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Semiclassical dynamics of strongly driven systems

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A nonperturbative analytical semiclassical approach describing the interaction of a quantum system with strong oscillating fields is presented, including the limit where the external high-frequency field destroys the classical trajectories of a field-free system. Applied to ionization of a Rydberg atom, our approach allows us to describe the so-called ''interference'' and ''adiabatic'' mechanisms of laser-induced stabilization of atomic Rydberg states in a unified way. [S1050-2947(98)50208-9]

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Approximate analytical solutions of the Schrödinger equation for a system interacting with a strong oscillating field are the cornerstones of our understanding of quantum dynamics in intense laser fields. For example, the Keldysh-type [1] solutions, reincarnated in recent recollision models [2], have been crucial to our understanding of intense laser-atom physics.

The major problem in finding analytical solutions is the difficulty in treating both the binding potential and the external field equally and nonperturbatively. For example, Keldysh-type theories ignore atomic potential in the final state of a laser-driven atom. We develop a semiclassical approach that deals with this difficulty, and that can be applied to a broad class of problems in different areas of physics.

The most interesting and general applications of our method are to the class of problems in which a quantum system with slow field-free dynamics faces fast oscillating external fields. An atomic physics example would be photoionization of a Rydberg atom by a strong laser field of frequency $\omega_L > 1/2n^2$ (n is the principal quantum number). A molecular optics example would be rotational heating of a molecule trapped and aligned in a focus of an intense infrared laser beam [3].

We have also extended our solution to include the limit at which the high-frequency field completely destroys the trajectories of the field-free system. This allows us to show that for Rydberg atoms two different mechanisms of atomic stabilization—the so-called ''interference'' [4] and ''adiabatic'' [5] stabilization models—are two limits of the same expression. As the field increases, the approximately constant ionization rate $\gamma \sim 1/n^3$, found in the region of interference stabilization, gives way to a decreasing γ as the amplitude of electron oscillations in the external field approaches the characteristic size n^2 of the Kepler orbit.

Let a particle move in a time-independent potential $U_1(x)$ and interact with an external time-dependent field described by the potential $U_2(x,t)$ (e.g., $x\mathcal{E}\cos\omega_L t$ for the laser field). We assume that the solutions for the potentials $U_1(x)$ and $U_2(x,t)$ are known separately.

In the semiclassical approximation we look for the wave function in a form $\Psi(x,t)=\exp[iS(x,t)]$, with the initial condition $\Psi(x,t_0)\equiv\Psi_{in}(x)=\exp[iS_{in}(x)]$, which describes a field-free system at some initial moment t_0. For a standing wave, such as an eigenstate, the initial problem is solved for each of the two counterpropagating waves independently.

We look for $S(x,t)$ in a form $S(x,t)=S_1(x,t)+S_2(x,t)+\sigma(x,t)$. Here $\Psi_1(x,t)=\exp[iS_1(x,t)]$ satisfies the Schrödinger equation for the potential U_1 only, and $\Psi_2(x,t)=\exp[iS_2(x,t)]$ satisfies the Schrödinger equation for U_2 only. The initial condition is satisfied by setting $S_1(x,t_0)=S_{in}(x)$ and $S_2(x,t_0)=\sigma(x,t_0)=0$, where t_0 is the moment at which the external field is turned on. The condition $S_2(x,t_0)=0$ is met automatically if the external field is initially zero.

The semiclassical equation for $\sigma(x,t)$ is

$$-\frac{\partial\sigma}{\partial t}=\frac{1}{2m}\frac{\partial\sigma}{\partial x}\left[\frac{\partial\sigma}{\partial x}+2\frac{\partial S_1}{\partial x}+2\frac{\partial S_2}{\partial x}\right]+\frac{1}{m}\frac{\partial S_1}{\partial x}\frac{\partial S_2}{\partial x}, \tag{1}$$

where we dropped all terms linear in $\hbar$ and higher. Here we discuss a one-dimensional problem, but for periodic motions the method can also be used in a three-dimensional (3D) case.

Equation (1) is the Hamilton-Jacobi equation for $S(x,t)$ written in the form $S(x,t)=S_1(x,t)+S_2(x,t)+\sigma(x,t)$. The terms $\partial S_1/\partial x=p_1(x)$ and $\partial S_2/\partial x=p_2(x,t)$ are the momenta in each of the potentials U_1 and U_2 separately, and $\partial\sigma/\partial x=\delta p$ is the correction to these momenta in the exact expression $p=p_1+p_2+\delta p$. As long as $\delta p \ll p_1+p_2$, we can neglect $\partial\sigma/\partial x$ in square brackets in Eq. (1). There are at least two cases when δp is small. The first is a high-frequency external field in which fast and slow motions are well separated and hence $p\approx p_1+p_2$. Second, $p_2 \ll p_1$ results in $\delta p \ll p_1$.

Let $\tilde{x}(t')$ be a trajectory that arrives at point x, at the moment t, and satisfies the characteristic equation for Eq. (1) $d\tilde{x}/dt'=[p_1(\tilde{x})+p_2(\tilde{x},t')]/m$. We introduce

$$\tau(x)=m\int_{x_0}^{x}\frac{dx'}{p_1(x')}, \tag{2}$$

a classical time accumulated along the field-free trajectory between an arbitrary point x_0 and the point x. Denoting the

inverse function as $x_{cl}(\tau)$, we can write the *field-free* trajectory that arrives at point x at moment t as $x_{cl}[\tau(x)+t'-t]$.

Now, for fast-oscillating $p_2(x,t)$ the standard procedure of separating fast and slow motions [6] can be used to write $\tilde{x}(t')$ as a superposition of slightly modified field-free trajectory and oscillations $x_{osc}(t')$ around this trajectory:

$$\tilde{x}(t')=x_{cl}[\tau(x-x_{osc}(t))+t'-t]+x_{osc}(t'). \qquad (3)$$

Here $x_{osc}(t')$ is determined by $m\dot{x}_{osc}(t')=p_2(x_{cl}(\tau+t'-t),t')$. Modification $\tau(x)\to\tau(x-x_{osc})$ in x_{cl} in Eq. (3) is to ensure $\tilde{x}(t'=t)=x$. The solution Eq. (3) is valid if the amplitude of oscillations x_{osc} is small compared to both the size of the field-free orbit and the scale of the inhomogeneity of the external field: $p_{1,2}(x\pm x_{osc})\approx p_{1,2}(x)$.

Using the trajectory $\tilde{x}(t')$, one can verify that the approximate solution of Eq. (1) is

$$\sigma(x,t,t_0)=-\frac{1}{m}\int_{t_0}^{t}dt'\,p_1[\tilde{x}(t')]p_2[\tilde{x}(t'),t'], \qquad (4)$$

provided that $p_{1,2}(x)\approx p_{1,2}(x\pm x_{osc})$, that is, both fields are sufficiently homogeneous on the scale of x_{osc}. Then one can also replace $\tilde{x}$ in Eq. (4) with the field-free trajectory x_{cl} [see Eq. (3)], simplifying practical calculations of $\sigma(x,t,t_0)$.

Thus, $\sigma(x,t,t_0)$ is determined by the product p_1p_2 accumulated along the trajectory $\tilde{x}(t')$ that must arrive at a point x at time t with $p_1(x)=\partial S_1/\partial x$; initial momentum p_2 due to the external field (e.g., drift momentum in the laser field), is equal to zero. The separation of fast and slow motions can also be done for a 3D system, in which case p_1p_2 in Eq. (4) is replaced by a scalar product.

The solution of Eq. (4) is valid if (i) $p_{1,2}(x\pm x_{osc})\approx p_{1,2}(x)$, that is, sufficiently small oscillation amplitude; and (ii) $\delta p=\partial\sigma/\partial x\ll p_1+p_2$, that is, sufficiently small change in the zero-order momentum p_1+p_2 due to coupling of the two motions. None of these conditions explicitly requires high-field frequencies, and both can be satisfied at frequencies comparable to that of the system, provided $p_2\ll p_1$. In general, for short times $|\partial\sigma/\partial x|$ is always small. Its increase with time determines for how long our solution is applicable.

According to Eq. (4), $\delta p=\partial\sigma/\partial x$ is due to the work of the external field along the field-free trajectory and the work of the field $U_1(x)$ along the oscillating part x_{osc} of the trajectory $\tilde{x}(t')$ Eq. (3). Since this is less than $p_1^2/2m$, this work can still be large compared to the photon energy, ensuring that multiphoton processes dominate over single-photon and conventional perturbation theory is inapplicable.

The wave function evolution is given by $\Psi(x,t)=e^{i\sigma(x,t)}\Psi^{(0)}(x,t)$ with $\Psi^{(0)}(x,t)=\exp[i(S_1(x,t)+S_2(x,t)]$, which describes the evolution that ignores coupling of the two fields. Note that since an eigenstate corresponds to two counterpropagating waves, for such an initial condition one has two $\sigma(x,t,t_0)$ that differ by the direction of $p_1(x)$ on the classical trajectory.

Since σ does not have to be small compared to unity, the deviation $\Psi(x,t)-\Psi_{in}$ can be large—a useful property of perturbation theory *in action* compared to the standard quantum-mechanical perturbation theory.

Consider the laser field $\mathcal{E}f(t)\cos\omega_L t$, with envelope $f(t)$ sufficiently long to include many oscillations. Substituting $p_2=-\mathcal{E}f(t)\sin\omega_L t/\omega_L$ into Eq. (4) neglecting x_{osc} in the argument of p_1, and integrating by parts, we find that $\sigma=\mathcal{E}xf(t)\sin\omega_L t/\omega_L+\tilde{\sigma}$, where

$$\tilde{\sigma}(x,t,t_0)=-\int_{t_0}^{t}dt'\;\mathcal{E}x_{cl}(\tau-t+t')f(t')\cos\omega_L t'. \qquad (5)$$

The term $\mathcal{E}xf(t)\sin\omega_L t/\omega_L$ is gauge related and is cancelled by the identical term with a negative sign that appears in the Volkov propagator $\exp(iS_2)$ for the external field in the length gauge. Real absorption and/or emission of energy is described by $\exp[i\tilde{\sigma}(x,t,t_0)]$, which can be used to obtain simple expressions for multiphoton transitions.

To make the discussion more specific (yet keep the derivation general), consider a Rydberg atom in a state with principal quantum number $n\gg 1$ and an orbit with the eccentricity $\epsilon\approx 1$. Such a one-dimensional system can be realized experimentally [7]. Let the orbit be aligned with the electric field of the laser, and let the laser frequency be high: $1/2n^2<\omega_L\ll 1$ a.u.

The pulse duration T_L can be either long or short compared to the classical (Kepler) period of the system $T_n=2\pi n^3$. For $T_L\gg T_n$ we assume that complete ionization requires many Kepler periods and calculate the ionization probability $P_{ion}(T_n)$ over one Kepler period T_n. Obviously, for long pulses $T_L\gg T_n$, $P_{ion}(T_n)$ should be less than unity. In the opposite case of fast ionization short pulses $T_L\ll T_n$ have to be considered.

For $T_L\gg T_n$ the pulse envelope does not change significantly during one Kepler period. We set $f(t)=1$ and calculate $\exp(i\tilde{\sigma})$ over *one Kepler period* for the state $|n\rangle$, finding its depletion after one Kepler period. (In general, the wave function coincides with $|n\rangle$ only at t_0 and becomes a superposition of many $|n\rangle$ states once the pulse is on. However, the Shrödinger equation is linear and the propagator can be applied to each state in the superposition independently.) Changing the integration variable in Eq. (5) to $t''=\tau-t+t'$, we obtain

$$\tilde{\sigma}(x,t,t-T_n)=-\oint_{T_n}dt''\;\mathcal{E}x_{cl}(t'')\cos[\omega_L t''+\varphi(x,t)], \qquad (6)$$

where $\varphi(x,t)=\omega_L[t-\tau(x)]$ and the integral is calculated along the closed orbit arriving at point x at time t.

Equation (6) can be written as $\tilde{\sigma}=-\alpha\cos\varphi+\beta\sin\varphi$. Since $\exp(-i\alpha\cos\varphi+i\beta\sin\varphi)=\Sigma_k J_k(\sqrt{\alpha^2+\beta^2})\exp(+ik\varphi-ik\theta)$, where $\tan\theta\equiv\alpha/\beta$, we see that up to a common phase

$$\Psi(x,t)=\sum_m J_m(Z)e^{-im\theta}e^{+im\omega_L[t-\tau(x)]}\Psi_{in}(x), \qquad (7)$$

where

$$Z=\left| \oint_{T_n} dt''\ \mathcal{E}x_{cl}(t'')\exp(-i\omega_L t'') \right| = T_n\mathcal{E}|x_{cl}(\omega_L)|. \quad (8)$$

By projecting $\Psi(x,t)$ onto the field-free wave functions and using a standard semiclassical substitution of variables $x \rightarrow \tau(x)$ in the overlap integrals (that are accumulated at $x \ll 2n^2$) one can check what is already clear from the time dependence in Eq. (7): $A_m = e^{-im\theta}J_{-m}(Z)$ are the amplitudes of m-photon absorption after one round-trip along the Kepler orbit. The survival amplitude of the initial state $|n\rangle$, i.e., the amplitude of the initial wave function exactly reproducing itself after one Kepler period is $A_0 = J_0(Z)$. The probabilities of m-photon absorption after one Kepler period are $P_m(T_n) = |A_m|^2 = J_m^2(Z)$ and the total ionization probability is

$$P_{ion}(T_n) = \sum_{m \geqslant 1} P_m(T_n) = \frac{1 - J_0^2(Z)}{2} = \frac{1 - |A_0|^2}{2}. \quad (9)$$

We note that similar expressions for P_m were obtained for the case of laser-assisted bremsstrahlung in the pioneering paper [8] of Bersons, using a completely different approach. An analytical estimate for Z is [9] $Z \approx 2.58\mathcal{E}/\omega_L^{5/3}$. For $\mathcal{E} \ll \omega_L^{5/3}$ ionization requires many Kepler periods, and one can introduce the average ionization rate as $\Gamma_{ion} = P_{ion}(T_n)/T_n = [1 - J_0^2(Z)]/2T_n$.

Equation (8) is generalized for a 3D case by replacing $\mathcal{E}x_{cl}$ with the scalar product. The ionization probability [Eq. (9)] should then be averaged over the orientation of the Kepler trajectory with respect to the laser field axis, smearing out oscillations of $J_0^2(Z)$.

There is a well-known correspondence between semiclassical matrix elements x_{nm} and the Fourier components $x_{cl}(\omega_{nm})$ on the classical trajectory $x_{cl}(t)$. For a bound-free transition from the state $|n\rangle$ to the continuum state $|E\rangle$, $x_{cl}(\omega_{nE}) = \sqrt{2\pi/T_n}\,x_{nE}$ [9]. Using this relationship, in the limit $Z \ll 1$ one can easily see the equivalence of Γ_{ion} and the Fermi golden rule.

At $Z \sim 1$ ($\mathcal{E} \sim \omega_L^{5/3}$) complete depletion of the initial state occurs in one or fewer Kepler periods. Then it is logical to consider short pulses $T_L \leqslant T_n$. The correction $\exp(i\tilde{\sigma})$ is then calculated over the complete pulse duration. For $T_L \ll T_n$ we find that the ionization probability depends linearly on pulse duration [10] and the ionization rate can still be introduced. The rate is still given by the same formula $\Gamma_{ion} = [1 - J_0^2(Z)]/2T_n$, stabilizing around $\Gamma_{ion} \sim 1/2T_n$ at $Z \gg 1$ (the so-called ''death valley'').

The physical reason for a linear time dependence of the ionization probability in short pulses $T_L \ll T_n$ is simple: ionization of a Rydberg state occurs near the origin, while the wave function is delocalized over the whole orbit. Thus, initial conditions are evenly (in time) distributed along the Kepler orbit, and the number of trajectories that pass the origin during $T_L \ll T_n$ depends linearly on T_L.

In the field-free system classical actions for the states $|n\rangle$ and $|n+1\rangle$ differ by unity, and hence $\tilde{\sigma}, Z \geqslant 1$ means a strong mixing of adjacent Rydberg states, which forms the physical basis of the ''interference'' stabilization model [4]. The critical field for the onset of stabilization $\mathcal{E} \sim \omega_L^{5/3}$ agrees with the predicition of [4], as well as the minimum ionization lifetime $\tau_{min} \sim T_n$. The ''death valley'' of field strengths where $\tau_i \sim T_n$ is also predicted by the interference stabilization model when l mixing is taken into account [11]. Our calculation assumes an aligned orbit, which requires mixing of many l.

The applicability of the above results is limited by requiring small oscillation amplitude $\alpha = \mathcal{E}/\omega_L^2$ and small $\partial\tilde{\sigma}/\partial x$. To quantify these conditions we note that absorption and/or emission of photons by a Rydberg atom occurs at distances $x_{int} \sim \omega_L^{-2/3}$ [4], where $p_1 \sim \omega_L^{1/3}$. Using Eq. (6) and an estimate $Z \sim \mathcal{E}/\omega_L^{5/3}$, one finds that both δp and α are small as long as $\mathcal{E} \ll \omega_L^{4/3}$. We also note that our approximate semiclassical solution does not correctly describe the long-term ($t > T_n$) dynamics of outgoing above-threshold wave packets at $x \gg x_{int}$, since for them the correction to the field-free propagator is no longer small at $x \sim n^2$. However, this does not affect the ionization rates, which are determined at $x \sim x_{int}$.

Let us now address the problem of generalizing the results to the case of large oscillation amplitude $\alpha \gg x_{int}$ and relating ''interference'' and ''adiabatic'' stabilization pictures for Rydberg atomic states. Adiabatic, or Kramers-Henneberger, stabilization is associated with the Kramers-Henneberger (KH) transformation to the reference frame oscillating with the electron. In this frame the exact potential for the electron motion is $V(x - \alpha\cos\omega_L t)$, where $V(x)$ is the field-free (e.g., Coulombic) potential. Using the Fourier expansion, one can write

$$V(x - \alpha\cos\omega_L t) = V_0(x) + \sum_{k \geqslant 1} V_k(x)\cos k\omega_L t. \quad (10)$$

In high-frequency fields the second term on the right-hand side is argued to be a weak perturbation [5]. Eigenstates of the potential $V_0(x)$, which can be interpreted as an effective potential of a field-dressed system, are expected to be long-lived. This constitutes the main idea of ''adiabatic'' stabilization. For Rydberg states of an atom these effects are expected around $\alpha \sim n^2$ [13]. Adiabatic (or Kramers-Henneberger) stabilization is not identical to interference stabilization since the former can also occur in ground states of short-range potentials, even before the distortion of the short-range potential induces new bound states in the potential well $V_0(x)$ [12]. However, for Rydberg states we find that two stabilization pictures appear as two limits of the same general expression.

The key idea in generalizing our approach to the case of $\alpha \sim n^2$ is to treat $V_0(x)$ as $U_1(x)$ and the remaining part of Eq. (10) as the fast oscillating potential $U_2(x,t)$, directly including the major aspect of orbit distortion into the ''slow'' part of the trajectory. Separation of fast and slow motions in the KH frame requires that local oscillation amplitudes $a_k(x) = |dV_k/dx|/k^2\omega_L^2$ are small compared to the characteristic length scale of V_0 and V_k, which in the limit $\alpha \gg n^2$ is given by α. With increasing $\mathcal{E}$ a_k decreases and α increases and, hence, the condition $a_k \ll \alpha$ is better satisfied.

Following the same procedure as described above, we find that the semiclassical propagator describing absorption and/or emission of energy is $\exp[i\tilde{\sigma}(x,t,t_0)]$, where

$$\tilde{\sigma}=-\sum_{k\geqslant 1}\int_{t_0}^{t}dt'\ V_k(x_{KH}[t'+\tau_{KH}(x)-t])\cos(k\omega_L t'), \tag{11}$$

where $x_{KH},\tau_{KH}(x)$ refer to the trajectory in the potential $V_0(x)$. We change the integration variable to $t''=t'+\tau-t$ and introduce $\tilde{\sigma}(x,t,t-T_n^{(KH)})$, where $T_n^{(KH)}$ is the round-trip period in the KH potential $V_0(x)$. Since $\tilde{\sigma}(x,t,t-T_n^{(KH)})$ is a periodic function of $\varphi(x,t)=\omega_L[t-\tau_{KH}(x)]$, one can expand $\exp(i\tilde{\sigma})$ in the Fourier series in φ. The zero-order component of the expansion gives the amplitude of the initial state $|n^{(KH)}\rangle$ exactly reproducing itself after one classical period $T_n^{(KH)}$ (the survival amplitude):

$$A_0=\frac{1}{2\pi}\int_0^{2\pi}d\varphi\ \exp\left(i\sum_{k\geqslant 1}\tilde{\sigma}_k(\varphi)\right),$$

$$\tilde{\sigma}_k(\varphi)=-\oint dt''\ V_k(t'')\cos(k\omega_L t''+k\varphi). \tag{12}$$

Using the equivalence of the norm in φ and the Fourier domain, we find that $|A_0|^2+2\Sigma_{m\geqslant 1}|A_m|^2=1$ and, similar to the previous result, the ionization probability is $P_{ion}^{(KH)}(T_n^{(KH)})=(1-|A_0|^2)/2$.

Intereference stabilization is easily obtained as the limit of $P_{ion}^{(KH)}(T_n^{(KH)})$ at $\alpha\ll x_{int}$. Using the Taylor expansion we write $V(x-\alpha\cos\omega_L t)\approx V(x)-V_x'\alpha\cos\omega_L t$. Then Eq. (12) gives $A_0=J_0(Z^*)$ with $Z^*=T_n\alpha|V_x'(\omega_L)|$. The Fourier component $V_x'(\omega_L)$ is calculated on the field-free trajectory described by the Newton equation $\ddot{x}=-V_x'(x)$. Hence, $|V_x'(\omega_L)|=\omega_L^2|x(\omega_L)|$, yielding $Z^*=T_n\mathcal{E}|x(\omega_L)|$, identical to that given by Eq. (8). This establishes the connection between interference stabilization and the KH picture.

Adiabatic stabilization should appear as the limit of Eq. (12) at $\alpha\sim n^2$. For estimates we used a model potential $V(x)=-1/\sqrt{x^2+1}$. We found that for $\alpha\sim n^2$ ionization of a Rydberg state $n^{(KH)}\gg 1$ between the wells of $V_0(x)$ (i.e., $|x|\ll\alpha$) is negligible. Indeed, for $\alpha\sim n^2\gg 1$ the Rydberg electron motion between the wells of $V_0(x)$ is very slow and the Fourier integrals of this motion at frequencies $k\omega_L\gg\omega_n^{(KH)}$ are very small. [For $k|x|\ll\alpha$, $V_k\sim\cos(kx/\alpha-\pi k/2)V_0(x)$ with very flat $V_0(x)\sim-2\ln\alpha/(\pi\sqrt{\alpha^2-x^2})$ [14]. For $n^2\sim\alpha$, all $\tilde{\sigma}_k$ remain very small compared to unity as long as $x_{KH}(t')\ll\alpha$; $\tilde{\sigma}_k\ll 1$ indicates low ionization.]

Since ionization can only occur from the wells of $V_0(x)$, the ionization lifetime is determined by (i) the modification of $T_n^{(KH)}$ with increasing α and (ii) the efficiency of ionization during one pass of the well. Ionization cannot occur faster than in $\tau\sim T_n^{(KH)}$, which increases as the potential $V_0(x)$ is stretched with increasing α.

In the vicinity of the wells where $\Delta x\equiv\alpha-x\ll\alpha$ we have $V_k(x)\approx V_0(x)$ [14] as long as $k^2|\Delta x|\ll\alpha$. An estimate using this approximation shows that when the trajectory enters the well, $\exp(i\tilde{\sigma})$ becomes fast oscillating and A_0 quickly deviates from unity. Hence, for a Rydberg state $n^{(KH)}\gg 1$, ionization during one pass through the well is efficient and the wave function is depleted in $\tau\sim T_n^{(KH)}$. The change in $T_n^{(KH)}$ with increasing α determines the partial stabilization of the system.

In conclusion, the physical situation of having two different time scales for coupled (fast and slow) motions is quite typical in many areas of physics. Although the survival amplitude of the initial state was calculated here for a Rydberg atom, the derivation is general and valid for periodic trajectories in other systems, as long as the region of efficient photon absorption is small compared to the size of the field-free orbit.

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