Time ordering in kicked qubits

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Abstract

We examine time ordering effects in strongly, suddenly perturbed two-state quantum systems (kicked qubits) by comparing results with time ordering to results without time ordering. Simple analytic expressions are given for state occupation amplitudes and probabilities for singly and multiply kicked qubits. We investigate the limit of no time ordering, which can differ in different representations.

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I. INTRODUCTION

There are two reasons to consider time ordering in kicked qubits. First, the behavior of a two-state quantum system interacting with a rapidly changing external field, i.e. a diabatically changing qubit, may be described analytically. While such solutions were examined some forty years ago in the context of Landau-Zener transitions in atomic and molecular reactions [1], relatively little attention has been paid to this problem in the context of more recent work using two-state systems [2] ranging from quantum computing [3, 4] to quantum control of atomic and molecular reactions [5] to manipulation of matter waves [6], where this class of analytic solutions may be useful. The second reason is that time ordering has been used recently [7, 8, 9] to formulate an understanding of time correlation in multi-particle systems (or, in the context of this paper, systems of interacting qubits). The central question here is how one particle (or qubit) is connected with other particles (or qubits) in the time domain. This problem has previously been formulated [8] using second order perturbation theory, where observable time correlations between different particles arise from time ordering of weak, external interactions in atomic scattering [10]. The kicked qubit gives us an opportunity to study the nature of time ordering for a simple, analytically tractable system in a strongly non-perturbative regime.

Except for relatively simple $e^{-iE_jt/\hbar}$ phases, where the $E_j$ are simple eigenvalues of a time independent Hamiltonian, there are only two ways in which time enters the time evolution of a quantum system. The first is through the explicit time dependence of an external interaction $\hat{V}(t)$, and the second is through the constraint of time ordering imposed by the time dependent Schrödinger equation itself. This time ordering, discussed below, imposes a causal-like constraint that places operators such as $\hat{H}(t_n)\ldots\hat{H}(t_2)\hat{H}(t_1)$ in order of increasing time. This confining condition interrelates the influence of the time parameters $t_n\ldots t_1$. In second order perturbation theory it has been shown [10] that the time ordering constraint has negligible effect if either $\hat{V}(t)$ or its variation with time is sufficiently small over the time of the experiment (perturbative or constant potential limit) or if the energy levels of the system before perturbation are all nearly the same (degeneracy limit). In either case, principal value contributions from energy fluctuations in short-lived intermediate states vanish [11].

In this paper we formulate the problem of time ordering in a non-perturbative two-state quantum system, i.e. a qubit. After describing the basic formalism in Section II A...
in Section II B we define the limit without time ordering and show that time ordering disappears in either the constant potential limit or in the limit of degeneracy of the two unperturbed states, where $\hat{H}(t')$ and $\hat{H}(t'')$ commute. We discuss the relationship between time ordering and the adiabatic approximation. Analytic solutions for singly and multiply kicked qubits are presented in Section II C with and without time ordering. In the case of a single kick, we show how time ordering affects the transition probability from one state to another. In the subsequently discussed case of a double kick, any transition is due entirely to time ordering effects. We discuss corrections for pulses of finite duration and in Section III provide calculations illustrating our results. We present most calculations and some key formulas in both the Schrödinger and intermediate (or interaction) pictures, and discuss some differences between time ordering effects in the two pictures.

II. THEORY

A. Basic formulation

Consider a two-state system, whose states are coupled by a time dependent external interaction, e.g. a qubit with “on” and “off” states. The time dependent Hamiltonian for this system may be expressed as

$$\hat{H}(t) = \hat{H}_0 + \hat{V}(t)$$

$$= \begin{bmatrix} -\Delta E/2 & 0 \\ 0 & \Delta E/2 \end{bmatrix} + \begin{bmatrix} 0 & V(t) \\ V(t) & 0 \end{bmatrix}$$

$$= -\frac{\Delta E}{2}\sigma_z + V(t)\sigma_x,$$

where $\Delta E = E_2 - E_1$ is the energy difference of the eigenstates of $\hat{H}_0$. Here $\sigma_x$ and $\sigma_z$ are the usual Pauli spin matrices.

Two simplifying, but removable, assumptions have been made in the second line. First, we assume that all of the time dependence in the interaction operator $\hat{V}(t)$ is contained in a single real function of $t$, which is often justifiable on experimental grounds [12, 13, 14]. Secondly, in this paper we assume for convenience that the interaction does not contain a term proportional to $\hat{H}_0$. Obviously, an interaction operator $\hat{V}$ having the form of a combination of $\sigma_x$ and $\sigma_y$ (i.e. a complex time dependent field) is equivalent to the above
form after rotation of coordinates. As we shall discuss later, there are other choices of how
to separate $\hat{H}$ into $\hat{H}_0 + \hat{V}$, and these choices have consequences.

For a qubit with “on” and “off” states $\begin{bmatrix} 1 \\ 0 \end{bmatrix}$ and $\begin{bmatrix} 0 \\ 1 \end{bmatrix}$, the probability amplitudes then evolve according to

$$\frac{\hbar}{i} \frac{d}{dt} \begin{bmatrix} a_1 \\ a_2 \end{bmatrix} = \begin{bmatrix} -\Delta E/2 & V(t) \\ V(t) & \Delta E/2 \end{bmatrix} \begin{bmatrix} a_1(t) \\ a_2(t) \end{bmatrix}.$$  

The solution to Eq. (2) may be written in terms of the time evolution matrix $\hat{U}(t)$ as

$$\begin{bmatrix} a_1(t) \\ a_2(t) \end{bmatrix} = \hat{U}(t) \begin{bmatrix} a_1(0) \\ a_2(0) \end{bmatrix} = \begin{bmatrix} U_{11}(t) & U_{12}(t) \\ U_{21}(t) & U_{22}(t) \end{bmatrix} \begin{bmatrix} a_1(0) \\ a_2(0) \end{bmatrix},$$  

where an experiment is begun at a time $t = 0$ and completed at $t = T_f$. Since we assume the two-state system is closed, $P_1(t) + P_2(t) = |a_1(t)|^2 + |a_2(t)|^2 = 1$.

The time evolution operator $\hat{U}(t)$ may be expressed here as

$$\hat{U}(t) = Te^{-\frac{i}{\hbar} \int_0^t \hat{H}(t')dt'} = Te^{-\frac{i}{\hbar} \int_0^t (-\Delta E \sigma_z + V(t') \sigma_z) dt'}$$  

$$= T \sum_{n=0}^{\infty} \frac{(-i/\hbar)^n}{n!} \int_0^t \hat{H}(t_n) dt_n \ldots \int_0^t \hat{H}(t_2) dt_2 \int_0^t \hat{H}(t_1) dt_1.$$  

The only non-trivial time dependence in $\hat{U}(t)$ arises from time dependent $\hat{H}(t)$ and time ordering $T$. The Dyson time ordering operator $T$ specifies that $\hat{H}(t_i)\hat{H}(t_j)$ is properly ordered:

$$T \hat{H}(t_i)\hat{H}(t_j) = \hat{H}(t_i)\hat{H}(t_j) + \theta(t_j - t_i) \left[ \hat{H}(t_j), \hat{H}(t_i) \right].$$  

Time ordering imposes a connection between the effects of $\hat{H}(t_i)$ and $\hat{H}(t_j)$ and leads to observable, non-local, time ordering effects [12, 13, 14] when $[\hat{H}(t_j), \hat{H}(t_i)] \neq 0$.

1. **Pulses**

In this paper we regard $V(t)$ as having the form of a smoothly varying pulse, or sequence of pulses, each of duration $\tau$ and peaked at $T_k$. We define phase angles

$$\alpha = \int_0^{T_f} V(t') dt'/\hbar$$  

$$\beta = \tau \Delta E/2\hbar$$  

$$\gamma t = t \Delta E/2\hbar.$$  

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The angle $\alpha$ is a measure of the strength of the interaction $V(t)$ over the duration of a given pulse. In this paper we are mostly interested in the non-perturbative regime corresponding to $\alpha \geq 1$, so that substantial changes in the state occupation probabilities, $P_1$ and $P_2$, may occur. The angle $\beta$ is a measure of the influence of $\hat{H}_0$ during the interaction interval $\tau$. The angle $\gamma t$ is the phase accumulation of the propagation due to $\hat{H}_0$ over a time $t$. The diabatic (kicked) limit corresponds to $\beta \ll 1$, the perturbative limit corresponds to $\alpha \ll 1$, and the adiabatic (slow) limit generally corresponds to $\tau \rightarrow \infty$.

B. Time ordering

Since time ordering effects can be defined as the difference between a result with time ordering and the corresponding result in the limit of no time ordering, it is useful to specify carefully the limit without time ordering. Removing time ordering corresponds to replacing $T \rightarrow 1$ in Eq. (4). This corresponds to the zeroth order term in an eikonal-like, Magnus expansion in commutator terms [15]. In the limit of no time ordering, a multi-particle time evolution operator factorizes into a product of single-particle evolution operators [8].

1. Limit of no time ordering

Replacing $T$ with 1 in Eq. (4), in the Schrödinger picture we have,

$$\hat{U}(t) = T e^{-\frac{1}{\hbar} \int_0^t \hat{H}(t')dt'} \rightarrow \sum_{n=0}^{\infty} \left(\frac{-i}{\hbar}\right)^n \left[ \int_0^t \hat{H}(t')dt' \right]^n = \sum_{n=0}^{\infty} \left(\frac{-i}{\hbar}\right)^n \left[ \hat{H}_0 t + \int_0^t \hat{V}(t')dt' \right]^n = \sum_{n=0}^{\infty} \left(\frac{-i}{\hbar}\right)^n \left[ (\hat{H}_0 + \hat{V}) t \right]^n = e^{-i\hat{H}_0 t/\hbar} = \hat{U}^0(t),$$

(6)

where

$$\hat{V} t = \int_0^t \hat{V}(t')dt' = \int_0^t V(t')dt' \sigma_x = \alpha \hbar \sigma_x,$$

$$\hat{H} = \hat{H}_0 + \hat{V},$$

and $[\hat{H}_0, \hat{V}]$ terms are non-zero. By expanding in powers of $[\hat{H}(t''), \hat{H}(t')]$, it is straightforward to show that to leading order in $\hat{V}$ and $\hat{H}_0$ the time ordering effect is given by

$$\hat{U} - \hat{U}^0 \simeq -\frac{1}{2\hbar^2} \int_0^t dt'' \int_0^{t''} dt' \left[ \hat{H}(t''), \hat{H}(t') \right] = -\frac{1}{2\hbar^2} \left[ \hat{H}_0, \hat{V}_0 \right] \int_0^t dt'(t - 2t') f(t'),$$

(7)
where $\hat{V}(t') = \hat{V}_0 f(t')$. This leading term disappears if the pulse centroid $T_k = t/2$ and $f(t')$ is symmetric about $T_k$. Furthermore, $\hat{U} - \hat{U}^0$ vanishes identically in the special cases of $V(t') = 0, V(t') = \hat{V}$, or $\Delta E = 0$, as will be discussed below in Section IIIB2.

In general there is no simple analytic form for the exact result $\hat{U}(t)$. For the result without time ordering, we have

$$\hat{U}^0(t) = e^{i\gamma t \sigma_z - i\alpha \sigma_x} = \begin{bmatrix} \cos \xi + i\gamma t \sin \xi \frac{\xi}{\xi} & -i\alpha \sin \xi \frac{\xi}{\xi} \\ -i\alpha \sin \xi \frac{\xi}{\xi} & \cos \xi - i\gamma t \sin \xi \frac{\xi}{\xi} \end{bmatrix},$$

where $\xi = \sqrt{\alpha^2 + (\gamma t)^2}$. Here we have used the well known identity $e^{i\phi \sigma \cdot \hat{n}} = \cos \phi + i\sigma \cdot \hat{n} \sin \phi$, following from $(\sigma \cdot \hat{n})^n = 1$ (or $\sigma \cdot \hat{n}$) if $n$ is even (or odd).

Similarly, in the intermediate, or interaction, picture, $\hat{U}_I(t) = e^{i\hat{H}_0 t} \hat{U}(t)$, and one has

$$\hat{U}_I(t) = Te^{-\frac{i}{\hbar} \int_0^t \hat{V}_I(t') dt'} e^{-\frac{i}{\hbar} \int_0^t \hat{V}_I(t') dt'} \Rightarrow \sum_{n=0}^{\infty} \frac{(-i/\hbar)^n}{n!} \left[ \int_0^t \hat{V}_I(t') dt' \right]^n \Rightarrow \sum_{n=0}^{\infty} \frac{(-i/\hbar)^n}{n!} \left[ \hat{V}_I(t) \right]^n = \hat{U}_I^0(t),$$

where $\hat{V}_I(t') = e^{i\hat{H}_0 t'/\hbar} \hat{V}(t') e^{-i\hat{H}_0 t'/\hbar}$ and $\hat{V}_I t = \int_0^t \hat{V}_I(t') dt'$. For a Gaussian pulse of the form discussed in Section IIIB

$$\hat{V}_I t = \alpha \hbar e^{-\beta^2} [\sigma_x \cos 2\gamma T_k + \sigma_y \sin 2\gamma T_k]$$

(10)

and

$$\hat{U}_I^0(t) = \begin{bmatrix} \cos (\alpha e^{-\beta^2}) & -i \sin \left(\alpha e^{-\beta^2}\right) e^{-i\gamma T_k} \\ -i \sin \left(\alpha e^{-\beta^2}\right) e^{i\gamma T_k} & \cos (\alpha e^{-\beta^2}) \end{bmatrix}$$

(11)

as long as the measurement time $t$ is after the completion of the pulse, i.e. $t - T_k \gg \tau$.

It has been shown previously [10] that to second order in perturbation theory, $\hat{U}_I - \hat{U}_I^0 \sim [\hat{V}_I(t''), \hat{V}_I(t')]$, somewhat similar to the commutator in the Schrödinger picture above. From this we immediately see that time ordering effects do not appear until second order in a perturbative expansion in $\alpha$. Again, $\hat{U}_I - \hat{U}_I^0 \to 0$ in the special limits $\hat{V}_I \to 0, \hat{V}_I \to \hat{V}_I$, or $\Delta E \to 0$, to be discussed immediately below. However, we will also find in Section IIIB that $\hat{U}_I - \hat{U}_I^0$ vanishes in the diabatic limit of a single ideal kick, $V(t) \sim \delta(t - T_k)$, whereas $\hat{U} - \hat{U}^0$ is non-zero. Thus, in principle, the definition of the limit of no time ordering depends on the picture (representation) used. In the discussion we shall relate this difference to the
gauge choice of how one separates $\hat{H}$ into $\hat{H}_0 + \hat{V}(t)$. As shown in calculations presented below, this difference can be negligibly small under some conditions.

In both pictures, time ordering effects are associated with the fluctuation of a time-dependent interaction about its time averaged value.

2. Relation to other limiting cases

Now we compare the limit without time ordering with the degenerate, weakly varying potential, and adiabatic limits. The connection with the diabatic (kicked) limit appears in Section II C, where we discuss the analytic solution for $\hat{U}$ in the case of a short pulse.

We noted above that for a general pulse, there is no analytic solution for $\hat{U}(t)$. However, in the limit when the unperturbed states become nearly degenerate, i.e. $\Delta E \ll \hbar/T_f$, we obtain

$$\hat{U}(t) \rightarrow \hat{U}^D(t) = e^{-i\int_0^t \hat{V}(t')dt'/\hbar} = e^{-i\alpha}\begin{bmatrix} \cos \alpha & -i\sin \alpha \\ -i\sin \alpha & \cos \alpha \end{bmatrix}.$$ (12)

This result may be obtained either from the coupled state equations of Eq. (2), or from Eq. (4) with $\Delta E \rightarrow 0$. In this degenerate limit the mathematical complexity of the qubit simplifies significantly, as may be seen by comparing Eq. (12) with Eqs. (15) and (17). Most of the complex time connections have been removed. We call this degenerate qubit a dit. In such a dit if the phase angle $\alpha$ equals $\pi/2$, then an “on” state is turned off and an “off” state is turned on with probability 1. Then the dit is further reduced in complexity to a trivial classical bit.

We notice that Eq. (12) may also be obtained from Eq. (8) by taking $\gamma \rightarrow 0$. Thus, time ordering effects vanish in the degenerate limit.

A second situation in which time ordering effects generically become small is the case of a constant or weakly varying potential, $|\hat{V} - \hat{\bar{V}}| \ll \hbar/T_f$. Then the full evolution matrix approaches the non-time-ordered expression given by Eq. (8) or Eq. (9). Clearly the perturbative limit $\alpha \ll 1$ is a special case of this, but $\hat{U} - \hat{U}^0$ vanishes also for a large average external potential $\hat{\bar{V}}$, as long as fluctuations around $\hat{\bar{V}}$ are small. Such a situation may sometimes be addressed more transparently by absorbing the average part of $\hat{\bar{V}}$ into $\hat{H}_0$. In
the extreme case $\dot{V} = 0$, Eq. (8) reduces to

$$\hat{U}^0(t) \to e^{-i\hat{H}_0 t/\hbar} = \begin{bmatrix} e^{i\gamma t} & 0 \\ 0 & e^{-i\gamma t} \end{bmatrix},$$  \hspace{1cm} (13)$$

which of course agrees with the exact evolution matrix $\hat{U}(t)$.

In summary, time ordering effects disappear either when (i) $\alpha \ll 1$ or when (ii) $\beta \ll 1$ ($\gamma T_f \ll 1$) in the intermediate (Schrödinger) picture. The physics becomes especially simple in the overlap of regimes (i) and (ii). There, one easily finds that in the Schrödinger picture, for example,

$$\hat{U}(t) \simeq \hat{U}^0(t) \simeq \begin{bmatrix} 1 + i\gamma t & -i\alpha \\ -i\alpha & 1 - i\gamma t \end{bmatrix}. \hspace{1cm} (14)$$

It can be shown that corrections and time ordering effects start at $O(\alpha \gamma^2 t^2)$ and $O(\alpha^2 \gamma t)$ for a symmetric pulse centered at $T_k = t/2$ (see Eq. (23)). The situation is similar in the intermediate picture, except that time ordering effects vanish identically at leading order in $\alpha$, and begin at $O(\alpha^2 \gamma t)$ only.

Strictly speaking, time ordering effects also vanish if (iii) $[\hat{H}_0, \dot{V}] = 0$; however this situation is of little practical interest due to the fact that no transition or population transfer is possible.

In addition to the degenerate, perturbative, and diabatic regimes, a fourth limit exists in which analytic solutions for $\hat{U}(t)$ are generally available. In the adiabatic limit where the external interaction $V(t)$ changes slowly in time, it is useful to define the instantaneous level splitting $\Omega(t) = \sqrt{(\Delta E)^2 + 4V^2(t)}$ and the accumulated phase $\theta = \int_0^t \Omega(t') dt'/2\hbar$. Then

$$\hat{U}(t) \to \hat{U}^A(t)$$ \hspace{1cm} (15)

$$= \begin{bmatrix} \cos\theta \cos\phi_- + i\sin\theta \cos\phi_+ & \cos\theta \sin\phi_- - i\sin\theta \sin\phi_+ \\ -\cos\theta \sin\phi_- - i\sin\theta \sin\phi_+ & \cos\theta \cos\phi_- + i\sin\theta \cos\phi_+ \end{bmatrix}.$$  \hspace{1cm} (16)

Here $\phi_{\pm} = (\phi(t) \pm \phi(0))/2$ where $\phi(t') = \tan^{-1}(2V(t')/\Delta E)$. In the special case when $V(t) = V(0)$, relevant for a pulse,

$$\hat{U}^A(t) = \begin{bmatrix} \cos\theta(t) + i\frac{\Delta E}{\Omega(t)} \sin\theta(t) & -2i\frac{V(t)}{\Omega(t)} \sin\theta(t) \\ -2i\frac{V(t)}{\Omega(t)} \sin\theta(t) & \cos\theta(t) - i\frac{\Delta E}{\Omega(t)} \sin\theta(t) \end{bmatrix}. \hspace{1cm} (16)$$

We note that this solution is similar mathematically to the rotating wave approximation (RWA), which is widely used to describe atomic transitions using external fields tuned to
frequencies near the resonant transition frequency between two states [19, 20, 21]. Inserting Eq. (15) into Eq. (2), one finds that the leading correction is small when $\hbar \dot{V}(t') \Delta E \ll \Omega^3(t')$. When the splitting $\Delta E$ of the unperturbed qubit is not small, i.e. $\Delta E \geq V$, then this adiabatic validity condition reduces to the Landau-Zener criterion [22], namely, $\hbar \dot{V}(t') \ll (\Delta E)^2$. The leading correction to Eq. (15) or Eq. (16) is then given by non-adiabatic transitions at the avoided level crossings, where $V(t')$ and thus the level splitting $\Omega(t')$ goes through a minimum. More generally, for a pulse having a smooth shape, such as the Gaussian pulses discussed in Section III, the criterion $\hbar \dot{V}(t') \Delta E \ll \Omega^3(t')$ reduces to the union of $\alpha \ll \beta^2$ and $\beta \ll \alpha^2$.

Remarkably, the adiabatic regime overlaps both with the degenerate limit (when $\beta, \gamma T_f \to 0$ at fixed $\alpha$) and with the perturbative limit (when $\alpha \to 0$ at fixed $\beta, \gamma T_f$). Thus, Eq. (15) reduces either to Eq. (12) or Eq. (13) as $\Delta E \to 0$ or $V \to 0$, respectively. In these overlap regions, time ordering effects are small. More generally, however, time ordering effects in the purely adiabatic regime ($\alpha \gg 1$ and $\beta \geq 1$ or $\beta \gg 1$ and $\alpha \geq 1$) are large. Time ordering effects are also large when $\alpha$ and $\beta$ are both of order unity, where no simple analytic solutions exist for the full evolution matrix $\hat{U}(t)$.

C. Kicked qubits

Now we consider time ordering in kicked qubits, i.e. the diabatic limit where $V(t) \sim \delta(t - T_k)$. First, we present analytic expressions [23, 24] for a kicked qubit, i.e. a two state system subject to an external interaction, $V(t)$, that changes rapidly with respect to $T_{\Delta E} = \pi/\gamma = 2\pi \hbar/\Delta E$, the period of oscillation of the free system. The corrections needed for finite pulses will be briefly analyzed. We also discuss the extension to multiple kicks, using the double kick as an example. Finally, we consider the influence of time ordering in these kicked systems.

1. Single kick

Here we consider a two-state system where the interaction $V(t)$ may be expressed as a sudden “kick” at $t = T_k$, namely $V(t) = \alpha \hbar \delta(t - T_k)$. For such a kick the integration over
time is trivial and the time evolution matrix in Eq. (4) becomes
\[ \hat{U}_{K}(t) = e^{i\Delta E_{2}(t-T_{k})/\hbar} e^{-i\int_{T_{k}}^{t} V(t')dt'/\hbar} e^{i\Delta E_{2}T_{k}\sigma_{z}/\hbar} \]
\[ = e^{i\gamma(t-T_{k})} \begin{bmatrix} 0 & e^{-i\gamma(t-T_{k})} \\ e^{i\gamma(t-T_{k})} & 0 \end{bmatrix} \begin{bmatrix} \cos \alpha & -i \sin \alpha \\ -i \sin \alpha & \cos \alpha \end{bmatrix} \begin{bmatrix} e^{i\gamma T_{k}} & 0 \\ 0 & e^{-i\gamma T_{k}} \end{bmatrix} \]
\[ = \begin{bmatrix} e^{i\gamma t} \cos \alpha & -ie^{i\gamma T_{k}} \sin \alpha \\ -ie^{-i\gamma T_{k}} \sin \alpha & e^{-i\gamma t} \cos \alpha \end{bmatrix} \]
\[ = \begin{bmatrix} \cos \alpha' & -i \sin \alpha' \\ -i \sin \alpha' & \cos \alpha' \end{bmatrix} \]
(17)
for \( t > T_{k} \). The second line follows from the identity given below Eq. (8) above. As explained below this solution is valid when \( \beta \ll 1 \) so that there is little effect from \( \hat{H}_{0} \) during the short time when \( \hat{V} \) is active.

From Eqs. (3) and (17) we have for a kicked qubit initially found in state 1:
\[ P_{1}(t) = |a_{1}(t)|^{2} = |U_{11}^{K}(t)|^{2} = \cos^{2} \alpha \]
\[ P_{2}(t) = |a_{2}(t)|^{2} = |U_{12}^{K}(t)|^{2} = \sin^{2} \alpha . \]
(18)
The corresponding probabilities for a kicked qubit without time ordering are discussed below in Section II C 4.

2. Finite pulse corrections for a single pulse

When the pulse width is finite the corrections to Eq. (17) are \( O(\beta) \) and corrections to Eq. (18) are \( O(\beta^{2}) \). These corrections result from the commutator of the free Hamiltonian \( \hat{H}_{0} \) with the interaction \( \hat{V} \) during the time \( \tau \) when the pulse is active. This is related to the series expansion that arises in the split operator method [25]. For example, in the case of a rectangular pulse of width \( \tau \), the exact time evolution is given by
\[ \hat{U}_{\text{rectangular}}(t) = e^{i\gamma t-i\beta \left( \cos \alpha' + i\beta \frac{\sin \alpha'}{\alpha} \right) -ie^{i\gamma T_{k} \alpha \sin \alpha'} + e^{-i\gamma T_{k}} -ie^{-i\gamma (t-2T_{k})} \alpha \sin \alpha'} -e^{-i\gamma t+i\beta \left( \cos \alpha' - i\beta \frac{\sin \alpha'}{\alpha} \right) \alpha} \],
(19)
where \( \alpha' = \sqrt{\alpha^{2} + \beta^{2}} \). To leading order in \( \beta \), i.e. in the width of the pulse, the error in the kicked approximation is given by
\[ \delta \hat{U}(t) = \hat{U}_{\text{rectangular}}(t) - \hat{U}_{K}(t) = i\beta \left( \sin \frac{\alpha}{\alpha} - \cos \alpha \right) \begin{bmatrix} e^{i\gamma t} & 0 \\ 0 & e^{-i\gamma t} \end{bmatrix} . \]
(20)
For a narrow pulse having a generic symmetric shape, the leading correction to the kicked approximation will still have the form

\[
\delta \hat{U}(t) = \hat{U}(t) - \hat{U}^K(t) = i\beta g(\alpha) \begin{bmatrix}
e^{i\gamma t} & 0 \\
0 & -e^{-i\gamma t}
\end{bmatrix},
\]

(21)

where \(g(\alpha)\) is now a function that depends on the shape of the pulse. By comparing Eqs. (4) and (17) at leading order in \(\Delta E\), after some algebra one obtains

\[
g(\alpha) = \frac{2}{\tau} \int dt \left[ \cos^2 \left( \int_{t_k}^{t} V(t') dt'/\hbar \right) - \cos^2(\alpha/2) \right].
\]

(22)

Expanding \(\hat{U}(t)\) of Eq. (4) and \(\hat{U}^K(t)\) of Eq. (17) simultaneously in \(\Delta E\) and \(V\), or equivalently in \(\beta\) and \(\alpha\), we find

\[
\delta \hat{U}(t) = \hat{U}(t) - \hat{U}^K(t) = i\frac{\Delta E}{\hbar} \int dt \left[ \left( \frac{\alpha}{2} \right)^2 - \left( \int_{t_k}^{t} V(t') dt'/\hbar \right)^2 \right] \begin{bmatrix}
e^{i\gamma t} & 0 \\
0 & -e^{-i\gamma t}
\end{bmatrix}
\]

\[
+ \frac{i(\Delta E)^2}{2} \int V(t')(t' - T_k)^2 dt'/\hbar^3 \begin{bmatrix}
0 & e^{i\gamma(t-2T_k)} \\
-2\gamma(t-2T_k) & 0
\end{bmatrix},
\]

so the two leading correction terms scale as \(\beta\alpha^2\) and \(\beta^2\alpha\).

3. **Multiple kicks**

A series of either identical or non-identical pulses can easily be handled by multiplication of several matrices of the form of Eq. (17). For example, one may consider a sequence of two kicks of opposite sign at times \(t = T_1\) and \(t = T_2\), namely, \(V_{\text{kick antikick}}(t) = \alpha \hbar \delta(t - T_1) - \alpha \hbar \delta(t - T_2)\). Following the procedure given in Eq. (17) one obtains the time evolution matrix for \(t > T_2\),

\[
\hat{U}_{\text{kick antikick}}(t) = e^{i\gamma(t-T_2)\sigma_z} e^{i\alpha\sigma_x} e^{i\gamma(T_2-T_1)\sigma_z} e^{-i\alpha\sigma_x} e^{i\gamma T_1 \sigma_z}
\]

(24)

\[
= \begin{bmatrix}
e^{i\zeta(\cos \gamma T_s + i \sin \gamma T_s \cos 2\alpha)} & e^{i\gamma(t-2T)} \sin \gamma T_s \sin 2\alpha \\
-2\gamma(t-2T) \sin \gamma T_s \sin 2\alpha & e^{-i\zeta(\cos \gamma T_s - i \sin \gamma T_s \cos 2\alpha)}
\end{bmatrix},
\]

where \(\zeta = \gamma(t - T_s), \bar{T} = (T_1 + T_2)/2\), and \(T_s = T_2 - T_1\). As \(\gamma T_s \to 0\), \(\hat{U}_{\text{kick antikick}}(t)\) reduces to Eq. (13).
For a double kick with \( V = 0 \), we have from Eqs. (3) and (24),

\[
P_1(t) = |a_1(t)|^2 = |U_{11}^{\text{kick antikick}}(t)|^2 = \cos^2 \gamma T_s + \sin^2 \gamma T_s \cos^2 2\alpha \\
P_2(t) = |a_2(t)|^2 = |U_{12}^{\text{kick antikick}}(t)|^2 = \sin^2 \gamma T_s \sin^2 2\alpha .
\]  

(25)

The single-kick result of Eq. (17) remains valid for two or more kicks of combined strength \( \alpha_1 + \ldots + \alpha_n = \alpha \) if the total phase associated with the inter-kick free evolution, \( \gamma(T_n - T_1) \), is small. A mathematical analysis for multiple kicks separated by arbitrary time intervals is straightforward, but not included here. In the case of a periodic series of pulses with period \( T \), the time evolution may be obtained by diagonalizing the matrix of Eq. (17) and finding the Floquet eigenstates and eigenphases. The two Floquet eigenphases are then given by \( e^{\pm i\chi} \), where \( \chi = \cos^{-1}[\cos \alpha \cos \gamma T] \).

4. **Time ordering for single and multiple kicks or pulses**

We first consider the case of a single kick or pulse. In the Schrödinger picture, time ordering effects are present even for a single ideal kick, specifically the time ordering between the interaction and the free evolution preceding and following the kick. Thus, in the absence of time ordering, the time evolution \( \hat{U}^0(t) \) is given by Eq. (3), which differs from the exact expression \( \hat{U}^K(t) \) of Eq. (17) when \( \alpha \) and \( \gamma t \) are both non-zero. The time ordering effect \( \hat{U}^K(t) - \hat{U}^0(t) \) vanishes in either the degenerate limit \( \gamma t \to 0 \) or in the perturbative limit \( \alpha \to 0 \). For small \( \alpha \) and \( \gamma t \) and assuming \( T_k = t/2 \), the time ordering effect at leading order takes the form of a sum of \( O(\alpha(\gamma t)^2) \) and \( O(\alpha^2\gamma t) \) terms. For a Gaussian-shaped pulse, the transition probability in the Schrödinger picture without time ordering is given by the second line of Eq. (29).

In the intermediate picture, time evolution without time ordering for an ideal kick is obtained by substituting \( \beta = 0 \) into Eq. (11), and agrees perfectly with the exact expression of Eq. (17), when the latter is transformed into the intermediate picture. Thus, time ordering effects disappear for a single ideal kick in the intermediate picture, in contrast with the Schrödinger case. This is easily understood by considering that in the intermediate picture, time ordering is only between interactions at different times, \( \hat{V}_I(t') \) and \( \hat{V}_I(t'') \), not between the interaction \( \hat{V}(t') \) and the free Hamiltonian \( \hat{H}_0(t'') \), as in the Schrödinger case. For a single ideal kick, all the interaction occurs at one instant, and no ordering is needed. Of
course, for a finite-width pulse, i.e. \( \beta \neq 0 \), time-ordering effects do begin to appear even in the intermediate picture. To leading order, \( \hat{U}^K_f(t) - \hat{U}^\gamma_f(t) = O(\alpha^2 \beta) \). We note that the time ordering effect in the intermediate picture is independent of the measurement time \( t \), though it does depend on the pulse width \( \tau \) through the \( \beta \) parameter. For a Gaussian-shaped pulse, the transition probability in the intermediate picture without time ordering is given by the third line of Eq. (29).

We are now ready to examine the time ordering effect for a multi-pulse sequence, focusing on the pulse-antipulse scenario of Section II C 3. In the limit of no time ordering one has \( P^0_2(t) = 0 \) in the Schrödinger picture as seen from Eqs. (3) and (13) with \( \bar{V} = 0 \). In the intermediate picture, however, the transition probability is non-zero even without time ordering. For a Gaussian pulse-antipulse sequence, one may show that \( \bar{V}_I t = 2\alpha e^{-\beta^2} \sin \gamma T_s [\sigma_x \sin 2\gamma \bar{T} - \sigma_y \cos 2\gamma \bar{T}] \), where \( \bar{T} = (T_1 + T_2)/2 \). Note that for \( t - T_2 \gg \tau \), \( \bar{V}_I t \) depends on \( T_1 \) and \( T_2 \) but not \( t \). Then, \( e^{-i\bar{V}_I t/\hbar} = \cos [2\alpha e^{-\beta^2} \sin \gamma T_s/2] - i [\sigma_x \sin 2\gamma \bar{T} - \sigma_y \cos 2\gamma \bar{T}] \cdot \sin [2\alpha e^{-\beta^2} \sin \gamma T_s] \). Consequently,

\[
\hat{U}^0_f(t) = \begin{bmatrix}
\cos[2\alpha e^{-\beta^2} \sin \gamma T_s] & e^{-2i\gamma \bar{T}} \sin[2\alpha e^{-\beta^2} \sin \gamma T_s] \\
-e^{2i\gamma \bar{T}} \sin[2\alpha e^{-\beta^2} \sin \gamma T_s] & \cos[2\alpha e^{-\beta^2} \sin \gamma T_s]
\end{bmatrix}.
\] (26)

We use this result in the next section to study the effect of time ordering on the transfer of population from one state to another.

We note that as either \( \gamma \to 0 \) or \( \alpha \to 0 \), one has \( \hat{U}^0_f(t) \to \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \) in contrast to
\( \hat{U}^0(t) \to \begin{bmatrix} e^{i\gamma t} & 0 \\ 0 & e^{-i\gamma t} \end{bmatrix} \). For simplicity, we may consider the case where each pulse is an ideal kick, i.e. \( \beta = 0 \). Then in the perturbative regime, expanding in \( \alpha \), one finds \( U^0_{11} \approx 1 - 2\alpha^2 \sin^2 \gamma T_s \), which can be compared to \( e^{-i\gamma t} U^\text{kick}_{11} \approx 1 - 2i\alpha^2 e^{-i\gamma T_s} \sin \gamma T_s \), so that except for special values of \( \gamma \) (including 0), these matrix elements differ by \( O(\alpha^2) \). Similarly, \( U^0_{12} \approx 2\alpha \sin \gamma T_s e^{-2i\gamma \bar{T}} + O(\alpha^3) \) compared to \( e^{-i\gamma t} U^\text{kick}_{12} \approx 2\alpha \sin \gamma T_s e^{-2i\gamma \bar{T}} + O(\alpha^3) \). Hence \( \hat{U}_I \) and \( \hat{U}^0 \) agree to leading order in \( \alpha \). This should not be surprising, as time ordering has no effect at leading order in perturbation theory.
III. CALCULATIONS

As an illustrative specific example we present in this section the results of numerical calculations for $2s \rightarrow 2p$ transitions in atomic hydrogen caused by a Gaussian pulse of width $\tau$. The occupation probabilities of the $2s$ and $2p$ states are evaluated by integrating two-state equations using a standard fourth order Runge-Kutta method. This enables us to verify the validity of our analytic solutions for kicked qubits in the limit $\tau \rightarrow 0$ and also to consider the effects of using finite-width pulses. In this system, the unperturbed level splitting is the Lamb shift, $\Delta E = E_{2p} - E_{2s} = 4.37 \times 10^{-6}$ eV. The corresponding time scale is the Rabi time, $T_{\Delta E} = 2\pi \hbar / \Delta E = 972 \times 10^{-12}$ s, which gives the period of oscillation between the states.

For any practical system, the pulse duration $\tau$ can neither be too large nor too small. If $\tau$ is larger than $T_{\Delta E}$, then the pulse will not be sudden and the kicked approximation will fail. On the other hand if $\tau$ is too small, then the interaction will have frequency components that couple the initial state to other levels. Specifically, if $\tau$ is less than $2\pi \hbar / (E_{3p} - E_{2s}) \approx 10^{-15}$ s, then the interaction will induce transitions into the $3p$ level and the system will not be well approximated by a two-state system. Also there is another constraint in our case. If the experiment lasts longer than the lifetime of the $2p$ state, $1.6 \times 10^{-9}$ s, then we lose population from our two-state system, i.e. dissipation cannot be neglected. Similar calculations can be done in many other applications, including, for example, Josephson junctions [4].

In the first part of this section we present results for the target state occupation probability, $P_2$, as a function of time. We shall examine how well the approximations we use are satisfied for a $2s - 2p$ transition caused by a pulse of finite width. We shall do this first for a single pulse and then for a double pulse. In the second part of this section we examine effects of time ordering. Here we shall evaluate $P_2(t)$ both with and without time ordering for pulses of finite width. This will be done in both the Schrödinger and intermediate pictures.

A. Pulsed two-state system

In our numerical calculations we use for convenience an interaction of the form $V(t) = (\alpha \hbar / \sqrt{\pi \tau}) e^{-(t-T_k)^2/\tau^2}$, i.e. a Gaussian pulse centered at $T_k$ with width $\tau$. The evaluation of the integrated pulse strength $\alpha$ in terms of the dipole matrix element for the $2s - 2p$
transition is discussed in a previous paper \[18\]. When \( \tau \) is small enough for the sudden, kicked approximation to hold, \( V(t) \rightarrow \alpha \hbar \delta(t - T_K) \), and the analytic expressions of Eqs. (17) and (24) apply. Here we shall determine how the occupation probability \( P_2(t) \) depends on the pulse width \( \tau \), to find where the kicked results are approximately valid for finite pulses.

We do this first for a single pulse chosen so that an ideal kick would transfer the occupation probability \( P_2(t) \) suddenly from zero to one at \( t = T_k \). Then we consider two equal and opposite pulses occurring at times \( T_1 \) and \( T_2 \). We study this doubly kicked system as a function of both pulse width \( \tau \) and separation interval \( T_s = T_2 - T_1 \).

1. Single pulse

![Graph](image.png)

**FIG. 1:** Occupation probability of the target state as a function of time for a qubit interacting with a single pulse. The heavy solid line corresponds to \( \tau = 1 \) ps (almost an ideal kick), the thin dashed line to \( \tau = 10 \) ps (where small deviations from an ideal kick occur), and the thin dotted line to \( \tau = 100 \) ps (where the kicked approximation is breaking down). The Rabi time for the oscillation between the states is 972 ps.

In Fig. 1 we show results of a calculation for the probability \( P_2(t) \) that a hydrogen atom initially in the 2s state makes a transition into the 2p state when strongly perturbed by a single Gaussian pulse applied at \( t = T_k \). We have obtained our results by numerically integrating the two-state coupled equations,

\[
i \hbar \dot{a}_1 = -\frac{1}{2} \Delta E a_1 + \frac{\alpha}{\sqrt{\pi \tau}} e^{-(t-T_k)^2/\tau^2} a_2
\]
\begin{equation}
\dot{a}_2 = \frac{i \hbar}{2} \Delta E a_2 + \frac{\alpha}{\sqrt{\pi \tau}} e^{-(t-T_k)^2/\tau^2} a_1.
\end{equation}

Here the pulse is applied at \(T_k = 150\) ps and we have chosen \(\alpha = \pi/2\) so that in the limit of a perfect kick all of the population will be transferred from the 2s to the 2p state after \(t = T_k\).

In Fig. 1 one sees that the ideal kick results are very nearly achieved by choosing \(\tau\) to be a factor of \(10^{-3}\) times smaller than the Rabi time, \(T_{\Delta E}\), in which the population oscillates between the 2s and 2p states. When \(\tau/T_{\Delta E} \approx 10^{-2}\), a small deviation from an ideal kick can be seen in the figure. In this case \(P_2(T_f) = 0.9977\). When \(\tau/T_{\Delta E} \approx 10^{-1}\), the transition takes a few tenths of a nanosecond to occur and only 82% of the population is transferred at 300 ps. The error in \(P_2(T_f)\) resulting from the kicked approximation grows as \((\tau/T_{\Delta E})^2 \sim \beta^2\), as expected from Eq. (23).

2. A positive followed by a negative pulse

\begin{figure}
\centering
\includegraphics{fig2}
\caption{Occupation probability of the target state as a function of time for a double pulse that returns the system to its initial state in the kicked limit. The heavy solid line corresponds to \(\tau = 1\) ps (almost ideal kicks), the thin dashed line to \(\tau = 10\) ps (where small deviations from ideal kicks occur), and the thin dotted line to \(\tau = 100\) ps (where the kicked approximation is breaking down).}
\end{figure}

In Figs. 2 and 3 we show results of a calculation for the probability \(P_2(t)\) that a hydrogen atom initially in the 2s state makes a transition into the 2p state when acted on by a double Gaussian pulse. Two Gaussian-form pulses are applied, the first at \(t = T_1\) and the second
FIG. 3: Occupation probability of the target state as a function of time for a double pulse that fully transfers population in the kicked limit. The heavy solid line corresponds to $\tau = 1$ ps (almost ideal kicks), the thin dashed line to $\tau = 10$ ps (where small deviations from ideal kicks occur), and the thin dotted line to $\tau = 100$ ps (where the kicked approximation is breaking down).

at $t = T_2$. The separation interval between pulses is $T_s = T_2 - T_1$. The final occupation probability of the target state is measured at $t = T_f$. The pulses are opposite in sign, but otherwise identical, so the interaction integrated over the whole interval $[0, T_f]$ is zero.

Our results for this double pulse have been obtained by numerically integrating

\begin{align*}
  i\hbar \dot{a}_1 &= -\frac{1}{2} \Delta E a_1 + (\alpha/\sqrt{\pi \tau}) \left[ e^{-\left(\frac{t-T_1}{\tau}\right)^2} - e^{-\left(\frac{t-T_2}{\tau}\right)^2} \right] a_2 \\
  i\hbar \dot{a}_2 &= \frac{1}{2} \Delta E a_2 + (\alpha/\sqrt{\pi \tau}) \left[ e^{-\left(\frac{t-T_1}{\tau}\right)^2} - e^{-\left(\frac{t-T_2}{\tau}\right)^2} \right] a_1 .
\end{align*}

In Fig. 2, the first pulse is applied at $T_1 = 100$ ps and the second at $T_2 = 586$ ps, giving a separation time $T_s = 486$ ps = $\pi/2\gamma$. From Eqs. (5) and (25) one sees that this is precisely the value of $\gamma T_s$ required to yield complete transfer from the 2s to the 2p state at $T_1$ and then full transfer back to the 2s state at time $T_2$ in the limit of an ideal double kick. Moreover, we have chosen an integrated pulse strength $\alpha = \pi/2$ for each pulse for the same reason.

The parameters for Fig. 3 are the same except that $\alpha = \pi/4$, so that the 2p target state is fully populated after $T_2$ for an ideally kicked system.

As in Fig. 1 we see that when $\tau/T_{\Delta E} < 10^{-3}$, the kicked limit is well satisfied, and when $\tau/T_{\Delta E} \approx 10^{-2}$, small deviations from an ideal kick can be seen in Fig. 2. In this case $P_2(T_f) = 1.1 \times 10^{-5}$. Again deviations from the ideal kick limit in the transfer probability $P_2$ grow as $(\tau/T_{\Delta E})^2$. 

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The results in Fig. 3 are for a double pulse that first takes the population halfway from $2s$ to $2p$, and then the rest of the way for an ideal kick-antikick sequence. When $\tau/T_{\Delta E} \approx 10^{-2}$, small deviations from an ideal kick can be observed in the figure near both steps. At 700 ps, $P_2(T_f) = 0.99934$, i.e. the population is nearly, but not quite perfectly transferred to the target state. When $\tau/T_{\Delta E} \approx 10^{-1}$, the transition takes a few tenths of a nanosecond to occur and only 80% of the population is transferred, i.e. the transfer is not ideal.

### B. Time ordering

In this subsection we consider the more complex issue of time ordering in $2s - 2p$ transitions in atomic hydrogen caused by a single or double Gaussian pulse. Finite-width pulse effects are again considered. The effect of time ordering is evaluated by comparing results of calculations with and without time ordering for the probability $P_2$ of transferring an electron population from the $2s$ launch state to the $2p$ target state. Since the limit without time ordering is different in the Schrödinger and intermediate pictures, we include results for both pictures.

The equations including time ordering are given by Eqs. (27) and (28) above. The analogous equations without time ordering are found by taking $V(t) \rightarrow \bar{V}$ in the Schrödinger picture and $V_I(t) \rightarrow \bar{V}_I$ in the intermediate picture.

#### 1. Single pulse

In Fig. 4 we show the effects of time ordering for a single pulse. We have chosen our parameters so that in the limit of an ideal kick the population is completely transferred from the $2s$ to the $2p$ state of hydrogen at time $T_k$, as described above. On the left hand side of Fig. 4 we show how the occupation probability of the $2p$ target state varies as a function of the Gaussian pulse width $\tau$ for three different values of the observation time $T_f$. For sharp pulses the exact transfer probability $P_2(T_f)$ and the transfer probability without time ordering in the intermediate picture $P^0_I(2p)$ are quite similar, but differences appear, as expected, when $\tau/T_{\Delta E} = \beta/\pi$ becomes large.

However in the Schrödinger picture there are very large differences between the results with and without time ordering, $P_2(T_f)$ and $P^0_2(T_f)$, even for an ideal kick. This occurs
FIG. 4: Target state probability as a function of the pulse width $\tau$ (on the left), and as a function of the observation time $T_f$ (on the right). Here $T_{\Delta E} = 2\pi \hbar / \Delta E = \pi / \gamma$ is the Rabi time for oscillations between the states, where $\Delta E = E_{2p} - E_{2s}$. The heavy line denotes probability including time ordering, the dashed line denotes the probability in the intermediate picture without time ordering, and the dotted line represents the probability in the Schrödinger picture without time ordering. On the right, the lines begin at the midpoint of the pulse, $T_f = T_k$. The Schrödinger results damp out for large $T_f$ on the right as explained in the text.

because the energy splitting $\Delta E$ is non-zero, and for $T_f > \alpha \hbar / \Delta E = \alpha T_{\Delta E}/2\pi$, the average potential $V = \alpha / T_f$ becomes smaller than the energy splitting $\Delta E$. Thus, for a given pulse, the influence of the potential necessarily decreases at large $T_f$, and any transfer probability becomes exponentially small. In effect, the free propagation before and after the pulse diminishes the effect of the pulse itself in the Schrödinger picture, when time ordering is
removed. This behavior contrasts with the intermediate picture result (Eq. (11)), where $P^0_{I_2}(T_f)$ depends on $\beta = \gamma \tau$ but not on $T_f$, as seen also on the left side of Fig. 4. The contrast is evident on the right hand side of Fig. 4 where, after the pulse has died off, the value of $P^0_2(T_f)$ dies out as $T_f$ increases, while $P^0_{I_2}(T_f)$ approaches a constant.

For a single narrow pulse one may compare the time-ordered result for the transfer probability using the kicked approximation (Eqs. (17) and (23)) with the exact expressions in the absence of time ordering in the Schrödinger and intermediate pictures, given by Eqs. (8) and (11),

$$
P^0_2(T_f) = \sin^2 \alpha + O(\alpha^2 \beta^2)
$$

$$
P^0_2(T_f) = \frac{\alpha^2}{\alpha^2 + (\gamma T_f)^2} \sin^2 \sqrt{\alpha^2 + (\gamma T_f)^2}
$$

These three equations are consistent with the numerical results shown in Fig. 4. As $\Delta E \to 0$, $\gamma T_f$ and $\beta$ become small, time ordering effects disappear, and all three results coincide at $\sin^2 \alpha$.

2. A positive followed by a negative pulse

Finally, we consider the role of time ordering in the case of two equal and opposite pulses separated by an interval $T_s$. In the limit of ideal kicks, the kick-antikick evolution operator has been expressed analytically above with and without time ordering in Eqs. (23) and (26). This yields for the probability $P_2(T_f)$ at times $T_f$ after the second kick,

$$
P_2(T_f) = \sin^2 \gamma T_s \sin^2 2\alpha + O(\beta^2)
$$

$$
P^0_{I_2}(T_f) = \sin^2 \left[ 2\alpha e^{-\beta^2} \sin^2 (\gamma T_s) \right]
$$

The existence of these analytic results is helpful in studying the role of time ordering in $P_2$.

In Fig. 5 we compare $P_2$ with $P^0_{I_2}$ as a function of the separation time $T_s$ between the pulses. Here the measurement time $T_f$ is taken to be well after the second pulse has decayed, $T_f - T_2 \gg \tau$. We note that, as expected, in all cases the occupation probability goes to zero as the two opposite pulses coalesce, i.e. as $T_s \to 0$. On the left side we show the occupation
FIG. 5: Target state probability as a function of the separation time $T_s = T_2 - T_1$ between the two pulses, for integrated pulse strength $\alpha = \pi/2$, $\pi/4$, and $3\pi/8$. On the left the pulse width is a rather narrow 10 ps, while on the right the pulse width is 100 ps, where the kicked approximation is breaking down. Again the heavy line denotes the exact result including time ordering, while the thin dashed line denotes the intermediate picture result without time ordering. The Schrödinger picture result without time ordering is identically zero for all values of $T_s$ and $\alpha$.

Probability for a Gaussian pulse of width $\tau = 10$ ps for three values of the pulse strength $\alpha$, namely $\alpha = \pi/2$ corresponding to a kick that turns a qubit from off to on at $T_1$ and back to off at $T_2$, $\alpha = \pi/4$ where a qubit is on after $T_2$, and an intermediate strength $\alpha = 3\pi/8$. On the left side $\tau/T_{\Delta E} = 10^{-2}$, so that the kicked result is accurately obtained. We note that $P^0_{12} \approx P_2$, and time ordering effects vanish, when $T_s/T_{\Delta E} = \gamma T_s/\pi$ is an integer or half-integer, consistent with Eq. (30). Away from these special values, large time ordering
effects are present. On the right side, the pulses are quite broad ($\tau/T_{\Delta E} = 10^{-1}$), and the kicked approximation is clearly breaking down.

IV. DISCUSSION

In this paper we have studied the role of time ordering in a strongly perturbed two-state quantum system. We have defined the time ordering effect as the difference between a calculation with time ordering and one without time ordering. This is the way correlation, entanglement, and non-random processes are also defined [26]. In all of these cases it is useful to define carefully the limit without the effect (time ordering, correlation, or non-randomness). In the case of time ordering we have seen that the limit without time correlation depends on how one separates $\hat{H}$ into $\hat{H}_0 + \hat{V}$, i.e. what we call a choice of gauge. In practice this choice sometimes rests on the choice of the time averaged interaction. Precisely this same issue occurs in defining correlation [27], namely the somewhat arbitrary choice of a mean field interaction. A similar problem can arise in defining entanglement [28] and non-random processes. In any case the problem is not new.

For clarity and simplicity we have chosen $\hat{H}_0 = \text{const} \times \sigma_z$ and $\hat{V} = f(t)V_0\sigma_x$ in this paper. However other gauge choices are possible and in some cases may be more sensible. Specifically, in many cases experimental conditions can lead to a sensible gauge choice where, for example, the asymptotic state of an unperturbed atom is an eigenstate of $\hat{H}_0$, and $\hat{V}$ corresponds to an external electric or magnetic field imposed on the atom during part of the experiment. Such a $\hat{V}$ may sensibly contain $\sigma_z$ and/or $\sigma_y$ components.

We have shown above that the limit of no time ordering depends on the picture (representation) used. While this difference is small when time ordering effects are small, the differences can be large otherwise. Hence it might be argued that our time ordering analysis is useful primarily to determine if time ordering effects are small or large. This argument sometimes occurs in the use of correlation (although it is not common to use different representations for correlation). In any case the dependence on representation is not new [29].

Convergence properties of the Magnus expansion in the Schrödinger and interaction pictures have been known for some time to differ widely [30]. While the Schrödinger picture result (that in a sense corresponds to an especially simple gauge choice of $\hat{H}_0 = 0$) is formally easier to write down, it often does not yield predictions as reliable as the more complete
results found in the intermediate picture. We have illustrated this above with calculations and analysis of \( \hat{U}_0 \) and \( \hat{U}_I \) for single and multiple pulses. The intermediate picture takes maximum advantage of knowledge about the eigenstates and spectrum of \( \hat{H}_0 \). In other words the intermediate picture is generally more complete than the Schrödinger picture and often more sensible. It is useful to separate \( \hat{H} \) into \( \hat{H}_0 + \hat{V} \) in such a way as to include as much of the problem as possible in \( \hat{H}_0 \), whose solutions are known. In the extreme limit of the Heisenberg picture (which can be thought of as a gauge choice where \( \hat{H} = \hat{H}_0 \) and \( \hat{V} = 0 \)), we have \( \hat{U}_H = \hat{U}_0^H = 1 \), and there is never any time ordering in the time evolution.

Here we have worked in the time domain and formulated the question of time ordering by explicitly working with the Dyson time ordering operator, \( T \). Equivalently one may work in the energy domain, as has been done recently in the context of atomic scattering to analyze experimental data and identify time ordering effects \([10, 14]\). A key transformation for time ordering from the time domain to the energy domain is the Fourier transform of the step function, namely,

\[
\int e^{iE't} \Theta(t)e^{-iEt} dt = \frac{1}{E-E'+i\eta} = \pi \delta(E-E') + iP \nu E \frac{1}{E-E'},
\]

where the effect of time ordering is associated with \( i\eta \), which gives rise to the principal value term. The \( i\eta \) carries the effect of the boundary condition on the Green’s function in energy space. This is discussed in more detail elsewhere \([8, 10]\). Since \( [\hat{H}(t''), \hat{H}(t')] \) provides a connection between interactions at \( t'' \) and \( t' \), the quantum time propagator includes non-local effects in time. An example of a counter-intuitive time sequence occurs in stimulated Raman adiabatic passage (STIRAP), where efficient and robust population transfer is attained using two pulsed radiation fields in a three-level system \([31]\).

The time ordering effects considered in this paper are associated with a sequential ordering of interactions. The normal boundary condition imposed on the evolution operator is that the sequence proceeds in the direction of increasing time (or alternatively decreasing time to study time reversal). Hence effects of time ordering are associated with a direction of the flow of time. In this paper we have not included dissipation; hence all amplitudes explicitly satisfy invariance under time reversal, e.g. in Eqs. \( (17) \) and \( (24) \) which include effects due to time ordering. Effects of time ordering have been observed in systems that satisfy time reversal invariance \([12, 13, 14]\). This means that observable evidence of the direction of time (time ordering) can be obtained without violating the symmetry of time reversal invariance.
V. SUMMARY

In this paper we have given a definition of time ordering in a strongly perturbed quantum system, namely that time ordering is the difference between calculations with and without time ordering. This definition is similar to the definition of correlation. In both cases effects arise from differences between an instantaneous interaction and its averaged value. When the effect of time ordering is small, the dependence on representation is weak. However, when time ordering effects are large, the difference between representations can also be large. We have considered in detail time ordering for qubits that are strongly and suddenly perturbed by an external interaction. We have illustrated our methods for a $2s-2p$ transition in atomic hydrogen caused by a Gaussian pulse of finite width in time. Other diabatically changing qubits may also be analyzed with our methods. Simple analytic expressions have been given for the occupation amplitudes and probabilities for kicked qubits, including single and multiple kicks. We think that it should be possible to find analytic solutions for correlated kicked qubits, so that time coupled interacting qubits may also be studied analytically.

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