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Universal energy scaling law for optimally excited nonlinear oscillators

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We compute the optimal temporal profile for an external driving force $F(t)$ that can maximize the energy absorption of any driven non-linear oscillator. The technique is based on constraining the maximum amplitude of the force field such that optimal control theory can provide quasi-analytical solutions. We illustrate this computational technique for the undamped Duffing oscillator as well as for a driven quantum mechanical two-level system. We find that under optimal force conditions the asymptotic time-dependence of the maximum amplitude growth is given by a power law $X(t) \sim t^{2/\alpha}$, where the (possibly non-integer) exponent is determined by the highest degree of the oscillator’s nonlinearity $\alpha$. As a universal result, this predicts that the maximal energy absorption of any non-linear oscillator grows (under an optimized force field) quadratically in time. We also find for the two-level system that even under optimized excitation conditions the maximally achievable inversion does not monotonically increase with the force amplitude. It is characterized by an interesting sequence of $n$-cycle thresholds as well as a self-termination of the growth.

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I. INTRODUCTION

The search for the optimal energy transfer from an external source into a classical or quantum mechanical nonlinear dynamical system is an important challenge in many areas of science and engineering. To construct a perfect temporal or spatial dependence of an external force field that under specific constraints can maximize a certain objective, such as the excitation of a system variable, is a very common application of optimal control theory [1–8].

A historical example of such a temporal control mechanism from accelerator physics is the synchrocyclotron, where the frequency of the driving rf electric field is varied in time to compensate for relativistic effects due the particles’ velocity [9, 10]. In the corresponding spatial control mechanism (isochronous cyclotron) the magnetic field has a chosen inhomogeneity to keep the particles focused.

In other fields of physics, for excellent reviews on numerous applications of autoresonances in general nonlinear systems, see the work by Friedland [11]. A nice tutorial on the application of optimal control theory to atomic and molecular systems was provided by Werschnik and Gross [12]. Recently, optimal control theory was examined for quantum field theoretical mechanisms to increase the yield in laser assisted pair creation processes [13–16]. In many theoretical analyses of these nonlinear dynamics, the effect on the external force field was often limited to the perturbative regime so that analytical methods could be employed [17]. As soon as the system leaves the linear regime, these solutions are not necessarily very reliable.

In order to avoid the, from a practical perspective, uninteresting infinite forces, one usually has to restrict some properties of the force field [18–21]. A very common constraint limits the total energy in the pulsed force field. Unfortunately, the determination of the optimal time-dependence of the energy-constrained forces usually requires extensive computational analysis.

In this work, we will show that if we examine amplitude-restrained force fields, then quasi-analytical approaches are available for estimating the optimal time-dependence of the force. Using the example of classical nonlinear oscillators as well as quantum mechanical two-level systems, we suggest that an oscillatory force whose frequency instantaneously matches the natural eigenfrequency of the system at each time is not necessarily the most efficient means to excite the system. In fact, if the degree of nonlinearity $\alpha$, for the corresponding potential $V(x) \sim x^{\alpha}$ exceeds 2, an appropriate blue shift is required for the optimal driving frequency to accommodate for the dressing of the system by the non-perturbative excitation force.

The work is structured as follows. In Sec. II we show for any general classical mechanical oscillator the fastest energy transfer is provided by a discontinuous (telegraph-like) force field with appropriately chosen sign switching times. We outline an iterative scheme to determine these switching times for any oscillator. In Sec. III we illustrate this scheme for concrete physical systems pointing out that the energy-scaling of the optimal energy growth rate is universal and therefore independent of the degree of nonlinearity of the oscillator. In Sec. IV we predict that the dressing effects associated with nonperturbative forces require either a red or blue shift in the optimal chirping frequency compared to the instantaneous natural frequency. In Sec. V we illustrate the universality of our findings for a quantum mechanical system [22–24]. We close in Section Sec. VI with a discussion and an outlook to future challenges.
II. METHODOLOGIES TO DETERMINE THE TEMPORAL SHAPE OF THE OPTIMAL FORCE

The optimal (amplitude-limited) force \( F(t) \), that leads to the maximal energy growth of any oscillator is given by a telegraph-like signal with characteristic moments in time \( T_n \) when \( F(t) \) changes its sign. The sequence of the exact switching times can be obtained from numerical solutions to an iterative set of coupled transcendental equations. These solutions suggest remarkable scaling laws for these times as well as the associated maximal elongations of the oscillators as time progresses. We show that the optimal force field can be also obtained from the solution to a single ordinary differential equation in time based on some instantaneous feedback.

A. The optimal force field

The principal goal of this Section is to construct the optimal time-dependence of an external time-dependent field \( F(t) \) that can maximize the amount of energy transferred to a general nonlinear oscillator after a final time from that force. The equation of motion for the amplitude \( x(t) \), is given by \( m \ddot{x} + V'(x) + F(t) = 0 \). This problem is related to maximizing directly the final amplitude of the oscillator. For simplicity, in this work we assume that the particle of mass \( m \) is initially at rest and located at the origin. In order to exclude the trivial solution \( F(t) = \infty \) and to find a possibly experimentally realizable shape, we have confined the maximal permitted magnitude of \( F(t) \) to \( -F_0 \leq F(t) \leq F_0 \). In Appendix A we review the usual variational approach used in optimal control theory. Here the optimal force field \( F(t) \), as well as the Lagrangian co-state and state variables, have to satisfy consistently a set of nonlinear coupled differential equations that for a general potential \( V(x) \) are not possible to be solved analytically. However, we can obtain a helpful, simple property about \( F(t) \). It turns out that a necessary condition for \( F(t) \), to lead to either a local or global maximum \( x(T) \), is that \( F(t) \) must take the form of a telegraph-like signal, i.e., \( F(t) = F_0 \ g(t)/|g(t)| \) for any arbitrary function \( g(t) \). In other words, except for the two temporally trivial constant cases \( F(t) = -F_0 \) or \( F(t) = F_0 \), the optimum force field \( F(t) \) has to be a discontinuous function of time. The optimum force field \( F(t) \) has to jump back and forth between its only two permitted values \(-F_0 \) and \( F_0 \). This key observation opens the door to a remarkably easy analysis based on a straightforward determination of those moments in time when the optimal force function changes its sign. Alternatively, in Sec. II B we will also show that by implementing a simple feedback mechanism the optimal force field can be obtained from the solution of a simple autonomous differential equation that determines automatically the periodicity changes of the pulse train.

The sequence of those moments in time, when the optimal force switches its sign, are denoted by \( T_n \) with \( n = 0, 1, 2, \ldots, \) can be constructed from an iterative sequence \( T_{n+1} = H(T_n, A_{n+1}, A_n) \), where \( A_{n+1} \) and \( A_n \) denote the maximal excursions of the particle associated with the turning point times \( T_{n+1} \) and \( T_n \). We set \( T_0 \) and \( A_0 \) to zero. The first switch time \( T_1 \) for \( F(t) \) is given by the time the particle takes to travel in the potential \( V(x) - F_0 x \) from its initial location \( x(0) = 0 \) to \( A_1 \), i.e., \( T_1 = (m/2)^{1/2} \int_0^{A_1} dx[ F_0 x - V(x) ]^{-1/2} \). The value of the particle’s first (positive) turning point \( A_1 \) follows from a similar consideration based on energy conservation, i.e., \( 0 = -F_0 A_1 + V(A_1) \). The next switch time \( T_2 = T_1 + (m/2)^{1/2} \int_{A_1}^{A_2} dx[ F_0 A_1 + V(A_1) - F_0 x - V(x) ]^{-1/2} \) is associated with the travel time from location \( A_1 \) to the second turning point \( A_2 \), which is the (negative) real solution to \( F_0 A_2 + V(A_2) = F_0 A_1 + V(A_1) \). In summary, we obtain the following set of coupled recursion relationships for \( A_{n+1} \) and \( T_{n+1} \),

\[
T_{n+1} = T_n + (m/2)^{1/2} \int_{A_n}^{A_{n+1}} dx[-F_0 A_n + V(A_n)] + F_0 x - V(x)]^{-1/2}, \quad \text{for } n \text{ even (1)}
\]

\[
T_{n+1} = T_n - (m/2)^{1/2} \int_{A_n}^{A_{n+1}} dx[F_0 A_n + V(A_n)] - F_0 x - V(x)]^{-1/2}, \quad \text{for } n \text{ odd (2)}
\]

\[
V(A_{n+1}) - F_0 A_{n+1} = V(A_n) - F_0 A_n, \quad \text{for } n \text{ even (3)}
\]

\[
V(A_{n+1}) + F_0 A_{n+1} = V(A_n) + F_0 A_n, \quad \text{for } n \text{ odd (4)}
\]

Let us consider the general class of those potentials \( V(x) \) that can be characterized by a single nonlinearity, i.e., \( V(x) = \lambda |x|^\alpha \), where the real variable \( \alpha \) with \( 1 \leq \alpha \leq \infty \) denotes the degree on nonlinearity of the oscillator and the positive constant \( \lambda \) is the amplitude of the potential. We do not consider the case of those binding potentials for \( 0 \leq \alpha \leq 1 \), as the restoring force would be less than the one provided by the external time-dependent force. As a side issue, we note that for the special case \( \alpha = 2 \) we recover the usual harmonic oscillator \( V(x) = m \Omega^2 x^2 / 2 \), (if \( \lambda = m \Omega^2 \)), while for the largest possible degree of nonlinearity, \( \alpha = \infty \) we obtain the infinite potential well, which restricts the particle’s position to \(-1 \leq x(t) \leq 1 \) and to a force-free motion between these turning points. Under this set of single- nonlinearity oscillators, one can examine the following set of approximate solutions to Eqs.(1) to (4),

\[
T_n = N_T(\alpha) \left( \frac{m}{\lambda} \right)^{1/4(\alpha-2)/(\alpha-1)} \left( \frac{m}{F_0} \right)^{\alpha-2} n^{\alpha/(\alpha-1)}, \quad \text{(5)}
\]

\[
A_n = (-1)^{n+1} N_A(\alpha) \left( \frac{F_0}{\lambda m} \right)^{1/\alpha} T_n^{2/\alpha}, \quad \text{(6)}
\]
where $N_A$ and $N_T$ are dimensionless parameters to be determined. The functional form of these two approximate expressions are suggested from a dimensional analysis and also by generalizing two special limiting cases (see below).

We have illustrated the remarkable accuracy of this set of solutions for any $\alpha$, $\lambda$ and $F_0$ in Fig. 1, where we compare the predictions of Eqs. (5) and (6) with the exact numerical solutions to Eqs. (1) to (4) for oscillators with four degrees of nonlinearity $\alpha = 1.5, 2, \pi$ and 4.

The agreement for sufficiently large $n$ is remarkable. We also see that for $\alpha < 2$ the durations between the optimal switching times $\tau_n \equiv T_n - T_{n-1}$ increase with the number $n$ of sign changes, whereas for $\alpha > 2$ the optimal driving frequency for the oscillators increases with $n$.

The dimensionless numerical factors $N_A(\alpha)$ and $N_T(\alpha)$ in Eqs. (5) and (6) were obtained such that the values of $A_n$ and $T_n$ in the equations match that of the solutions of Eqs.(1) to (4). These coefficients do not depend on the particle’s mass $m$, the external force amplitude $F_0$, nor the strength $\lambda$ of the potential. They are therefore unique to each oscillator. We have graphed them in Fig. 2.

Quite remarkably, for a certain range $\alpha > 4$, $N_A(\alpha)$ is practically constant and depends only very weakly on the degree of nonlinearity.

There are two interesting, special cases for which $A_n$ and $T_n$, and therefore the numerical values for $N_A(\alpha)$ and $N_T(\alpha)$, can be easily derived analytically. The first special case is the simple harmonic oscillator $V(x) = m\Omega^2x^2/2$. The increase of the turning points with $F_0$ can be determined analytically as $A_n = (-1)^{n+1}(2n)F_0m^{-1}\Omega^{-2}$. Also, the corresponding integrals can be evaluated analytically as well, leading to the interesting result $T_n = (n/2)\pi/\Omega$, independent of $F_0$. This means that we obtain $N_A(2) = 2/\pi$ and $N_T(2) = \pi$. We note that also the natural period of the corresponding unforced oscillator ($F_0 = 0$), $2\pi/\Omega$, is independent of the distance between the two turning points. While in the driven case ($F_0 \neq 0$) the particle is accelerated more between the two turning points, the resulting distance of travel also increases with $F_0$.

The second fully analytically accessible system occurs for $\alpha = \infty$. Here the maximal excursion amplitude at each turning point is given by $A_n = (-1)^{n+1}$, such that $N_A(\infty) = 1$ in Eqs. (5) and (6). Similarly, we obtain $T_n = 2(m/F_0)^{1/2}n^{1/2}$, such that $N_T(\infty) = 2$.

### B. Alternative numerical determination of the optimal force field $F(t)$

As an alternative method to determine the switch times $T_n$ for the optimal force field, we can obtain $F(t)$ directly from the solution to a single differential equation. This approach can be computationally more efficient than the tedious recursive method. The key observation is that the optimal force is always aligned parallel to the instantaneous velocity, i.e., $F(t) = F_0\text{Sign}(dx/dt)$. This means, to obtain the optimized temporal evolution for any oscillator, we can remove this explicit dependence on the independent variable $F(t)$ and simply use this specific functional form in the equation of motion, i.e., we need to solve

$$m\frac{d^2x}{dt^2} = -\frac{dV}{dx} + F_0\text{Sign}\left(\frac{dx}{dt}\right),$$

with the initial conditions $x(0) = dx/dt(0) = 0$. This technique of constructing the optimal $F(t)$ from the solution of single differential equation is certainly more efficient and computationally reliable than trying to solve consistently the corresponding set of Eqs.(1) to (4). It also incorporates the easiest form of an instantaneous feedback mechanism [25–41], where the force is instantly determined by an internal variable, which is here the velocity $dx/dt$. Including this particular instantaneous feedback mechanism removes any external time-dependent force and the dynamics becomes autonomous, but as a result, the equation of motion becomes even more nonlinear in addition to that provided by $V(x)$. The term $F_0\text{Sign}(dx/dt)$ could be interpreted as a “negative drag,” where the (negative) drag coefficient is inversely proportional to the particle’s speed $|dx/dt|$.

### III. THE SCALING OF THE ENERGY WITH TIME

We can generalize the scaling laws predicted for systems with a single-degree of nonlinearity to multiple degrees, such as the well-known Duffing oscillator [42–44]. We examine the temporal growth of the oscillator’s energy and suggest a new scaling law that is universal to any driven classical mechanical system under optimal force conditions. We compare the energy conversion efficiency of the discrete telegraph-like pulse trains with monochromatic forces that are naturally continuous in time.

#### A. Effective instantaneous degree of nonlinearity

Here we illustrate our computational method for the special example of the (nonlinear) Duffing potential $V(x) = m\Omega^2x^2/2 + \lambda x^4/4$. Using the technique outlined above, we can easily compute the elongation $x(t)$ under optimal force conditions, by solving the corresponding differential equation $md^2x/dt^2 = -m\Omega^2x - \lambda x^3 + F_0\text{Sign}(dx/dt)$ numerically.

In Fig. 3(a) we have graphed (on a logarithmic scale) the corresponding absolute value of the amplitude $x(t)$ for the parameters $m = 1$, $\Omega = 2$, $\lambda = 0.04$ and $F_0 = 1$. For convenience we use atomic units. For comparison, we have also added the single-nonlinearity solutions $|A_n|$ obtained from Eq.(6) for $\alpha = 2$ and $\alpha = 4$ where we used the numerically found values of $T_n$. For our parameters, they grow like $x(t) \sim t^{3}$ (for $\alpha = 2$) and $x(t) \sim t^{1/2}$ (for
region of the transition domain.

The transition between the different temporal scaling regions suggests that for shorter times the harmonic portion of $V(x)$ determines the overall dynamical response $x(t)$, whereas for longer times (and the corresponding larger excursion distances $x$) the nonlinear portion of $V(x)$ becomes more relevant. This temporally changing degree of effective nonlinearity can be captured quantitatively if we define $\alpha_{eff}(t) = 2[\log(A_n) - \log(A_{n-1})]/[\log(T_n) - \log(T_{n-1})]$. This instantaneous nonlinearity represents the inverse of the slope of the maximal excursion amplitudes of $|x(t)|$ versus time on double logarithmic axes. It is based on the scaling law $|A_n| \sim T_n^{2/\alpha}$ characteristic for single-nonlinearity oscillators, see Eq.(6). In order to test if the definition of this new dynamical characteristics $\alpha_{eff}(t)$ is indeed physically meaningful, we have graphed it in Figure 3c. Complementary to the increasing instantaneous frequency, it nicely shows that it starts at the expected value $\alpha_{eff}(t) = 2$ for the linearity dominated early time regime and then changes to approach $\alpha_{eff}(t) = 4$ for the longer times. This nicely illustrates how one can recover
the oscillator’s degree of nonlinearity from its dynamical response to an optimized force.

B. The temporal growth of the free- and total energy

The $t^{2/\alpha}$ scaling of the maximum amplitude characteristic of each cycle has some remarkably universal consequences for the corresponding growth of the force-free energy as well as total energy at those moments in time $T_n$. The force-free energy is given by $e \equiv (m/2)(dx/dt)^2 + V(x)$. Its temporal growth satisfies the equation of motion $de/dt = F(t)dx/dt$, which due to the in-phase relationship between the velocity and the (telegraph-like) optimal force $F(t) = F_0 \text{Sign}[dx/dt]$, reduces to $de/dt = F_0|dx/dt|$. As $dx/dt$ is a nontrivial, the exact temporal growth $e(t)$ can only be obtained numerically, unless $\alpha = 2$ or $\alpha = \infty$. But is clear that $e(t)$ is a monotonically growing function of time.

In contrast, the corresponding total energy, denoted by $E \equiv (m/2)(dx/dt)^2 + V(x) - F_0 \text{Sign}[dx/dt]x$, is constant in time during each cycle when the force is constant. The total energy during the n-th cycle, denoted by $E_n$, changes abruptly from $E_n$ to $E_{n+1}$ at those moments in time $T_n$, when the force switches its sign. This sudden increase at times $T_n$ is given by $E_{n+1} - E_n = 2F_0|A_n|$.

During each cycle there is a distinct moment in time when the instantaneous values of the two energies $e$ and $E$ agree with each other. These times occur precisely when the particle’s position crosses the origin at $x = 0$ such that the potential energy $V(x)$ vanishes and both energies reflect the kinetic energy.

In Fig. 4 we have graphed $e(t)$ and $E(t)$ for the optically pumped sextic oscillator together with the analytical prediction $V(A_n)$. While the growth of $e(t)$ takes a nonlinear but monotonically growing shape, at the turning points it matches the analytical prediction

$$e(T_n) = V(A_n) = \lambda|A_n|^\alpha = N_\alpha(\alpha)^\alpha \frac{m}{\lambda F_0} T_n^2, \quad (8)$$

where we have used Eq.(6) for the dashed line as well as the open circles. As the $\alpha$-dependence in the exponent for the time cancels out, we have here a remarkably universal finding. This predicts that under optimal driving conditions the energy growth of any driven oscillator with a single-nonlinearity $\alpha$ is always quadratic in time, independent of $\alpha$. However, due to the possible time-dependence of the effective instantaneous degree of nonlinearity $\alpha(t)$ (see Sec. III A) for oscillators with mixed nonlinearities, the prefactor $N_\alpha(\alpha)^\alpha/\lambda$ in Eq.(8) can vary in time, such that this universal power-law behavior $e(T_n) \sim T_n^2$ is only approximately valid for oscillators with mixed nonlinearities.

For example, for the harmonic oscillator the corresponding to the energy $e(T_n) = (2/\pi^2) F_0 m^{-1} T_n^2$, independent of $\Omega$. 

FIG. 3: (a) The maximal elongations $|A_n|$ for the Duffing oscillator as a function of time under optimal driving force $F(t)$ for the first 6000 cycles. For comparison, the two reference lines are $|A_n|(T_n) \sim T_n^{2\alpha}$ from Eqs. (5) and (6) for $\alpha = 2$ and $\alpha = 4$, but $T_n$ was used from the mixed-$\alpha$ system; (b) The instantaneous frequency $\omega(t)$ of the optimal driving force; (c) The instantaneous effective value of the nonlinearity $\alpha_{\text{eff}}(t) \equiv 2[\text{Log}(T_n) - \text{Log}(T_{n-1})]/[\text{Log}(A_n) - \text{Log}(A_{n-1})]$. [The parameters are $\Omega = 2$, $\lambda = 0.04$, $F_0 = 1$.]

FIG. 4: The growth of the force-free energy $e(t)$ and the total energy $E(t)$ as a function of time for the optimally driven sextic oscillator ($\alpha = 6$) for the first 4 cycles. The dashed line and the four open circles are the associated analytical prediction according to Eq.(8). [The parameters are ($\alpha = 1$), $F_0 = 0.5$]
C. Energy conversion efficiency

The finding that the optimal force field \( F(t) \) has to be a discontinuous function of time seems to contradict at first the intuitive expectation that, for example, in the case of the linear oscillator, a fully resonant situation is usually discussed for a (continuous) sinusoidal excitation force. In fact, one could argue that the higher harmonic spectral frequency components of a (telegraph) square wave function are actually ‘wasted’ as they do not match the natural resonance frequency \( \Omega \). To illustrate this point for a simple example, we have compared the temporal growth of \( x(t) \) obtained from \( m \dot{x}^2 x/dt^2 = -m\Omega^2 x + F(t) \), for the two driving forces \( F(t) = F_0 \text{Sign} [\sin (\Omega t)] \) and for \( F_{\text{trig}}(t) = F_0 \sin (\Omega t) \). While both forces lead to a linear growth of the turning points \( x(t) \) in time, the prefactor of \( x_{\text{opt}}(t) \) [\(~ (2/\pi) t F_0 \omega^{-1} \), see Sec. III A] is by a factor of \( 4/\pi \) larger than the one for \( x_{\text{trig}}(t) [\sim (1/2) t F_0 \omega^{-1}] \), confirming that for an amplitude constraint force field, a discontinuous \( F(t) \) is indeed superior to the traditional sinusoidal resonant force.

However, this does not mean that the energy transfer efficiency of \( F_{\text{opt}}(t) \) is automatically higher than that of \( F_{\text{trig}}(t) \). As both forces are external, any energy depletion mechanism is obviously neglected in this treatment. For example, in electrodynamics the energy contained in the electric field pulse is directly proportional to the time-

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For example, \( \text{in electrodynamics the energy contained in the electric field pulse is directly proportional to the time-} \)

integration of the source field. One could therefore consider here the integral \( \int dt F(t)^2 \) as a possible measure for the available energy reservoir provided by the external force field, as a possible measure for the available energy reservoir. In this case, for a fair comparison, we should have compared the effect of the two equally ‘energy normalized’ forces \( F_{\text{opt}}(t) = F_0 \text{Sign} [\sin (\Omega t)] \) and \( F_{\text{trig}}(t) = 2^{1/2} F_0 \text{Sign} [\sin (\Omega t)] \) on the growth of \( x(t) \). In this case, we find that the growth of the elongation \( x(t) \) exceeds that of \( F_{\text{opt}}(t) \) by 11\%, associated with a factor of \( x_{\text{trig}}(t)/x_{\text{opt}}(t) = 2^{1/2} (\pi/4) = 1.11 \). This confirms quite interestingly that if we had fixed the total energy of the force \( F(t) \), i.e., \( \int dt F(t)^2 \), (instead of the amplitude) then a sinusoidal force does indeed lead to a larger amplitude \( x(t) \) as expected.

IV. OPTIMAL FREQUENCY OF \( F(T) \) RELATIVE TO THE INSTANTANEOUS NATURAL FREQUENCY

As discussed in Sec. II B, we can also examine the properties of the optimal force \( F(t) \) by solving first the autonomous differential equation \( m \ddot{x} x/dt^2 = -m\Omega^2 x - \lambda x^3 + F_0 \text{Sign} [dx/dt] \) numerically for \( x(t) \) and then computing \( dx/dt \) from this solution, allowing us to construct the optimal \( F(t) = F_0 \text{Sign} [dx/dt] \). In order to better interpret the meaning of \( F(t) \), we have determined the inverse of the difference between two consecutive switching times \( \pi/(T_n - T_{n-1}) \), which we interpreted in Sec. III A as the instantaneous frequency of \( F(t) \), denoted by \( \omega(t) \). This frequency \( \omega(t) \) is graphed in Fig. 3(b) for \( m = 1 \), \( \Omega = 4 \) and \( \lambda = 0.04 \). We found that \( F(t) \) is periodic with frequency \( \Omega \) for early times, while for longer times the frequency increases with time.

One could conjecture that the switching time durations, i.e., \( \tau_n = T_n - T_{n-1} \), could follow the instantaneous half periods of the associated natural (unforced) motion associated with this amplitude. While this correspondence is exactly true for the harmonic oscillator, i.e., \( T_n - T_{n-1} = \pi/\Omega \), it can only serve as a rough guidance for the exact switching times.

For example, one can also estimate the amplitude dependence of the corresponding non-driven purely quartic oscillator (with \( V(x) = \lambda x^4/4 \)) that oscillates periodically between \( x \pm A \), given by \( \tau_{\alpha=4} = (2m)^{-1/2} \int_{-A}^{A} dx [\lambda A^4/4 - \lambda x^4/4]^{-1/2} \). While rescaling and numerical evaluation of the integral \( \int_{-A}^{A} dx [1 - x^4]^{-1/2} \), we obtain \( \tau_{\alpha=4} \approx 7.416(\lambda m)^{-1/2} A^{-1} \). On the other hand, the true switching time \( (T_n - T_{n-1}) \) is given in Eqs. (1) to (4) by the integral \( (2m)^{-1/2} \int_{-A}^{A} dx [-F_0 A + \lambda A^4/4 + F_0 x - \lambda x^4/4]^{-1/2} \), where \( A_{\text{right}} \) follows from \(-F_0 A_{\text{right}} + V(A_{\text{right}}) = F_0 A + V(-A) \). We see that the two integrals differ in two ways, the expressions in the second square root contains the additional (positive) term \( F_0 (A + x) \), and also its upper integration limit is not \( A \) but a larger amplitude \( A_{\text{right}} \). While the first mechanism would decrease the switching time relative to the natural half period \( \tau_{\alpha=4} \), the larger upper limit \( A_{\text{right}} \) would increase the switching time. In order to examine which of these two mechanisms is the dominant one, we have compared in Fig. 5 the \( \tau_{\alpha=4} \) (given by the open circles) with the true switching times for \( F_0 = 0.01, 0.1 \) and 0.3.

There are two interesting observations. First, the required switch time is a non-monotonic function of the amplitude \( A \). For \( \alpha = 4 \) and small \( A \) the time increases whereas for large amplitudes it decreases. This immediately suggests that it is not possible to obtain an optimal driving force where the instantaneous frequency is a monotonic function of time, i.e., a linear chirp is not the best choice. For larger excursions \( A \), the match is expected as the (spatially constant) force \( F_0 \) becomes negligible compared to the much larger restoring force provided by \( V(x) \). Also, in the same limit, for large values of \( A \) the solution to \(-F_0 A_{\text{right}} + V(A_{\text{right}}) = F_0 A + V(-A) \) becomes \( A_{\text{right}} = A \). This finding suggests that for early times (when \( A \) is small) the switching time for the optimal \( F(t) \) can only be approximated by the natural half period in the perturbative limit, when \( F_0 = 0 \) is small.

The second observation suggests that the optimal switching time should always be chosen shorter than the instantaneous period for oscillators with \( \alpha > 2 \). This means that the corresponding instantaneous driving frequency should, at least for short excitation times, be chosen above the instantaneous natural frequency of the quartic oscillator. For longer times (associated with larger amplitudes \( A \)), the value of the natural frequency can serve as a good guidance for the frequencies of the
optimal driving force.

To provide a complete picture, we have compared the optimal switching durations with the corresponding natural (amplitude) dependent half-periods also for oscillators with $\alpha < 2$. Here the natural periods decrease with increasing amplitudes. In addition, the optimal switching durations have to be chosen even longer. In other words, the optimum driving frequencies need to be redshifted relative to the increasing (force-free) oscillation periods.

V. QUANTUM MECHANICAL TWO-LEVEL SYSTEM UNDER OPTIMIZED EXCITATION

A. The model system and the optimal force to maximize the final inversion

We show that the same ideas with regard to computing the optimal external force field for a classical oscillator can be easily generalized to a nonlinear quantum mechanical system. It turns out that a suitable chirped telegraph signal provides also here the optimum force field to maximize the inversion of this system if the external force is amplitude constrained.

The time-evolution of the quantum state $|\Psi(t)\rangle$ of a two-level system [22–24] is described (in the absence of any decoherence) by the two complex probability amplitudes $C_g(t)$ and $C_e(t)$ such that $|\Psi(t)\rangle = C_g(t)|g\rangle + C_e(t)|e\rangle$. The Hamiltonian describing the energy transfer between the ground state $|g\rangle$ and the excited state $|e\rangle$ is given (in atomic units) by

$$H(t) = \omega_0 |e\rangle\langle e| + F(t)\{|e\rangle\langle g| + |g\rangle\langle e|\},$$

where $\omega_0$ is the energy difference between the two levels and $\int F(t)\, dt$ denotes the product of the external time-dependent electric field, whose amplitude is limited by $-F_0 < F(t) < F_0$ and the dipole moment of this transition.

The Schrödinger equation $i\hbar \frac{d|\Psi(t)\rangle}{dt} = H(t)|\Psi(t)\rangle$ leads to two coupled equations of motion for $C_g(t)$ and $C_e(t)$ which preserve $|C_e(t)|^2 + |C_g(t)|^2 = 1$.

We assume that the atom is initially in its ground state, i.e., $C_g(t=0) = 1$ and try to construct the optimal force $f(t)$, such that the final inversion (population difference), defined as $S_3 = |C_e|^2 - |C_g|^2$, is maximal after the interaction. It is therefore appropriate to construct first the corresponding equations of motion for $S_3$. They are given by the coupled set of three equations

$$\frac{dS_1}{dt} = -\omega_0 S_2(t),$$

$$\frac{dS_2}{dt} = \omega_0 S_1(t) - F(t) S_3(t),$$

$$\frac{dS_3}{dt} = F(t) S_2(t),$$

where $S_1 \equiv C_g(C_e^* + C_g^* C_e)$ and $S_2 \equiv -i(C_g C_e^* + C_g^* C_e)$ are the real and imaginary parts of the polarization and $F(t)$ (with $-F(0) \leq F(t) \leq F(0)$) is the scaled external force containing the dipole moment between both states. The set of three equations for the three (pseudo-spin) Bloch vector variables preserve $S_1(t)^2 + S_2(t)^2 + S_3(t)^2 = 1$, as the corresponding $3 \times 3$ matrix generator is a real antisymmetric matrix, which has the eigenvalues 0 and $\pm i\Omega$. Here we denote with $\Omega \equiv \sqrt{\omega_0^2 + F_0^2}/\omega_0$ the laser dressed frequency if $F(t) = F_0$. It should not be confused with any generalized Rabi-frequency $\omega_L = \omega_0 F_0^2/\omega_0^2$, which is very different and characterizes the time-scale for an inversion for a monochromatic driving force with frequency $\omega_L$ in those regions, where the rotating wave approximation is valid.

Applying the methodology laid out in App. A, for the quantum system the coupling term added to the Hamiltonian is given (in atomic units) by

$$\int_0^\infty F(t)\, dt = \alpha n - \alpha m,$$

where $\alpha = 4$ oscillator (associated with $F_0 = 0$ and the true required switching durations $\tau_n = T_n - T_{n-1}$ for the optimal driving force $F(t)$ as a function of the left elongation of the particle $x(T_n = -A)$.[other parameters are $m = 1$ and $\lambda = 1$]
tonian of optimal control theory is given by
\[ h = \lambda_1 [-\omega_0 S_2] + \lambda_2 [\omega_0 S_1 - S_3 F(t)] + \lambda_3 S_2 F(t), \] (13)

If we re-parametrize the amplitude constrained force again as \( F(t) = F_0 \sin[\omega_0(t)t] \), then the corresponding main equation of optimal control theory, i.e., \( dh/d\omega = 0 \) requires for this quantum system that any local or global force field also has to be again discontinuous, in full agreement with the findings for the classical oscillators discussed in the Sections above. In other words, our problem is once again reduced to computing the best possible set of switch times \( T_n \) for the telegraph signal \( F(t) \).

### B. Analytical estimates of the switching times \( T_n \)

We find an analytical expression for the perfect driving frequency for optimal energy absorption, which is always larger (blue shifted) than the eigenfrequency associated with the energy difference between upper and lower state.

The specific structure of the equations Eqs. (10) to (12) suggests also for the quantum mechanical system a constructive technique for determining the optimal switch times to maximize \( S_3(t) \). While in Sec. III this was guaranteed by keeping the force \( F(t) \) parallel to the velocity of the particle, for the two-level system we have to keep the force parallel to the imaginary part of the polarization \( S_2(t) \). In other words, if we choose \( F(t) = F_0 \text{Sign}[S_2(t)] \), then \( S_3(t) \) is required to grow monotonically as the right hand side (rhs) of Eq. (12) has to be non-negative at all times. In addition to constructing the optimal switch times, we also have to examine under which conditions for \( \omega_0, F_0 \), and the total interaction time we can achieve an optimum inversion \( S_3 \) and by how much this maximally achievable inversion differs from its theoretical upper limit of \( S_3 = 1 \).

In App. B we derive explicit expressions for the switching times \( T_n \). For the sequence of switching times for the optimal telegraph-like force \( F(t) \) are given by \( T_n = \pi n / \Omega \), with \( n = 1, 2, 3 ..., N \). The total number \( N \) of required (equidistant) switching times depends on the amplitude \( F_0 \) of the force. The smaller the force, the larger is the number of required switching cycles. Quite remarkably, this means that each value of \( F_0 \) dictates a precise number of switching cycles after which the dynamics self-terminates.

### C. Numerical results for n-cycle thresholds under optimal force conditions

In this section we illustrate the unusual halt of the temporal growth of \( S_3(t) \) under optimal force conditions and also compare it with the data for a monochromatic (and therefore continuous) driving force whose frequency is either at exact resonance with \( \omega_0 \) or at \( \Omega \).

In Fig. 6 we have graphed the inversion \( S_3(t) \) for seven values of \( F_0 \) and a level energy spacing \( \omega_0 = 2\pi \), which sets the natural time scale of the system \((2\pi/\omega_0)\) to unity.

We see that the number of cycles to approach the final inversion (when the dynamics self-terminates) decreases with increasing \( F_0 \). For the lowest force \((F_0/\omega_0 = 1.5)\), the quasi-oscillatory growth takes about seven cycles until the dynamics self-terminates at \( S_3(t) \) close to unity. For the next two larger forces \((F_0/\omega_0 = 2 \text{ and } 3)\) it takes 5 and 4 cycles and the final inversion decreases. The next largest force \((F_0/\omega_0 = 4.56)\) matches exactly the second threshold value \( F_{\text{thre},2} \) and we enter the region where the maximal inversion is reached after two cycles. For an even larger force \((F_0/\omega_0 = 7)\) the final inversion increases again until for \((F_0/\omega_0 = 10.88)\) which matches exactly the first threshold \( F_{\text{thre},1} \), and we enter the 1-cycle region, in which the inversion increases again with increasing \( F_0 \).

In order to obtain a more quantitative and complete view of this complicated sequence of increasing and decreasing inversions as a function of the force amplitude \( F_0 \), we have graphed in Fig. 7 the final \( S_3 \). While in the limits of small as well as large force amplitudes \( F0 \) we can always reach the theoretical upper limit of \( S_3 = 1 \), the maximally achievable inversion is indeed a highly non-monotonic function of \( F_0 \), characterized by several threshold values for \( F_0 = F_{\text{thre},n} \), where the inversion takes a local minimum. These threshold values describe exactly the transitions from the n-cycle to the \((n + 1)\) cycle regions in \( F_0 \). For example, consistent with our analytical derivations above, we find the first threshold occurs at \( F_{\text{thre},1} = 1.732\omega_0 \) (with \( S_3 = 0.500 \)). This numerical value matches the exact analytical value \( 3^{1/2}/\omega_0 \), which describes the onset of final inversions that self-terminate after only a single cycle, i.e., after a time of \( t = \pi/\omega_0 \).
$T_1 = \pi/\left(\omega_0^2 + F_0^2\right)^{1/2}$. The transition to the 2-cycle domain (i.e., $T_2 = 2\pi/\left(\omega_0^2 + F_0^2\right)^{1/2}$) occurs for $F_{\text{thre.2}} = 0.726\omega_0$ (with $S_3 = 0.809$), which matches again with the analytical estimate $F_{\text{thre.2}} = (5 - 2\cdot5^{1/2})^{1/2}\omega_0$ derived above. While it is very difficult to obtain analytical data for the other thresholds, their numerical values can be determined from the figure. For example, we find $F_{\text{thre.3}} = 0.482\omega_0$ (with $S_3 = 0.902$), $F_{\text{thre.4}} = 0.364\omega_0$ (with $S_3 = 0.940$) and $F_{\text{thre.5}} = 0.293\omega_0$ (with $S_3 = 0.960$).

As a final point of discussion and to motivate possible future research work, we want to briefly compare the temporal growth of the inversion $S_3(t)$ obtained for the optimal telegraph signal with the corresponding response to the two monochromatic signals $F(t) = F_0 \sin(\omega_0 t)$ and $F(t) = F_0 \sin(\Omega t)$.

As expected, we find that the telegraph signal minimizes the required time to reach the maximal inversion. We also see that the blue-shifted frequency is most advantageous for the telegraph signal, whereas the bare resonant condition (driving force frequency $= \omega_0$) is favored over the blue shifted frequency for the monochromatic case. As a side remark, we note that in this highly nonlinear domain the response is far from what one could have expected for the rotating wave approximation where the high-frequency components of the force-field are neglected in a rotating frame.

**VI. SUMMARY AND OUTLOOK TO OPEN QUESTIONS**

We have examined the time-dependence of the optimal external force field $F(t)$ that can maximize the energy transfer to any classical nonlinear oscillator as well as a quantum mechanical two-level atom. We have shown that we can obtain an excellent estimate for $F(t)$ if we constrain its amplitude and require $-F_0 \leq F(t) \leq F_0$. Optimal control theory predicts that under this constraint the optimal $F(t)$ must be discontinuous and alternating at specific times between the values $\pm F_0$. This means, that the problem of constructing $F(t)$ can be reduced to determining the optimal switch times $T_n$ for the sign of $F(t)$. As the force is piece-wise constant, we can obtain analytical estimates for $T_n$ from the corresponding Newton or Schrödinger equation. In case of an (energy) unbounded oscillator, the telegraph signal leads to an unlimited growth of the energy while for the two-level system it can self-terminate the quantum dynamics.

It turns out that the traditional belief that the optimum chirp of the force field is the one that perfectly matches the instantaneous frequency of the force-free system, is only correct if the amplitude of the force is sufficiently small [11]. In the more general nonperturbative limit, the dressing of the force field has to be taken into account. This nonlinear mechanism (associated with Stark-shifts due to $F(t)$) effectively blue-shifts the optimally chirped frequency for those classical ($\alpha > 2$) and quantum systems that we have investigated. For classical oscillators with a nonlinearity ($\alpha < 2$) a red-shifted instantaneous driving frequency is required.

In addition to the analytical estimates of the switch time, we have also proposed a feedback mechanism where the values of the optimal force field are determined from either the velocity (classical system) or the imaginary part of the polarization (quantum system). While in any real experimental system an instantaneous feedback is not possible, the unavoidable time delay between the...
measurement of the dynamical values and the adoption by the force field would naturally deteriorate the efficiency of the energy transfer.

It might be interesting to examine if the exact data regarding the red- or blue-shifted instantaneous driving frequencies for the optimal telegraph signal can be used as a quantitative guidance for other forces that are continuous in time. As the analytical predictions rely heavily on the precise validity of the in-phase relationship between the velocity and force, they might provide a robust guidance for continuously driven systems only at either earlier times, or for those forces whose amplitudes are perturbative compared to $dV/dx$, such that the red- or blue-shifts are negligible. To the best of our knowledge, there is presently no reliable analytical guidance for the optimal chirping of the frequency for strong quasi-monochromatic driving fields. In this case one could possibly try to generalize the present approach based on the velocity-force in-phase relationship for each single force-cycle. However, as the force’s amplitude will be time-dependent in this case, the corresponding equations that relate two consecutive maximal elongation amplitudes $A_n$ and $A_{n+1}$ will be nontrivial.

We would expect that the power law behavior can be applied for those potentials with $V(x \to \infty) \neq \infty$ to estimate the smallest possible time duration required to eject a particle to infinity. In the case of molecular optical physics, the inter-atomic Morse-potential $[1 - \exp(-x)]^2$ or the Lennard-Jones potential $(|x|^{-12} - |x|^{-6})$ can be used to examine the scaling of the shortest possible dissociation time with system parameters under optimal driving conditions.

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Appendix A: Optimal control theory for an amplitude-constraint force field

Here we briefly review how the usual tools of optimal control theory can be applied to a driven nonlinear oscillator, given by the Hamilton function $H = p^2/(2m) + V(x) - F(t)x$. Its Hamilton equations of motion, i.e., $dx/dt = p/m$ and $dp/dt = -V' + F(t)$ serve as a constraint together with the limitation of the amplitude of $F(t)$. In order to rewrite this constrained problem to maximize the final amplitude $x(T)$ as an equivalent but unconstrained problem, we can introduce the two Lagrangian multipliers (co-state functions) $\lambda_1(t)$ and $\lambda_2(t)$. As a result, we have to maximize the objective $J$, defined as

$$J = x(T) + \int_0^T dt[\lambda_1(p/m - dx/dt) + \lambda_2(-V' + F - dp/dt)], \quad (A1)$$

This objective $J$ is a functional of $F(t)$, $x(t)$, $\lambda_1(t)$ and $\lambda_2(t)$. It turns out that the notation can be simplified if we define a Hamiltonian $h$ according to $h = \lambda_1 p + \lambda_2 (-V' + F)$ leading to

$$J = x(T) + \int_0^T dt(\lambda_1 dx/dt + \lambda_2 dp/dt) + \int dt \, h, \quad (A2)$$

To account for the force field $F(t)$ that is amplitude-limited, i.e., $-F_0 \leq F(t) \leq F_0$, we can parameterize the force as $F(t) = F_0 \cos[\omega(t)t]$, where the introduced time-dependent ‘frequency’ $\omega(t)$ is an unknown real function of time. The optimization goal therefore translates to the determination of the time-dependence of $\omega(t)$, such that the resulting $F(t)$ can optimize the final amplitude of our nonlinear oscillator.

If we compute the variation of the objective $\delta J$, we obtain

$$\delta J = \delta x(T) + \int dt[\delta \lambda_1 dx/dt + \lambda_1 \delta dx/dt] + \lambda_2 \delta dp/dt + \int dt \, \delta h, \quad (A3)$$

If we integrate the terms containing $\lambda_1 \delta dx/dt$ and $\lambda_2 \delta dp/dt$ by parts, and using $\delta x(t = 0) = 0$ and $\delta p(t = 0) = 0$ due to the fixed initial conditions, we obtain

$$-\int dt[\lambda_1 (dx/dt) + \lambda_2 dp/dt] \int dt(\lambda_1 dx/dt + \lambda_2 dp/dt) + \lambda_1(T) \delta x(T) - \lambda_2(T) \delta p(T), \quad (A4)$$

Next, the variation of $h$ amounts to $\delta h = (\partial h/\partial \omega) \delta \omega + (\partial h/\partial x) \delta x + (\partial h/\partial \lambda_1) \delta \lambda_1 + (\partial h/\partial \lambda_2) \delta \lambda_2$. Using the specific definition of $h$, the partial derivatives simplify to $\partial h/\partial \omega = p/m$ and $\partial h/\partial \lambda_2 = -V' + F$.

If we require that the variation $\delta J$ vanishes for the optimal solutions, we have to require that each co-factor of each variation, $\delta \omega$, $\delta \lambda_1$, $\delta \lambda_2$, $\delta x$, $\delta p$, $\delta x(T)$ and $\delta p(T)$ vanishes, i.e., we obtain,

$$\partial h/\partial \omega = 0, \quad (A5)$$

$$dx/dt = p/m, \text{ with } x(0) = 0, \quad (A6)$$

$$dp/dt = -V' + F, \text{ with } p(0) = 0, \quad (A7)$$

$$d\lambda_1/dt = -\partial h/\partial x = \lambda_2 d^2V/dx^2, \text{ with } p(0) = 0. \quad (A8)$$
Eq.(A5) is the main control equation and can be solved. As \( h \equiv \lambda_1 p/m + \lambda_2 (-V' + F(t)) \) we have \( \partial h/\partial \omega = \lambda_2 F \partial \omega = -\partial F \partial \omega = -\lambda_2 \omega_0 \sin(\omega(t) t) = 0 \). From Eq.(A8) and Eq.(A9) it follows that neither of the two Lagrange parameters \( \lambda_1 \) or \( \lambda_2 \) can be zero. Therefore, the frequency \( \omega(t) \) can take only discrete values of either \( \omega(t) t = 0 \) or \( \omega(t) = \pi \). This means that the optimal \( F(t) \) is a telegraph-like signal oscillating between \( CF_0 \) or \( F_0 \). In other words, it can be expressed as \( F(t) = \text{Sign}[g(t)] = F_0 g(t)/|g(t)| \), where \( g(t) \) can be any function of time. We also note that the equations Eq.(A6) and Eq.(A7) for the state variables \( x(t) \) and \( p(t) \) do not contain the co-state variables, and the co-state equations Eq.(A8) and Eq.(A9) for \( \lambda_1(t) \) and \( \lambda_2(t) \) do not contain explicitly \( F(t) \). But as \( \lambda_1(t) \) and \( \lambda_2(t) \) depend on \( x(t) \), which depends on \( F(t) \), the set of equations have non-trivial solutions.

**Appendix B: Analytical estimates for the switching times**

In this appendix we derive the switching times for the two-level system. Let us first examine the first time interval from \( t = 0 \) to \( t = T_1 \) with \( F(t) = F_0 \) during which \( S_2(t) \) is positive. While there are in general no analytical solutions for \( S_1(t) \), \( S_2(t) \) and \( S_3(t) \), here \( F(t) \) is piecewise constant and solutions can be found for each cycle. We obtain

\[
S_1(t) = F_0 \omega_0 [-1 + \cos(\Omega t)]/\Omega^2, \quad (B1)
\]

\[
S_2(t) = F_0 \sin(\Omega t)/\Omega, \quad (B2)
\]

\[
S_3(t) = -[\omega_0^2 + F_0^2 \cos(\Omega t)]/\Omega^2, \quad (B3)
\]

This means that \( S_2 \) remains positive from \( t = 0 \) to \( t = T_1 = \pi/\Omega \).

In order to construct the second switch time \( T_1 \), we have to use \( S_1(T_1) = -2F_0 \omega_0 \), \( S_2(T_1) = 0 \), and \( S_3(T_1) = -[\omega_0^2 + F_0^2]/\Omega^2 \) as the new initial conditions and the negative force \( F(t) = -F_0 \) for the continued time evolution. Here we obtain the slightly more complicated solutions

\[
S_1(t) = F_0 \omega_0 [-3F_0^2 + \omega_0^2 + (F_0^2 - 3\omega_0^2) \cos(\Omega(t - T_1))]/\Omega^4, \quad (B4)
\]

\[
S_2(t) = F_0 (F_0^2 - 3\omega_0^2) \sin[\Omega(t - T_1)]/\Omega^3, \quad (B5)
\]

\[
S_3(t) = [3F_0^2 \omega_0^2 - \omega_0^4 + F_0^2 (F_0^2 - 3\omega_0^2) \cos(\Omega(t - T_1))]/\Omega^4, \quad (B6)
\]

which, in general, would predict the second switch time to be \( T_2 = T_1 + \pi/\Omega \).

However, \( S_1(t) \) is only negative in this interval \( T_1 < t < T_2 \) if the prefactor in Eq.(B5) is negative, in other words, if \( F_0 < 3^{1/2} \omega_0 \). If, however, it turns out that the magnitude of the force \( F_0 \) is actually larger than \( 3^{1/2} \omega_0 \), then \( S_2(t) \) would continue to be positive also in this second interval and -as a result- the rhs of Eq.(A12) is negative (as \( F(t) < 0 \)). This would lead to an (undesired) decrease of \( S_3(t) \) in this second cycle. This means that, in retrospect, we should not have switched the sign of \( F(t) \) at \( T_1 \). We could therefore (incorrectly) conjecture that we should have kept the sign of \( F(t) \) even for \( t > T_1 \) as positive. Quite interestingly though, the corresponding solution (for positive \( F(t) \)) in this cycle [given by \( S_2(t) = F_0 \sin(\Omega t)/\Omega \) is actually negative for \( t > T_1 \). This means that unfortunately the rhs of Eq.(12) is also negative in this case. This means that *neither sign* of \( F(t) \) can maintain the desired growth of \( S_3(t) \) in the second cycle. In other words, if \( F_0 > F_{thre,1} \), where the first threshold is given by \( F_{thre,1} = 3^{1/2} \omega_0 \), then this particular excitation scheme under a telegraph-like force field reaches its optimum inversion already after a single cycle of duration \( t = \pi/\Omega \), given by the positive amount

\[
S_{3, opt}(t) = [F_0^2 - \omega_0^2]/\Omega^2, \quad \text{if } F_0 > 3^{1/2} \omega_0 \quad (B7)
\]

which takes the value \( S_{3, opt} = 0.5 \) at the threshold for \( F_0 = F_{thre,1} \). Quite interestingly, we will show in Sec. V C that this particular value is actually the smallest possible inversion for any optimized telegraph-like excitation.

As a side issue, we mention that this abrupt halt of any further evolution of \( S_3(t) \) is quite fascinating when viewed from the perspective of the numerical solution to the Eq.(13), where we have used \( F(t) = F_0 \text{Sign}[S_2(t)] \). At time \( t = T_1 \), \( S_2(t) \) is numerically extremely close to zero and therefore the sign of \( F(t) \) basically becomes random and determined by numerical noise. However, \( F(t) \) remains a function of the sign of this noisy \( S_2(t) \). As a result, the functional form of Eq.(11) guarantees that \( S_2(t) \) remains close to zero and therefore \( dS_1(t)/dt \) as well as \( dS_3(t)/dt \) remain practically equal to zero. This occurrence of high frequency oscillations is analogous to the behavior of a simple electronic comparator with a slow moving, noisy input voltage. Here one usually employs a positive feedback in the form of a Schmitt trigger to eliminate the oscillations, which are unwanted in this case.

Let us now return to the case, where \( F_0 < F_{thre,1} \), such that the sign change of \( F(t) \) to negative for the second cycle actually continues the desired increase of \( S_3(t) \) and the evolution does not yet self-terminate. We would then find that (after the above-mentioned time \( T_2 = T_1 + \pi/\Omega \)) we should switch the sign of \( F(t) \) back to positive for the third cycle. In this interval we would obtain the solution for the polarization

\[
S_3(t) = F_0 (F_0^4 - 10F_0^2 \omega_0^2 + 5\omega_0^4) \sin[\Omega(t - T_2)]/\Omega^5 \quad (B8)
\]

If \( F_0 \) is sufficiently small such that the prefactor \( (F_0^4 - 10F_0^2 \omega_0^2 + 5\omega_0^4) \) is positive, we have \( F(t)S_2(t) \) positive as desired and the third switch time would be \( T_3 = T_2 + \pi/\Omega \).
On the other hand, if the prefactor \((F_0^3 - 10F_0^2\omega_0^2 + 5\omega_0^4) = (F_0^3 - z_i\omega_0^2)(F_0^3 - z_j\omega_0^2)\) [with \(z_1 \equiv (5 - 2 \times 5^{1/2})^{1/2} \approx 0.727\) and \(z_2 \equiv (5 + 2 \times 5^{1/2})^{1/2} \approx 3.078\)] happens to be negative, (then similar to the scenario above) \(S_3(t)\) is negative and leading to an undesired decrease of \(S_3(t)\). As the alternative solutions for \(T_2 \leq t < T_3\) (with a negative sign of the force) would also lead to a decrease of \(S_3(t)\), the evolution would come to a halt at time \(t = T_3\) with the optimum value \(S_3(T_2)\). In order to reach the second cycle, we had already required that \(F_0 < 3^{1/2}\omega_0 \approx 1.732\omega_0\). This means that \((F_0^3 - z_i\omega_0^2)(F_0^3 - z_j\omega_0^2)\) is negative if \(F_0 > z_1\omega_0\). This means that

\[
S_{3,\text{opt}} = \frac{(6F_0^2\omega_0^2 - \omega_0^4 - F_0^4)}{\Omega^4}
\]

if \((5 - 2 \times 5^{1/2})^{1/2} \omega_0 < F_0\) \hspace{1cm} (B9)

which takes the value \(S_{3,\text{opt}} = (5 + 5^{1/2})/4\) at the second threshold for \(F_0 = F_{thr,2} \equiv (5 - 2 \times 5^{1/2})^{1/2} \omega_0\). This particular threshold value is actually the second local minimum of the inversion for any optimized telegraph-like excitation. If the force amplitude \(F_0\) is smaller than \(F_{thr,2}\), this iterative scheme can be continued as the telegraph signal given by \(F(t) = F_0\text{Sign}[\sin(\Omega t)]\) will continue to increase \(S_3(t)\).

[25] For excellent works on very sophisticated feedback mechanisms used in laboratory settings in attoscience and for coherent control of atomic and molecular systems, see [26-41] and many references therein.
[42] G. Duffing, “Erzwungene Schwingungen bei

J. Guckenheimer and P. Holmes, “Nonlinear oscillations, dynamical systems, and bifurcations of vector fields” (Springer-Verlag, 1983).
