f(\nu) \sin\theta$, this formula must be valid asymptotically also. As we have shown that the asymptotic Fourier transforms of the Lorentzian and hyperbolic secant are proportional for large detunings, the Lorentzian must induce a transition amplitude that obeys a formula similar to Eq. (4). From Eqs. (7), we see that to construct the Lorentzian and hyperbolic secant Fourier transforms so that they are asymptotically identical, it is necessary to choose the Lorentzian pulse area β_L to be twice that of β_L. This immediately gives the large detuning scaling law for the Lorentzian

$$a_{2L} = -i\sqrt{2\pi}\, 2\tilde{f}_L(\alpha) \sin\frac{\beta}{2}. \tag{8a}$$

This result has been independently obtained by carrying out an asymptotic solution of Eqs. (3).[12] One can also show that for the pulse $A_C = \beta_C$ cosechπx, the appropriate scaling law is

$$a_{2C} = -i\frac{\sqrt{2\pi}}{2}\tilde{f}_C(\alpha)\sin 2\beta. \tag{8b}$$

For the hyperbolic secant pulse, the transition amplitude vanishes for pulse areas $\beta = n\pi$, n integral for all detunings. The Lorentzian, on the other hand, has eigenvalues $\beta = n\pi$ for zero detuning, while those for large detuning are $\beta = 2n\pi$. The eigenvalues of A_C go from $n\pi$ at $\alpha = 0$ to $\frac{n\pi}{2}$ as $\alpha \to \infty$.

The existence of a pole at x=i is a sufficient, but not a necessary condition that the asymptotic Fourier transform of a coupling pulse $\sim p(\omega)e^{-|\omega|}$. For example, the function $(1+x^2)^{-3/2}$ has an asymptotic Fourier transform proportional to $\nu^{1/2}\, e^{-\nu}$. The factor $\nu^{1/2}$ precludes deducing the asymptotic transition amplitude from the Rosen-Zener formula. Similarly, the squares of the hyperbolic secant and of the Lorentzian each have poles of second order at x=i, with the consequence that, for both of these, $\tilde{V}_a \sim \nu^1\, e^{-|\nu|}$, so that while these will have asymptotic transition amplitudes that are related to each other, they cannot be obtained by scaling from Eq. (4). In our next paper, we shall show

how to calculate asymptotic transition amplitudes when the coupling pulse has second- and higher-order poles at x=i. For now, we merely present the formulae for the transition amplitudes generated by the squares of the hyperbolic secant and Lorentzian

$$a_2(H2) = -i\,\frac{2\pi}{C^2}\,e^{-|\alpha|}\sin\left[C\sqrt{\frac{|\alpha\beta|}{\pi}}\right]\sinh\left[C\sqrt{\frac{|\alpha\beta|}{\pi}}\right] \tag{9a}$$

$$a_2(L2) = -i\,\frac{2\pi}{C^2}\,e^{-|\alpha|}\sin\left[C\sqrt{\frac{|\alpha\beta|}{2\pi}}\right]\sinh\left[C\sqrt{\frac{|\alpha\beta|}{2\pi}}\right], \tag{9b}$$

where $C = 1 + \frac{1}{6} + \frac{1}{56} + \frac{1}{182} + \approx 1.194$. Equation (9a) can be obtained from Eq. (9b) by scaling techniques derived in this paper.

III. Summary and Conclusion

In this paper, we have demonstrated that pulse shapes $A(t)$ whose Fourier transforms asymptotically approach the form $\phi(\nu)e^{-|\nu|}$, where ϕ is slowly varying, may be categorized into families which differ according to the function ϕ. Within each family, the transition amplitudes $a_2(\infty)$ are related by simple scaling laws, so that if one is able to derive an expression for the transition amplitude generated by one member of the family, corresponding formulae for all other members of the family may be written down by inspection.

A sufficient condition that the Fourier transform be of the required form is that it be obtainable in the asymptotic region as a contour integral evaluated from the residue at a single pole on the imaginary time axis. For the case where $A(t)$ has simple poles, $a_2(\omega)$

may be inferred from the solution of the Rosen-Zener problem[2,3], known for fifty years, by a trivial scaling operation.

Our results were obtained by examining the structure of the terms in perturbation expansions for transition amplitudes. (We have demonstrated that these sequences always converge in two-level problems provided that the pulse areas are finite. Low-order approximations, however, are frequently not useful for $t \to \infty$ even when they are valid at finite times.) With suitable choices of ratios of pulse areas, corresponding terms in the series for different members of the same family will be identical.

In a future paper[12], we shall present methods for explicitly calculating transition amplitudes that apply to higher-order, as well as simple poles. Thus, we are not restricted in practice to writing scaling laws for pulses which may be compared in the asymptotic region to the hyperbolic secant.

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Appendix - Convergence of Perturbation Theory for the Transition Amplitude

We demonstrate here that the perturbation series for a_2 converges for all finite pulse areas. The contribution of order $(2k+1)$ is

$$b_1^{(k)} = -i\beta^{2k+1} a_2^{(2k+1)} =$$

$$-i\beta^{2k+1}(-1)^k \int_{-\infty}^{\infty} f(x_1) e^{-i\alpha x_1} dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} f(x_j) e^{i(-1)^j \alpha x_j} dx_j . \qquad \text{(A-1)}$$

Now assume that $A(x)$ is of a single algebraic sign. Without loss of generality we may take this to be positive. We compare the series with the corresponding expression for $\alpha = 0$.

$$b_{10}^{(k)} = -i\beta^{2k+1}(-1)^k \int_{-\infty}^{\infty} f(x_1) dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} f(x_j) dx_j = \qquad \text{(A-2)}$$

$$-i\beta^{2k+1}(-1)^k \int_{-\infty}^{\infty} |f(x_1)| dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} |f(x_j)| dx_j . \qquad \text{(A-2')}$$

Invoking the theorems on repeated integrals of the same function

$$b_{10}^{(k)} = \frac{-i\beta^{2k+1}}{(2k+1)!}(-1)^k \left(\int_{-\infty}^{\infty} f(x)\, dx \right)^{2k+1},$$

and the terms are recognized as identical to those for the series -i sin...

Now consider the series

$$F(\beta) = \sum \left| b_{10}^{(k)} \right| = \sum \frac{|\beta^{2k+1}|}{(2k+1)!} \left(\int_{-\infty}^{\infty} f(x)\,dx \right)^{2k+1}$$

$$= \sum \frac{|\beta^{2k+1}|}{(2k+1)!} .$$

This is evidently the series for sinh β, which converges so long as β is finite. Hence, the series of Eq. (A-2) is absolutely convergent. Now

$$\left| b_1^{(k)} \right| = |\beta^{2k+1}| \left| \int_{-\infty}^{\infty} f(x_1) e^{-i\alpha x_1} dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} f(x_j) e^{i(-1)^j \alpha x_j} dx_j \right|$$

$$\leq |\beta^{2k+1}| \left| \int_{-\infty}^{\infty} |f(x_1)|\,dx_1 \prod_{j=2}^{2k+1} \int_{-\infty}^{x_{j-1}} |f(x_j)|\,dx_j \right|$$

$$\leq \left| b_{10}^{(k)} \right| ,$$

so that the series, Eq. (A-1) is also absolutely convergent, and our result is established.

We note that the same arguments will apply to perturbation series at finite times, provided merely that $\int_{-\infty}^{x} f(x')dx' = F(x)$ is of one sign and finite. If f(x) changes sign, the results will still be valid provided the generalized area $\int_{-\infty}^{x} |f(x')|dx'$, is finite.

A simple case where the convergence theorem does not apply is the coupling function $A(x) = (\text{const})\,(\tanh \pi x/2)/x$, since β is logarithmically divergent. In addition, since the pulse area is proportional to the

Fourier transform at zero frequency, the multiple integrals in the frequency domain for the third- and higher-order contributions to the perturbation series contain regions where the integrands blow up, so that the individual terms beyond first order may not even exist. (The first-order contribution will be finite, since the Fourier transform for this pulse exists for $\nu \neq 0$. In this case, we note that the infinite area does not imply a pulse of infinite energy, so that it theoretically could exist. One evidently cannot use the methods developed here to describe the dynamics. At the very least, decay would have to be included in the analysis, and a completely non-perturbative treatment utilized.)

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