Ionization in strong low-frequency fields: from quantum S-matrix to classical pictures

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Abstract

This text gives a coarse overview of the physics of strong field ionization and one formalism that can be used to describe it – the so-called strong-field S-matrix. It also deals with the basics of the Strong Field Approximation (SFA). The formalism is then used to develop the theory of high harmonic generation, and crudely extended to multi-electron systems. Much work remains to be done to go beyond the SFA, and develop consistent theory of HHG in multi-electron systems that does not rely on the SFA.

I. BASIC FORMALISM FOR IONIZATION

We shall begin with the single active electron approximation, which has worked extremely well in atoms driven by low-frequency fields. We will discuss how to include many electrons later – and this is far from done at the moment.

Moreover, we shall first focus on the ionization part, because it is the key component of what we need to do for the harmonic project.

The formal solution of the TDSE

$$i\dot{|}\Psi\rangle = \hat{H}(t)|\Psi\rangle \tag{1}$$

is

$$|\Psi(t)\rangle = e^{-i\int_{t_i}^{t}\hat{H}(\tau)d\tau}|\Phi_i\rangle = \hat{U}(t,t_i)|\Phi_i\rangle \tag{2}$$

Evaluating the exponential operator $\hat{U}(t,t_i)$ is a tedious task, no easier than solving the TDSE. However, we will find good use to this formal solution very shortly.

Let us re-write Eq.(1) in a slightly different form,

$$i\dot{|}\Psi\rangle = \hat{H}_0|\Psi\rangle + \hat{V}|\Psi\rangle \tag{3}$$

What I have done here is broken the full Hamiltonian H into two parts – $H = H_0 + V$. Often, H_0 is taken as the field-free Hamiltonian and $V(t)$ is taken as the interaction with the laser field, $\hat{V} = -\hat{\mathbf{d}}\mathbf{F}(t)$, where $\hat{d}$ is the dipole operator and $F(t)$ is the electric laser field. However, I could have broken H into any other pair, for example $H = H_V + U$ where

$$\hat{H}_V = \frac{\hat{\mathbf{p}}^2}{2} + \hat{V}(t) \tag{4}$$

describes the interaction of a free electron with the laser field and U is the potential of the ionic core. The separation of the Hamiltonian $H(t)$ into two parts is known as 'partitioning' of the Hamiltonian.

Our immediate goal is to relate the solution for the full Hamiltonian H to the solution for the Hamiltonian H_0, the latter presumably known. Let us write the TDSE with the Hamiltonian H_0

$$i\dot{|}\Psi^{(0)}\rangle = \hat{H}_0|\Psi^{(0)}\rangle \tag{5}$$

Its formal solution is

$$|\Psi^{(0)}\rangle = e^{-i\hat{H}_0 t}|\Psi(t=0)\rangle = e^{-i\hat{H}_0 t}|\Phi_i\rangle \tag{6}$$

where $|\Phi_i\rangle$ is the same initial state of the system as above in Eq.2 and the index (0) means that this solution applies to the Hamiltonian H_0.

What is the relationship between $\Psi(t)$ and $\Psi^{(0)}(t)$? Direct substitution into the TDSE Eq.(1) shows that its exact solution can be written as

$$|\Psi(t)\rangle = |\Psi^{(0)}(t)\rangle + |\Delta\Psi(t)\rangle = e^{-i\hat{H}_0 t}|\Phi_i\rangle + |\Delta\Psi(t)\rangle$$
$$|\Delta\Psi(t)\rangle = -i\int_{t_i}^{t} dt' e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau}\hat{V}(t')e^{-i\hat{H}_0(t'-t_i))}|\Phi_i\rangle \quad (7)$$

Substituting this into the TDSE shows that it does indeed work.

Let's be frank: Eq.(7) does not look very inviting. However, it this this general – and exact – expression where interesting approximations can be explicitly tried, sometimes based on rigorous math and sometimes based on physical reasoning.

Let us look at the physics behind the expression Eq.(7). The system starts in the state $|\Phi_i\rangle$. During the time-interval before some moment t' it evolves without interacting with the laser field, as if the field was not there. This may look weird, but one can't argue with the exact expression. Besides, it is very convenient: if the initial state is an eigenstate of the Hamiltonian H_0 with energy E_i, all this evolution does is accumulates the phase due to the energy, $\exp[-iE_i(t'-t_i)]$.

This quiet evolution ends at a moment t' when the system is kicked by the instantaneous laser field $V(t')$. To which state the transition occurs at this moment is anybody's guess. It is called a virtual transition and it can go anywhere – the energy conservation law need not to be satisfied until the interaction is over. The single dipole operator hiding behind $\hat{V} = \hat{r}\mathbf{F}$ can induce single-photon transitions to the continuum states even when the photon energy is much less than the binding energy (ionization potential) I_p. Such transitions do not yet mean that the electron has become free – only that it is transiently populating continuum states and may become free at some point when (or if) this virtual population becomes real, that is, satisfying the energy conservation law.

From the moment t' to the moment of observation t the evolution is under the action of the full Hamiltonian, including both the laser field and the field-free potential.

At the moment we are going to look at ionization, which means that our final state is the continuum state. If we are interested to find the transition amplitude from the initial bound state Φ_i to some final continuum state Ψ_f, then the wavefunction must be projected onto that state. The projection of $\Psi^{(0)}(t)$ onto Ψ_f is zero, and the transition amplitude a_{fi}

is

$$a_{fi}(t) = \langle \Psi_f | \Psi(t) \rangle = \langle \Psi_f | \Delta\Psi(t) \rangle =$$
$$= -i \int^t dt' \langle \Psi_f | e^{-i \int_{t'}^t \hat{H}(\tau) d\tau} V(t') e^{-i\hat{H}_0(t'-t_i))} | \Phi_i \rangle \quad (8)$$

Once we decide to take a look at HHG, we will need to use the same amplitudes, as we shall see later.

This expression is often refereed to as strong-field S-matrix. It is exact. No approximations have been made yet. Now let's turn to approximations.

II. THE STRONG FIELD APPROXIMATION

Let us think about the physics of the situation in the strong low frequency field. "Low frequency" means "compared with the characteristic response frequency" of the system – the inverse of the excitation frequency. For most systems of interest it is in the deep UV, so that the response time is few atomic units, or about 100 asec. Thus, the interaction with the IR field would be in the low-frequency regime.

While the electron is in the initial – ground – state, not much is happening until it manages to escape to the continuum at some t', which could be pretty much any instant of time. At this point in time the strong field takes the electron over and it starts to oscillate in the field, possibly scattering on the parent ion. Can we put this physical picture into the mathematical terms? Or, rather, can we use this picture to do something to the formal expressions for the amplitude a_{fi}?

Once we realize that the ground state and the continuum are the two main participants in the dynamics, the plan is rather clear. Indeed, in Eqs.(7,8) first the electron sits in the ground state until the moment t', at which point the laser field $V(t')$ kicks it to the continuum. Now, while in the continuum, the electron is dominated by the laser field. Therefore, instead of the exact propagator

$$\exp(-i \int_{t'}^t \hat{H}(\tau) d\tau)$$

we will use an approximate propagator that includes the laser field fully and exactly but completely ignores the field-free potential of the system.

$$a_{fi}(t) \sim -i \int^t dt' \langle \Psi_f | e^{-i \int_{t'}^t \hat{H}_{\rm v}(\tau) d\tau} V(t') e^{-i\hat{H}_0(t'-t_i))} | \Phi_i \rangle \quad (9)$$

where $H_{\rm v}$ is given by Eq.(4). This is the essence of the Strong Field Approximation. One of the main reasons to make such as approximation – completely neglect the atomic (or molecular) potential in the continuum – is that the propagator for the free electron in the laser field

$$U_{\rm V}(t,t') = e^{-i\int_{t'}^{t}\hat{H}_{\rm V}(\tau)d\tau} \tag{10}$$

is known exactly. It is called the Volkov propagator. It corresponds to solving the TDSE for the Hamiltonian $H_{\rm V}(t)$, which in the length gauge is:

$$\hat{H}_{\rm v}(t) = \frac{\mathbf{p}^2}{2} + xF(t) \tag{11}$$

with x being the polarization direction of the laser field.

A. The Volkov Propagator and the classical connection

To understand how the Volkov propagator acts on the electron in the laser field, let us see what would it do with the plane wave $\exp(i\mathbf{kr})$, which describes the free electron which has the kinetic momentum $\mathbf{k}$ along the laser polarization.

All the free electron does in the laser field is oscillates. Classically, if at time t' the electron has kinetic momentum $k' = k(t')$ (kinetic momentum $k(t') = mv(t')$ is proportional to the instantaneous velocity), then its kinetic momentum at any other time is

$$\mathbf{k}(t) = \mathbf{k}(t') - \mathbf{A}(t') + \mathbf{A}(t) \tag{12}$$

where $A(t)$ is the vector-potential of the electromagnetic field, which in the dipole approximation does not depend on spatial coordinate and is related to the electric field of the laser pulse as $F(t) = -\dot{A}(t)$.

Note that

$$\mathbf{k}(t) - \mathbf{A}(t) = \mathbf{k}(t') - \mathbf{A}(t') = \mathbf{p} = \text{const} \tag{13}$$

The conserved quantity $\mathbf{p}$ is called the canonical momentum.

This purely classical picture shows up when we apply the quantum Volkov propagator to the plane wave with the kinetic momentum k' at the moment t':

$$e^{-i\int_{t'}^{t}\hat{H}_{\rm V}(\tau)d\tau}|\mathbf{k}'\rangle = e^{-i\int_{t'}^{t}E(\tau)d\tau}|\mathbf{k}\rangle \tag{14}$$

Here the plane waves $|k\rangle, |k\rangle$ have different momenta related by the Eq.(12), and the instantaneous kinetic energy is

$$E(\tau) = \frac{1}{2}[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 = \frac{1}{2}[\mathbf{k}' - \mathbf{A}(t') + \mathbf{A}(\tau)]^2$$
$$\mathbf{k} = \mathbf{k}' - \mathbf{A}(t') + \mathbf{A}(t) \tag{15}$$

In terms of the conserved canonical momentum $\mathbf{p}$, which is also equal to the kinetic momentum of the electron after the laser field is switched off, the result can be re-written as

$$e^{-i\int_{t'}^{t}\hat{H}_{\rm V}(\tau)d\tau}|\mathbf{p} + \mathbf{A}(t')\rangle = e^{-i\int_{t'}^{t}E(\tau)d\tau}|\mathbf{p} + \mathbf{A}(t)\rangle \tag{16}$$

Thus, the coordinate part of the wavefunction has been changed, but the temporal phase added is the same for all coordinates because the interaction (the laser field) is homogeneous. This is the only reason we could replace the Hamiltonian operator in the exponent with the energy.

The result above is exact and can be obtained in many ways. Let me now derive it using the classical action and the semiclassical approach. While this route is not the simplest, it gives you a very clear feeling for the quantum-classical connection. For simplicity of notations, let me assume that the motion is one-dimensional, parallel to the laser field polarization. Generalizing to 3D is easy and is a good exercise.

Our initial condition – the plane wave $\Psi(x,t') \propto \exp(ik'x)$ at the moment t' has the nice semiclassical form $\Psi(x,t') \propto \exp(iS)$ with the initial condition for the action $S_F(k',x,t=t',t') = k'x$. So let us look for the solution of the TDSE in the form

$$\Psi(x,t) = e^{iS_F(k',x,t,t')}$$
$$S_F(k',x,t',t') = k'x \tag{17}$$

The many arguments in S_F denote the initial momentum, the initial time, and the coordinate x at the moment t. The subscript F stands for 'full' – it stresses that we have full classical action that depends on initial and running times, coordinates, and momenta. The reason to use this notation will become clear later, when a part of S_F will be denoted as S, following the tradition that has (unfortunately) entrenched itself in the strong field community and leads to a lot of confusion when the component S of the full action S_F is referred to as the classical action.

Substituting this expression into the TDSE for the free electron in the laser field yields exact equation for the action

$$-\frac{\partial S_F}{\partial t} = \frac{1}{2}\left[\frac{\partial S_F}{\partial x}\right]^2 + xF(t) - \frac{i}{2}\frac{\partial^2 S_F}{\partial x^2} \tag{18}$$

This equation differs from the classical Hamilton-Jacobi (HJ) equation for the action due to the presence of the last term, which involves the second-order spatial derivative. It coincides with the HJ equation if this term is equal to zero. This is precisely the case for the potentials that depend on x linearly, which allows for solutions that also linearly depend on x.

Exact solution of Eq.(18), subject to the initial conditions above, is

$$S_F = k'x + [A(t) - A(t')]x - \frac{1}{2}\int_{t'}^{t} d\tau \left[k' - A(t') + A(\tau)\right]^2 = k(t)x - \frac{1}{2}\int_{t'}^{t} d\tau k^2(\tau) \tag{19}$$

and is identical to the classical action for the free electron in the laser field.

B. Transition amplitudes in the SFA

Let the system start at the moment t_i in the ground state $\Phi_i \equiv \Phi_g$ with the energy $E_g = -I_p$ (and hence $\exp(-i\hat{H}_0(t' - t_i)) = \exp(+iI_p(t' - t_i))$. The amplitude to find the system with the momentum $\mathbf{k}$ at the time t, according to the general equation Eq.(8), is

$$a(\mathbf{k}, t) = -i\int^{t} dt' \langle \mathbf{k}| e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} V(t') e^{i\hat{I}_p(t'-t_i))}|\Phi_g\rangle \tag{20}$$

In the SFA, we replace the exact propagator between t' and t with the Volkov propagator,

$$a(\mathbf{k}, t) = -i\int^{t} dt' \langle \mathbf{k}| e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau} V(t') e^{i\hat{I}_p(t'-t_i))}|\Phi_g\rangle \tag{21}$$

Now we use the fact that have just learned how to propagate the plane wave in the laser field, and apply this knowledge to the 'bra':

$$\langle \mathbf{k}| e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau} = e^{-i\int_{t'}^{t}E(\tau)d\tau}\langle \mathbf{k} + \mathbf{A}(t') - \mathbf{A}(t)| = e^{-i\int_{t'}^{t}E(\tau)d\tau}\langle \mathbf{k}(t')| \tag{22}$$

with the instantaneous kinetic energy

$$E(\tau) = \frac{1}{2}[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 = \frac{1}{2}[\mathbf{k}' - \mathbf{A}(t') + \mathbf{A}(\tau)]^2 \tag{23}$$

The SFA expression for the amplitude to find the system with the momentum $|\mathbf{k}\rangle$ at an instant t becomes

$$a_{\mathbf{k}}(t) = -i\int_{t_i}^{t} dt' e^{-i\int_{t'}^{t}E(\tau)d\tau + iI_p(t'-t_i)}\langle \mathbf{k} + \mathbf{A}(t') - \mathbf{A}(t)|V(t')|\Phi_g\rangle \tag{24}$$

where

$$E(\tau) = \frac{1}{2}[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 \tag{25}$$

since we have fixed the momentum $\mathbf{k}$ at the instant t.

In the literature, the $\mathbf{k}, t, t'$-dependent phase of this integral,

$$S(t, t', \mathbf{k}) = \frac{1}{2}\int_{t'}^{t} d\tau[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 - I_p t' \tag{26}$$

is often referred to as the classical action of the free electron in the laser field. It is a stretch of the term, since it also includes the $-I_p t'$, and misses the coordinate-dependent part. Often you will find it written in terms of the canonical momentum $\mathbf{p} = \mathbf{k} - \mathbf{A}(t)$, which is the conserved quantity and will stay unchanged at all times — that's within the SFA, of course. In this case the phase will look like

$$S(t, t', \mathbf{p}) = \frac{1}{2}\int_{t'}^{t} d\tau[\mathbf{p} + \mathbf{A}(\tau)]^2 - I_p t' \tag{27}$$

In terms of action and the canonical momentum $\mathbf{p}$, the SFA transition amplitude is

$$a_{\mathbf{p}}(t) = -i\int^{t} dt' e^{-iS(t,t',\mathbf{p})-iI_p t_i}\langle \mathbf{p} + \mathbf{A}(t')|V(t')|g\rangle \tag{28}$$

In future, if and when you come across such expressions, pay special attention to the meaning of the momentum – is it canonical or kinetic.

Eqs.(24,28) are intuitive and clear. The electron sits in the ground state until t' when it makes (at this point still virtual) transition to the continuum. Then the electron moves in the laser field, converting the virtual transition into real and oscillating – as the free electron should. It accumulates the phase given by the integral of its instantaneous energy $E(\tau)$, performed between the moment of birth t' and the moment of observation t. The electron finishes with the canonical momentum $\mathbf{p}$, which dictates the initial kinetic momentum that the electron populates at t', $\mathbf{k} = \mathbf{p} + \mathbf{A}(t')$.

There are several major problems with this result, all stemming from the main approximation of the theory - to neglect the effect of the Coulomb potential.

(1) During the transition to the continuum the electron will be liberated differently if we include its interaction with the atomic core. After all, if it has to tunnel through the barrier, the shape of this barrier is important – and it is heavily affected by the binding potential. Thus, the ionization amplitude will be different. This problem can be corrected by incorporating the effect of the Coulomb tail into the electron action.

(2) The propagation in the continuum is also different: the electron not only oscillates in the laser field, it can also scatter off the atomic core. This is not present in the SFA formalism, but can be included additionally: The SFA result can be considered as the first term in a perturbative expansion, with the atomic potential being a perturbation. The corresponding new terms are often referred to as SFA2, and have been derived and analyzed by M. Lewenstein et al, see Ref.[1].

(3) The Volkov propagator is sensitive to the gauge. The one in these notes is written in the length gauge. In the velocity gauge where $\hat{V} = \hat{\mathbf{p}}\mathbf{A} + \mathbf{A}^2/2$ the plane wave state $|\mathbf{p}\rangle$ with canonical momentum $\mathbf{p}$ stays the same between t' and t. Physically, this is simply because in the velocity gauge $\mathbf{p}$ refers to *canonical*, not *kinetic* momentum, and in the laser field the *canonical* momentum of the free electron is a constant of motion. This does not lead to any problems in the exact theory - i.e. if the electron was indeed free at all times. But in the approximate theory this is not the case - the initial field-free ground state is not a state of the free electron. *The result is that SFA is not gauge invariant, which is bad news for a theory.*

However, the good news is that the gauge problem directly affects only the pre-exponential terms in the amplitude, and keeps the major piece – which is the fast oscillating exponent – intact. So with the exponential part we are more-or-less safe, at least as far as gauges go.

(4) Finally, by using the plane waves as continuum states, we have selected a basis which is not orthogonal to the initial state of the system. In other words, in addition to all other problems our basis set is also overcomplete: it includes all plane waves that already make up a complete basis *plus* the extra state. The pre-exponential factor which include the bound-free transition matrix elements suffers greatly – but the key exponential dependence stays the same.

To summarize, SFA is wrong in so many ways that it violates every rule in the theory book. But the physical picture that stands behind it is intuitive, clear, and compelling. As a result, the SFA is used very widely, and it works very well for gaining qualitative and – with proper modifications along the lines described first in Ref. [2] and further developed by [3] – even quantitative insight into the physics of intense laser-matter interaction.

III. STRONG-FIELD IONIZATION: EXPONENTIAL VS POWER LAW

Let us now use it to look at the dynamics of strong-field ionization, and see how exponential dependence on the laser field strength typical for tunnelling turns into a power law dependence commonly associated with multi-photon ionization.

Up to the global phase factor $e^{iI_p(t-t_i)}$ the SFA probability amplitude of populating the field-free continuum state labelled by the canonical momentum $|\mathbf{p}\rangle$ is

$$a_{\mathbf{p}}(t) = -i\int^t dt' e^{-iS(t,t',\mathbf{p})}\langle \mathbf{p}+\mathbf{A}(t')|V(t')|g\rangle$$
$$S(t,t',\mathbf{p}) = \frac{1}{2}\int_{t'}^t d\tau[\mathbf{p}+\mathbf{A}(\tau)]^2 + I_p(t-t') \tag{29}$$

Let us assume that the field is linearly polarized, $\mathbf{F}\cos\omega t$. Then the vector-potential is

$$A_\parallel = -\frac{\mathbf{F}}{\omega}\sin\omega t = -v_0\sin\omega t$$
$$A_\perp = 0 \tag{30}$$

Then, keeping only exponential accuracy, we obtain

$$a(\mathbf{p},t) \sim \int_{-\infty}^t dt' \exp(-iS(\mathbf{p},t,t')) \tag{31}$$

where

$$S(\mathbf{p},t,t') = \left(I_p + \frac{1}{2}p_\perp^2\right)(t-t') +$$
$$+\frac{1}{2}\int_{t'}^t d\tau\left[p_\parallel - v_0\sin\omega\tau\right]^2 \tag{32}$$

is commonly refereed to in the literature as the action integral (even though, rigorously speaking, it is not quite that), $v_\parallel$ and $v_\perp$ are the velocity components parallel and perpendicular to $\mathbf{F}$ and $v_0 = \mathbf{F}/\omega$ is the velocity amplitude of electron oscillations.

You already see that having non-zero perpendicular momenta is like increasing Ip. Hence, for now $p_\perp$ can be set to zero. If we are interested in the probabilities of populating non-zero $p_\perp$, all we need to do is take the formula for $p_\perp = 0$ and replace I_p with $I_p + p_\perp^2/2$.

This expression is the standard SFA expression for the ionization amplitudes. The next step is to evaluate the integral.

A. The saddle point approximation and the classical connection

To evaluate the integral, we need to understand what does the phase $S(\mathbf{p}, t, t')$ in this integral do. The best way to approach this is to re-write S in terms of dimensionless variables: let us pull v_0 out of the brackets of Eq.(32), and also introduce phase $\phi = \omega\tau$ as a new dimensionless integration variable:

$$S(\mathbf{p}, t, t') = \frac{I_p}{\omega}(\omega t - \omega t') + \frac{v_0^2}{2\omega}\int_{\omega t'}^{\omega t} d\phi \, [u - \sin\phi]^2 \tag{33}$$

where the dimensionless momentum is $u = p_\parallel / v_0$.

We see that there are two important parameters in the phase – $N = I_p/\omega$ and

$$\frac{v_0^2}{2\omega} = 2\frac{U_p}{\omega} = 2Z \tag{34}$$

where $U_p = \mathbf{F}^2/4\omega^2$ is the ponderomotive energy – the laser-cycle-average kinetic energy of the oscillating electron, and Z is known as the Reiss parameter. The number $N = I_p/\omega$ tells us roughly how many photons are required for ionization.

In strong low-frequency fields both parameters are very large, meaning that the phase is changing very rapidly with time. For example, at intensities around $10^{14}\mathrm{W/cm}^2$ and for $w = 1.56\mathrm{eV}$, which corresponds to the laser wavelength of $800nm$, the characteristic value of the second term in the phase, over one laser cycle, is about 10 π, which is a very large phase change over one laser cycle.

This is very good news. If the phase is large and oscillates quickly, we can use the saddle-point method to calculate the integral.

The saddle-point method proceeds as follows. First, one looks for the values of the integration variable t' where the phase of the integrand is stationary, that is, the derivative with respect to t' is zero. In our case we have

$$\begin{aligned} S(\mathbf{p}, t, t') &= I_p(t - t') + \frac{v_0^2}{2}\int_{t'}^{t} d\tau \, [u - \sin\omega\tau]^2 \\ \frac{\partial S(\mathbf{p}, t, t')}{\partial t'} &= -I_p - \frac{v_0^2}{2}[u - \sin\omega t']^2 = 0 \end{aligned} \tag{35}$$

Solving this equation is the key component of the calculation, and we will turn to it shortly. Once we find the stationary point t'_{in} which satisfies the above equation, we can move to the second step. (In case you are wondering about the sub-script 'in', hold on a bit).

To make notations simpler, I will drop the arguments $\mathbf{p}$ and t from $S(\mathbf{p}, t, t')$, keeping only t'. The second step of the calculation is to expand $\exp(-iS(t'))$ in Taylor series around the stationary phase point $t'_{\rm in}$. Since the first derivative w.r.t t' at this point is zero, the second derivative S'' is kept and the integral becomes

$$\int dt' e^{-iS(t')} \simeq e^{-iS(t'_{\rm in})} \int dt' e^{-i\frac{S''(t'_{\rm in})}{2}(t'-t'_{\rm in})^2} \tag{36}$$

Now one uses the fact that the phase changes quickly and changes a lot. Hence, the integral converges quickly and the limits of integration with respect to $\xi = t' - t'_{\rm in}$ in the vicinity of the stationary point are extended to $\pm\infty$. The integral is then well-known, and the answer is

$$\int dt' e^{-iS(t')} \simeq e^{-iS(t'_{\rm in})} \sqrt{2\pi/iS''(t'_{\rm in})} \tag{37}$$

The pre-exponential factor is not important at this stage, since we have already dropped many pre-exponential terms when writing the amplitude as in Eq.(29). Moreover, the pre-exponential term one finds in the SFA is always quantitatively wrong. It is the exponential dependence that gives the qualitative insight we are looking for.

There could be many stationary phase points, and one needs to sum the contributions from all of them, so the full answer is

$$\int dt' e^{-iS(t')} \sim \sum_{t'_{\rm in}} e^{-iS(t'_{\rm in})} \tag{38}$$

Each stationary phase point corresponds to an ionization burst that leads to the specific final momentum u (in dimensionless units) at the detector.

The phase $S(t'_{\rm in})$ has real and imaginary parts, so let us write

$$S(\mathbf{p}, t, t'_{\rm in}) = {\rm Re}S(\mathbf{p}, t, t'_{\rm in}) - i\sigma(\mathbf{p}, t, t'_{\rm in}) \tag{39}$$

In a long laser pulse where all cycles are the same, different stationary points $t'_{\rm in}$ separated by an integer number of laser cycles yield the same imaginary part. Thus, the sum of the contributions of all stationary points to the total ionization amplitude is:

$$a(p, t) \propto e^{-\sigma(\mathbf{p})} \sum_{t'_{\rm in}} \exp\left[-i{\rm Re}S(p, t, t'_{\rm in})\right] \tag{40}$$

In a long laser pulse, where many cycles are the same, and many ionization bursts are identical, the interference of many stationary phase points separated by an integer number

of laser cycles leads to minima and maxima in the electron spectra. Since the ionization bursts repeat every laser cycle, they form a train of electronic wavepackets with the laser cycle being the 'repetition rate' in the train. In the energy domain, this train leads to peaks separated by the photon energy. The maxima are nothing but the above threshold ionization (ATI) peaks.

Now, having understood the time-domain picture of strong-field ionization for many laser cycles, let us finally turn to the contribution from a single stationary point, i.e. from a single ionization burst that produces electrons with the canonical momentum $p = v_0 u$. To find the ionization rate for each value of $p = v_0 u$, we will need to do as follows.

First, we need to solve the stationary point equation

$$\frac{v_0^2}{2}\left[u - \sin\omega t'\right]^2 = -I_p$$
$$\left[u - \sin\omega t'\right]^2 = -\frac{I_p}{2U_p} = -\gamma^2 \tag{41}$$

where γ is the Keldysh parameter, and find the stationary point $t'_{\rm in}$. It is clear that the solutions of this equation are complex-valued: no real-valued t' can give negative square, and we should look for the complex solutions $t'_{\rm in} = t'_{\rm out} + i\tau_T$.

Second, we need to calculate the value of the phase S at this point, and then find the imaginary part of this phase $\sigma(p)$ which will give us the corresponding rate,

$$\Gamma(p) \propto e^{-2\sigma(p)} \tag{42}$$

This expression is written with only exponential accuracy, and it is a standard semi-classical expression for the transmission probability – but written for the time-dependent and not for the time-independent problem.

What is the meaning of Eq.(41) and these complex times? The left-hand side of the equation is the kinetic energy of the electron. The kinetic energy has negative value when the particle enters the classically forbidden region as it tunnels from the binding potential well through the barrier created by the oscillating electric field.

Appropriately, the velocity under the barrier is imaginary,

$$u - \sin\omega t' = \pm i\gamma \tag{43}$$

Since the motion under the barrier occurs with imaginary velocity, it must proceed in imaginary time, so that the product of velocity and time (distance) has a chance of being real,

or at least having a growing real part, allowing the electron to come out of the barrier. The associated semiclassical trajectory enters the classically forbidden region at $t'_{\rm in} = t'_{\rm out} + i\tau_T$. As we move along the imaginary time axis from $t' = t'_{\rm in} = t'_{\rm out} + i\tau_T$ towards $t' = t'_{\rm out}$ on the real time axis, decreasing the imaginary part from $i\tau_T$ to zero and keeping the real part $t'_{\rm out}$ fixed, the associated electron trajectory

$$x(t') = \int_{t'_{\rm in}}^{t'} d\tau v_0[u - \sin\omega\tau] \tag{44}$$

evolves towards the exit point from the barrier. Which of the two signs on the left hand side of Eq.(43) should we take? The decision must ensure that the semiclassical transmission amplitude $\exp(-\sigma(p))$ is exponentially small and not exponentially large, dictating that τ_T is positive, and hence we write

$$u - \sin[\omega t'_{\rm out} + i\omega\tau_T] = -i\gamma \tag{45}$$

Eq.(45) can be solved for any u, but for the moment I will limit the discussion to $u = 0$. The reason is simple: these values dominate the overall ionization rate, integrated over all $u = p/v_0$. Indeed, we will see very quickly that for $u = 0$ the complex time $t'_{\rm in} = t'_{\rm out} + i\tau_T$ has $t_{\rm out} = 0$. Thus, for $u = 0$ the electron emerges from the classically forbidden region, where it has been travelling in complex (imaginary) time, at the maxima of the instantaneous electric field $\mathbf{F}\cos\omega t_{\rm out} = \mathbf{F}$. This is precisely when the strong-field ionization is peaked. Thus, the rate for $u = 0$ corresponds to the rate at the peaks of the field, and it will dominate the total, cycle-averaged, ionization rate. With exponential accuracy, this is all we need to know.

Our equation becomes

$$\sin\omega(i\tau_T) = i\gamma \tag{46}$$

or

$$\sinh(\omega\tau_T) = \gamma \tag{47}$$

Remembering that

$$\sinh(\omega\tau_T) \equiv \frac{e^{\omega\tau_T} - e^{-\omega\tau_T}}{2} \tag{48}$$

and denoting $\exp(\omega\tau_T) = z$, we find the quadratic equation

$$z - \frac{1}{z} = 2\gamma \tag{49}$$

with the solution

$$z = \gamma + \sqrt{\gamma^2 + 1}$$
$$\omega\tau_T = \ln[\gamma + \sqrt{\gamma^2 + 1}] \tag{50}$$

Let us now look at the two limits of this expression. For $\gamma \ll 1$ we have

$$\tau_T = \frac{\gamma}{\omega} \tag{51}$$

This expression gives a clear meaning to the γ parameter in terms of the so-called 'tunnelling time' τ_T: small γ mean that during tunnelling the barrier has no time to oscillate: $\omega\tau_T \ll 1$. Once we found the stationary phase point, we can now calculate the corresponding action integral:

$$S(\mathbf{p}, t, t') = I_p(t - i\tau_T) + \frac{v_0^2}{2}\int_{i\tau_T}^{0} d\tau \left[\sin\omega\tau\right]^2 + \frac{v_0^2}{2}\int_0^t d\tau \left[\sin\omega\tau\right]^2 \tag{52}$$

We note that by introducing the new integration variable $\tau = i\xi$ the integral term can be re-written as

$$\int_{i\tau_T}^{0} d\tau \left[\sin\omega\tau\right]^2 = i\int_0^{\tau_T} d\xi \sinh^2(\omega\xi) \tag{53}$$

and hence the imaginary part $\sigma = -\mathrm{Im}S$ is

$$\sigma = I_p\tau_T - \frac{\mathbf{F}^2}{2\omega^2}\int_0^{\tau_T} d\xi \sinh^2(\omega\xi) \tag{54}$$

Once we calculate the remaining integral, we find that in the limit $\gamma \ll 1$ we get exact analogue of a DC tunnelling exponent:

$$\Gamma \propto \exp\left[-\frac{4}{3}I_p\tau_T\right] = \exp\left[-\frac{2}{3}\frac{[2I_p]^{3/2}}{F}\right] \tag{55}$$

Let us now look at the opposite limit of very large $\gamma \gg 1$. The integral we need to calculate and the imaginary part of the action are given by the same general expression as before,

$$\sigma = I_p\tau_T - \frac{\mathbf{F}^2}{2\omega^2}\int_0^{\tau_T} d\xi \sinh^2(\omega\xi) \tag{56}$$

but now

$$\tau_T = \frac{1}{\omega}\ln[\gamma + \sqrt{\gamma^2 + 1}] \approx \frac{1}{\omega}\ln(2\gamma) \tag{57}$$

Calculating the integral, we find that in the limit $\gamma \gg 1$ the ionization rate is given by the expression familiar from the time-dependent perturbation theory for the multi-photon ionization process:

$$\Gamma \propto \mathbf{F}^{2I_p/\omega} \propto I^{I_p/\omega} \tag{58}$$

If we do the calculation for the general case of arbitrary γ, we can obtain a general expression for the ionization rate

$$\Gamma \propto \exp\left[-\frac{\mathbf{F}^2}{\omega^3}\left[\left(\gamma^2+\frac{1}{2}\right)\omega\tau_T - \frac{1}{4}\sinh(2\omega\tau_T)\right]\right] \tag{59}$$

where $\omega\tau_T = \ln[\gamma + \sqrt{\gamma^2+1}] = \mathrm{Arcsh}(\gamma)$.

IV. SEMICLASSICAL PICTURE OF HIGH HARMONIC GENERATION

Now let us see how the SFA theory can be applied to such highly non-perturbative phenomenon as the generation of very high harmonics of intense incident radiation.

First, let us recall where these harmonics come from. For an experimentalist, they come from a little gas jet sitting in a vacuum chamber and irradiated by a laser. For a theorist, harmonics come from the Fourier components of laser-induced polarization $P(t)$ in the gas,

$$\mathbf{P}(t) = n_0\mathbf{d}(t) = n_0\langle\Psi(t)|\mathbf{d}|\Psi(t)\rangle \tag{60}$$

where n_0 is the number density. All we need is $\Psi(t)$, the wavefunction we already know from the SFA theory:

$$\Psi(t) = a_g(t)|\Phi_g\rangle + \int d\mathbf{k}|\mathbf{k}\rangle a_{\mathbf{k}} \tag{61}$$

where $|g\rangle$ is the ground state.

In terms of the continuum amplitudes for different kinetic momenta $\mathbf{k}$, the expression for the dipole can be re-written as

$$\mathbf{d}(t) = \int d\mathbf{k}\langle\Phi_g|\mathbf{d}|\mathbf{k}\rangle a_{\mathbf{k}}(t) + c.c. \tag{62}$$

There is no need to assume SFA in this expression – $\mathbf{k}$ are simply states with asymptotic momentum k at the detector.

Now, using the SFA expressions for the amplitudes $a_k(t)$, and assuming that the ionization is not too strong and hence most of the amplitude always resides in the ground state, hence

$a_g \approx \exp[+iI_p(t-t_i)]$, we get

$$\mathbf{d}(t) = -i\int^t dt' \int d\mathbf{k}\langle\Phi_g|\mathbf{d}|\mathbf{k}\rangle e^{-i\int_{t'}^t E(t'')dt''-iI_p(t-t')}$$
$$\mathbf{F}\cos\omega t'\langle\mathbf{k}+\mathbf{A}(t')-\mathbf{A}(t)|\mathbf{d}|\Phi_g\rangle + c.c. \tag{63}$$

This rather lengthy and cumbersome expression can be simplified quite a bit if we do three things:

(1) recall that the pre-exponential factors are wrong anyway and drop them;

(2) recall that the exponential dependence on k is quadratic and hence the integral over d$\mathbf{k}$ is from a Gaussian function;

(3) recall that the integrals from Gaussians, even with very involved quadratic expressions, can be easily performed once we find the points at which the phase of the exponent is stationary with respect to all the integration variables.

Before doing that, I will add a third integral into this already involved expression. I want a specific Fourier component of the dipole, at a frequency Ω:

$$\mathbf{d}(\Omega) \sim -i\int dt \int^t dt' \int d\mathbf{k}$$
$$e^{-i\int_{t'}^t E(t'')dt''-iI_p(t-t')+i\Omega t}\langle\mathbf{k}+\mathbf{A}(t')-\mathbf{A}(t)|\mathbf{dF}\cos\omega t'|\Phi_g\rangle \tag{64}$$

where I dropped all matrix elements, and also left only the positive Ω part of the spectrum. This $d(\Omega)$ is the harmonic response of single atom.

The triple integral (actually, five-fold is you count that there are three integrals over the momentum) is very transparent and logical. The electron can be promoted to the continuum at any time t' – thus the integral over t'. It can emit a photon with frequency Ω at any moment t – thus the integral over t. In principle, it can be in any state $|\mathbf{k}\rangle$ at the moment of emission – thus the integral over $d\mathbf{k}$. The $\exp(+i\Omega t)$ corresponds to the emission of the photon Ω, while the rest of the phase is related to the electron absorbing (changing) energy while moving in the continuum. The energy is measured from the ground state (thus the I_p) part, which is logical since the harmonic emission concludes in the ground state.

Let us look at the phase in this integral: is it fast oscillating or not? If the phase accumulation is many π over one cycle of the driving field, that would be fast. This is indeed the case when

$$U_p = \mathbf{F}^2/4\omega^2 \gg \omega \tag{65}$$

where U_p is the so-called ponderomotive energy – the average energy of free electron oscillations in the laser field. Since the phase

$$\begin{aligned}\Theta\ (t,t',p) &= \int_{t'}^{t} E(\tau)d\tau + I_p(t-t') - \Omega t \\ &= \int_{t'}^{t} d\tau \frac{1}{2}[\mathbf{p}+\mathbf{A}(\tau)-\mathbf{A}(t)]^2 + I_p(t-t') - \Omega t \end{aligned} \tag{66}$$

is oscillating fast, let us look for its stationary points. The phase depends on all three integration variables. Hence, we have to find first derivatives with respect to all these three variables and find those points where all are equal to zero. Calculating the partial derivatives of Θ, we obtain the following conditions:

$$\begin{aligned}&\int_{t'}^{t} d\tau[\mathbf{k}+\mathbf{A}(\tau)-\mathbf{A}(t)] = 0 \\ &\frac{1}{2}[\mathbf{k}+\mathbf{A}(t')-\mathbf{A}(t)]^2 + I_p = 0 \\ &\frac{1}{2}\mathbf{k}^2 + I_p = \Omega \end{aligned} \tag{67}$$

These equations can be re-written in a very transparent form:

$$\begin{aligned}&\mathbf{x}(t) = \mathbf{x}(t') \\ &\frac{1}{2}\mathbf{k}^2(t') + I_p = 0 \\ &\frac{1}{2}\mathbf{k}^2(t) + I_p = \Omega \end{aligned} \tag{68}$$

The first condition says that, in order to emit the photon, the electron must come back to the same spot from where it left as it started to tunnel out. Making a very reasonable assumption that the electron departed from the core, we see that the electron must come back to the same core of the same parent ion, after oscillating in the laser field.

The second condition is already familiar to us. It cannot be satisfied classically, and describes tunnelling. The corresponding moment $t' = t'_{\rm in}$ is the moment when the electron enters the classically forbidden region, just like in the case of ionization which we have considered above. Its imaginary part τ_T determines the tunneling time, and its real part $t'_{\rm out}$ is determined by the condition

$$\mathbf{k}(t_{\rm out}) = \mathbf{k}(t) + \mathbf{A}(t_{\rm out}) - \mathbf{A}(t) \approx 0 \tag{69}$$

in the limit of large U_p, when $\gamma^2 \equiv I_p/2U_p \ll 1$.

The third condition is nothing but the energy conservation law for the emission of the harmonic photon Ω.

So now we have a simple classical physical picture emerging from the five-fold integral. This picture is of an electron tunnelling out and appearing in the continuum at some time $t_{\rm out}$. It emerges from under the barrier with nearly zero velocity. Then it moves in the laser field until it comes back to the same parent ion. Here its instantaneous kinetic energy $\mathbf{k}^2(t)/2$ is converted into the photon by recombining into the ground state. The photon energy is $\Omega = \mathbf{p}^2(t)/2 + I_p$.

The most important result that follows from the classical equations and its quantum counterpart for harmonic generation is that the maximum electron energy at the return scales with U_p as $3.17U_p$. One can easily check it by taking different possible moments of birth $t_{\rm out}$, setting classical initial conditions to zero (both position and velocity) and finding the instantaneous kinetic energy at the moment of return t, defined as the moment when the electron coordinate is equal to zero again. The upper limit on the energy at the moment of return implies that classically harmonic spectra cannot extend beyond the cut-off at $I_p + 3.17U_p$. Quantum mechanically, harmonics at higher energies are still possible - but require complex t to satisfy the energy conservation law. As the stationary phase point for t-integral moves into the complex plane, the contribution dies out exponentially.

Turning back to the quantum analysis, once we know the stationary phase points that satisfy the equations above, we can easily find the answer for the full integral. In the limit $\gamma^2 < 1$, the five-fold integral reduces to the product of three amplitudes for ionization (tunnelling), propagation, and recombination:

$$d(\Omega) \propto A_{rec}(t(\Omega))A_{prop}(t_{\rm out}(t) \to t)A_{ion}(t_{\rm in} \to t_{\rm out}(t)) \tag{70}$$

This factorized form of the expression for the induced dipole $d(\Omega)$ appears as a natural consequence of the saddle point method, which implies that the result is proportional to the integrand taken at the stationary point. The ionization amplitude is given by the imaginary part of the phase, $\exp(-\sigma)$, just like for ionization. It corresponds to time-integral for the phase $S(k,t,t')$ from $t' = t_{\rm in}$ to $t_{\rm out}$. The propagation amplitude arises from the momentum integral and depends on t, t'. It contains the next component of the factor $\exp(-iS(k,t,t'))$, which corresponds to the integral from $t' = t_{\rm out}$ to t. It also contains the factor $(t - t_{\rm in})^{-3/2}$, which describes the spreading of the continuum wavepacket and results from integration over

the momenta k. The recombination amplitude is given by the transition dipole between the continuum state with momentum $k(t)$ and the ground state.

V. BEYOND THE SFA AND SINGLE ACTIVE ELECTRON

The Strong Field Approximation (SFA) has many drawbacks and "wrongs" that fly right into the face of any rigorous quantum theory, and it continues to amaze me that one of the key original papers – that by L. V. Keldysh from 1965 [5] – was published in such a puritan and picky Soviet journal as the Journal of Experimental and Theoretical Physics. Fortunately, the SFA gets most of the basic physics right, providing excellent basis for understanding the fundamentals of strong-field processes. Moreover, it can be modified and turned into a quantitatively accurate theory, see [2, 3, 13]

Importantly, the SFA allows us to see simple classical pictures that underlie the seemingly very complex strong-field dynamics. This classical picture is due to P. Corkum and F. Brunel [16, 17], and it was pre-dated by the quantum insight developed by M. Kuchiev [14] and by the key numerical results of K. Schafer, J. Krause and K. Kulander [15]. By now, the new students in the field begin with the simple classical picture, taking it for granted. It is, however, important to know how this picture emerged from the combination of quantum and classical analysis and the experimental results. I strongly recommend to begin with the papers of F. Brunel [17] and M. Kuchiev [14] and K. Schafer et al [15], then turning to the seminal paper by P. Corkum.

In addition to the SFA [5–9], more accurate and versatile but technically more complicated approach has been developed in the 60-th by V. S. Popov, A. M. Perelomov and M. V. Terent'ev – the so-called PPT theory [10, 11]. Finally, I must mention the effective range theory (ERT) of M. Frolov, N. Manakov, and A. Starace [12], which grew out of the earlier work of L. Rapoport, N. Manakov, and B. Zon. It is essentially exact for the short-range binding potentials (such as those of negative ions). It also shows that the PPT theory, when applied to the short-range potentials, is almost exact – the only thing it is missing is the Stark shift of the ground state. Unlike the SFA, the ERT and the PPT are gauge-invariant.

The insight gained from these theories is instrumental in turning the highly approximate SFA models into quantitatively accurate. The corresponding ideas were suggested in [2] and very successively used by T. Brabec, F. Krausz, and V. Yakovlev already in late 90-th

and early 2000 to describe experiments on high harmonic generation and the production of attosecond pulses in atomic gases (see e.g. [13]). The version known as the quantitative rescattering theory, developed by C. D. Lin, T. Morishita and co-workers (probably without knowing about the work of T. Brabec and co-workers a decade earlier) has been very successful in describing many aspects of strong-field experiments in molecules [3]. It shows how well one can do when armed with clear understanding of the basic underlying physical mechanisms. For example, in case of high harmonic generation, the approximate SFA expression can be turned into quantitatively accurate by substituting correct ionization amplitudes and proper recombination dipoles, which take into account the structure of the atom or the molecule.

VI. HHG DIPOLE FOR MANY ELECTRONS, INCLUDING LASER-INDUCED DYNAMICS IN THE IONIC CORE BETWEEN IONIZATION AND RECOMBINATION

In multielectron systems there are multiple ways of energy sharing between the liberated electron and ion left behind. The ion can be left in its ground or excited electronic states. On the formal language these options correspond to different ionization channels. Multiple ionization channels give raise to multiple HHG channels: the returning electron recombines with the ion in the ground or excited state. Multiple HHG channels present different pathways connecting the same initial and final state - ground state of the neutral system - via different electronic states of the ion (see Fig.**??**). Thus, high harmonic emission in multielectron systems results from multichannel interference [**?**], i.e. the interference of harmonic lights emitted in each channel. These interference naturally records multielectron dynamics excited upon ionization and probed by recombination [**?**]. How important are these multiple channels? How hard it is to excite the ion upon strong field ionization?

In strong-field ionization is exponentially sensitive to the ionization potential I_p, suggesting that after ionization the molecular ion is left in its ground electronic state (electron removal from highest occupied molecular orbital (HOMO) in the Hartree-Fock picture). However, multiple ionization channels can be very important in molecules due to geometry of molecular orbitals and proximity of excited electronic states in the ion to the ground state.

The formalism described above in sections 1.1-1.7 is essentially a single-channel picture of HHG. Therefore, it can easily be extended to multiple channels.

We first introduce the Hamiltonian of an N-electron neutral molecule interacting with a laser field:

$$H^N = T_e^N + V_C^N + V_{ee}^N + V_l^N, \tag{71}$$

$$V_C^N = \sum_{m,i=1}^{i=N} 1/|\mathbf{R}_m - \mathbf{r}_i|, \tag{72}$$

$$V_{ee}^N = \sum_{i\neq j}^{N} 1/|\mathbf{r}_i - \mathbf{r}_j|, \tag{73}$$

$$V_l^N = \sum_i E(t)\cdot \mathbf{r}_i. \tag{74}$$

Here the nuclei are frozen at their equilibrium positions $\mathbf{R}_m$, index m enumerates the nuclei, superscript N indicates the number of electrons involved, T_e^N is electron kinetic energy operator, V_C^N describes the Coulomb potential of the nuclei, V_{ee}^N describes the electron-electron interaction, and V_l^N describes the interaction with the laser field. We will also use the Hamiltonian of the ion in the laser field H^{N-1} and the Hamiltonian of an electron H^e interacting with the laser field, the nuclei, and the $(N-1)$ electrons of the ion, $H^e = H^N - H^{N-1}$. The exact solution of the Schroedinger equation for N-electron wave-function of the molecule, which is initially in the ground electronic state $\Psi_g^N(\mathbf{r})$:

$$i\frac{\partial}{\partial t}\Psi^N(\mathbf{r},t) = H^N\Psi^N(\mathbf{r},t), \tag{75}$$

$$\Psi^N(\mathbf{r},t=0) = \Psi_g^N(\mathbf{r}) \tag{76}$$

can be written as [**?**]

$$\Psi^N(\mathbf{r},t) = -i\int_0^t dt'\, U^N(t,t')V_l^N(t')\, U_0^N(t',0)\Psi_g^N(\mathbf{r}) + U_0^N(t,0)\Psi_g^N(\mathbf{r}). \tag{77}$$

Here the N-electron propagators U_0^N and U^N are determined by $i\partial U_0^N/\partial t = H_0^N U_0^N$, where H_0^N is the field-free Hamiltonian of the molecule $H_0^N = H^N - V_l^N$ and $i\partial U^N/\partial t = H^N U^N$, is a full propagator. The harmonic dipole is

$$D(t) = -i\langle U_0^N(t,0)\Psi_g(\mathbf{r})|\mathbf{r}|\int_0^t dt'\, U^N(t,t')V_l^N(t')\, U_0^N(t',0)\Psi_g^N(\mathbf{r})\rangle + cc. \tag{78}$$

Just like in one-electron case, the propagation without the laser field can be easily solved if the energy $|E_g|$ and the wave-function of the initial state of a neutral molecule or an atom

FIG. 1: Left panel: Subcycle dynamics in the N_2^+ ion aligned at $\theta = 50^o$ to the laser field polarization: populations of the field-free ionic states X (blue), A (red), and B (green) in I=0.8 10^{14} W/cm^2, 800 nm laser field. Right panel: Electronic states of N_2^+ ion.

FIG. 2: Left panel: Diagonal channel in HHG, associated with ionization from and recombination to the same orbital. Right panel: Cross-channel in HHG associated with ionization from and recombination to different orbitals. This channel is due to real excitations induced by the laser field between ionization and recombination.

are known:

$$U_0^N(t,0)\Psi_g(\mathbf{r}) = e^{-iE_g t'}\Psi_g(\mathbf{r}), \tag{79}$$

The full propagation $U^N(t,t')$ is just as hard as the solution of the original equation (**??**) with the multielectron Hamiltonian (**??**). To simplify the analysis we will make the following two approximations. First, we will neglect the correlations between the electrons in the ion and in the liberated electron after ionization. In this case the full propagator factorizes into two independent parts describing the evolution of the continuum electron and the evolution of the ion in the laser field between ionization and recombination: $U^N(t,t') \simeq U^{N-1}(t,t')\,U^e(t,t')$. Second, we will keep the analysis at the level of the SFA for the continuum electron, just like we did in the single electron case considered above: $U^e(t,t') \simeq U_V^e(t,t')$. These two approximations can be further improved by including electron-electron correlations perturbatively [**?**] and using the eikonal-Volkov states [**?**] for the continuum electron. These states include the laser field fully, the interaction of continuum electron with the core in the eikonal approximation and the coupling between them [**?**].

Just like in one-electron formalism considered above, we will introduce identity resolved on the momentum states of continuum electron and electronic states of the ion [**?**]

$$I = \int d\mathbf{k} \sum_n \mathbb{A}|\mathbf{n}^{(N-1)} \otimes \mathbf{p}_t^n\rangle\langle\, \mathbf{n}^{(N-1)} \otimes \mathbf{p}_t^n|\mathbb{A}, \tag{80}$$

The harmonic dipole reduces to

$$\begin{aligned} D(t) = -i \int_0^t dt' \int d\mathbf{p} \langle e^{iE_g(t-t')}\Psi_g(\mathbf{r})|\mathbf{r}|\, U^{N-1}(t,t')|\mathbf{n}^{(N-1)}\rangle\, U_V^e(t,t')|\mathbf{p}_t\rangle \\ \times \langle p_t^n \mathbf{n}^{(N-1)}|V_l^N(t')|\Psi_g^N(\mathbf{r})\rangle + cc. \end{aligned} \tag{81}$$

The laser-induced dynamics of bound states of the ion is described by the propagator $U^{N-1}(t,t')|\mathfrak{n}^{(N-1)}\rangle$. It can be easily solved if the dipole couplings d_{nm} between these states and the eigen energies E_m of all essential states are known. The time-dependent amplitude $a_{mn}(t)$ of excitation from state n at moment t' to state m at the moment t given by $a_{mn}(t) = \langle\mathfrak{m}^{(N-1)}|\, U^{N-1}(t,t')|\mathfrak{n}^{(N-1)}\rangle$ is a solution of the equation:

$$\frac{d\mathbb{A}_n}{dt} = [\mathbb{H} + \mathbb{V}(t)]\,\mathbb{A}_n, \tag{82}$$

where $\mathbb{H} = \begin{bmatrix} E_1 & 0 & 0 \\ 0 & E_2 & 0 \\ 0 & 0 & E_3 \end{bmatrix}$ is the Hamiltonian of the ion, including several (three in the present case) ionic states, $\mathbb{V}(t) = \begin{bmatrix} 0 & V_{12} & V_{13} \\ V_{21} & 0 & V_{23} \\ V_{31} & V_{32} & 0 \end{bmatrix}$ is the matrix describing coupling between the different essential states, $V_{nm} = d_{nm}F(t)$ and $\mathbb{A}_n = \begin{bmatrix} a_{1n}(t) \\ a_{2n}(t) \\ a_{3n}(t) \end{bmatrix}$ is the vector representing the amplitudes of laser induced excitations of all the essential states starting from state n.

Introducing the channel specific Dyson orbital $\Psi_n^D(\mathbf{r}) \equiv \langle\mathfrak{n}^{(N-1)}|\Psi_g^N(\mathbf{r})\rangle$ we can re-write the multielectron dipole D_{mn} corresponding to leaving the ion in the state n after ionization and recombination into the state m in the form very similar to one -electron dipole:

$$D_{mn}(t) = -i\int_0^t dt' \int d_m\mathbf{p}d^*(\mathbf{p}+A(t))a_{mn}(t)e^{-iS(\mathbf{p},t,t')}\mathcal{E}(t')d_n(\mathbf{p}+A(t')), \tag{83}$$

$$d_n(\mathbf{p}+A(t)) \equiv \langle e^{i(\mathbf{p}+\mathcal{A}(t))\mathbf{r}}|\mathbf{r}|\Psi_n^D(\mathbf{r})\rangle, \tag{84}$$

$$S(\mathbf{p},t,t') \equiv \frac{1}{2}\int_{t'}^t (\mathbf{p}+\mathcal{A}(\tau))^2 d\tau + I_p(t-t'). \tag{85}$$

Total harmonic signal results from coherent superposition of dipoles associated with each ionization-recombination channel:

$$D(t) = \sum_{mn} D_n(t) \tag{86}$$

note that substantial subcycle transitions (Fig.**??**) corresponding to laser-induced dynamics of the ion between ionization and recombination have crucial impact on harmonic radiation. They lead to the appearance of the cross-channels in HHG (off-diagonal channels D_{mn} in

equation (**??**)) since the state of the ion changes between the ionization and recombination. These channels are substantial in high harmonic generation from N_2 molecules [**?**].

In recent HHG literature the language of molecular orbitals arising in the Hartree-Fock picture is often used. Loosely speaking removing electron from the highest occupied molecular orbital (HOMO) creates the ion in the ground state and removing electron from lower lying orbital (e.g. HOMO_1, HOMO-2) creates the ion in the excited state. Electron removal from an orbital, creates a hole in this orbital. Laser induced dynamics in the ion can move the hole between the orbitals between ionization and recombination (Fig.**??**).

VII. THE MULTICHANNEL MODEL OF HHG

The application of the saddle point method in each channel leads to the following half-cycle dipole:

$$D^j_{mn}(t) = a^m_{rec}(\mathbf{p}_s, t) a^{mn}_{prop}(t, t_i) a^n_{ion}(\mathbf{p}_s, t_i), \tag{87}$$

$$a^n_{ion}(\mathbf{p}_s, t_i) = \frac{(2\pi)^{1/2}}{S''_{t_i,t_i}} e^{-iS(\mathbf{p}_s, t'_i, t_i)} \mathcal{E}(t_i) d_n(\mathbf{p}_s + A(t_i)), \tag{88}$$

$$a^{mn}_{prop}(t, t_i) = \frac{(2\pi)^{3/2}}{(t - t_i)^{3/2}} e^{-iS(\mathbf{p}_s, t, t'_i)} a_{mn}(t), \tag{89}$$

$$a^m_{rec} = \frac{(2\pi)^{1/2}}{S''_{t_r,t_r}} d^*_m(\mathbf{p}_s + A(t)). \tag{90}$$

Here we have considered the dipole on the real axis. Note that the propagation amplitude is modified to include the laser -induced dynamics in the ion $a_{mn}(t)$. Now once we have factorized the dipole we can use improved amplitudes for each step.

Improving ionization. the improved ionization amplitude can be taken from semi-analytical and/or numerical approaches. The improved ionization amplitude consists of two parts:

$$\widetilde{a}^n_{ion}(\mathbf{p}_s, t_i) = \mathbb{R}_{\mathfrak{lm}}(I_p, F) e^{-iS(\mathbf{p}_s, t'_i, t_i)}. \tag{91}$$

the exponent describes the sub-cycle dynamics of strong-field ionization, the pre-factor $\mathbb{R}_{\mathfrak{lm}}(I_p, F)$ describes the influence of the core potential and shape of the initial state on the ionization rate.

Improving propagation. The most important modification of the propagation amplitude is due to the presence of nodal planes in bound states of molecular systems. The nodal

planes leave the imprints on electron wave-packet after ionization and thus the shape of the returning wave-packet will also change affecting recombination. Below we describe the simple way of including the nodal planes.

Improving recombination. Recombination step can be significantly improved beyond the SFA, if one uses the recombination dipoles $d_m^*(\mathbf{p}_s + A(t))$ calculated using ab-initio approaches, such as Schwinger variational method [**?**] or R-matrix [**?**]. Refs[**?**] use the eikonal approximation for continuum states to obtain the improved dipoles.

How to introduce depletion?

The full dipole for a specific channel $d_{mn}(t)$ obtains as the sum over different half-cycles and the harmonic spectrum results from the FFT of the full dipole $d_{mn}(N\omega)$:

$$d_{mn}(t) = \sum_j D_{mn}^j(t), \tag{92}$$

$$d_{mn}(N\omega) = \int dt e^{-(N\omega)t} d_{mn}(t) e^{iN\omega t}. \tag{93}$$

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[] Here for simplicity we will use field-free states of the ion, however since the ion is polarized by the laser field it is more convenient to use the basis of quasistatic states.

Theory of Intense Laser Matter Interaction. Lecture I

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This Lecture gives a general outline of the problems we will look at.

INTRODUCTION

The purpose of these lectures is to provide a brief introduction into the methods and ideas in the theory of intense laser-atom and laser molecule interaction. The land of intense laser-matter interaction is vast, and I will explore but a small corner of it.

Let us start by deciding upon the observables of interest. For this particular course, these include:

1. Light induced polarization and the properties of light emitted by molecules interacting with the strong laser field;

2. Products of laser-induced ionization: electrons, ions, molecular fragments, final states of the ions and the survived neutrals.

When it comes to electrons, we could be interested in their energy and angle-resolved spectra: how many electrons with a given energy fly in a given direction?

When it comes to molecular ions, we are interested in their quantity, energies, and – if ionization breaks the molecule into many different pieces (fragments) – the composition of those fragments. For example, let us take a molecule such as HCl and ionize it, making HCl^+. We may be interested in finding out how many intact HCl^+ we have made, and how many of the ions broke into H^+ and Cl, or into H and Cl^+, in which direction these fragments went, and with what energy.

In this short course I will only cover few bits and pieces related to these questions, and the discussion will be weighted rather heavily towards the theory of the dynamics associated with laser-induced polarization, which is responsible for the high harmonic generation.

POLARIZATION

Why are we interested in laser induced polarization? Because it tells us about how the charges move in the system. The polarization induced in the medium is $\mathbf{P}(t) = \mathcal{N}\mathbf{d}(t)$, where $\mathcal{N}$ is the number density and the dipole moment $\mathbf{d}(t) = q\mathbf{r}(t)$ describes the motion of charges. Here $\mathbf{r}(t)$ is the trajectory of the charge denoted by q. The dipole moment is the laser-induced microscopic polarization of an individual atom or molecule, and it tells us about the response of the charges in this atom or molecule to the external field.

Fourier transform $\mathcal{P}(\Omega)$ describes nonlinear response of the medium at various frequencies. Taken, say, at $\Omega = 3\omega_L$, it ultimately leads to third harmonic generation in the medium. Measuring these harmonics, as well as all other emission which comes out of the medium – the amplitudes, phases, and polarizations – can help us to fully characterize $\mathcal{P}(\Omega)$ and, through its Fourier transform, $P(t) = \mathcal{N}\mathbf{d}(t)$. This will tell us a lot about how the charges move in molecules and atoms interacting with the laser fields.

Emission of radiation means that the system is giving back some energy it has absorbed from the laser field. What is the origin of this emission?

Instantaneous energy (kinetic plus potential) can go down when the binding potential of the system and the laser field conspire to reduce the excitation of the system. In this case we are dealing with stimulated emission. Just as stimulated absorption, it is a natural part of system's dynamics. This stimulated emission can only occur at the frequency of the simulating field – the driving laser field.

But what if the driven system emits light at frequencies which are very different from that of the driving laser field? Textbooks on classical electrodynamics teach us that accelerated charge, whose motion is described by $d(t)$, emits radiation. Only for a harmonic oscillator $d(t)$ will contain the frequencies identical to that of the driving field. Typically, the Fourier transform of $d(t)$ will include frequencies which are very different from that of the driving field.

This emission cannot be stimulated in the standard quantum-mechanical sense, since there are no external laser fields acting at the frequencies other than the fundamental frequency ω_L. Thus, we have to conclude that this emission is spontaneous, even though it is induced by the laser field. But this spontaneous emission is very different from the conventional spontaneous emission which is not coherent from atom to atom, or from a molecule to another molecule. The emission we are dealing with, being induced and controlled by the common external driving force, has a well-defined phase for various atoms, and is coherent from one atom to another.

There are many examples of such coherent spontaneous emission, from Dicke super-radiance to photon echoes and other coherent emissions of light, which can be generated by the coherently excited medium long after the inducing laser pulses have left the interaction volume.

As long as the laser field is described classically, dealing with spontaneous emission requires an eclectic approach

– no matter whether an atom or a molecule is described classically or quantum mechanically.

One relies on the classical description of emission by an oscillating dipole $\mathbf{d}(t)$:

$$W(t) = \frac{2}{3c^3}|\ddot{\mathbf{d}}(t)|^2 \quad (1)$$

Here W is the total energy emitted by a dipole per unit time, in all angles – that is, the power of emission. If we want the emission spectrum, all we need is to Fourier transform the acceleration:

$$\mathcal{W}(\Omega) = \frac{2}{3c^3}\left|\int dt e^{i\Omega t}\ \ddot{\mathbf{d}}(t)\right|^2 = \frac{2\Omega^4}{3c^3}|\mathcal{D}(\Omega)|^2 \quad (2)$$

Now we only need to find the induced dipole. For a quantum system with a wavefunction $\Psi(t)$

$$\mathbf{d}(t) = \langle\Psi(t)|\hat{\mathbf{d}}|\Psi(t)\rangle \quad (3)$$

This is the most general way to find the polarization. It is not based on any kind of perturbation theory, it is valid for arbitrarily strong or complex laser field, but it means that we need to solve the time-dependent Schroedinger equation to find the wavefunction.

What are the questions that we may want to ask about the polarization? One most obvious question is: how does the spectrum look like? How far does it extend? We will discuss these questions in the following lectures. In Lecture II, we will study high harmonic generation in bound systems and we will see that it can tell us a lot about the underlying charge dynamics. After looking at strong field ionization in Lecture III, we will come back to high harmonic generation in Lecture IV, this time dealing with continuum dynamics and ionization.

IONIZATION: PHYSICAL PICTURES

If we know the wavefunction, we know everything – in principle. Extracting this knowledge is generally a lot of pain, however. But with ionization it is not that difficult if what you want is the ionization probability. All you need is to collect the wavefunction norm that is far enough.

What are the questions that we may want to ask about ionization? Here are some:

1. How does ionization depend on the laser intensity?
2. How does ionization depend on the laser frequency?
3. How does ionization depend on the structure of the ionizing orbital?

There are two distinctly different regimes, and the physical pictures associated with them are also very different.

Modest Fields

One is that of weak to modest fields, and relatively few photons needed for ionization. This is where we talk about the importance of resonances, and think in terms of making a few $\hbar\omega_L$ jumps via a set of intermediate virtual states onto the continuum.

The theoretical method that works very well here is the time-dependent perturbation theory, and we might have a brief look at it down the road.

The perturbative picture breaks down when the fields approach intensities in the range of 10^{13}W/cm^2 or so. Of course, there is no hard line here and nor one can say that 10^{13}W/cm^2 is universal. The boundary between perturbative and non-perturbative regimes depends on the laser frequency, ionization potential, initial state, energy spectrum, etc. These lectures will skip over the high/moderate frequency and modest intensity regime of the usual time dependent perturbation theory.

Strong Low-Frequency Fields

Dealing with ionization, I will focus on the case when the photon frequency is small compared to the ionization potential Ip, $\omega_L \ll Ip$. This is the regime where we need to absorb a lot of photons to ionize the system, and therefore the required field intensity will be high.

This is the standard situation we deal with when we use near IR lasers and noble gasses, and this is what you will face with Ti:Sapph or its sub-harmonics and most of the targets.

For the ionization potentials in the range of 10eV or so, or higher, and near-IR or mid-IR or even for the visible red light, we won't see much signal until the laser intensity approaches and exceeds 10^{13}W/cm^2 or so. In this strong-field regime the picture of ionization is rather complex.

Let the field be polarized along the x axis. The interaction potential is $V(x) - d_x F(t) = V(x) + x\mathcal{E}\cos\omega_L t$, where I have set the electron charge to $q = -1$ and used that $d_x = qx$, and my field is $\mathcal{E}\cos\omega_L t$. This potential oscillates up and down as shown in Figure 1.

There are two physical pictures associated with ionization. One is the smooth extension of the perturbative picture into the strong-field domain, see Fig.2

The second appeals to the idea of tunnelling. If there is a barrier created by the laser field and the binding potential, then we can tunnel through it, see Fig.3

How do these two pictures co-exist? do they co-exist peacefully, or is there a cold war between them? Which of these two pictures is correct? How does ionization really proceed? These issues will be the subject of Lecture III.

But before we move on to the math, let us take a closer look at the pictures, see Fig.1. Two things must be mentioned.

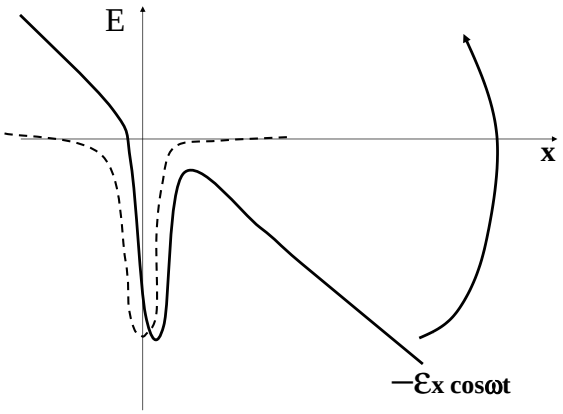

FIG. 1: Potential created by an instantaneous laser field and a potential well.

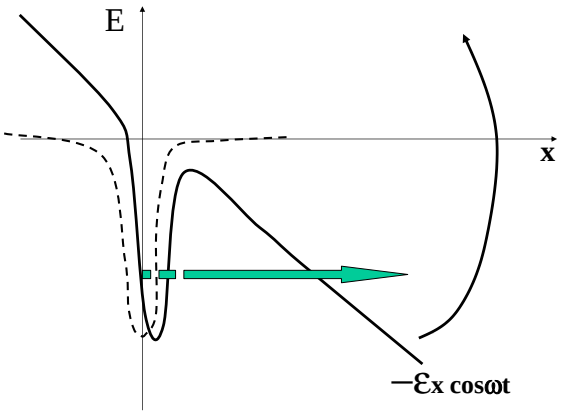

FIG. 3: Horizontal ionization channel: tunnel ionization from the standard perspective.

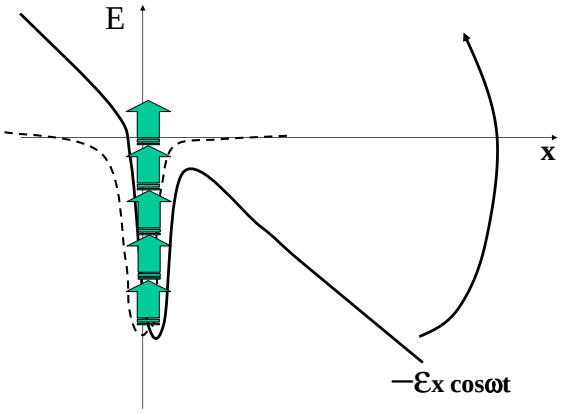

FIG. 2: Vertical ionization channel: multi-photon absorption from standard perspective.

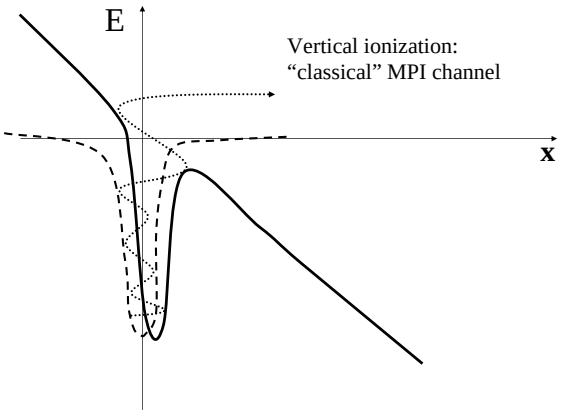

FIG. 4: Vertical ionization channel without tunnelling – multi-photon ionization in the classical limit

(1) First, the potential well itself, in the classically allowed region, is being distorted and modified every cycle. Only if it has a reasonable size, though. There is absolutely no distortion of the infinitely narrow and infinitely deep hole which is called the delta-function potential and is a favorite toy of strong-field theorists. Thus, the physical mechanisms of ionization in a short-range (delta-function) potential, which is realistic for the negative ions, and in the long-range potential (everything else) will likely be different.

(2) Second, the barrier is oscillating up and down, opening a possibility for tunneling out every half-cycle. This is true for any values of the frequency, intensity, etc. But whether the chance for tunnelling can be used, and how – this remains to be seen and is the main subject of the discussion in Lecture III. Note that chance of tunnelling through a barrier (oscillating fast or not so fast) appears for all potentials, long-and short-range.

With this overview in mind, we now turn to the bound state dynamics in laser driven systems.

TDSE AND EQUATION FOR AMPLITUDES

Let us now be specific and look at a system with the Hamiltonian $\hat{H}_0$, placed in an external time-dependent field $\hat{V}(t)$? We will take the interaction potential in the length form: $\hat{V}(t) = -\mathbf{d}\mathcal{E}(t)$. The total Hamiltonian is now $\hat{H} = \hat{H}_0 + \hat{V}(t)$. We will refer to the unperturbed Hamiltonian $\hat{H}_0$ as 'field-free', and to the time-dependent perturbation $\hat{V}(t)$ as 'field'.

The corresponding TDSE is

$$i\frac{\partial}{\partial t}\Psi = [\hat{H}_0 + \hat{V}(t)]\Psi \tag{4}$$

The eigenstates ψ_n of the field-free Hamiltonian H_0 form a complete basis set. Therefore, we can always write the wavefunction $\Psi(t)$ as

$$\Psi(t) = \sum_n a_n(t)\psi_n \tag{5}$$

Our amplitudes a_n will depend on time, reflecting the dynamics of population transfer between different states. This time-dependence is precisely what we are interested in.

We now insert Eq.(5) into the TDSE and recall that $\hat{H}_0\psi_n = E_n\psi_n$. The result is

$$i\sum_n \dot{a}_n(t)\psi_n = \sum_n a_n(t)E_n\psi_n + \sum_n a_n(t)\hat{V}(t)\psi_n \tag{6}$$

Next, we use the standard trick to get rid of the sum on the left-hand side. We recall that all eigenstates of $\hat{H}_0$ are orthogonal to each other,

$$\langle\psi_k|\psi_n\rangle = \int_{-\infty}^{\infty} d\mathbf{r}\psi_k^*(\mathbf{r})\psi_n(\mathbf{r}) = \delta_{kn} \tag{7}$$

(where $\delta_{kn} = 1$ for $k = n$ and $\delta_{kn} = 0$ for $k \neq n$), multiply the left and right-hand sides of the equation Eq.(6) with ψ_k^*, and integrate over $d\mathbf{r}$. The result is

$$i\dot{a}_k(t) = E_k a_k(t) + \sum_n a_n(t)V_{kn}(t) \tag{8}$$

where the transition matrix elements of the perturbation $\hat{V}(t)$ are

$$V_{kn}(t) = \langle\psi_k|\hat{V}(t)|\psi_n\rangle = \int_{-\infty}^{\infty} d\mathbf{r}\ \psi_k^*(\mathbf{r})V(\mathbf{r},t)\psi_n(\mathbf{r}) \tag{9}$$

As for the initial conditions, it is frequently useful to assume that only one field-free state is initially populated, while all others are empty, taking the opportunity to follow and analyze the dynamics of each initial state individually. We can always make such an assumption, thanks to the linearity of the Schroedinger equation. For several initially populated states $n_1, n_2, ...$ with the initial amplitudes $a_{n_1}^{(0)}, a_{n_2}^{(0)}, ...$ the linearity of TDSE allows one to solve several initial value problems for each of the states $n_1, n_2, ...$ separately, assuming all other states empty. Then the solutions, which we will denote $a_k^{n_1}(t), a_k^{n_2}(t), ...$, should be added coherently, weighted with the initial amplitudes:

$$a_k(t) = \sum_{n_i} a_k^{n_i}(t)\ a_{n_i}^{(0)} \tag{10}$$

The differential equations Eq.(8) deserve a few comments. They are just as exact as the original TDSE. No assumption has been made about the strength of the perturbation. Importantly, all spatial dependence has been integrated out – together with the second-order spatial derivatives; only the first-order time derivatives are left in these equations. Therefore, they are potentially much simpler that the original TDSE, both for numerical and analytical analysis.

Note, that the equations remain the same no matter how many electrons are involved. All the complexity of the underlying multi-electron dynamics is hidden in the transition matrix elements $V_{kn}(t)$ between the *field-free* eigenstates ψ_n. Thus, from the theoretical perspective, the onus is on calculating the field-free excited states and the transition matrix elements between them.

There has to be a price to pay for such possible simplifications. Indeed, a second glance at Eqs.(8) discovers the problem: the equations are all coupled to each other. The approach based on Eqs.(8) is efficient when the number of states involved in the dynamics, and hence the number of equations, is limited – solving infinitely many coupled equations is rarely an option. The problem becomes particularly acute when dealing with ionization, ubiquitous in strong laser fields. As soon as unbound (continuum) motion gets involved, the number of participating eigenstates becomes, strictly speaking, infinite. There are routes to deal with this problem, associated with replacing the true continuum with a dense but still discrete manifold of eigenstates, but the warning yellow flags have been raised. The stronger the laser field, the more important the ionization, the larger the range of energies involved in the dynamics, and the more costly and computationally intense the method becomes. We will only use this method in the next Lecture II for bound states, and will switch to a different approach when dealing with ionization.

Theory of Intense Laser Matter Interaction. Lecture II

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This Lecture focuses on some general properties of bound state dynamics in strongly driven systems.

INTRODUCTION

The purpose of these lectures is to provide a brief introduction into the general theoretical methods and to illustrate some interesting and rather general effects in bound electron dynamics during intense laser-atom and laser molecule interaction.

TIME-PERIODIC HAMILTONIANS AND THE FLOQUET THEOREM

Let us consider a quantum system interacting with a periodic laser field, so that its Hamiltonian is

$$H(t) = H_0 - \hat{\mathbf{d}}\mathbf{F}\cos\omega t \tag{1}$$

This Hamiltonian is periodic with the period $T = 2\pi/\omega$. Also periodic is the linear operator $\hat{U}(t) = H(t) - i\partial/\partial t$ offered to us by the time-dependent Schroedinger equation

$$\hat{U}\Psi(t) = 0 \quad \hat{U}(t+T) = \hat{U}(t) \tag{2}$$

What can be said about the solutions of such equation? Well, the initial impetus might be to say that the solutions are periodic. This is, of course, wrong. One does not need to go far for a counter-example - how about ionization of an atom in a periodic (cw) laser field? There you have it – an irreversible and completely aperiodic process induced by a periodic Hamiltonian.

There is analogy with time-independent Hamiltonians that are periodic in space. In fact, for a mathematician looking at the linear differential equation $\hat{U}\Psi = 0$ there is not much difference between time and space. The operator $i\partial/\partial t$ is not much different from the momentum operator - same imaginary unity, same first derivative.

In space-periodic systems we have the so-called Bloch states, which are nearly (that is, up to a phase) periodic solutions of the time-independent Schroedinger equation, i.e. its solutions that satisfy periodic boundary conditions. But of course not all solutions will be of the Bloch type. The Bloch states form a complete basis set, and then any solution can be expanded into it. Any solution is a wavepacket of the Bloch states, which are defined not only by the differential equation, but also by the periodic boundary conditions.

So, three important words were said: (1) wavepacket, (2) periodic boundary conditions, (3) basis set. The same applies to time-periodic Hamiltonians, the Floquet states, and the general solutions of initial value problems. Namely, (1) the Floquet states are defined by the equation 2 and the periodic boundary conditions in time (up to a phase factor; the conditions will show up in a minute); (2) the Floquet states form a complete basis set; (3) general solution is a wavepacket of the Floquet states.

Just as spatial periodicity is enforced onto the Bloch states by setting periodic boundary conditions in space, temporal periodicity is enforced upon the Floquet states by requiring that they obey periodic boundary conditions in time.

Now it is time to specify the boundary conditions. Suppose that at some moment $t_0 + T$ the wavefunction happened to match what it looked like at an instant t_0, up to a factor λ:

$$\Psi(t_0 + T) = \lambda\Psi(t_0) \tag{3}$$

If this happened for at least one single moment t_0 and its partner $t_0 + T$, the wavefunction is doomed to be periodic forever. Now (and only now) the periodicity of the Hamiltonian will force the wavefunction to reproduce between $t_0 + T$ and $t_0 + 2T$ the exact same evolution it experienced between t_0 and $t_0 + T$. Indeed, the initial condition at $t_0 + T$ was the same as at t_0 (up to a factor λ), and the differential equation is the same.

What about this factor λ? If $\Psi(t+T) = \lambda\Psi(t)$, then $\Psi(t+nT) = \lambda\Psi(t+nT-T) = ... = \lambda^n\Psi(t)$. Clearly, an additional time-dependent function is showing up here. Since the norm must conserve, λ can only be a phase factor - the phase factor which is multiplying every laser cycle. We already see the answer: $\lambda = \exp(-i\epsilon T)$, a snapshot of a time-dependent phase factor $\exp(-i\epsilon t)$. Now the boundary condition for the Floquet state can be set as:

$$\Psi(t) = e^{-i\epsilon t}\Phi(t), \quad \Phi(t+T) = \Phi(t) \tag{4}$$

The value ϵ is called the quasi-energy, and the periodic function Φ is the corresponding quasi-energy state – or the Floquet state. These states form a complete basis set, just like the field-free states do. All possible values of ϵ which are allowed by Eqs.(2,4) form the spectrum (the Floquet spectrum) of the *dressed quantum system* (dressed by the laser field).

Since Φ is periodic in time, we can expand it into the

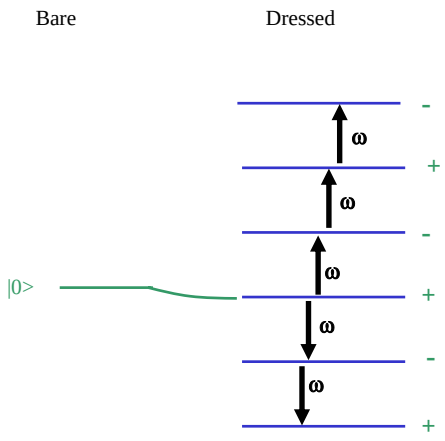

FIG. 1: The Floquet Ladder

Fourier series:

$$\Phi(x,t) = \sum_k a_k \phi_k(x) e^{-ik\omega t}$$
$$\Psi(x,t) = \sum_k a_k \phi_k(x) e^{-i(\epsilon + k\omega)t} \quad (5)$$

Here the coordinate parts $\phi_k(x)$ are normalized to unity, and the coefficients a_k carry the weights of each Fourier component.

Each $\phi_k(x)$ corresponds to the absorption of k photons, as is clear from the second of the two equations above: the energy ϵ has been raised by $k\omega$. Each Floquet state forms an infinite ladder of states "quantum system $+k\omega$", corresponding to having k photons in the "dress", see Fig.1.

Of course, the ladder is only formally infinite - the weights $|a_k|^2$ of each step in the ladder will eventually go down, making higher and higher steps of the ladder less and less sturdy.

If the laser field is turned on adiabatically, each field-free state $\Psi_n^{(0)} = \exp(-iE_n t)\Phi_n^{(0)}$ will go into a corresponding Floquet state $\Psi_n = \exp(-i\epsilon_n t)\Phi_n(x,t)$, and each energy E_n will go into a corresponding quasi-energy ϵ_n.

$$\Psi_n^{(0)} = e^{-iE_n t}\Phi_n^{(0)} \to e^{-i\epsilon_n t}\Phi_n(x,t)$$
$$\Phi_n(x,t) = \sum_k a_{n,k}\phi_{n,k}(x)e^{-ik\omega t} \quad (6)$$

If the initial field-free state had a definite parity, the same will be true for the harmonics of the Floquet state. The one which corresponds to the *net* absorption of zero photons, $\phi_{n,k=0}(x)$, will retain the same parity as its field-free counterpart. Each photon absorption or emission will change the parity to the opposite. So, for example, if the field-free state was even, then its $k = 0$ Floquet harmonic is even, $k = +1$ is odd, and so on, see Fig.1.

Are there any other general statements that one can make about the Floquet spectrum?

(1) Any general solution of the initial value problem - time-dependent Schroedinger equation for some initial condition – is a superposition of the Floquet states, with their respective phase factors. If an irreversible dynamical process such as ionization (no matter how slow or weak) is happening in the system dressed by the laser field, then rigorously speaking the spectrum of various ϵ's in this superposition *must be continuous*. Indeed, discrete spectrum means that the process shows some repeated events, like the Poincare returns, incompatible with irreversibility.

(2) Although rigorously the quasi-energy spectrum is continuous, in practice it can be treated as discrete. As a price, one has to deal with complex quasi-energies which describe decay: $\epsilon = \varepsilon - i\Gamma$. The norm of $\Psi = \exp(-i\epsilon t)\Phi$ now decays exponentially with time, as $\exp(-2\Gamma t)$ (factor 2 because we have to square the function to get the norm). The same happens for a Hydrogen atom in a constant electric field. Rigorously, it has continuous spectrum - any energy is allowed. But in practice we deal with quasi-stationary bound states which decay.

FLOQUET ANALYSIS OF COHERENT EMISSION SPECTRA

Let us now use the Floquet approach to look at the time-dependent dipole moment $\mathbf{d}(\mathbf{t})$, induced by a laser field in a quantum system, and the spectrum of the coherent emission associated with it, which is given by the Fourier transform of $d(t)$.

In most strong-field experiments on high harmonics to-date, even with 10 fs pulses, the turn-on of the field is sufficiently adiabatic with respect to the electronic response of the system (which is on 10^2 attosecond scale). This means that when the system starts in a single quantum state - ground - only a single Floquet state will be created (populated) - the state that adiabatically connects to the field-free ground state.

Thus, we will have a ladder of photon steps as shown in Fig.1, with the distance between the steps equal to the laser frequency ω.

If the ground state has, say, even parity (usually the case), then the first step in the ladder is odd, the next is even, and so on. Emission of the harmonic photons $K\omega$ will correspond to the transitions between the different steps of the Floquet ladder, as shown in Fig.2. Since the emission of the harmonic photon changes the parity, transitions can only occur between the steps of the ladder that have opposite parity and, hence, separated by the odd number of laser photons ω. So far, nothing surprising.

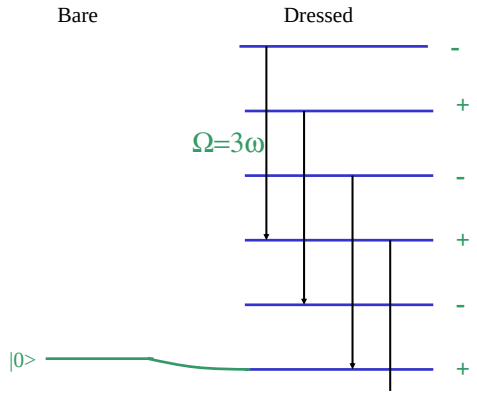

FIG. 2: The Floquet state and the transitions within it that lead to harmonic emission. If the ground state is even parity (usually is), then the first step in the ladder is odd, the next is even, and so on.

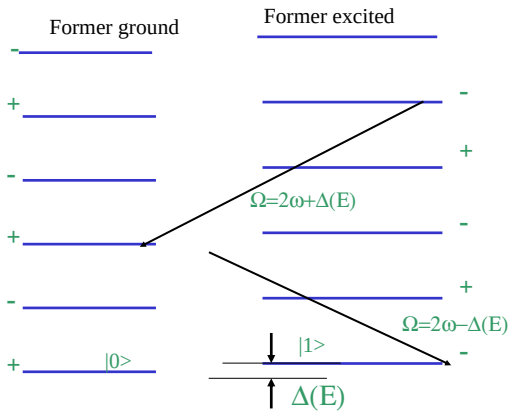

FIG. 3: Two Floquet states are populated coherently during fast non-adiabatic turn-on. Arrows mark transitions allowed by the selection rules which lead to the emission of "even harmonic" doublets (2-nd harmonic in the figure). $\Delta(E)$ is the splitting of the Floquet quasi-energies, which depends on the laser field strength E.

Before we move on to the surprises, let us get a bit more technical and look at the same emission in a slightly more mathematical manner to see how these jumps between the steps of the ladder come about.

If the system is in a Floquet state $\Psi_0(x,t)$, which originated from the (ground) state of the field-free system $|0\rangle$, then the polarization of the medium is

$$\mathbf{d}(t) = \langle \Psi_0(x,t)|\mathbf{d}|\Psi_0(x,t)\rangle \qquad (7)$$

Let us put into this equation the expression for the Floquet state,

$$\Psi_0(x,t) = e^{-i\epsilon t}\sum_k a_{0,k}\phi_{0,k}(x)e^{-ik\omega t} \qquad (8)$$

Here the coordinate parts $\phi_{0,k}(x)$ are normalized to unity, and the coefficients a_k carry the weights of each Fourier component. Putting Eq.8 into the Eq.7, we obtain

$$\mathbf{d}(t) = \sum_k \sum_n a^*_{0,n}a_{0,k}\langle\phi_{0,n}|\mathbf{d}|\phi_{0,k}\rangle e^{i(n-k)\omega t} \qquad (9)$$

Each term in the double sum corresponds to the emission of a photon $\Omega = (n-k)\omega$. It comes with the matrix element $\mathbf{d}_{n,k} = \langle\phi_{0,n}|\mathbf{d}|\phi_{0,k}\rangle$ and corresponds to the jump from the step number k to the step number n in Fig.2. Of course, these steps must have opposite parity – otherwise $\mathbf{d}_{n,k} = 0$. This means, once again, that $n-k$ must be odd.

There are plenty of terms with various n and k but the same difference $n-k$. All these correspond to the emission at the same frequency and have to be added coherently. In other words, all possible jumps between different pairs of steps on the Floquet ladder, which are separated by the same distance, have to be added coherently.

Consider now a case when the strong field is turned on very quickly, so that the system cannot adiabatically follow the changes in the field. In this case more than one Floquet state will be populated, especially if for some reason a second field-free state happened to be fairly close to the ground state.

Suppose that the two Floquet states, Ψ_0 and Ψ_1, are coherently populated, so that the total wavefunction of the system includes both: $\Psi = \alpha\Psi_0 + \beta\Psi_1$. Then the polarization of the medium is:

$$\begin{aligned}\mathbf{d}(t) &= [|\alpha|^2\langle\Psi_0|\mathbf{d}|\Psi_0\rangle + |\beta|^2\langle\Psi_1|\mathbf{d}|\Psi_1\rangle] \\ &+ [\alpha^*\beta\langle\Psi_0|\mathbf{d}|\Psi_1\rangle + \alpha\beta^*\langle\Psi_1|\mathbf{d}|\Psi_0\rangle] \qquad (10)\end{aligned}$$

The last two terms, complex conjugated to each other, describe jumps between the steps of the two Floquet ladders (Ψ_0 and Ψ_1) standing side-by-side. Once again, the selection rules require that the parity changes upon each jump. If the two original states which started these two ladders had opposite parity, the parity of each step is as shown in the Figure 3. The selection rules will then dictate that each allowed jump corresponds to the transition energy $\Omega = 2L\omega \pm \Delta(E)$, where $\Delta(E)$ is the splitting of the two quasi-energies - the shift between the two ladders. It depends on the field strength E.

Now, as a result of a quick turn-on, the coherent emission spectrum will look like shown in Fig.4 It contains "even harmonic" doublets (2-nd harmonic in the figure).

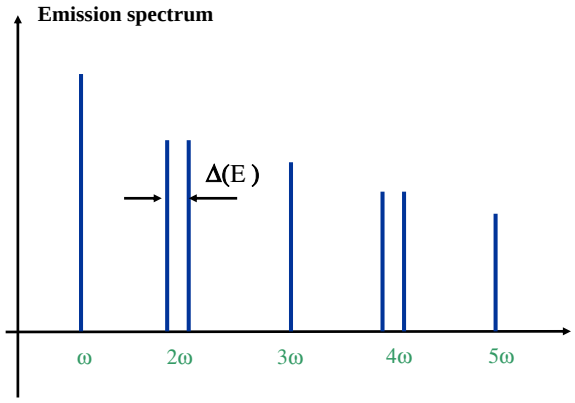

FIG. 4: Two Floquet states of the opposite parity are populated coherently during the turn-on. Coherent emission spectrum in this case includes the doublets around even harmonic lines.

The doublets are in reality nothing but the result of Raman transitions of various orders.

Now, finally, here comes the intriguing question: Is it possible that the two lines in the doublet merge into one? In other words, are there any conditions when the laser field induces degeneracy of the two Floquet states of the opposite parity, $\Delta(E) = 0$ in mathematical terms?

Before we go to the next section which contains positive answer to this question, let us for a second speculate about what such degeneracy would mean.

Second harmonics are never emitted in symmetric media. If we have managed to generate even harmonics - we have managed to break the symmetry in the medium.

How is this possible with a laser field $\mathbf{E}\cos\omega t$ which is linearly polarized, periodic, and seems to have no preference between left and right (positive and negative cosine values)? Is it possible to displace the electric charges and induce a permanent dipole (that is, break the symmetry) when the laser field is perfectly symmetric. And what would be the preferential direction that the system would choose?

A very general answer to all these questions is as follows. Suppose L is a characteristic size of the system and EL is the amplitude of the interaction strength, the voltage put across the system. Intuitively, it is clear that the interaction between the system and the field should be very different in the limits $EL \ll \Delta_0$ and $EL \gg \Delta_0$.

Suppose the field is strong and is turned on fast, so that it passes from the regime $EL \ll \Delta_0$ and into to the regime $EL \gg \Delta_0$ in a time which is much shorter than the laser cycle. In this case the field will have a specific direction at the moment when it passes through the demarcation line $EL \sim \Delta_0$. This direction could (and would) be special, and might be picked by the symmetric medium loosing its symmetry.

Of course, this special direction is determined not only by the peak intensity of the laser pulse but also, at a fixed peak intensity, by the absolute carrier phase of the oscillations under the envelope. For pulses $f(t)\cos\omega t$ and $-f(t)\cos\omega t$ the directions of the symmetry breaking will be opposite.

We now move to the example of such surprising effect, providing a much more accurate discussion of its physical details.

SYMMETRY BREAKING AND THE DESTRUCTION OF TUNNELING IN A DOUBLE WELL POTENTIAL

First of all, let me assure you that the effect is not limited to double-well systems and exists in multiple-well systems as well. The only reason for selecting a double-well potential is the simplicity of the treatment.

In a double well potential, each well on its own supports a state, see Fig.5. We denote these states $|L\rangle$ and $|R\rangle$, for "Left" and "Right". On their own, they are degenerate if the two wells are perfectly symmetric, which they are by our choice. However, since quantum particles tunnel, the addition of the second well makes sure that the two states local to each well are no longer the eigenstates of the system. Tunneling couples and splits them, removing the degeneracy, see Fig.5. If all other states are sufficiently far away (compared to the coupling between these two states), one can very easily diagonalize the 2x2 matrix with equal energies on the diagonal and a tunneling coupling off the diagonal.

The new eigenstates are the symmetric and antisymmetric superpositions of the local states:

$$\begin{aligned} |0\rangle &= \frac{|L\rangle + |R\rangle}{\sqrt{2}} \\ |1\rangle &= \frac{|L\rangle - |R\rangle}{\sqrt{2}} \end{aligned} \tag{11}$$

Conversely, the localized states are the symmetric and antisymmetric superpositions of the delocalized states:

$$\begin{aligned} |L\rangle &= \frac{|0\rangle + |1\rangle}{\sqrt{2}} \\ |R\rangle &= \frac{|0\rangle - |1\rangle}{\sqrt{2}} \end{aligned} \tag{12}$$

An example of such a system would be a diatomic molecular ion, such as H_2^+ - each nucleus makes a well, and the electron is running in between. Of course, to make analogy even better one has to localize the nuclei - that is, make a narrow vibrational wavepacket at appropriate internuclear separation, which determines the distance

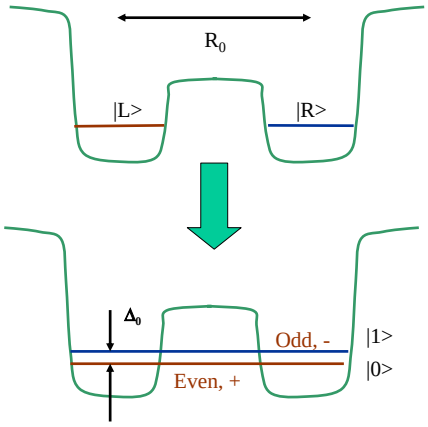

FIG. 5: In a double well potential, two delocalized "ground" states, even and odd, are approximately the symmetric and anti-symmetric superpositions of the localized states in each potential well. The splitting between them is due to tunneling, the tunneling time is $\tau_T = \pi/\Delta_0$.

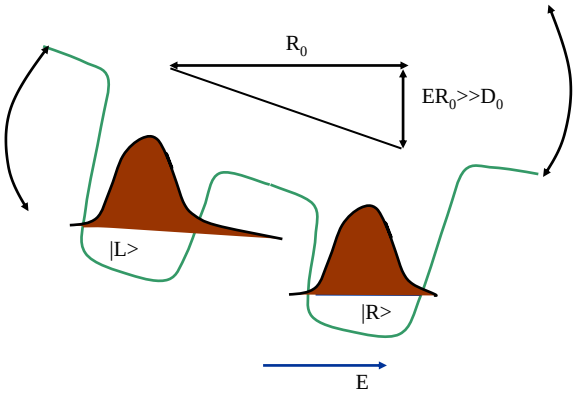

FIG. 6: Destruction of tunneling in a double well potential, for either a constant or an oscillating electric field. In a constant electric field the potential is biased by the field potential $-Fx$. The curved double-sided arrows indicate the rocking motion of the potential in the oscillating electric field. Tunnelling is only possible when the two wells are level with each other, and hence the electron moving, say, from left to right, can be admitted into the right well by the energy quantization.

between the wells. In H_2^+ the two localized states correspond to the electron being on the left or on the right nucleus, and the delocalized states $|0\rangle, |1\rangle$ are the familiar σ_g, σ_u states of H_2^+.

Before we proceed further, let me make a brief, simple, but very important remark about the physical meaning of Δ_0 - the splitting between the states $|0\rangle$ and $|1\rangle$. Let us put the system in a localized state $|L\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$, which is *not* an eigenstate. How will it evolve? Well, each eigencomponent of $|L\rangle$ will accumulate its own phase:

$$|\Psi(t)\rangle = \frac{|0\rangle e^{-iE_0 t} + |1\rangle e^{-iE_1 t}}{\sqrt{2}} = \\ = e^{-iE_0 t}\frac{|0\rangle + |1\rangle e^{-i\Delta_0 t}}{\sqrt{2}} \qquad (13)$$

and after a time $\tau_T = \pi/\Delta_0$ the state $|\Psi(t)\rangle$ will coincide with the right-localized state $|R\rangle$

$$|\Psi(t = \frac{\pi}{\Delta_0})\rangle = e^{-iE_0\frac{\pi}{\Delta_0}}\frac{|0\rangle - |1\rangle}{\sqrt{2}} = e^{-iE_0\frac{\pi}{\Delta_0}}|R\rangle \qquad (14)$$

Thus, the time $\tau_T = \pi/\Delta_0$ is the time it takes a particle (electron) to tunnel from one well to another.

If we want to break the symmetry and generate even harmonics, we need to make this splitting in the laser field equal to zero, $\Delta(E)$=0. Thus, we need to do two things: (1) destroy the tunneling and (2) catch the electron in one of the wells. Which one? – this will depend on the absolute carrier phase.

How do we destroy the tunneling? It is very easy with a constant electric field - you have to apply the voltage exceeding the tunneling splitting: $ER_0 \gg \Delta_0$. This is shown in Fig. 6, where the bias of the total potential is due to the electric field. The states localize in each well. Now the electron sitting in the left well can no longer tunnel efficiently to the right well because the quantization does not support it (at the energy of the left state). In this case they say that the resonant tunneling is no longer possible.

With an oscillating field the double-well potential will rock back and force around the middle every laser cycle: ER_0 is replaced now by $ER_0 \cos\omega t$. If ER_0 is sufficiently large, the tunneling will only happen at the brief instances when the two localized states R and L have the same energy - i.e. while $ER_0 \cos\omega t < \Delta_0$. At all other times tunneling is not possible. Hence, the tunneling time will grow and the tunneling splitting - now of the Floquet quasi-energies - will shrink. The "even harmonic" doublets will come closer.

But there is even more: there are some special values of the field where the tunneling is completely suppressed. What is the origin of such special values of the field? It is quantum destructive interference. Let us make a very crude qualitative estimate of when we should expect such interference and how it comes about.

Let's say the wavefunction is initially localized in the left well. At some point, while $ER_0 \cos\omega t < \Delta_0$, a small portion tunnels to the right well. During the next half-cycle the two parts of the wavefunction stay in their respective wells - one going up in energy and

one going down, acquiring different energy phase-shifts $\exp(\pm i(1/2)ER_0 \sin\omega t/\omega)$, with the total phase difference something like $\delta\phi \sim ER_0/\omega$). This phase difference becomes important when the next portion of the wavefunction tunnels to the right at the end of the half-cycle. The new fragment has to be added coherently to the half-cycle earlier escapee, and their interference will be destructive if $\delta\phi \sim ER_0/\omega \sim \pi(2n+1)$. The exact condition on the field is somewhat different, and is derived below. But the physics is exactly as described. And it means that over many cycles, on average, tunneling is destroyed and hence the Floquet states become degenerate.

Once we have found the points of the degeneracy of the Floquet states, the only question left is: where will the electron get stuck? This is completely determined by how we cross the demarcation line from the weak field to the strong-field regime, and where the electron was at that point. Initially, in weak fields, the electron was following the field oscillations, tunneling back and force between the wells and trying to keep minimal energy - i.e. stay in the lower well. But when the transition to the strong-field limit occurs, and occurs very quickly - the electron is no longer able to tunnel and it gets caught in the well it was in at this time. And where it was - this depends on the exact evolution of the electric field - that is, on the absolute carrier phase.

I conclude this chapter with a formal treatment of the problem. The wavefunction is written as

$$|\Psi(t)\rangle = a_0(t)|0\rangle + a_1(t)|1\rangle \tag{15}$$

and, when plugged into the Schroedinger equation, gives two differential equations for the amplitudes

$$\begin{aligned} i\dot{a}_0 &= -\frac{\Delta_0}{2}a_0 + V\cos\omega t a_1 \\ i\dot{a}_1 &= \frac{\Delta_0}{2}a_1 + V\cos\omega t a_0 \end{aligned} \tag{16}$$

where I have set the energies of the two field-free states to $\pm\Delta_0/2$ and denoted $V = V_{10} = V_{01} = d_{01}E \approx (R_0/2)E$ the coupling strength. We cannot use any rotating wave approximation here - the effect is way beyond it since it requires $V \gg \Delta_0$.

The way to deal with very strong fields is to try to include the field exactly, treating the field-free part as a perturbation. In this spirit, we look for the solutions in a form that would have worked for $\Delta_0 = 0$. In this case the local states in the left and right well would have been decoupled, and would have rocked up and down with the oscillations of the field, out of phase with each other.

So, we change to the amplitudes

$$a_L = \frac{a_0 + a_1}{\sqrt{2}} \qquad a_R = \frac{a_0 - a_1}{\sqrt{2}} \tag{17}$$

for which the equations are decoupled if $\Delta_0 = 0$:

$$\begin{aligned} i\dot{a}_L &= +V\cos\omega t a_L - \frac{\Delta_0}{2}a_R \\ i\dot{a}_R &= -V\cos\omega t a_R - \frac{\Delta_0}{2}a_L \end{aligned} \tag{18}$$

Now, "overlooking" the second term, we look for the solutions in a form

$$\begin{aligned} a_L &= b_L e^{-i\frac{V}{\omega}\sin\omega t} \\ a_R &= b_R e^{+i\frac{V}{\omega}\sin\omega t} \end{aligned} \tag{19}$$

For $\Delta_0 = 0$ this would have been exact solution with *constant* values of b_L and b_R determined by initial conditions. But in our case $\Delta_0 \neq 0$ and hence b_L and b_R are functions of time. If we put these expressions Eq.19 into Eq.18, we get

$$\begin{aligned} i\dot{b}_L &= -\frac{\Delta_0}{2}b_R e^{i\frac{2V}{\omega}\sin\omega t} \\ i\dot{b}_R &= -\frac{\Delta_0}{2}b_L e^{-i\frac{2V}{\omega}\sin\omega t} \end{aligned} \tag{20}$$

The problem is virtually done. Each differential equation should be integrated. We have very fast oscillating exponents on the right hand side, which do not contribute to such integrals unless they oscillate around non-zero constant background. Let us average the fast-oscillating terms over one laser period. This is, in fact, a perfectly sensible procedure if we are interested in a long-term evolution over many laser cycles - that's exactly tunneling given by a small splitting of merging quasi-energies. We get

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt e^{\pm i\frac{2V}{\omega}\sin\omega t} = J_0(2V/\omega) \tag{21}$$

Thus, the zero-order Bessel function J_0 gives the constant background that accumulates in the long-term integrals. The slow evolution is described by the averaged equations

$$\begin{aligned} i\dot{b}_L &= -\frac{\Delta_0}{2}J_0(2V/\omega)b_R \\ i\dot{b}_R &= -\frac{\Delta_0}{2}J_0(2V/\omega)b_L \end{aligned} \tag{22}$$

This is, in fact, the final answer. The field-free splitting (at V=0) between the levels Δ_0 has been replaced by the field-dependent splitting $\Delta_0 J_0(2V/\omega)$. The equations are virtually identical to the field-free ones up to this single simple substitution. Just like Δ_0 determined the splitting of the field-free energies, the splitting of the quasienergies is:

$$\Delta(E) = \Delta_0 J_0(2V/\omega) \tag{23}$$

The special points where exact degeneracy occurs, the tunneling is destroyed and the symmetry can be broken by a rapid turn-on correspond to the zeroes of the Bessel function, which in the limit of large argument are pretty close to our simple and crude estimates made above.

Theory of Intense Laser Matter Interaction. Lectures III-IV

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These Lectures introduce strong-field S-matrix and the strong-field approximation, and then use this formalism to describe strong-field ionization.

INTRODUCTION

The purpose of these lectures is to provide a glimpse into the theory of strong-field ionization. It is absolutely not possible to give a full account of this very rich problem, but I will point out some key ideas.

Strong-field ionization is a term that is used rather loosely. In these lectures, I will use this term to describe ionization in the regime where the laser frequency is much smaller than the ionization potential of the system, $\omega_L \ll I_p$, and when the characteristic excitation energy from the ground state of the neutral system ω_0 is also large compared to ω_L. What this means is that the electrons can respond very quickly to the oscillating laser field $\mathcal{E}\cos\omega_L t$, and from the electron's perspective these field oscillations are nearly adiabatic.

What does this condition mean in practice is that the modifications of the 'inside' part of the binding potential well are slow, from the electron's point of view, and that the classical-like vertical absorption mechanism is not efficient.

Quantum-mechanically, it means that excitations inside the potential well do not develop, and that the most efficient way for the electron to escape is via the classically forbidden region. Note that the key parameter here was the laser frequency and its ratio to the characteristic response frequencies of the system. Note also, that nowhere it is required that the barrier has to be static while the electron is going through it.

Now we will try to put this physical picture into math.

BASIC FORMALISM

Hamiltonians and Gauges

Let us start with the time-dependent Schroedinger equation, which reads:

$$i\dot{|}\Psi\rangle = [\hat{H}_0 + \hat{V}(t)]|\Psi\rangle = \hat{H}(t)|\Psi\rangle \tag{1}$$

Here $V(t)$ describes the interaction with the laser field and H_0 is the field-free hamiltonian. In this lecture I will assume that the wavelength of light is much bigger than the size of our quantum system, and so I will use the dipole approximation, which means that the spatial dependence of the electromagnetic field across the size of the system is ignored.

In the dipole approximation, laser-system interaction can be written in the so-called length gauge as $\hat{V} = -\hat{\mathbf{d}}\mathcal{E}(t)$. The Hamiltonian has the form

$$\hat{H}_{\rm LG}(t) = \frac{\hat{\mathbf{p}}^2}{2m} + U(\mathbf{r}) - \hat{\mathbf{d}}\mathcal{E}(t) \tag{2}$$

Here $U(r)$ is the interaction potential between the ionizing active electron and the ionic core. The mass of the electron is $m = 1$ in the atomic units which are used everywhere below. The electron charge $q = -e = -1$ in the same units.

There are two more approximations here. The first is to describe the laser field classically – this is fine given large number of photons (high intensity). The second is to use the single active electron approximation in Eq.(2) – this is a major approximation but it has been very successful in a majority of cases of interest.

Before we proceed further, let me stress that the length form of the interaction is approximate and can only be used in the dipole approximation. There is another gauge that is often used in treating laser-matter interaction, the so so-called velocity gauge. This one is general and works beyond the dipole approximation. The Hamiltonian in the velocity gauge is

$$\hat{H}_{\rm VG}(t) = \frac{[\hat{\mathbf{p}} - q\mathbf{A}]^2}{2m} + U(\mathbf{r})$$
$$\hat{H}_{\rm VG}(t) = \frac{[\hat{\mathbf{p}} + \mathbf{A}]^2}{2m} + U(\mathbf{r}) \tag{3}$$

where in the last line $q = -1$ was used and the vector potential $\mathbf{A}$ is defined as

$$\mathcal{E}(t) = -\frac{\partial \mathbf{A}(t)}{\partial t} \tag{4}$$

These two forms of the Hamiltonian are equally good, meaning that one can go from the TDSE in the velocity form to the TDSE in the length form by a unitary transformation

$$\Psi_{\rm LG}(t) = e^{-iq\hat{\mathbf{r}}\mathbf{A}(t)}\Psi_{\rm VG}(t) = e^{i\hat{\mathbf{r}}\mathbf{A}(t)}\Psi_{\rm VG}(t) \tag{5}$$

and the corresponding transformation of the Hamiltonian, which you are welcome to try to derive. All observable quantities are invariant under this unitary gauge transformation – that is, as long as the TDSE is solved exactly.

From now on I will stick mostly to the length gauge and I will drop the subscript LG from $H_{\rm LG}$.

Formal solutions

The discussion below applies equally to both the length and the velocity gauge, or any other gauge.

The formal solution of the TDSE

$$i\dot{|}\Psi\rangle = [\hat{H}_0 + \hat{V}(t)]|\Psi\rangle = \hat{H}(t)|\Psi\rangle \tag{6}$$

is

$$|\Psi(t)\rangle = e^{-i\int_{t_i}^{t}\hat{H}(\tau)d\tau}|\Phi_i\rangle = \hat{U}(t,t_i)|\Phi_i\rangle \tag{7}$$

Here Φ_i is the initial state of the system at a fixed moment $t = 0$. The exponential operator is the propagator $U(t, t_i)$. This solution seems to be of little use at this very moment: evaluating exponential operators is a tedious task, no easier than solving TDSE. However, we will find a good use to this formal solution very shortly.

The separation of the Hamiltonian $H(t)$ into the two parts, $H = H_0+V$ is also knows as the partitioning of the Hamiltonian. Here our partitioning is into the field-free part and the interaction with the laser field, but one can come up with many other ways for such partitioning. It turns out that one can use such partitioning to relate the TDSE solution for the full Hamiltonian to the solution for the part of the Hamiltonian H_0.

Let us write the TDSE with the Hamiltonian H_0

$$i\dot{|}\Psi^{(0)}\rangle = \hat{H}_0|\Psi^{(0)}\rangle \tag{8}$$

Its formal solution is

$$|\Psi^{(0)}\rangle = e^{-i\hat{H}_0 t}|\Psi(t=0)\rangle = e^{-i\hat{H}_0 t}|\Phi_i\rangle \tag{9}$$

where $|\Phi_i\rangle$ is the same initial state of the system as above in Eq.7 and the index (0) means that this solution applies to the Hamiltonian H_0.

What is the relationship between $\Psi(t)$ and $\Psi^{(0)}(t)$? Direct substitution into the TDSE Eq.(6) shows that its exact solution can be written as

$$\Psi(t) = \Psi^{(0)}(t) + \Delta\Psi(t) = e^{-i\hat{H}_0 t}|\Phi_i\rangle + \Delta\Psi(t)$$
$$\Delta\Psi(t) = -i\int_{t_i}^{t} dt' e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} V(t') e^{-i\hat{H}_0(t'-t_i))}|\Phi_i\rangle \tag{10}$$

Try to substitute this into the TDSE and check that it does indeed work. Here t_i is the initial moment of time when we know the wavefunction.

Let's see where we have come. We have started with a single exponential operator in Eq.(7) and we have replaced it with an integral over exponential operators taken from all times t'. Let's be frank: Eq.(10) does not look very inviting. However, it this this general – and exact – expression where interesting approximations can be explicitly tried, sometimes based on rigorous math and sometimes based on physical reasoning.

Let us look at the physics behind the expression Eq.(10). The system starts in the state $|\Phi_i\rangle$. During the time-interval before some moment t' it evolves without interacting with the laser field. If the initial state is an eigenstate of the Hamiltonian H_0 with energy E_i, all this evolution does is accumulates the phase due to the energy, $\exp[-iE_i(t' - t_i)]$.

This quiet evolution ends at a moment t' when the system is kicked by the instantaneous laser field $V(t')$. To which state the transition occurs at this moment is anybody's guess. It is called a virtual transition and it can go anywhere – the energy conservation law need not to be satisfied until the interaction is over. Then, from the moment t' to the moment of observation t the evolution is under the action of the full Hamiltonian, including both the laser field and the field-free potential.

If we are interested to find the transition amplitude from the initial field-free state Φ_i to some final state Ψ_f, then at the moment of observation the wavefunction must be projected onto the state of interest, Ψ_f. If we are interested in ionization and our initial state is a bound state, then the final (continuum) state must be orthogonal to the initial state. Then the projection of $\Psi^{(0)}(t)$ onto Ψ_f is zero, and the transition amplitude a_{fi} is

$$a_{fi}(t) = \langle\Psi_f|\Psi(t)\rangle = \langle\Psi_f|\Delta\Psi(t)\rangle =$$
$$= -i\int^{t} dt'\langle\Psi_f|e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau} V(t') e^{-i\hat{H}_0(t'-t_i))}|\Phi_i\rangle \tag{11}$$

This expression is often refereed to as strong-field S-matrix, as it reminds one time-dependent treatment of Scattering problems. It is exact. No approximations have been made yet. Now let's turn to the approximation.

THE VOLKOV PROPAGATOR AND THE STRONG FIELD APPROXIMATION

Let us think about the physics of the situation in the strong low frequency field. "Low frequency" means "compared with the characteristic response frequency" of the system. While the electron is in the initial – ground – state, not much is happening until it manages to escape to the continuum at some t', which could be pretty much any instant of time. At this point in time the strong field takes the electron over and it starts to oscillate in the field, possibly scattering on the parent ion. Can we put this physical picture into the mathematical terms? Or, rather, can we use this picture to do something to the formal expressions for the amplitude a_{fi}?

Once we realize that only the ground state and the continuum are involved in the dynamics, the plan is rather clear. Indeed, in Eqs.(10,11) first the electron sits in the ground state until the moment t', at which point the laser field $V(t')$ kicks it to the continuum. Now, while in the continuum, the electron is dominated by the laser field.

Therefore, instead of the exact propagator

$$\exp(-i\int_{t'}^{t}\hat{H}(\tau)d\tau)$$

we will use an approximate propagator that includes the laser field fully and exactly but completely ignores the field-free potential of the system. This is the essence of the Strong Field Approximation.

One of the main reasons to make such as approximation – completely neglect the atomic (or molecular) potential in the continuum – is that the propagator for the free electron in the laser field is known exactly. It is called the Volkov propagator. It corresponds to solving the TDSE for the Hamiltonian $H_{\rm v}(t)$:

$$\hat{H}_{\rm v}(t) = \frac{p^2}{2} + \mathbf{r}\mathcal{E}(t) \tag{12}$$

and is formally written as

$$\hat{U}_{\rm v}(t,t') = e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau} \tag{13}$$

To understand how this propagator – this exponential operator – acts on the electron in the laser field, let us see what would it do with the plane wave $\exp(i\mathbf{kr})$, which describes the free electron which has the kinetic momentum $\mathbf{k}$.

All the free electron does in the laser field is oscillates. Classically, if at time t' the electron has kinetic momentum $k' = k(t')$ (kinetic momentum $k(t') = mv(t')$ is proportional to the instantaneous velocity), then its kinetic momentum at any other time is

$$\mathbf{k}(t) = \mathbf{k}(t') - \mathbf{A}(t') + \mathbf{A}(t) \tag{14}$$

In other words,

$$\mathbf{k}(t) - \mathbf{A}(t) = \mathbf{k}(t') - \mathbf{A}(t') = \mathbf{p} = \text{const} \tag{15}$$

The conserved quantity $\mathbf{p}$ is called the canonical momentum. The instantaneous kinetic momentum (velocity) is expressed as

$$\mathbf{k}(t) = \mathbf{p} + \mathbf{A}(t) \tag{16}$$

What is important is that the plane wave $\exp(i\mathbf{kr})$ stays the plane wave, with the same canonical momentum, while its kinetic momentum changes as described by Eq.(16). The instantaneous kinetic energy during these oscillations is

$$E(t) = \frac{1}{2}[\mathbf{k}(t') - \mathbf{A}(t') + \mathbf{A}(t)]^2 \tag{17}$$

Now we can write the answer for the action of the Volkov propagator on the plane wave with kinetic momentum k' at the moment t':

$$e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau}|\mathbf{k}(t')\rangle = e^{-i\int_{t'}^{t}E(\tau)d\tau}|\mathbf{k}(t)\rangle \tag{18}$$

Here the plane waves $|k(t)\rangle, |k(t')\rangle$ have different momenta related by the Eq.(14). In terms of the conserved canonical momentum $\mathbf{p}$, which is also equal to the kinetic momentum of the electron after the laser field is switched off, the result can be re-written as

$$e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau}|\mathbf{p} + \mathbf{A}(t')\rangle = e^{-i\int_{t'}^{t}E(\tau)d\tau}|\mathbf{p} + \mathbf{A}(t)\rangle \tag{19}$$

Thus, the coordinate part of the wavefunction has been changed, but the temporal phase added is the same for all coordinates because the interaction (the laser field) is homogeneous. This is the only reason we could replace the Hamiltonian operator in the exponent with the energy.

Before we use this expression, let us turn it around a bit and apply the propagator to the bra- and not the ket. Let us specify the kinetic momentum at the instant t to be equal to $\mathbf{k}$, and back-propagate it in time:

$$\langle\mathbf{k}|e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau} = e^{-i\int_{t'}^{t}E(\tau)d\tau}|\langle\mathbf{k}(t')\rangle \tag{20}$$

where the instantaneous kinetic energy is

$$E(\tau) = \frac{1}{2}[\mathbf{k} + \mathbf{A}(t') - \mathbf{A}(\tau)]^2 \tag{21}$$

Now we write the SFA approximate expression for the amplitude to find the system with the momentum $|\mathbf{k}\rangle$ at an instant t.

We assume that the system has started at the moment t_i in the ground state $\Phi_i \equiv \Phi_g$ with the energy $E_g = -I_p$ (and hence $\exp(-i\hat{H}_0(t'-t_i)) = \exp(+iI_p(t'-t_i))$. The amplitude to find the system with the momentum $\mathbf{k}$ at the time t, according to the general equation Eq.(11), is

$$a(\mathbf{k},t) = -i\int^{t} dt'\langle\mathbf{k}|e^{-i\int_{t'}^{t}\hat{H}(\tau)d\tau}V(t')e^{i\hat{I}_p(t'-t_i))}|\Phi_g\rangle \tag{22}$$

In the SFA, we replace the exact propagator between t' and t with the Volkov propagator,

$$a(\mathbf{k},t) = -i\int^{t} dt'\langle\mathbf{k}|e^{-i\int_{t'}^{t}\hat{H}_{\rm v}(\tau)d\tau}V(t')e^{i\hat{I}_p(t'-t_i))}|\Phi_g\rangle \tag{23}$$

Now we use the fact that we know how to back-propagate the plane wave in the laser field:

$$a_{\mathbf{k}}(t) = -i\int^{t} dt' e^{-i\int_{t'}^{t}E(\tau)d\tau + iI_p(t'-t_i)} \langle\mathbf{k} + \mathbf{A}(t') - \mathbf{A}(t)|V(t')|g\rangle \tag{24}$$

where the instantaneous energy $E(\tau)$ is now given by

$$E(\tau) = \frac{1}{2}[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 \tag{25}$$

since we have fixed the momentum $\mathbf{k}$ at the instant t.

In the literature, the $\mathbf{k}, t, t'$-dependent phase of this integral,

$$S(t,t',\mathbf{k}) = \frac{1}{2}\int_{t'}^{t} d\tau[\mathbf{k} - \mathbf{A}(t) + \mathbf{A}(\tau)]^2 - I_p t' \tag{26}$$

is often referred to as the classical action of the free electron in the laser field. It is a bit of a stretch of the term, since it also includes the $-I_p t'$. Often you will find it written in terms of the canonical momentum $\mathbf{p} = \mathbf{k} - \mathbf{A}(t)$, which is a conserved quantity and will stay unchanged at all times — that's within the SFA, of course. In this case the phase will be

$$S(t, t', \mathbf{p}) = \frac{1}{2}\int_{t'}^{t} d\tau [\mathbf{p} + \mathbf{A}(\tau)]^2 - I_p t' \qquad (27)$$

In terms of action and the canonical momentum $\mathbf{p}$, the SFA transition amplitude is

$$a_{\mathbf{p}}(t) = -i\int^{t} dt' e^{-iS(t,t',\mathbf{p})-iI_p t_i}\langle \mathbf{p} + \mathbf{A}(t')|V(t')|\psi\rangle \qquad (28)$$

In future, if and when you come across such expressions, pay special attention to the meaning of the momentum – is it canonical or kinetic.

Eqs.(24,28) are intuitive and clear. The electron sits in the ground state until t' when it makes (at this point still virtual) transition to the continuum. Then the electron moves in the laser field, converting the virtual transition into real and oscillating – as the free electron should. It accumulates the phase given by the integral of its instantaneous energy $E(t'')$, performed between the moment of birth t' and the moment of observation t. The electron finishes with the canonical momentum $\mathbf{p}$, which dictates the initial kinetic momentum that the electron populates at t', $\mathbf{k} = \mathbf{p} + \mathbf{A}(t')$.

There are several major problems with this result, all stemming from the main approximation of the theory - to neglect the effect of the Coulomb potential.

(1) During the transition to the continuum the electron will be liberated differently if we include its interaction with the atomic core. After all, if it has to tunnel through the barrier, the shape of this barrier is important – and it is heavily affected by the binding potential. Thus, the ionization amplitude will be different. This problem can be corrected by incorporating the effect of the Coulomb tail into the electron action.

(2) The propagation in the continuum is also different: the electron not only oscillates in the laser field, it can also scatter off the atomic core. This is not present in the SFA formalism, but can be included additionally: The SFA result can be considered as the first term in a perturbative expansion, with the atomic potential being a perturbation. The corresponding new terms are often referred to as SFA2, etc. But the accuracy and the convergence of such series is generally a major problem.

(4) The Volkov propagator is sensitive to the gauge. The one in these notes is written in the length gauge. In the velocity gauge where $\hat{V} = \hat{\mathbf{p}}\mathbf{A}$ the plane wave state $|p\rangle$ stays the same between t' and t. Physically, this is simply because in this new gauge $\mathbf{p}$ refers to *canonical*, not *kinetic* momentum, and in the laser field the *canonical* momentum of the free electron is a constant of motion. This does not lead to any problems in the exact theory - i.e. if the electron is indeed free all the time. Then the extra phase that differs canonical and kinetic momenta states carefully cancels out, as it is chosen consistently for all states one deals with. But in the approximate theory it is not the case - the initial (ground) state is not a free electron state, and no consistent phase enters it. *The result is that SFA is not gauge invariant, which is really bad news for any theory.*

However, the good news is that the gauge problem directly affects only the pre-exponential term in the amplitude, and keep the major piece – which is the fast oscillating exponent – intact. So with exponential part we are more-or-less safe, at least as far as gauges go.

(5) Finally, there is one more major "wrong" with SFA. By using the plane waves as continuum states, we have selected a basis which is complete and not orthogonal to the initial state of the system. In other words, in addition to all other problems our basis set is also overcomplete: it includes all plane waves that already make up a complete basis *plus* an extra state. Result: the pre-exponential factor which includes transition matrix elements suffers – but the key exponential dependence stays the same.

To summarize, SFA is wrong in so many ways that it violates every rule in the theory book. But the physical picture that stands behind it is so intuitive, clear, and compelling that SFA is used very widely, and it works very well for gaining qualitative and sometimes even quantitative insight into the physics of intense laser-matter interaction.

KELDYSH-TYPE THEORIES OF STRONG-FIELD IONIZATION

The seminal paper by L. V. Keldysh on strong-field ionization, which has provided a unified picture of this process for various ionization regimes, has been published in 1965, very soon after the invention of the laser. The intensities available at that time have been many orders of magnitude lower than those envisioned by Keldysh. His results remained under-appreciated for almost two decades, pushed out onto the fringe by the successes of a much more quantitative time-dependent perturbation theory.

At the end of 70-th, it looked like the problem of multi-photon ionization was done and over with, and statements to that effect could have been found in the literature. This is precisely when the time-dependent perturbation theory began to fail. The first call was the discovery of above-threshold ionization in 1979 by Pierre Agostini. Let N be the minimum number of photons that the atom has to absorb to release an electron, $N = \mathrm{Int}[I_p/\omega] + 1$, where Int stands for the (floor) integer part. Then the energy of the freed electrons would be $E = N\omega - I_p$. Above threshold ionization (ATI) refers

to the observation of electrons with additional energy, gained by absorbing extra photons, $E = (N+s)\omega - I_p$. A few years later people have realized that this has all been predicted theoretically in mid-sixties by several groups of Russian theorists including (1) Keldysh, (2) Perelomov, Popov and Terent'ev (PPT), (3) Nikishov and Ritus. In 1980 H. Reiss developed his SFA, which is not much unlike the earlier Keldysh papers (even though Reiss says otherwise). Prior to that, in 1973, F. Faisal developed his approach – which is similar in some respects to the earlier PPT theory, albeit for a different (velocity) gauge. The various versions of the strong-field approximation are now referred to as KFR-theory (Keldysh-Faisal-Reiss).

The PPT theory stands alone. There are three reasons for it.

First, it is both deeper and more general than KFR. Second, it is generally superior in terms of quantitative accuracy compared to the KFR, with the exception of one very simple ad-hoc improvement of the KFR already described by Keldysh in 1965 – but without derivation.

Third, the PPT papers are written in the best style of the Russian school of the theoretical physics of that time: 'if you, dear reader, don't get that it this obvious, too bad for you, you idiot'.

The simplest limiting case of the PPT theory for very low-frequency laser fields have then been used (plagiarized) by Krainov, who turned it into now popular Ammosov-Delone-Krainov theory (ADK) that everyone uses. But let me stress that this latter is only valid in the tunnelling regime, and is not original in any way compared to the PPT theory.

I will now stop on the features and the math which are common in PPT and Keldysh, and the particular form of the approach is taken from the SFA.

Up to the global phase factor $e^{iI_p(t-t_i)}$ the SFA probability amplitude of populating the field-free continuum state labelled by the canonical momentum $|\mathbf{p}\rangle$ is

$$a_{\mathbf{p}}(t) = -i\int^t dt' e^{-iS(t,t',\mathbf{p})}\langle \mathbf{p}+\mathbf{A}(t')|V(t')|g\rangle$$
$$S(t,t',\mathbf{p}) = \frac{1}{2}\int_{t'}^t d\tau[\mathbf{p}+\mathbf{A}(\tau)]^2 + I_p(t-t') \quad (29)$$

The phase factor $e^{iI_p(t-t_i)}$ introduced for symmetry reasons corresponds to shifting the zero energy level to the ground state. Of course, nothing in observables changes as a result.

Let us assume that the field is linearly polarized, $\mathcal{E}\cos\omega t$. Then the vector-potential is

$$A_\parallel = -\frac{\mathcal{E}}{\omega}\sin\omega t = -v_0\sin\omega t$$
$$A_\perp = 0 \quad (30)$$

Then, keeping only exponential accuracy, we obtain

$$a(\mathbf{p},t) \sim \int_{-\infty}^t dt' \exp(-iS(\mathbf{p},t,t')) \quad (31)$$

where

$$S(\mathbf{p},t,t') = \left(I_p + \frac{1}{2}p_\perp^2\right)(t-t') +$$
$$+\frac{1}{2}\int_{t'}^t d\tau\left[p_\parallel - v_0\sin\omega\tau\right]^2 \quad (32)$$

is the familiar from the previous lecture action integral, $v_\parallel$ and $v_\perp$ are the velocity components parallel and perpendicular to $\vec{\mathcal{E}}$ and $v_0 = \mathcal{E}/\omega_L$ is the velocity amplitude of electron oscillations.

You already see that having non-zero perpendicular momenta is like increasing Ip. Hence, for now $p_\perp$ can be set to zero. If we are interested in the probabilities of populating non-zero $p_\perp$, all we need to do is take the formula for $p_\perp = 0$ and replace I_p with $I_p + p_\perp^2/2$. From now on, I will absorb $p_\perp^2/2$ into I_p.

This expression is the standard SFA expression for the ionization amplitudes, in the length gauge. The next step is to use it to evaluate ionization probabilities.

Calculating the integral

To calculate the integral for the ionization amplitude $a(\mathbf{p},t)$, we need to understand how does the phase $S(\mathbf{p},t,t')$ in this integral behave.

To do this analysis, let us temporarily re-write the action in terms of dimensionless variables: let us pull v_0 out of the brackets of Eq.(32), and also introduce the phase $\phi = \omega\tau$ as a new dimensionless integration variable:

$$S(p,t,t') = \frac{I_p}{\omega}(\omega t - \omega t') + \frac{v_0^2}{2\omega}\int_{\omega t'}^{\omega t} d\phi\,[u-\sin\phi]^2 \quad (33)$$

where the dimensionless momentum is $u = p_\parallel/v_0$. The momentum p here refers to the parallel momentum, $p_\perp^2/2$ is included into I_p.

We see that there are two important parameters in the phase – $N = I_p/\omega$ and

$$2Z = \frac{v_0^2}{2\omega} = 2\frac{\mathcal{E}^2}{4\omega^3}2 = \frac{U_p}{\omega} \quad (34)$$

where $U_p = \mathcal{E}^2/4\omega^2$ is the ponderomotive energy – the laser-cycle-average kinetic energy of the oscillating electron. In strong low-frequency fields both parameters are very large, meaning that the phase is changing very rapidly with time. For example, at intensities around $10^{14}\mathrm{W/cm}^2$ and for $w = 1.56\mathrm{eV}$, which corresponds to the laser wavelength of $800nm$, the characteristic value of the second term in the phase, over one laser cycle, is about 10 π, which is a very large phase change over one laser cycle. Therefore, we can use the saddle-point method to calculate the integral for the amplitude,

$$a(p,t) \sim \int_{-\infty}^t dt' \exp(-iS(\mathbf{p},t,t')) \quad (35)$$

where

$$S(p,t,t') = I_p(t-t') + \frac{v_0^2}{2}\int_{t'}^{t} d\tau\,[u-\sin\omega\tau]^2 \quad (36)$$

The saddle-point method proceeds as follows. First, one looks for the values of the integration variable t' where the phase of the integrand is stationary. In our case these stationary points are given by the equation

$$\frac{\partial S(p,t,t')}{\partial t'} = -I_p t' - \frac{v_0^2}{2}\left[u - v_0\sin\omega\tau'\right]^2 = 0 \quad (37)$$

Second, one expands $\exp(-iS(t'))$ in Taylor series around the stationary phase point $t' = t_m$. The first derivative w.r.t t' at this point is zero, and the integral becomes

$$\int dt' e^{-iS(t')} = e^{-iS(t_m)}\int dt' e^{-i\frac{S''(t_m)}{2}(t'-t_m)^2} \quad (38)$$

where the second derivative is denoted S''. Now one uses the fact that the integral converges quickly, so that the limits of integration with respect to $\xi = t' - t_m$ in the vicinity of the stationary point are expended to $\pm\infty$. The integral is then well-known, and the answer is

$$\int dt' e^{-iS(t')} \simeq e^{-iS(t_m)}\sqrt{2\pi/iS''(t_m)} \quad (39)$$

There could be many stationary phase points, and one needs to sum contributions from all of them, so the full answer is

$$\int dt' e^{-iS(t')} \simeq \sum_{t_m} e^{-iS(t_m)}\sqrt{2\pi/S''(t_m)} \quad (40)$$

Let us go back to our integral. The equation for the stationary phase points Eq.(37) can be re-written in terms of the dimensionless Keldysh parameter γ:

$$\begin{aligned}(u-\sin\omega t_m)^2 &= -\gamma^2\\ \gamma^2 = \frac{2I_p}{v_0^2} &= \frac{2I_p\omega^2}{\mathcal{E}^2}\end{aligned} \quad (41)$$

The solutions of this equation are always complex: all t_m will have imaginary part. Complex time means motion in classically forbidden region, it means tunnelling. In SFA all ionization, no matter what γ, is always rooted in tunneling.

To make our calculation slightly more specific, let us set $u \geq 0$. This means we are looking at the electrons flying in the positive direction. For each value of the normalized parallel component u of the canonical momentum, there are plenty of stationary phase points that satisfy Eq.(41). Indeed, let $t_0^{(0)}$ be one such point, with the real part of $\omega t_0^{(0)}$ between 0 and $\pi/2$ and an imaginary part $i\tau_T$:

$$\begin{aligned}t_0^{(0)} = \mathrm{Re}t_0^{(0)} + i\tau_T &\equiv t_0 + i\tau_T\\ 0 \leq \omega\mathrm{Re}t_0^{(0)} &\leq \pi/2\end{aligned} \quad (42)$$

Then $t_m^{(0)} = t_0^{(0)} + 2\pi m/\omega = t_0 + 2\pi m/\omega + i\tau_T$ would also satisfy the same stationary phase equation. Moreover, there are two points per each laser cycle – these are the points where sin(...) has the same value. Thus, in addition to $t_0^{(0)}$, there will also be $t_0^{(1)}$ related to $t_0^{(0)}$ as $\omega t_0^{(1)} = \pi - \omega t_0^{(0)}$.

Question: Show that the imaginary part of the action integral is the same for all $t_m^{(0,1)}$, particularly:

$$\mathrm{Im}[S(p,t,t_0^{(0)})] = \mathrm{Im}[S(p,t,t_m^{(0,1)})] \quad (43)$$

Remember that an integral from a point $t' = \mathrm{Re}t' + i\tau_T$ on the complex plane to the point t on the real axis can be calculated by first going to the real axis and then integrating along the real axis.

Since the imaginary part of the action is the same for all stationary phase points, we can write

$$S(p,t,t_m^{(0,1)}) = \mathrm{Re}[S(p,t,t_m^{(0,1)})] - i\sigma(\mathbf{p}) \quad (44)$$

where

$$\sigma = -\mathrm{Im}[S(p,t,t_0^{(0)})] \quad (45)$$

and the sum of the contributions of all stationary points to the total ionization amplitude is:

$$a(p,t) \propto e^{-\sigma(p)}\sum e^{-i\mathrm{Re}}[S(p,t,t_m^{(0,1)})] \quad (46)$$

with the summation over all m and $0, 1$.

As we see, in time-domain, we have a sequence of ionization bursts, twice per each laser cycle. These contributions from each successive cycle interfere with each other, leading to constructive and destructive interference depending on the specific value of p. The interference is responsible for turning the electron spectrum $|a(p)|^2$ into a sequence of peaks corresponding to a different number of absorbed photons. The picture is similar to optics, where a sequence of pulses following each other with regular intervals results in a 'comb' structure in the spectrum. Here the ionization bursts repeat every laser cycle, and there is a double-burst-structure for the ionization burst corresponding to a particular cycle.

Now, having understood the time-domain picture of strong-field ionization, let us finally turn to the contribution from a single stationary point, i.e. from a single ionization burst that produces electrons with the canonical momentum $\mathbf{p}$. The contribution from a single stationary point will also give us the ionization probability per laser cycle (up to factor 2, for two such points per cycle), and hence the ionization rate:

$$\Gamma(p) \propto e^{-2\sigma(p)} \quad (47)$$

This expression is written with only exponential accuracy.

Thus, to find the rate for each value of $p = v_0 u$, we will need to solve the equation Eq.(41) for $t_0^{(0)} = t_0 + i\tau_T$, then

calculate the action integral, find its imaginary part and find the corresponding rate.

Eq.(41) can be solved for any u, but for the moment I will limit the discussion to $u = 0$. The reason is simple: these values dominate the overall ionization rate, integrated over all $u = p/v_0$. Indeed, we will see very quickly that for $u = 0$ the complex time $t_0^{(0)} = t_0 + i\tau_T$ has $t_0 = 0$. Thus, for $u = 0$ the electron emerges from the classically forbidden region, where it has been travelling in complex (imaginary) time, at the maxima of the instantaneous electric field $\mathcal{E}\cos\omega t_0 = \mathcal{E}$. This is precisely when the strong-field ionization is peaked. Thus, the rate for $u = 0$ corresponds to the rate at the peaks of the field, and it will dominate the total, cycle-averaged, ionization rate. With exponential accuracy, this is all we need to know.

Our equation becomes

$$\sin\omega(t_0 + i\tau_T) = i\gamma \tag{48}$$

or, separating real and imaginary parts, $t_0 = 0$ (as promised) and

$$\sinh(\omega\tau_T) = \gamma \tag{49}$$

Remembering that

$$\sinh(\omega\tau_T) \equiv \frac{e^{\omega\tau_T} - e^{-\omega\tau_T}}{2} \tag{50}$$

and denoting $\exp(\omega\tau_T) = z$, we find the quadratic equation

$$z - \frac{1}{z} = 2\gamma \tag{51}$$

with the solution

$$\begin{aligned} z &= \gamma + \sqrt{\gamma^2 + 1} \\ \omega\tau_T &= \ln[\gamma + \sqrt{\gamma^2 + 1}] \end{aligned} \tag{52}$$

Let us now look at the two limits of this expression. For $\gamma \ll 1$ we have

$$\tau_T = \frac{\gamma}{\omega} \tag{53}$$

This expression gives a clear meaning to the γ parameter in terms of the so-called 'tunnelling time' τ_T: small γ mean that during tunnelling the barrier has no time to oscillate: $\omega\tau_T \ll 1$. Once we found the stationary phase point, we can now calculate the corresponding action integral:

$$S(\mathbf{p}, t, t') = I_p(t - i\tau_T) + \frac{v_0^2}{2}\int_{i\tau_T}^{t} d\tau\,[\sin\omega\tau]^2 \tag{54}$$

We note that by introducing the new integration variable $\tau = i\xi$ the integral term can be re-written as

$$\int_{i\tau_T}^{0} d\tau\,[\sin\omega\tau]^2 = i\int_0^{\tau_T} d\xi\sinh^2(\omega\xi) \tag{55}$$

and hence the imaginary part $\sigma = -\mathrm{Im}S$ is

$$\sigma = I_p\tau_T - \frac{\mathcal{E}^2}{2\omega^2}\int_0^{\tau_T} d\xi\sinh^2(\omega\xi) \tag{56}$$

Question: Calculate the remaining integral and show that in the limit $\gamma \ll 1$ this rate is given by the exact analogue of a DC tunnelling exponent:

$$\Gamma \propto \exp\left[-\frac{4}{3}I_p\tau_T\right] = \exp\left[-\frac{2}{3}\frac{[2I_p]^{3/2}}{\mathcal{E}}\right] \tag{57}$$

Let us now look at the opposite limit of very large $\gamma \gg 1$. The integral we need to calculate and the imaginary part of the action are given by the same general expression as before,

$$\sigma = I_p\tau_T - \frac{\mathcal{E}^2}{2\omega^2}\int_0^{\tau_T} d\xi\sinh^2(\omega\xi) \tag{58}$$

but now

$$\tau_T = \frac{1}{\omega}\ln[\gamma + \sqrt{\gamma^2 + 1}] \approx \frac{1}{\omega}\ln(2\gamma) \tag{59}$$

Question: Calculate the integral and show that in the limit $\gamma \gg 1$ this rate is given by the expression familiar from the time-dependent perturbation theory for the multiphoton ionization process:

$$\Gamma \propto \mathcal{E}^{2I_p/\omega} \propto I^{I_p/\omega} \tag{60}$$

Finally, do the calculation for the general case of arbitrary γ and derive the general expression for the ionization rate

$$\Gamma = \exp\left[-\frac{\mathcal{E}^2}{\omega^3}\left[\left(\gamma^2 + \frac{1}{2}\right)\omega\tau_T - \frac{1}{4}\sinh(2\omega\tau_T)\right]\right] \tag{61}$$

where $\omega\tau_T = \ln[\gamma + \sqrt{\gamma^2 + 1}] = \mathrm{Arcsh}(\gamma)$.