Nonlinear level crossing models

N. V. Vitanov and K.-A. Suominen
Helsinki Institute of Physics, PL 9, FIN-00014 Helsingin yliopisto, Finland
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We examine the effect of nonlinearity at a level crossing on the probability for nonadiabatic transitions $P$. By using the Dykhne-Davis-Pechukas formula, we derive simple analytic estimates for $P$ for two types of nonlinear crossings. In the first type, the nonlinearity in the detuning appears as a perturbative correction to the dominant linear time dependence. Then appreciable deviations from the Landau-Zener probability $P_{LZ}$ are found to appear for large couplings only, when $P$ is very small; this explains why the Landau-Zener model is often seen to provide more accurate results than expected. In the second type of nonlinearity, called essential nonlinearity, the detuning is proportional to an odd power of time. Then the nonadiabatic probability $P$ is qualitatively and quantitatively different from $P_{LZ}$ because on the one hand, it vanishes in an oscillatory manner as the coupling increases, and on the other, it is much larger than $P_{LZ}$. We suggest an experimental situation when this deviation can be observed.

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I. INTRODUCTION

The Landau-Zener (LZ) model [1,2] is widely used in quantum physics to describe level crossing and avoided crossing transitions. It provides the probability of transition between two quantum states coupled by an external field of constant amplitude and time-dependent frequency which passes through resonance with the transition frequency. This level crossing, seen in the diabatic basis (the basis of the two bare states), appears as an avoided crossing in the adiabatic basis (the basis comprising the two eigenstates of the Hamiltonian). Cases of level crossings and avoided crossings can be met in many areas in physics, such as laser-atom interactions [3], magnetic resonance [4], slow and cold atomic collisions, molecular physics [5,6], optical atoms [7,8], atom lasers [9,10], solid-state physics [11,12], ultrasmall tunnel junctions [13,14], nuclear physics [15], and particle physics [16,17]. The LZ model is a basic tool for describing and understanding such phenomena.

Given that the LZ model presumes very crude time dependences for the coupling (constant) and the detuning (linear), it is somewhat surprising that it has been often found to provide rather accurate results when applied to specific cases involving time-dependent couplings (e.g., pulse-shaped) and nonlinear detunings. To the best of our knowledge, no satisfactory explanation of this fact has been given so far. In the present paper, we try to answer this question by considering models where the coupling is still constant but the detuning is nonlinear. We investigate two main classes of nonlinear detunings. In the first class, the nonlinearity appears as a correction to a dominant linear time dependence near the crossing; we call this perturbative nonlinearity. In the second class, the detuning is proportional to an odd power of time, $\Delta \propto t^{N}$, with $N = 3, 5, 7, \ldots$; hence, the detuning cannot be linearized in any vicinity of the crossing and we call this nonlinearity essential. We have found significant qualitative differences between these two cases. We study the nonlinearity effects both numerically and analytically and we are interested in the near-adiabatic regime which is the one of primary interest as far as level crossing models are concerned. The analytic approach is based on the Dykhne-Davis-Pechukas formula [25,26] and a generalization of it that accounts for multiple transition points [27,28].

The paper is organized as follows. The basic equations and definitions are given in Sec. II. The perturbatively nonlinear models are discussed in Sec. III and the essentially nonlinear models in Sec. IV. The conclusions are summarized in Sec. V. The numerical method used for highly accurate integration of the Schrödinger equation is nontrivial and it is presented in the Appendix.

II. BACKGROUND

A. Basic equations and definitions

We wish to solve the Schrödinger equation ($\hbar = 1$),

$$i \frac{d}{dt} \psi(t) = H(t) \psi(t), \quad (1)$$

with the Hamiltonian

$$H(t) = \begin{bmatrix} -\Delta(t) & \Omega(t) \\ \Omega(t) & \Delta(t) \end{bmatrix}, \quad (2)$$

and $\psi(t) = [\psi_1(t), \psi_2(t)]^T$, where $\psi_1(t)$ and $\psi_2(t)$ are the probability amplitudes of states $\psi_1$ and $\psi_2$. The detuning $\Delta(t)$ and the coupling $\Omega(t)$ are both assumed real. We are interested in the case when the coupling is constant and the detuning is an odd function of time,

$$\Omega(t) = \text{const}, \quad \Delta(-t) = -\Delta(t). \quad (3)$$

Such is the case for the Landau-Zener (LZ) model,
\[ \Omega(t) = \text{const}, \quad \Delta_{LZ}(t) = \beta^2 t. \] (4)

Since the transition probability is invariant upon the sign inversions \( \Delta \to -\Delta \) and \( \Omega \to -\Omega \), we assume for simplicity and without loss of generality that both \( \Omega \) and the slope \( \beta^2 \) of the detuning at the crossing \( (t = 0) \) are positive and that \( \beta \) itself is positive.

The level crossing, seen in the diabatic basis \([1]\), translates into an avoided crossing in the adiabatic basis because the eigenvalues \( \pm \mathcal{E}(t) \) of the adiabatic states (the quasienergies) do not cross but only come close to each other near \( t = 0 \). Here

\[ \mathcal{E}(t) = \sqrt{\Omega^2 + \Delta^2(t)}. \] (5)

If the system is initially in state \( \psi_1 \) \([c_1(-\infty) = 1, \ c_2(-\infty) = 0]\), the transition probability in the diabatic basis at \(+\infty\) is given by \( |c_2(+\infty)|^2 \). The transition probability in the adiabatic basis (the probability for nonadiabatic transitions) is equal to the probability of no transition in the diabatic basis, \( P = |c_1(+\infty)|^2 \), and its determination will be our main concern. For the LZ model,

\[ P = e^{-\pi \alpha^2}, \] (6)

where \( \alpha = \Omega/\beta \) is the normalized coupling.

In Secs. [II] and [IV] we consider two types of nonlinear detunings and we investigate how the nonlinearity affects the LZ probability \([1]\). In the first type, the nonlinearity appears as a perturbative correction to a dominant linear time dependence near the crossing; for the superlinear model (Sec. [III A]) this correction is positive whereas for the sublinear model (Sec. [III B]) the correction is negative. In the second type, the detuning is proportional to \( t^N \), with \( N = 3, 5, 7, \ldots \), and the detuning is essentially nonlinear. Like in the LZ model, the detuning diverges as \( t \to \pm\infty \) in all considered cases. The studied nonlinear detunings are shown in Fig. [I].

### B. Dykhne-Davis-Pechukas formula

The Dykhne-Davis-Pechukas (DDP) formula \([25,26]\) provides the asymptotically exact probability for nonadiabatic transitions in the adiabatic limit. It reads

\[ P \sim e^{-2\text{Im} \mathcal{D}(t_c)}, \] (7)

where

\[ \mathcal{D}(t_c) = 2 \int_0^{t_c} \mathcal{E}(t) dt. \] (8)

The point \( t_c \) is called the transition point and it is defined as the (complex) zero of the quasienergy,

\[ \mathcal{E}(t_c) = 0, \] (9)

which lies in the upper complex \( t \)-plane (i.e., \( \text{Im} t_c > 0 \)) and if there are more than one such zero points, \( t_c \) is the one closest to the real axis. Equation (7) gives the correct asymptotic probability for nonadiabatic transitions provided (i) the quasienergy \( \mathcal{E}(t) \) does not vanish for real \( t \), including at \( \pm\infty \); (ii) \( \mathcal{E}(t) \) is analytic and single-valued at least throughout a region of the complex \( t \)-plane that includes the region from the real axis to the transition point \( t_c \); (iii) the transition point \( t_c \) is well separated from the other quasienergy zero points (if any) and from possible singularities. Amazingly, for the LZ model, the DDP formula (7) gives the exact probability (6) not only in the adiabatic limit \( (\alpha \gg 1) \) but also for any \( \alpha \).

In the case of more than one zero points in the upper \( t \)-plane, Davis and Pechukas \([26]\) have suggested, following George and Lin \([27]\), that Eq. (7) can be generalized to include the contributions from all these \( N \) zero points \( t^k_c \) in a coherent sum. This suggestion has been later verified by Suominen, Garraway and Stenholm \([28,31]\). The generalized DDP formula has the form

![FIG. 1. The level crossing cases studied in this paper. Upper figure, the detunings \( \Delta(t) \) in the diabatic basis. Lower figure, the quasienergies \( \pm \mathcal{E}(t) \) in the adiabatic basis.](image-url)
\[ P \sim \left| \sum_{k=1}^{N} \Gamma_k e^{D \langle t_k^c \rangle} \right|^2 , \]  
(10)

where

\[ \Gamma_k = 4i \lim_{t \to t_k^c} (t - t_k^c) \dot{\delta}(t) \]  
(11)

and \( \dot{\delta}(t) \equiv [\hat{\Omega}(t)\Delta(t) - \Omega(t)\dot{\Delta}(t)]/2\mathcal{C}^2(t) \) is the nonadiabatic coupling, where an overdot means a time derivative. In principle, Eq. (11) should be used when there are more than one zero points which are closest to the real axis and have equal imaginary parts and Eq. (10) should include only the contributions from these zeroes. The contributions from the farther zeroes are exponentially small compared to the dominant ones and may therefore be neglected. Retaining the contributions from all transition points, however, may be beneficial and it has been shown elsewhere [29,31] that for the Demkov-Kunike models [32], the full summation in Eq. (10) leads to the exact result.

The motivation for developing good approximations, such as Eq. (10) for the transition probability in the adiabatic limit, does not arise only from the general wish to have analytic expressions. In fact, the numerical integration of the time-dependent Schrödinger equation for the level crossing models studied in the present paper (most of which are very slowly convergent from a numerical viewpoint) becomes increasingly difficult and time consuming as we approach this limit. In Appendix A, we present the numerical approach used in our studies, which has been specifically developed for highly accurate numerical integration of slowly convergent two-state problems, such as the present ones. The need for sophisticated numerical approaches emphasizes the usefulness of the DDP formulas (8) and (10), which become increasingly accurate as the adiabatic limit is approached.

III. PERTURBATIVE NONLINEARITY

We wish to estimate the probability for nonadiabatic transitions \( P \) in the case when the linear LZ detuning (11) is perturbed by a small cubic nonlinearity in the vicinity of the crossing (at \( t = 0 \)),

\[ \Omega(t) = \text{const}, \quad \Delta(t) = \beta^2 t + \Gamma \beta + \ldots . \]  
(12)

When \( \Gamma > 0 \), the detuning passes through resonance in a superlinear manner (\( |\Delta(t)| > |\Delta|_L(t)| \)), whereas when \( \Gamma < 0 \), the detuning passes through resonance in a sublinear manner (\( |\Delta(t)| < |\Delta|_L(t)| \)). It is more convenient to work with dimensionless quantities and we choose \( \beta \) to define our frequency and time scales. We define the dimensionless coupling \( \alpha \), the nonlinearity coefficient \( \gamma \), and the time \( \tau \) as

\[ \alpha = \frac{\Omega}{\beta}, \quad \gamma = \frac{|\Gamma|}{\beta^2}, \quad \tau = \beta t, \]  
(13)

both \( \alpha \) and \( \gamma \) being positive. Then Eqs. (12) become

\[ \tilde{\Omega}(\tau) = \alpha, \quad \tilde{\Delta}(\tau) = \tau (1 + \gamma^2 \tau^2 + \ldots) , \]  
(14)

where \( \tilde{\Omega}(\tau) = \Omega(t)/\beta, \tilde{\Delta}(\tau) = \Delta(t)/\beta \), and the plus (minus) sign is for the superlinear (sublinear) case.

The direct treatment of model (14) involves a third-order algebraic equation which is too cumbersome. Moreover, for a negative nonlinearity, model (14) involves two unwanted additional spurious crossings. We avoid these drawbacks of model (14) by using instead two other models which contain positive and negative cubic nonlinearities, have a single level crossing, involve dealing with quadratic equations, and allow simpler derivations.

A. Superlinear model

The first model we consider is

\[ \tilde{\Omega}(\tau) = \alpha, \quad \tilde{\Delta}(\tau) = \tau \sqrt{1 + 2 \gamma^2 \tau^2} . \]  
(15)

For \( \gamma = 0 \), model (15) reduces to the LZ model. Near the crossing (\( \tau = 0 \)), we have \( \tilde{\Delta}(\tau) = \tau (1 + \gamma^2 \tau^2 + \ldots) \), i.e., model (15) reduces to model (14) with a positive nonlinear term. Moreover, as \( \tau \to \pm \infty \), the detuning diverges quadratically, i.e., faster than the (linear) LZ detuning. Hence, we call model (15) the superlinear model.

This model has two transition points in the upper complex half-plane,

\[ \tau_c^\pm = \frac{i\alpha}{\xi} \left( \sqrt{1 + \xi} \pm \sqrt{1 - \xi} \right) , \]  
(16)

where \( \xi \equiv 2 \sqrt{2} \alpha \gamma \). For \( \gamma \to 0 \), we have \( \tau_c^- \to i\alpha \) and \( \tau_c^+ \to i\infty \), and we recover the (single) LZ transition point.

1. The case of small \( \alpha \gamma \)

For \( \xi \equiv 2 \sqrt{2} \alpha \gamma < 1 \), both transition points are purely imaginary, with \( \text{Im}\tau_c^- < \text{Im}\tau_c^+ \). Hence, we can take the contribution from \( \tau_c^- \) only. We make the substitutions \( \tau = iy \) and \( \tau_c^- = iy_c \) in Eq. (8) and obtain

\[ D\left(\tau_c^-ight) = 2i \int_{0}^{y_c} \sqrt{\alpha^2 - y^2 + 2 \gamma^2 y^2} \, dy \]
\[ = \frac{1}{2} \pi \alpha \tau_c^- F \left( \frac{1}{2}, -\frac{1}{2}; 2; 1 - \sqrt{1 - \xi^2} \right) \]
\[ = i \frac{\pi \gamma^2}{2} F \left( \frac{1}{4}, \frac{3}{4}; 2; \xi^2 \right) , \]  
(17)

where \( F(a, b; c; z) \) is the Gauss hypergeometric function. The DDP formula gives

\[ P \sim e^{-\pi \alpha^2 F \left( \frac{1}{4}, \frac{3}{4}; 2; \xi^2 \right)} . \]  
(18)
For $\xi \equiv 2\sqrt{2}\alpha \gamma \ll 1$, Eq. (18) reduces to
\[ P \sim e^{-\pi \alpha^2 (1 + \frac{1}{4}\alpha^2 \gamma^2 + \ldots)}. \] (19)

Comparison with the LZ formula (15) shows that the superlinearity reduces the probability for nonadiabatic transitions.

The DDP result (14) is formally valid in the nearadiabatic regime ($\alpha \gg 1$). Moreover, in the derivation of Eq. (19) we assumed that $2\sqrt{2}\alpha \gamma \ll 1$. Hence, we should have
\[ \frac{1}{2\sqrt{2} \gamma} \gg \alpha \gg 1. \] (20)

We have to account also for the fact that both the other zero point $\tau_c^+$ and the singularity point $\tau_c^-$, situated at $\tau_0 = i/\gamma \sqrt{2}$, should be well separated from the transition point $\tau^-$. It is easy to verify that in the present case ($2\sqrt{2}\alpha \gamma < 1$) we have $\mathrm{Im} \tau^- > \mathrm{Im} \tau^+ < \mathrm{Im} \tau_0$ and hence, we should only have $\mathrm{Im} \tau^+ - \mathrm{Im} \tau^- \gg 1$. This is indeed the case if condition (20) is satisfied.

Condition (20) limits the applicability of Eq. (19) to very small nonlinearities only. We have found, however, through extensive comparison with numerical simulations that this condition can be relaxed to
\[ \frac{1}{2\sqrt{2} \gamma} \lesssim \alpha \lesssim 1. \] (21)

2. The case of large $\alpha \gamma$

For $\xi \equiv 2\sqrt{2}\alpha \gamma > 1$, the transition points (14) are complex, with equal imaginary parts and real parts which are equal in magnitude but opposite in sign. Then, following Eq. (10), we have to take the contributions from both of them. The integrals $D(\tau_c^\pm)$ are given by
\[ D(\tau_c^\pm) = \frac{i}{4} \pi \alpha \tau_c^\pm F \left( \frac{1}{2}, -\frac{1}{2}, 2; 1 \pm \frac{i}{4} \sqrt{x^2 - 1} \right) \] (22)
where $\pm D_r$ and $D_i$ are the real and imaginary parts. The factors (14) are $\Gamma_\pm = -1$ and the generalized DDP formula (15) gives
\[ P \sim 4e^{-2D_i} \cos^2 D_r. \] (23)

In Fig. 2, the probability for nonadiabatic transitions for the superlinear model (15) is plotted against the dimensionless coupling $\alpha \equiv \Omega/\beta$ for $\gamma = 0.25$. The DDP approximations (19) and (24) are seen to fit the numerical results very well. As seen from the figure, this is the case even for small $\alpha$, while the DDP formula is supposed to be valid for $\alpha \gg 1$ only. This is a consequence of the fact that the DDP formula provides the exact result for the LZ model for any $\alpha$.

The condition of validity of Eq. (23) is expected to be $\xi = 2\sqrt{2}\alpha \gamma \gtrsim 1$. However, for large coupling $\alpha$ and nonlinearity $\gamma$, the singularity point $\tau_0 = i/\gamma \sqrt{2}$ gets closer to the real axis than the transition points $\tau_c^\pm$ which makes the DDP result (23) inaccurate for very large $\alpha \gamma$; indeed, we have verified this numerically (not shown in Fig. 2). We should emphasize that the case of large nonlinearity resembles more that of the essential nonlinearity (Sec. IV) and hence, it is not interesting in the context of perturbative nonlinearity, considered in this section.

B. Sublinear model

The sublinear model is defined as
\[ \tilde{\Omega}(\tau) = \alpha, \quad \tilde{\Delta}(\tau) = \frac{\tau}{\sqrt{1 + 4\gamma^2 \tau^2}}. \] (24)
For $\gamma = 0$, it reduces to the LZ model (15). Near the crossing the detuning behaves as $\Delta(\tau) = \tau (1 - \gamma^2 \tau^2 + \ldots)$, i.e., model (24) reduces to model (15) with a negative nonlinear term. As $\tau \to \pm \infty$, the detuning diverges as $|\tau|^{1/2}$, i.e., slower than the (linear) LZ detuning. Hence, the name sublinear model.

This model has a single transition point in the upper half plane given by
\[ \tau_c = i\alpha \sqrt{4\alpha^2 \gamma^2 + 1 - 2\alpha^2 \gamma^2}. \] (25)
The integral $D(\tau_c)$ is given by
The DDP formula (7) gives
$$P \sim e^{-2\mathcal{J}},$$
where
$$\mathcal{J} = 2 \int_0^{\gamma c} \sqrt{\alpha^2 - \frac{y^2}{1 - 4\gamma^2 y^2}} \, dy.$$  
(27)

where the substitutions $\tau = iy$ and $\tau_c = iy_c$ have been made. The DDP formula (28) gives
$$P \sim e^{-2\mathcal{J}}.$$  
(28)

In the limits of small and large $\alpha \gamma$, we find
$$P \sim e^{-\pi \alpha^2 (1 - \frac{1}{2} \alpha^2 \gamma^2 + \ldots)}, \quad (\sqrt{2} \alpha \gamma \lesssim 1),$$  
(29a)
$$P \sim e^{-2\alpha \gamma + \pi/(16 \alpha^2 \gamma^2) + \ldots}, \quad (\sqrt{2} \alpha \gamma \gtrsim 1).$$  
(29b)

Comparison with the LZ formula (2) shows that the sublinearity increases the probability for nonadiabatic transitions. Note also that due to the absence of other transition points in the upper half-plane, there are no oscillations in $P$.

In Fig. 3, the probability for nonadiabatic transitions $P$ for the sublinear model (24) is plotted against the dimensionless coupling $\alpha \equiv \Omega/\beta$ for $\gamma = 0.25$. The numerical results are shown by dots and the analytic approximation (28) by a solid curve. The LZ probability (4) is shown by a dashed curve.

FIG. 3. The probability for nonadiabatic transitions $P$ for the sublinear model (24) plotted against the dimensionless coupling $\alpha \equiv \Omega/\beta$ for $\gamma = 0.25$. The numerical results are shown by dots and the analytic approximation (28) by a solid curve. The LZ probability (4) is shown by a dashed curve.

that the DDP formula (28) should be accurate if the two points $\tau_c$ and $\tau_0$ are well separated. For large $\alpha \gamma$, however, they approach each other [see Eq. (23)], which explains the small inaccuracy of Eq. (28) seen in Fig. 3.

C. Discussion

Comparison of Eqs. (14), (19) and (29a) shows that the probability for nonadiabatic transitions for model (24) for small nonlinearity is given by
$$P \sim P_{LZ} e^{-2\Omega \Gamma / \beta^8}.$$  
(30)

This equation is valid for $\Omega \Gamma / \beta^6 \lesssim 1$. Since the deviation from the LZ probability $P_{LZ}$ depends exponentially on the coupling $\Omega$ and the nonlinearity coefficient $\Gamma$, this deviation can be very large. On the other hand, however, this difference emerges when $\Omega / \beta \gtrsim 1$; then $P$ is very small (virtually zero), which explains why the LZ formula is often found to be more accurate than anticipated.

In Fig. 4, the probability for nonadiabatic transitions $P$ is plotted against the nonlinearity coefficient $\gamma$ for the superlinear model (15) and the sublinear model (24). For $\gamma = 0$, both the superlinear and sublinear probabilities are equal to the LZ value $P_{LZ} \approx 0.043$. As $\gamma$ departs from zero, the superlinear probability decreases while the sublinear probability increases, in agreement with our analytic result (30). This qualitatively different behavior is readily explained by looking at the quasienergies $\pm \varepsilon(t)$. For the superlinear model (15), the avoided crossing between the quasienergies is sharper than in the LZ
case, while for the sublinear model \([24]\), it is flattter than the LZ one (see Fig. [1]). Consequently, the “duration” \(T\) of the avoided crossing for the superlinear model is shorter than for the LZ model, while for the sublinear model it is larger, \(T_{\text{super}} < T_{\text{LZ}} < T_{\text{sub}}\) which means that the same relation should apply for the probabilities, \(P_{\text{super}} < P_{\text{LZ}} < P_{\text{sub}}\). 

D. Comparison with the Allen-Eberly-Hioe model

An argument in favor of Eq. (38) can be derived from the Allen-Eberly-Hioe model \([34,35]\).

\[
\Omega(t) = \frac{A}{\cosh(t/T)}, \quad \Delta(t) = B \tanh(t/T), \quad (31)
\]

which is a particular case of the Demkov-Kunike model \([29,32]\). Model (31) can be solved analytically and gives

\[
P = \frac{\cosh^2(\pi \sqrt{B^2 - A^2 T})}{\cosh^4(\pi B T)}. \quad (32)
\]

It has been shown in \([30]\) that this model is a member of a class of infinite number of models, for all of which the nonadiabatic probability is given by Eq. (32). Another member of this class is the model

\[
\Omega(t) = A, \quad \Delta(t) = B \tan(t/T), \quad (33)
\]

\((-\frac{1}{2} \pi T < t < \frac{1}{2} \pi T).\)

We can make model (33) behave like the perturbatively nonlinear model \([12]\) (i.e., make the coupling duration infinite and the detuning divergent) by letting \(T \to \infty\) and \(B \to \infty\), while maintaining the slope at the crossing \(B/T\) constant. Then Eq. (32) reduces to

\[
P \sim \exp \left[ -\pi \frac{A^2 T}{B} \left( 1 + \frac{A^2}{4 B^2} + \ldots \right) \right]. \quad (34)
\]

The same result follows from the DDP approximation \([31]\) by accounting for the fact that for model (33), \(\alpha = A \sqrt{T/B}, \beta^2 = B/T, \) and \(\Gamma = B/3T^3\).

IV. ESSENTIAL NONLINEARITY

The essentially nonlinear model is defined by

\[
\Omega(t) = \text{const.}, \quad \Delta(t) = \beta^{N+1} t^N, \quad (35)
\]

where \(N = 3, 5, 7, \ldots\) is an odd number. Model (35) cannot be linearized in any vicinity of the crossing.

The zero points of the quasienergies \((\text{8})\) in the upper \(t\)-plane are

\[
t^k_c = \alpha^{1/N} e^{i \pi (2k-1)/(2N)}, \quad (k = 1, 2, \ldots, N), \quad (36)
\]

where \(\tau = \beta t\) and \(\alpha = \Omega / \beta\). With the exception of \(t^c_{c_{(N+1)/2}}\), which is imaginary, the transition points are grouped in pairs that have the same imaginary parts but opposite real parts. The significant difference between the LZ model and the essentially nonlinear model is that \(t^c_{c_{(N+1)/2}}\) is the only transition point in the former, while in the latter it is the one farthest from the real axis and hence, with the smallest contribution to the nonadiabatic probability. The largest contribution comes from the points \(t^c_k\) and \(t^c_{N}\) which are closest to the real axis.

The integrals \(D(t^k_c)\) are given by

\[
D(t^k_c) = 2 \nu_N \alpha^{(N+1)/N} e^{i \pi (2k-1)/(2N)}, \quad (37)
\]

where the number \(\nu_N\) denotes the value of the integral

\[
\nu_N = \int_0^1 \sqrt{1 - x^{2N}} \, dx. \quad (38)
\]

The first few values of \(\nu_N\) are \(\nu_1 = \pi/4 \approx 0.785, \nu_3 \approx 0.911, \nu_5 \approx 0.944, \nu_7 \approx 0.959\). The factors (11) are given by \(\Gamma_k = (-1)^k\). The generalized DDP formula (10) gives

\[
P \approx 2 \sum_{k=1}^{(N-1)/2} (-1)^k e^{-\nu_t \sin \frac{\pi k}{N} \cos \left( \frac{2k-1}{N} \pi \right)} + (-1)^{(N+1)/2} e^{-\nu_t}, \quad (39)
\]

with \(\eta = 2 \nu_N \alpha^{(N+1)/N}\). For large \(\alpha\), the main contribution to \(P\) comes from the first term \((k = 1)\) in the sum.

The probability (39) is plotted in Fig. [8] against the coupling \(\alpha\) and compared with the exact numerical results for \(N = 3\) (top figure), \(N = 5\) (middle figure), and \(N = 7\) (bottom figure). Obviously, the probability (39) is qualitatively different from the LZ probability \(P_{\text{LZ}}\). First, there are oscillations which appear due to the existence of multiple transition points. From another point of view, the oscillations appear because the nonadiabatic coupling \(\delta(t)\) has two peaks, in contrast to the single-peaked nonadiabatic coupling in the LZ model. Second, the transition probability is much larger than \(P_{\text{LZ}}\) which can be explained by the fact that in the quasienergy picture, the avoided crossing has a much longer duration (see Fig. [8]).

These differences can be verified experimentally. For example, an essentially nonlinear crossing arises when a two-state atom is excited by a frequency modulated laser pulse with a supergaussian time dependence \([i.e., \Omega(t) \propto e^{-t/T^2}]\) and the frequency modulation is produced by the self-phase modulation technique, in which the phase shift \(\varphi(t)\) is proportional to either the amplitude of the field \([i.e., \varphi(t) \propto \Omega(t)]\) or the intensity of the field \([i.e., \varphi(t) \propto \Omega^2(t)]\) \([37]\). Since the detuning is proportional to \(\varphi(t)\), we shall have \(\Delta(t) \propto t^{2n-1}\) with a crossing at \(t = 0\). As the hypergaussians are almost constant near \(t = 0\), this example reproduces model (35) very well.
V. SUMMARY AND CONCLUSIONS

We have examined the effect of nonlinearity in the detuning at the level crossing on the probability for nonadiabatic transitions $P$. Our analysis has been based upon both analytic approximations derived by using the Dykhne-Davis-Pechukas formula and numerical calculations. We have distinguished two types of nonlinearities: perturbative and essential. In the former type, the nonlinearity appears as a correction to a dominant linear time dependence near the crossing. In the latter, the detuning is proportional to an odd power of time, $\Delta \propto t^N$ with $N = 3, 5, 7, \ldots$.

For the perturbative nonlinearities, the probability for nonadiabatic transitions $P$ is larger than $P_{LZ}$ for sub-linear nonlinearity and smaller than $P_{LZ}$ for superlinear nonlinearity. For both the superlinear and sublinear cases, the appreciable deviations from $P_{LZ}$ emerge for large coupling when $P$ is already very small, virtually unobservable. This fact explains why the LZ model has often been found to provide more accurate results than anticipated. We have provided a simple analytic estimate for the deviation as a function of the nonlinearity, which should be a useful criterion for estimating the applicability of the LZ model to any level crossing case, as far as perturbative nonlinearity is concerned.

For the essential nonlinearity, we have found that the nonadiabatic probability $P$ is both quantitatively and qualitatively different from $P_{LZ}$ because on the one hand, it vanishes in an oscillatory manner as the coupling increases, and on the other, it is much larger than $P_{LZ}$. From a mathematical viewpoint, these differences derive from the existence of two complex transition points equally close to the real axis, both being closer than the single LZ transition point. From a physical viewpoint, the differences can be explained by the fact that the avoided crossing is flatter and of longer duration than the LZ one and the nonadiabatic coupling has two peaks, in contrast to the single-peaked LZ nonadiabatic coupling. We have suggested an experimental situation when this deviation can be observed.

We have limited our analysis to the case when the detuning is an odd function of time [Eq. (3)], which excludes asymmetric level crossings, e.g., quadratic corrections in the perturbative models. It would be interesting to estimate the effect of such quadratic terms although then the DDP treatment is more complicated. It should be noted that the parabolic level crossing model considered in [38] involves either two level crossings or no crossing and hence, it is different from the present case of a single level crossing.

Finally, we point out that the detuning nonlinearity is only one of the sources of possible inaccuracies in applications of the LZ model. Others include the finite coupling duration [36] and the nonzero transition times [38].

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APPENDIX A: NUMERICAL INTEGRATION OF SLOWLY CONVERGENT TWO-STATE PROBLEMS

In this Appendix, we describe the numerical algorithm we have used to integrate the Schrödinger equation \( \hat{H} \). The Landau-Zener model is notoriously known for its slow numerical convergence because the amplitude of the oscillations, which appear in the time evolution of the transition probability \( P \), only vanishes as \( |t|^\frac{-3}{2} \) in the diabatic basis \( \hat{\Sigma} \). This is a consequence of the slow (linear) divergence of the detuning. Of course, the final problem is to carry out the numerical integration in the time evolution is needed. The situation is even worse for the sublinear model \( \mathcal{H} \) in which the oscillations amplitude vanishes only as \( |t|^\frac{-3}{2} \). One of the possibilities to alleviate this problem is to carry out the numerical integration in the adiabatic basis, where the oscillation amplitude vanishes as \( |t|^\frac{-3}{2} \) for the LZ model \( \hat{\Sigma} \) and as \( |t|^\frac{-5}{2} \) for the sublinear model. This improvement may be insufficient if the transition probability is very small, as in the present paper. Yet another problem arises from the finite initial (large negative) time, because it introduces additional oscillations in \( P \). For the LZ model these problems can be resolved by starting the integration at the crossing \( (t = 0) \) and propagating the solution towards the desired (positive or negative) time \( \hat{\Sigma} \). The initial conditions in this approach require the values of \( P \) and a few of its derivatives at \( t = 0 \), which can be found exactly \( \mathcal{H} \). Unfortunately, this approach does not apply to the models in the present paper.

We have used a numerical approach which combines and generalizes ideas by Bambini and Lindberg \( \mathcal{H} \) and Berry and Lim \( \mathcal{H} \). The approach is based on two concepts. First, we perform the numerical integration in a superadiabatic (SA) basis \( \mathcal{H} \), rather than in the usual diabatic or adiabatic bases. The successive SA bases are obtained iteratively \( \mathcal{H} \) and they are not the more familiar superadiabatic bases obtained as truncated asymptotic series in the adiabatic parameter \( \mathcal{H} \). The \( n \)-th order SA states are defined as the instantaneous normalized eigenstates of the Hamiltonian in the SA basis of order \( n - 1 \). For instance, by diagonalizing the Hamiltonian in the diabatic basis (which is the SA basis of order \( n = 0 \)) we obtain the adiabatic basis (SA basis of order \( n = 1 \)). The recursive relations between the “couplings” \( \Omega_n(t) \) and the “detunings” \( \Delta_n(t) \) in two successive SA bases are given by

\[
\Delta_{n+1}(t) = \sqrt{\Omega_n^2(t) + \Delta_n^2(t)},
\]

\[
\Omega_{n+1}(t) = \frac{\Omega_n(t)\Delta_n(t) - \Omega_n(t)\Delta_n(t)}{2\Delta_{n+1}(t)}.
\]

Here \( \Delta_0(t) \equiv \Delta(t) \) and \( \Omega_0(t) \equiv \Omega(t) \) are the detuning and the coupling in the diabatic basis, whereas \( \Delta_1(t) \equiv \dot{\Delta}(t) \) and \( \Omega_1(t) \equiv \dot{\Omega}(t) \) are the quasienergy and the nonadiabatic coupling in the adiabatic basis. As far as the level crossing models in the present paper are concerned, the advantage of using the \( n \)-th SA basis is that the oscillations in the transition probability evolution, whose amplitude is proportional to the ratio \( \Omega_n(t)/\Delta_n(t) \) at large times, vanish much more quickly. This is so because at large times, the SA “detuning” \( \Delta_n(t) \) diverges in the same manner as \( \Delta(t) \), while the SA “coupling” \( \Omega_n(t) \) vanishes as \( |t|^{-n} \). Hence, the oscillation amplitude vanishes as \( |t|^{-n} \). We have used the third SA basis \((n = 3)\), in which the oscillation amplitude vanishes as \( |t|^{-5} \) for the LZ model \( \mathcal{H} \), as \( |t|^{-3} \) for the sublinear model \( \mathcal{H} \), as \( |t|^{-11} \) for the superlinear model \( \mathcal{H} \), and as \( |t|^{-3n-3} \) for the essentially nonlinear models \( \mathcal{H} \). Moreover, this approach provides the possibility to check the accuracy by calculating \( P \) in different SA bases.

As we have pointed out above, another problem, which cannot be resolved merely by the choice of basis, is the finite initial time. We have overcome it in the manner of Bambini and Lindberg \( \mathcal{H} \) by using the symmetry of the two-state problem in the \( n \)-th SA basis. The Bambini-Lindberg approach is based on a connection between the two-state evolution matrix \( \hat{U}(t, 0) \), describing the evolution from time \( t = 0 \) to time \( t \), and the evolution matrix \( \hat{U}(t, -t) \), describing the evolution from \( -t \) to \( t \). This approach allows to start the integration at \( t = 0 \), propagate it towards \( +\infty \), and stop the integration when some convergence criterion is fulfilled [for instance, we have required that the oscillation amplitude at time \( t \) is smaller than \( 10^{-4}P(t) \)]. It is easy to see that this approach is much faster than merely twice compared to the standard one (starting at large negative time), in which a convergence check would require to start the entire integration again at a larger negative time. The Bambini-Lindberg approach needs to be generalized because it applies to the case of a coupling and a detuning that are both even functions of time, while in the present case, the \( n \)-th SA “detuning” is an odd function in the diabatic basis and an even function in the SA bases, while the \( n \)-th SA “coupling” can be either an even function (for odd \( n \)) or an odd function (for even \( n \)). The relations between \( \hat{U}(t, -t) \) and \( \hat{U}(t, 0) \) for the four possible combinations of symmetries in the detuning and the coupling (odd or even functions) have been derived in \( \mathcal{H} \) in a similar manner as in \( \mathcal{H} \). We present the results in Table I. In the table, \( b_1 = b_1(+\infty) \) and \( b_2 = b_2(+\infty) \) are the so-called fundamental solutions in the corresponding \( n \)-th SA basis, i.e., the solutions for the initial conditions \( b_1(0) = 1 \) and \( b_2(0) = 0 \) at \( t = 0 \). As we can see from the table, in the different SA bases, the transition probability is expressed in terms of the fundamental solutions differently. Moreover, depending on the SA basis, the desired probability for nonadiabatic transitions is equal to the probability of no transitions (for \( n = 0, 2, 4, \ldots \)) or to the transition probability (for \( n = 1, 3, 5, \ldots \)).

The last case in Table I, in which both the “coupling” and the “detuning” are odd functions of time and which is given for the sake of completeness, is not interesting.
in the context of the present paper but it represents an interesting effect called symmetry forbidden transitions, in which the system returns to its initial state in the end of the interaction \[\Omega_n(-t) = \Omega_n(t)\]. This return is only determined by the symmetry of the Hamiltonian and does not depend on its particular details. Moreover, it remains valid in the general \(N\)-state case as well.

| Symmetry | \(U_{n}(+\infty, -\infty)\) | Cases | \(P\) |
|----------|-----------------|------|------|
| \(\Omega_n(-t) = \Omega_n(t)\) | \(U_{11} = b_1^2 + b_2^2\) | odd \(n\) | \(U_{12}^2\) |
| \(\Delta_n(-t) = \Delta_n(t)\) | \(U_{12} = 2i\text{Im}(b_1 b_2)\) | | |
| \(\Omega_n(-t) = -\Omega_n(t)\) | \(U_{11} = |b_1|^2 - |b_2|^2\) | \(n = 0\) | \(U_{11}^2\) |
| \(\Delta_n(-t) = -\Delta_n(t)\) | \(U_{12} = -2b_1 b_2\) | even \(n\) | \(U_{11}^2\) |
| \(\Omega_n(-t) = -\Omega_n(t)\) | \(U_{11} = 1\) | | |
| \(\Delta_n(-t) = -\Delta_n(t)\) | \(U_{12} = 0\) | | |

TABLE I. The relations between the fundamental solutions \(b_1(+\infty)\) and \(b_2(+\infty)\) in the \(n\)-th superadiabatic basis and the probability \(P\) for nonadiabatic transitions. In the elements of the evolution matrix \(U_{n}(+\infty, -\infty)\) in the \(n\)-th SA basis as well as in the fundamental solutions we have omitted for simplicity the arguments \(\pm \infty\) and the label \(n\).

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\(^{a}\) Electronic address: vitanov@rock.helsinki.fi

\(^{b}\) Permanent address: Department of Physics, University of Helsinki, PL 9, FIN-00014 Helsingin yliopisto, Finland

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