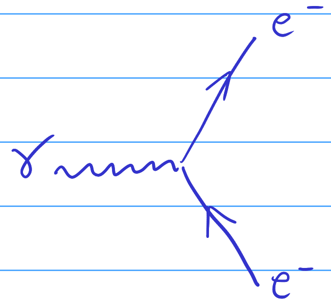
THEORY FOR ULTRAFAST DYNAMICAL PHOTOEMISSION

cases of dressed continuum, dressed atom
and dressed ion

(an analytical approach)



JAN MARCUS DAHLSTRÖM

TU WIEN Thu 14 Sep. 2023

Outline for tutorial:

$$e^{-i\chi}$$

- Light-matter interaction: (only atoms)

  Unitary transformations and gauges

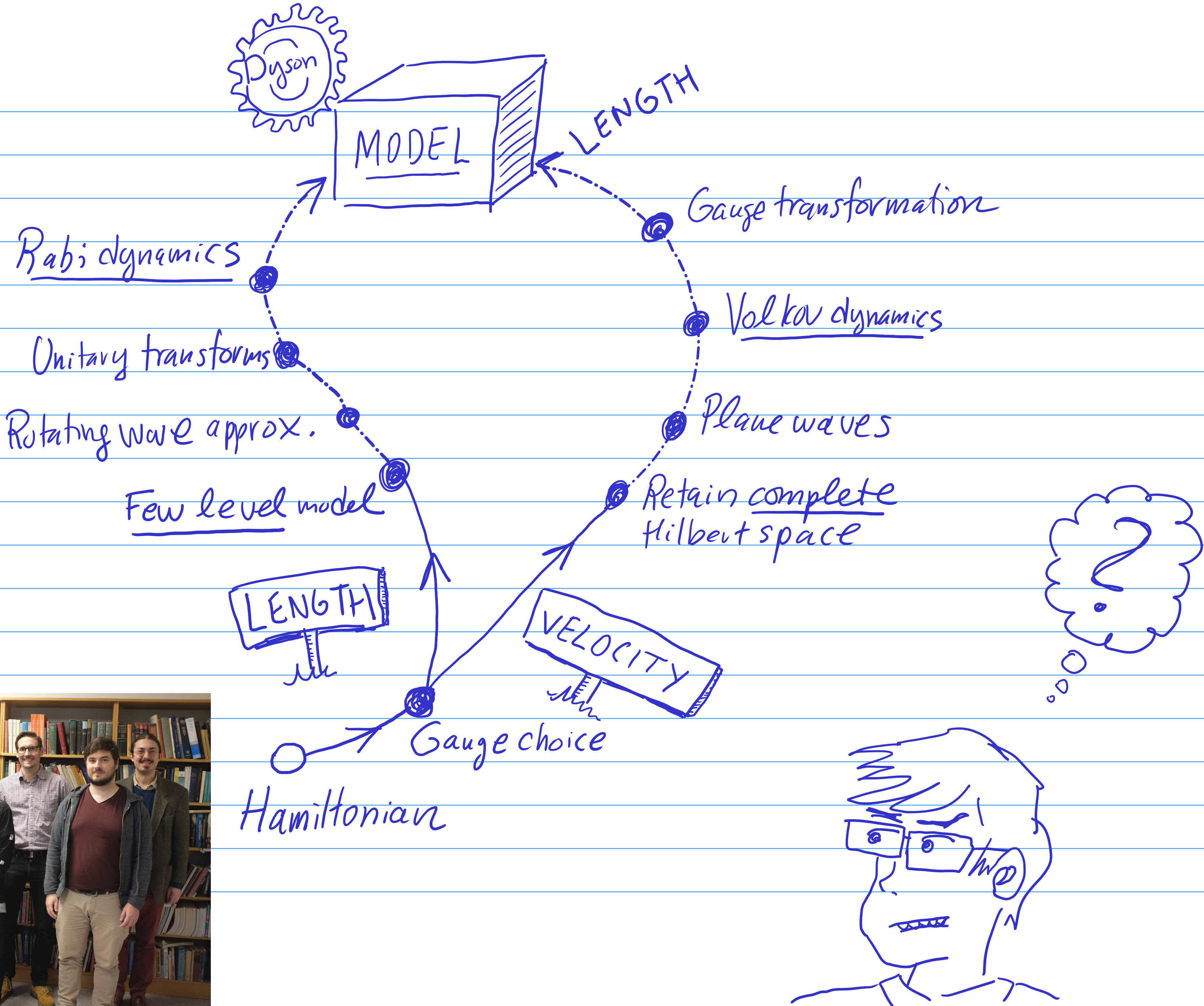
- Time evolution operator

  Analytical solutions for "simple" processes

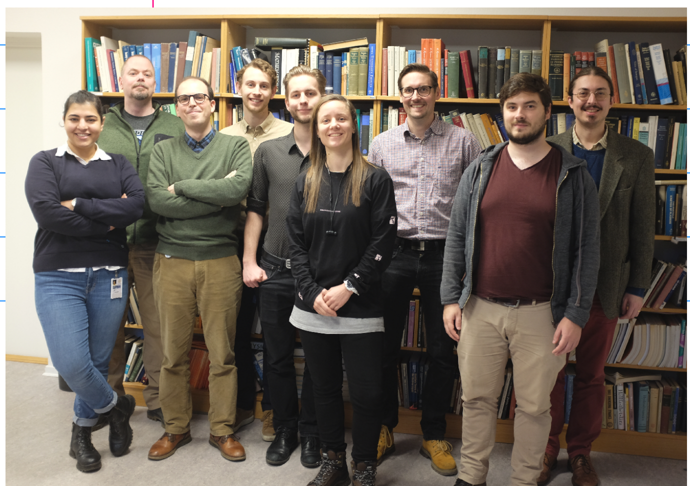
$$U(t, t_i)$$

- Dyson series expansion

  Approximate solutions for "complicated" processes



Dyson
MODEL
LENGTH
Gauge transformation
Rabi dynamics
Volkov dynamics
Unitary transforms
Rotating wave approx.
Plane waves
Few level model
Retain complete Hilbert space
LENGTH
VELOCITY
?
Gauge choice
Hamiltonian



Time-dependent Schrödinger equation (TDSE)

(homogeneous linear first-order differential equation)

$$\boxed{i \frac{d}{dt} |\Psi(t)\rangle = H(t) |\Psi(t)\rangle}$$

given initial state $|\Psi_i\rangle = |\Psi(t_i)\rangle$.

H is the Hamiltonian for the physical system.

Our task is to find analytical solutions to $|\Psi(t)\rangle$, $t \geqslant t_i$.

(or some good approximations)

Time-evolution operator ("propagator")

$$\boxed{|\Psi(t)\rangle = U(t, t_i)|\Psi_i\rangle}$$ (deterministic)

Property of propagator for TDSE:

LHS: $i\frac{d}{dt}|\Psi(t)\rangle = i\frac{d}{dt}U(t,t_i)|\Psi_0\rangle$

RHS: $H(t)|\Psi(t)\rangle = H(t)\,U(t,t_i)\,|\Psi_0\rangle$

$$\Rightarrow \boxed{\frac{\partial}{\partial t}\Big(U(t,t_i)\Big) = -i\,H(t)\,U(t,t_i)}$$

Formal solutions to the "propagator": I, II, III

Sakurai Modern Quantum Mechanics

CASE I: Time-independent Hamiltonian $\boxed{H(t) = H(t') = \hat{H}.}$

$$\boxed{U(t,t_i) = e^{-i(t-t_i)\hat{H}}}$$

exponential operator

Sketch:

$$\frac{\partial}{\partial t}\left(U(t,t_i)\right) = -i\hat{H}\, e^{-i(t-t_i)\hat{H}} = -i\hat{H}\, U(t,t_i) \quad \blacksquare$$

"Proof!"

Taylor series

USE: $$\boxed{e^{M} = \sum_{n=0}^{\infty} \frac{1}{n!} M^{n}}$$ M is a square matrix.

Assume $\hat{H}$ is a finite-dimensional Hermitian matrix.

$$M = -i(t-t_i)\hat{H} \longrightarrow M^n = \left(-i(t-t_i)\hat{H}\right)^n = (-i)^n (t-t_i)^n (\hat{H})^n$$

"*Proof*!"

Taylor series

USE: $$\boxed{e^{M} = \sum_{n=0}^{\infty} \frac{1}{n!} M^{n}}$$ M is a square matrix.

Assume $\hat{H}$ is a finite-dimensional Hermitian matrix.

$$M = -i(t-t_i)\hat{H} \longrightarrow M^{n} = \left(-i(t-t_i)\hat{H}\right)^{n} = (-i)^{n}(t-t_i)^{n}(\hat{H})^{n}$$

Take time derivate of each term:

$$\frac{\partial}{\partial t}\left\{(-i)^{n}(t-t_i)^{n}(\hat{H})^{n}\right\} = (-i)^{n}\, n\,(t-t_i)^{n-1}(\hat{H})^{n}$$

$$= (-i\hat{H})\left\{-i(t-t_i)\hat{H}\right\}^{n-1}, \quad n = 1, 2, 3, \ldots$$

$$\frac{\partial}{\partial t} e^{-i(t-t_i)\hat{H}} = (-i\hat{H})\sum_{n=1}^{\infty}\frac{n}{n!}\left\{-i(t-t_i)\hat{H}\right\}^{n-1} = (-i\hat{H})\, e^{-i(t-t_i)\hat{H}}$$

$(n-1)!$

Reconstruct exponential operator

CASE II:

- Time-dependent Hamiltonian: $H(t) \neq H(t')$ $\quad t' \neq t$,
- commutes at different times: $[H(t), H(t')] = 0$

behaves like "numbers"

$$\boxed{U(t,t_i) = e^{-i\int_{t_i}^{t} d\tau\, H(\tau)}}$$

Integrate all "energies" over time

Sketch:

$$\frac{\partial}{\partial t}\left(U(t,t_i)\right) = \left(-i\frac{\partial}{\partial t}\int_{t_i}^{t} d\tau\, H(\tau)\right) e^{-i\int_{t_i}^{t} d\tau\, H(\tau)} = -i H(t)\, U(t,t_i)$$

$\boxtimes$

"Proof":

Non-degenerate Hermitian matrices that commute share eigenvectors:

$$\begin{cases} [M,N]=0 \\ M=M^\dagger, \quad N=N^\dagger \end{cases} \implies M = U\Lambda_M U^\dagger \qquad N = U\Lambda_N U^\dagger, \qquad U^\dagger U = \mathbb{1}$$

Diagonal

Unitary

where U is the same unitary matrix of eigenvectors and $\Lambda_M = \mathrm{diag}(\lambda_M)$ and $\Lambda_N = \mathrm{diag}(\lambda_N)$.

"Proof":

Non-degenerate Hermitian matrices that commute share eigenvectors:

Diagonal

Unitary

$$\begin{cases} [M,N]=0 \\ M=M^\dagger, \quad N=N^\dagger \end{cases} \Longrightarrow \quad M = U\Lambda_M U^\dagger \qquad N = U\Lambda_N U^\dagger, \qquad U^\dagger U = \mathbb{1}$$

where U is the same unitary matrix of eigenvectors and $\Lambda_M = \mathrm{diag}(\lambda_M)$ and $\Lambda_N = \mathrm{diag}(\lambda_N)$.

Use diagonal form:

$$H(\tau) = U\Lambda(\tau)U^\dagger \; : \quad e^{-i\int_{t_i}^{t} d\tau\, H(\tau)} = \sum_{n=0}^{\infty} \frac{(-i)^n}{n!}\left(\int_{t_i}^{t} d\tau\, H(\tau)\right)^n = \ldots$$

Time-dependent eigenvalues on diagonal

$$\ldots = \sum_{n=0}^{\infty} \frac{(-i)^n}{n!}\left(U\left[\int_{t_i}^{t} d\tau\,\Lambda(\tau)\right]U^\dagger\right)^n = U e^{-i\int_{t_0}^{t} d\tau\,\Lambda(\tau)}U^\dagger \quad \square$$

Time independent

Exponent of diagonal

Useful!

CASE III:

- Time-dependent Hamiltonian : $H(t) \neq H(t')$ $t' \neq t$,
- doesn't commute at different times : $[H(t), H(t')] \neq 0$

not like "numbers"

$$\boxed{U(t,t_i) = T e^{-i\int_{t_i}^{t} d\tau H(\tau)}}$$

Time ordering

$t \geq t' \geq t'' \geq \cdots \geq t^{(n)} \geq t_i$

Dyson series expansion

$$= \mathbb{1} + \sum_{n=1}^{\infty} (-i)^n \int_{t_i}^{t} dt' \int_{t_i}^{t'} dt'' \cdots \int_{t_i}^{t^{(n-1)}} dt^{(n)} H(t') H(t'') \cdots H(t^{(n)})$$

Useful?

CASE III :

- Time-dependent Hamiltonian : $H(t) \neq H(t')$ $t' \neq t$,
- doesn't commute at different times : $[H(t), H(t')] \neq 0$

not like "numbers"

$$\boxed{U(t,t_i) = \mathcal{T} e^{-i\int_{t_i}^{t} d\tau H(\tau)}}$$

Time ordering

$t \geqslant t' \geqslant t'' \geqslant \cdots \geqslant t^{(n)} \geqslant t_i$

Dyson series expansion

$$= \mathbb{1} + \sum_{n=1}^{\infty} (-i)^n \int_{t_i}^{t} dt' \int_{t_i}^{t'} dt'' \cdots \int_{t_i}^{t^{(n-1)}} dt^{(n)} H(t')H(t'') \cdots H(t^{(n)})$$

$$\approx \mathbb{1} - i H(t_i) \Delta t$$

Small time steps in numerical simulations.
$\Delta t = t - t_i$

How to devolop analytical models using Dyson expansion:

Suppose that we know the analytical propagator for a "simple" problem: $H_0(t)$

$$|\Psi_0(t)\rangle = U_0(t,t_i)|\Psi_i\rangle, \quad \text{where } U_0(t,t_i) \text{ is known.}$$

where $H_0|\Psi_0(t)\rangle = i\frac{d}{dt}|\Psi_0(t)\rangle$

The total "complicated" Hamiltonian is:

$$H(t) = H_0(t) + V_0(t)$$

where $V_0(t)$ is the "perturbation" beyond the simple problem: $H_0(t)$.

Exact propagator rewritten using Dyson expansion:

$$\boxed{U(t,t_i) = U_0(t,t_i) - i\int_{t_i}^{t} dt'\, U(t,t')\, V_0(t')\, U_0(t',t_i)}$$

simple propagation

?? (unknown)

interaction at time t'

simple propagation $t_i \to t'$

Anatomy of strong field ionization: M. Y. Ivanov, M. Spanner and O. Smirnova J. Mod. Opt. 52, 165 (2005)

"Proof":

$$i\frac{d}{dt}|\psi(t)\rangle = H(t)|\psi(t)\rangle = \left(H_0(t) + V_0(t)\right)|\psi(t)\rangle \qquad \text{(RHS)}$$

Insert ansatz:

$$i\frac{d}{dt}|\psi(t)\rangle = i\frac{d}{dt}\left[U_0(t,t_i) - i\int_{t_i}^{t} dt'\, U(t,t')V_0(t')U_0(t',t_i)\right]|\psi_i\rangle = \dots$$

$$\dots = \left[H_0(t)U_0(t,t_i) - i^2\frac{d}{dt}\int_{t_i}^{t} dt'\, U(t,t')V_0(t')U_0(t',t_i)\right]|\psi_i\rangle = \dots$$

$$\dots = \Big[H_0(t)U_0(t,t_i) + \overbrace{U(t,t)}^{\mathbb{1}}V_0(t)U_0(t,t_i) + \dots$$

$$\dots + \int_{t_i}^{t} dt' \underbrace{\left(\frac{d}{dt}U(t,t')\right)}_{-iH(t)U(t,t')} V(t')U_0(t',t_i)\Big]|\psi_i\rangle$$

$$\ldots = \left[H_0(t) U_0(t,t_i) + V_0(t) U_0(t,t_i) - i \int_{t_i}^{t} dt' \, H(t) U(t,t') V_0(t') U_0(t',t_i) \right] |\psi_i\rangle$$

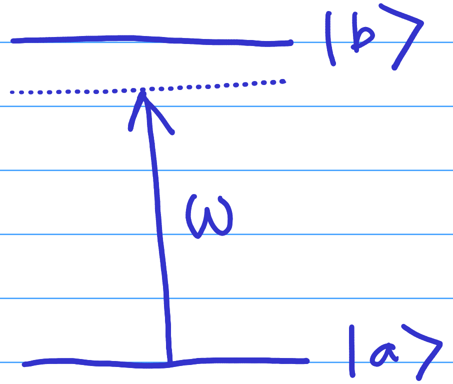
$$\ldots = \underbrace{\left[H_0(t) + V_0(t) \right]}_{H(t)} U_0(t,t_i) - i \int_{t_i}^{t} dt' \, H(t) U(t,t') V_0(t') U_0(t',t) \Big] |\psi_i\rangle$$

$$\ldots = H(t) \underbrace{\left\{ U_0(t,t_i) - i \int_{t_i}^{t} dt' \, U(t,t') V_0(t') U_0(t',t) \right\}}_{U(t,t_i)} |\psi_i\rangle$$

$$\ldots = H(t) U(t,t_i) |\psi_i\rangle = H(t) |\psi(t)\rangle \quad \blacksquare$$

"SIMPLE" PROBLEM 1. RABI OSCILLATIONS

(in two-level system)

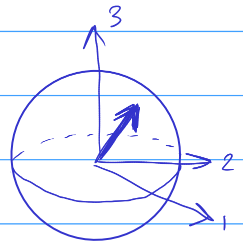


$$H(t) = H_0(t) + V_0(t)$$

$$\begin{cases} H_0|a\rangle = \varepsilon_a|a\rangle \\ H_0|b\rangle = \varepsilon_b|a\rangle \end{cases}$$

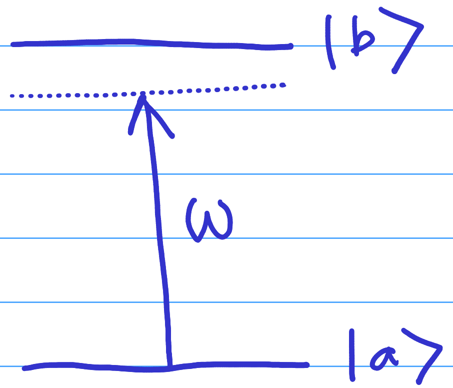
$$\langle b|V_0|a\rangle = E_0 \cos\omega t \, \langle b|z|a\rangle$$

Optical Resonance and Two-Level Atoms L. Allen and J.H Eberly (1987)



Alternative view with pseudo spin on Bloch sphere: Feynman et al. J. Appl. Phys. 28, 49 (1957)

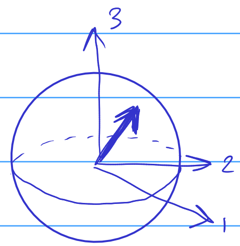
"SIMPLE" PROBLEM 1. RABI OSCILLATIONS (in two-level system)



$$H(t) = H_0(t) + V_0(t)$$

$$\begin{cases} H_0|a\rangle = \mathcal{E}_a|a\rangle \\ H_0|b\rangle = \mathcal{E}_b|a\rangle \end{cases}$$

$$\langle b|V_0|a\rangle = E_0 \cos\omega t \langle b|z|a\rangle$$



Optical Resonance and Two-Level Atoms L. Allen and J.H Eberly (1987)

Alternative view with pseudo spin on Bloch sphere: Feynman et al. J. Appl. Phys. 28, 49 (1957)

WARNING: There are <u>no</u> atoms with two levels. Must use <u>length</u> gauge.

Kobe and Smirl Am. J. Phys. 46(6) 624 (1976)

Hamiltonian:

$$H(t) = \underbrace{\varepsilon_a |a\rangle\langle a| + \varepsilon_b |b\rangle\langle b|}_{H_0 \text{ (Field-free atom)}} + \underbrace{E(t)\left[z_{ba}|b\rangle\langle a| + z_{ab}|a\rangle\langle b|\right]}_{V_0(t) \text{ (Interaction with field)}}$$

Transition matrix element is <u>real</u>: $z_{ba} = z_{ab}^* = z_{ab}$, $z_{ab} \in \mathbb{R}$.

Rabi frequency:

$$\Omega = E_0 z_{ba}$$

Detuning:

$$\Delta\omega = \omega - (\varepsilon_b - \varepsilon_a) = \omega - \omega_{ba}$$

Rotating Wave Approximation (RWA): $|\Delta\omega| \ll \omega$

$$\boxed{V_0^{RWA}(t) = \frac{\Omega}{2}\left[e^{-i\omega t}|b\rangle\langle a| + e^{+i\omega t}|a\rangle\langle b|\right]}$$

Unitary transformation:

$$H(t) = H_0 + V_0(t) \xrightarrow{T} \tilde{H} = \tilde{H}_0 + \tilde{V}_0$$

A <u>new</u> Hamiltonian $\tilde{H}$ found from TDSE:

$$H|\psi\rangle = i\frac{d}{dt}|\psi\rangle \longleftarrow \begin{cases} |\tilde{\psi}(t)\rangle = T|\psi(t)\rangle \\ |\psi\rangle = T^{\dagger}|\tilde{\psi}\rangle \end{cases}$$

Unitary transformation:

$$H(t) = H_0 + V_0(t) \xrightarrow{T} \tilde{H} = \tilde{H}_0 + \tilde{V}_0$$

A <u>new</u> Hamiltonian $\tilde{H}$ found from TDSE:

$$\begin{cases} |\tilde{\psi}(t)\rangle = T|\psi(t)\rangle \\ |\psi\rangle = T^\dagger|\tilde{\psi}\rangle \end{cases}$$

$$H|\psi\rangle = i\frac{d}{dt}|\psi\rangle$$

$$HT^\dagger|\tilde{\psi}\rangle = i\frac{d}{dt}\left(T^\dagger|\tilde{\psi}\rangle\right) = i\left(\frac{dT^\dagger}{dt}\right)|\tilde{\psi}\rangle + iT^\dagger\frac{d}{dt}|\tilde{\psi}\rangle$$

$$\underbrace{\left\{THT^\dagger - iT\left(\frac{dT^\dagger}{dt}\right)\right\}}_{\tilde{H}}|\tilde{\psi}\rangle = i\frac{d}{dt}|\tilde{\psi}\rangle$$

$\tilde{H}$: The <u>new</u> Hamiltonian has <u>two</u> terms.

How to go from a CASE $\underline{III}$ Hamiltonian to a CASE I ?

$$H^{RWA}(t) = \varepsilon_a |a\rangle\langle a| + \varepsilon_b |b\rangle\langle b| + \frac{\Omega}{2}\left[e^{-i\omega t} |b\rangle\langle a| + e^{+i\omega t} |a\rangle\langle b| \right]$$

CASE $\underline{III}$

Consider:

$$T_{co}(t) = \begin{bmatrix} 1 & 0 \\ 0 & e^{i\omega t} \end{bmatrix},$$

which is unitary: $T_{co}^{\dagger}(t) = T_{co}^{-1}$

How to go from a CASE III Hamiltonian to a CASE I ?

CASE III

$$H^{RWA}(t) = \mathcal{E}_a |a\rangle\langle a| + \mathcal{E}_b |b\rangle\langle b| + \frac{\Omega}{2}\left[e^{-i\omega t} |b\rangle\langle a| + e^{+i\omega t} |a\rangle\langle b| \right]$$

Consider:

$$T_{co}(t) = \begin{bmatrix} 1 & 0 \\ 0 & e^{i\omega t} \end{bmatrix}, \quad \text{which is unitary:} \quad T_{co}^{\dagger}(t) = T_{co}^{-1}$$

New Hamiltonian:

Due to time-dependence of transform

$$H^{co} = T_{co} H^{RWA} T_{co}^{\dagger} - i\, T_{co} \left(\frac{d T_{co}^{\dagger}}{dt} \right) = \begin{pmatrix} \mathcal{E}_a & 0 \\ 0 & \mathcal{E}_b \end{pmatrix} + \begin{pmatrix} 0 & \Omega/2 \\ \Omega/2 & 0 \end{pmatrix} + \begin{pmatrix} 0 & 0 \\ 0 & -\omega \end{pmatrix}$$

$$\Rightarrow \boxed{H^{co} = \begin{pmatrix} \mathcal{E}_a & \Omega/2 \\ \Omega/2 & \mathcal{E}_b - \omega \end{pmatrix}}$$

Time independent Hamiltonian: CASE I.
(quantum optics)

In "co-rotating" frame.

"Atom—Photon Interactions: Basic Process and Appilcations" Cohen-Tannoudji, Dupont-Roc and Grynberg (1998)

Application of Paulimatricies:

After another unitary transformation to move the zeropoint energy

$$\left(T_{shift} = \exp\left[-\frac{i}{2}(\omega - \varepsilon_b - \varepsilon_a)t\right] \right)$$

$$\boxed{H = \frac{\Delta\omega}{2}\sigma_z + \frac{\Omega}{2}\sigma_x}$$

We define the Pauli vector: $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

Rabi propagator:

CASE I:

$$U(t,t_i) = e^{-i(t-t_i)H} = e^{-i(t-t_i)\left[\frac{\Delta\omega}{2}\sigma_z + \frac{\Omega}{2}\sigma_x\right]}$$

Define: $A = -i\vec{v}\cdot\vec{\sigma} = -i\alpha$

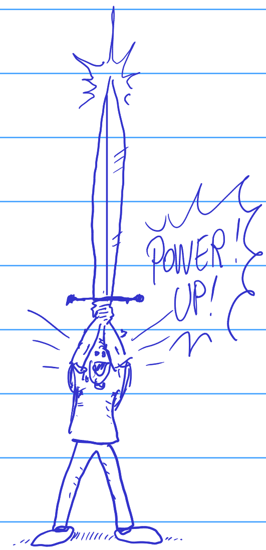
where $\vec{v} \in \mathbb{R}^3$

$$\begin{cases} v_x = (t-t_i)\Omega/2 \\ v_y = 0 \\ v_z = (t-t_i)\Delta\omega/2 \end{cases}$$

Use: $e^A = e^{-i\alpha} = \cos\alpha - i\sin\alpha$

$$= \sum_{n:\,\text{even}} \frac{1}{n!}(-i)^n \alpha^n + \sum_{n:\,\text{odd}} \frac{1}{n!}(-i)^n \alpha^n$$

Need to compute α^n for all powers n.



Powers of α:

$$\alpha^n = (\vec{v}\cdot\vec{\sigma})^n = (\vec{v}\cdot\vec{\sigma})(\vec{v}\cdot\vec{\sigma})\cdots(\vec{v}\cdot\vec{\sigma})$$

$$\alpha = \vec{v}\cdot\vec{\sigma} = v_x\sigma_x + v_y\sigma_y + v_z\sigma_z$$

$$\alpha^2 = (\vec{v}\cdot\vec{\sigma})(\vec{v}\cdot\vec{\sigma}) = (v_x\sigma_x + v_y\sigma_y + v_z\sigma_z)(v_x\sigma_x + v_y\sigma_y + v_z\sigma_z)$$

$$= v_x^2\overbrace{\sigma_x^2}^{1} + v_y^2\overbrace{\sigma_y^2}^{1} + v_z^2\overbrace{\sigma_z^2}^{1} + \dots$$

$$\dots + v_xv_y\underbrace{(\sigma_x\sigma_y + \sigma_y\sigma_x)}_{0} + v_xv_z\underbrace{(\sigma_x\sigma_z + \sigma_z\sigma_x)}_{0} + v_yv_z\underbrace{(\sigma_y\sigma_z + \sigma_z\sigma_y)}_{0} =$$

$$= (v_x^2 + v_y^2 + v_z^2)\,\mathbb{1} = |\vec{v}|^2\,\mathbb{1}$$

$$\alpha^3 = \alpha^2\alpha = |\vec{v}|^2\,\mathbb{1}\,(\vec{v}\cdot\vec{\sigma}) = |\vec{v}|^2(v_x\sigma_x + v_y\sigma_y + v_z\sigma_z)$$

The higher powers follow easily!

Conclusion:

- odd powers are proportional to $(\vec{\nu}\cdot\vec{\sigma})$
- even powers are proportional to $\mathbb{1}$.

General result:

$$e^{-i\vec{\nu}\cdot\vec{\sigma}} = \sum_{n:\text{even}} \frac{1}{n!}(-i)^n |\vec{\nu}|^n \mathbb{1} + \sum_{n:\text{odd}} \frac{1}{n!}(-i)^n |\vec{\nu}|^{n-1} (\vec{\nu}\cdot\vec{\sigma}) = \cdots$$

$$\cdots = \cos(|\vec{\nu}|)\,\mathbb{1} - i\sin(|\vec{\nu}|)\,\frac{(\vec{\nu}\cdot\vec{\sigma})}{|\vec{\nu}|}$$

In our case:

$$\begin{cases} |\vec{\nu}| = \sqrt{\nu_x^2 + \nu_y^2 + \nu_z^2} = (t-t_i)\dfrac{\sqrt{\Omega^2 + \Delta\omega^2}}{2} \equiv \underbrace{(t-t_i)}_{\text{Time evolution}} \dfrac{\overbrace{W}^{\boxed{\text{Generalized Rabi frequency}}}}{2} \\ (\vec{\nu}\cdot\vec{\sigma}) = (t-t_i)\left(\dfrac{\Omega}{2}\sigma_x + \dfrac{\Delta\omega}{2}\sigma_z\right) \end{cases}$$

"Flip" operator $\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$, "Split" operator $\sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$

Rabi "propagator":

Pauli matrix form:

Identity operator

$$\boxed{U(t,t_i) = \cos\left[\frac{W}{2}(t-t_i)\right]\mathbb{1} - i\sin\left[\frac{W}{2}(t-t_i)\right]\left(\frac{\Omega\sigma_x + \Delta\omega\sigma_z}{W}\right)}$$

Test on $|a\rangle$ at $t_i = 0$

$$U(t,0)|a\rangle = \cos\theta\,|a\rangle - i\sin\theta\left(\frac{\Omega\sigma_x + \Delta\omega\sigma_z}{W}\right)|a\rangle$$

$$= \underbrace{\left(\cos\theta - i\frac{\Delta\omega}{W}\sin\theta\right)}_{a(t)}|a\rangle - \underbrace{i\frac{\Omega}{W}\sin\theta}_{b(t)}\,|b\rangle = \ldots$$

Well-known Rabi amplitudes

$$\theta = Wt/2$$

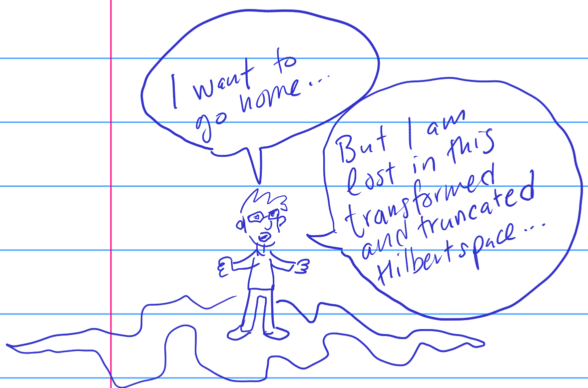
Dressed-state picture $\rightarrow$ two exponential functions

$$U(t,0)\,|a\rangle = \dots$$

$$\dots = \frac{1}{2}\left(\left(\left(1-\frac{\Delta\omega}{W}\right)e^{i\theta} + \left(1+\frac{\Delta\omega}{W}\right)e^{-i\theta}\right)|a\rangle - \frac{\Omega}{W}\left(e^{i\theta} - e^{-i\theta}\right)|b\rangle\right)$$

Asymmetric with detuning

Different signs



Dressed-state picture → two exponential functions

$$U(t,0)|a\rangle = \dots$$

$$\dots = \frac{1}{2}\left(\left(1-\frac{\Delta\omega}{W}\right)e^{i\theta} + \left(1+\frac{\Delta\omega}{W}\right)e^{-i\theta}\right)|a\rangle - \frac{\Omega}{W}\left(e^{i\theta} - e^{-i\theta}\right)|b\rangle$$

Asymmetric with detuning

Different signs

Let's go back to the co-rotating frame:

$$U_{co}(t,t_i) = T^{\dagger}_{shift}(t)\, U_{shift}(t,t_i)\, T_{shift}(t_i) \;, \quad T_{shift}(t) = e^{-\frac{i}{2}(\omega - \varepsilon_a - \varepsilon_b)t}$$

Let's go back to the original semi-classical Schrödinger frame:

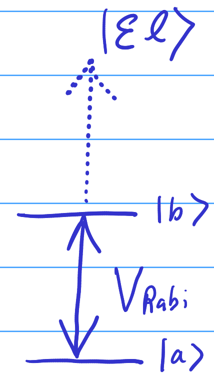
$$U(t,t_i) = T^{\dagger}_{co}(t)\, U_{co}(t,t_i)\, T_{co}(t_i) \;, \quad T_{co}(t) = \begin{bmatrix} 1 & 0 \\ 0 & e^{i\omega t} \end{bmatrix}$$

How to enter the complement of the essential states?

Consider the full Hamiltonian in length gauge:

$$H_{full} = \underbrace{\sum\!\!\!\!\!\!\int_n \mathcal{E}_n |n\rangle\langle n|}_{H_A} + \underbrace{\sum\!\!\!\!\!\!\int_{n \neq m} E(t)\, z_{mn} |m\rangle\langle n|}_{V_A(t)}$$

Atoms have infinitely many states and continua.

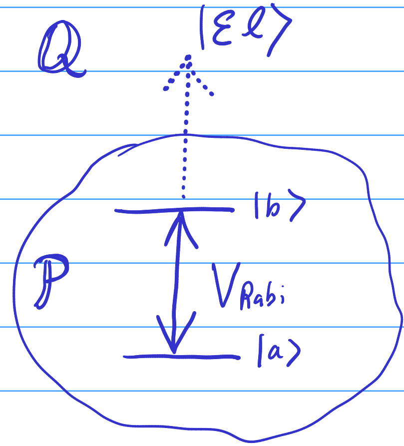


How to enter the complement of the essential states?

Consider the full Hamiltonian in length gauge:

$$H_{full} = \underbrace{\sum_n \mathcal{E}_n |n\rangle\langle n|}_{H_A} + \underbrace{\sum_{n\neq m} E(t)\, z_{mn} |m\rangle\langle n|}_{V_A(t)}$$

Atoms have infinitely many states and continua.



$$\dots = \underbrace{H_A + V_{Rabi}(t)}_{} + V_0(t) \quad ,$$

"strongly coupled": H_0

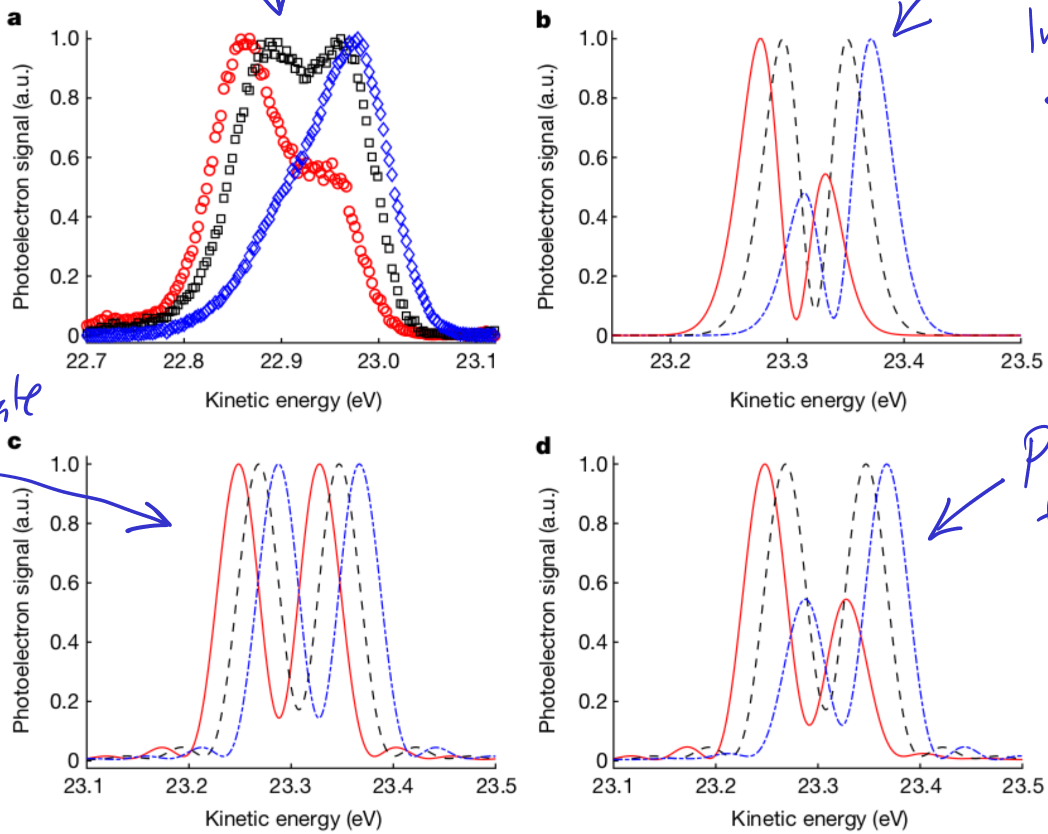
"perturbation": $V_0(t) = V_A(t) - V_{Rabi}(t)$

Beers and Armstrong Phys. Rev. A 12, 2447 (1975)

Roguś and Lewenstein J. Phys. B 19 3051 (1986)

Experiment at FERMI

Time-Dependent Configuration Interaction Singles (TDCIS)



Photoionization from excited state $|b\rangle$

Photoionization from ground state $|a\rangle$

"Autler-Townes" doublets

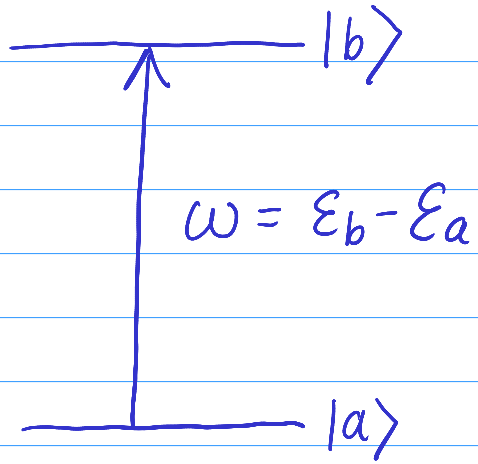
Fig. 2 | Asymmetry of the ultrafast AT doublet. a, Deconvoluted experimental photoelectron spectra with a symmetric AT doublet (black squares) at 23.753-eV photon energy, and the asymmetric ones at ±13-meV detuning (blue diamonds and red circles, respectively). **b**, Ab initio photoelectron spectra using TDCIS at three photon energies with a symmetric AT doublet at 24.157-eV photon energy (dashed black line) and the asymmetric ones at ±13-meV detuning. The red (blue) curve corresponds to red (blue) detuned light. **c**,**d**, The same as in **b**, but using the analytical model for 3/2 Rabi periods in the case of one-photon ionization from $|b\rangle$ (**c**) and two-photon ionization from $|a\rangle$ (**d**). The loss of contrast observed in the experimental spectra arises owing to macroscopic averaging of the target gas sample (see Supplementary Information for details). a.u., arbitrary units.

Nandi et al. Nature 608, 488 (2022)

"SIMPLE" PROBLEM 2 : AREA THEOREM

(here only one atom)

Resonant two-level system:



(Note: $\Delta\omega = 0$)

Laser pulse:

$$E(t) = \Lambda(t)\, E_0 \cos \omega t$$

Envelope

Interaction (RWA):

$$V(t) = \frac{\Omega(t)}{2}\left[e^{-i\omega t} |b\rangle\langle a| + e^{i\omega t} |a\rangle\langle b| \right]$$

Rabi fequency:

$$\begin{cases} \Omega_0 = E_0\, z_{ba} \\ \Omega(t) = E_0\, z_{ba}\, \Lambda(t) \end{cases}$$

"Flip" operator

$$\boxed{\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}}$$

Transform the Hamiltonian in Pauli matrix frame:

$$H(t) = \begin{pmatrix} \varepsilon_a & \Omega(t)/2 \, e^{i\omega t} \\ \Omega(t)/2 \, e^{-i\omega t} & \varepsilon_b \end{pmatrix} \longrightarrow H(t) = \begin{pmatrix} 0 & \Omega(t)/2 \\ \Omega(t)/2 & 0 \end{pmatrix}$$

$$H(t) = \frac{\Omega(t)}{2} \sigma_x \quad \leftarrow \text{Time-}\underline{\text{dependent}}\text{ Hamiltonian}$$

$$[H(t'), H(t)] = \frac{\Omega(t')\Omega(t)}{4} [\sigma_x, \sigma_x] = 0$$

which <u>commutes</u> with itself at <u>all times</u> $\Rightarrow$ <u>CASE II</u>

Good, news!

Propagator for resonant Rabi dynamics:

CASE II:

$$U(t,t_i) = e^{-i\int_{t_i}^{t} dt' H(t')} = e^{-i \overbrace{\int_{t_i}^{t} dt' \frac{\Omega(t')}{2}}^{\theta(t,t_i)\,:\,\text{scalar}} \sigma_x}$$

Pauli matrix $\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$

$$= \sum_{n=0}^{\infty} \frac{1}{n!} \left[-i\theta\right]^n \sigma_x^{\,n} \,, \qquad \sigma_x^{\,n} = \begin{cases} \sigma_x \,, & n \text{ is odd} \\ \mathbb{1} \,, & n \text{ is even} \end{cases}$$

Propagator for resonant Rabi dynamics:

CASE II:

$$U(t,t_i) = e^{-i\int_{t_i}^{t} dt' H(t')} = e^{-i\overbrace{\int_{t_i}^{t} dt' \frac{\Omega(t')}{2}}^{\Theta(t,t_i)\,:\,\text{scalar}}\,\sigma_x}$$

Pauli matrix $\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$

$$= \sum_{n=0}^{\infty} \frac{1}{n!}\left[-i\Theta\right]^n \sigma_x^n \,, \qquad \sigma_x^n = \begin{cases} \sigma_x \,, & n \text{ is odd} \\ \mathbb{1} \,, & n \text{ is even} \end{cases}$$

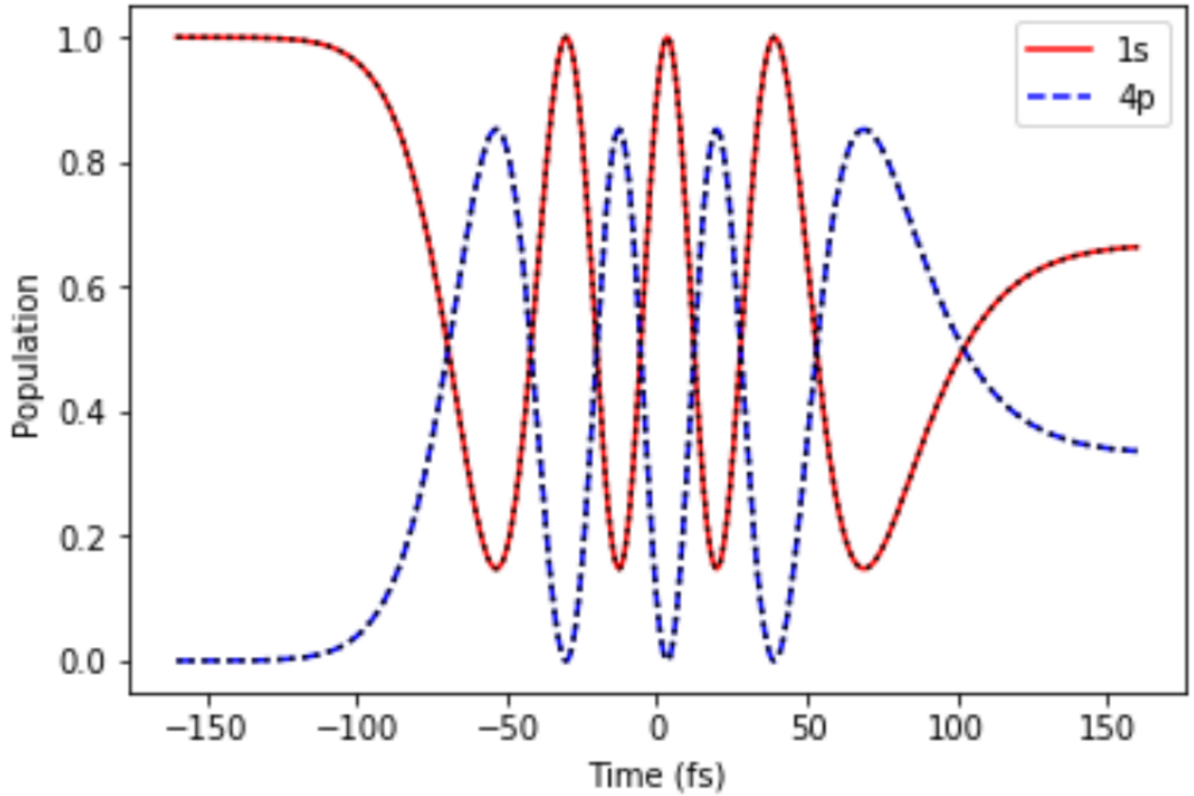
$$\ldots = \sum_{n:\,\text{even}} \frac{1}{n!}(-i)^n \Theta^n \,\mathbb{1} + \sum_{n:\,\text{odd}} \frac{1}{n!}(-i)^n \Theta^n \,\sigma_x$$

$$\ldots = \underbrace{\cos\Theta \;\mathbb{1}}_{\text{remain in same initial state}} \; - \; \underbrace{i\sin\Theta \;\sigma_x}_{\text{flip to the other state}} \,, \qquad \underbrace{\Theta = \int_{t_i}^{t} dt' \frac{\Omega(t')}{2}}_{\text{pulse "area" determines the amount of Rabi oscillations.}}$$

HOMEWORK:

Find the analytical time-evolution operator to the "detuned area theorem".

HINT: Assume that the detuning has the same time-dependence as the E-field envelope.



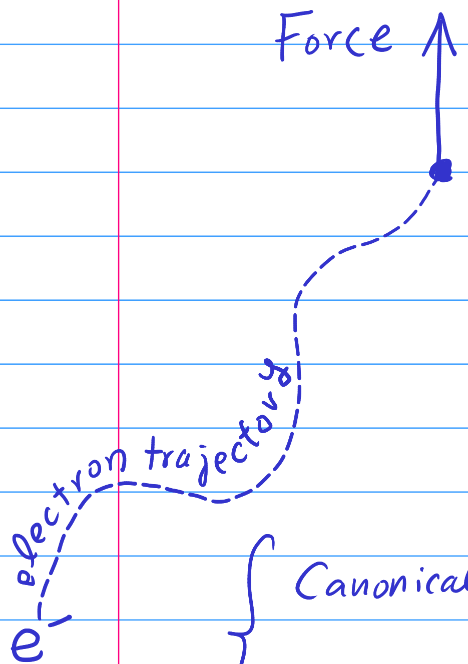
← Rabi oscillations driven at the time-dependent generalized Rabi frequency.

$W(t) = \Lambda(t)\,W_0$

There is no known general solution for any detuning.

"SIMPLE" PROBLEM 3 : VOLKOV WAVES

Free electron in a laser field



Consider minimal coupling Hamiltonian within the dipole approximation

$\Longrightarrow$ VELOCITY GAUGE

$$H_V^{\nu}(t) = H(t) = \frac{1}{2}\Big[\underbrace{\hat{p} + A(t)}\Big]^2$$

Kinetic momentum $\hat{\pi}(t) = \hat{p} + A(t)$

$$\begin{cases} \text{Canonical momentum: } & \hat{p} = -i\nabla \\ \text{Electric field: } & E(t) = -\dfrac{dA}{dt} \end{cases}$$

Vector potential

The Volkov Hamiltonian is a CASE II Hamiltonian.

- It is time dependent $H(t)$ due to $\mathbb{A}(t)$.
- It commutes at different times.

"Proof":

$$[H(t), H(t')] = H(t)H(t') - H(t')H(t) = \dots$$

$$\dots = \left(\frac{1}{2}\right)^2 \left((\hat{\mathbb{p}} + \mathbb{A}(t))^2 (\hat{\mathbb{p}} + \mathbb{A}(t'))^2 - (\hat{\mathbb{p}} + \mathbb{A}(t'))^2 (\hat{\mathbb{p}} + \mathbb{A}(t)) \right) = 0$$

due to the vector potential being only a function of time (not space) within the dipole approximation.

The Volkov propagator in velocity gauge:

CASE II:

$$U(t,t_i) = e^{-i\int_{t_i}^{t} dt' H(t')}$$

$$= \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} \left(\int_{t_i}^{t} dt' H(t') \right)^n$$

$$= \sum_{n=0}^{\infty} \frac{(-i)^n}{n!} \left(\frac{1}{2} \int dt' \left[\hat{\mathbb{P}} + \mathbb{A}(t') \right]^2 \right)^n \quad , \begin{cases} \hat{\mathbb{P}} = -i \nabla \\ \mathbb{A}(t) \in \mathbb{R}^3 \end{cases}$$

Plane waves:

Real vector $\mathbb{k} \in \mathbb{R}^3$

$$\hat{p}\,|\mathbb{k}\rangle = \mathbb{k}\,|\mathbb{k}\rangle \quad , \quad \langle \mathbb{r}|\mathbb{k}\rangle = \frac{1}{(2\pi)^{3/2}} e^{i\mathbb{k}\cdot\mathbb{r}}$$

Since we use velocity gauge we should use a complete basis:

$$\boxed{\mathbb{1} = \int d^3k \; |\mathbb{k}\rangle\langle\mathbb{k}| \; ,}$$

which is full filled by the plane wave basis.

Insert identities (best trick ever!)

$$U(t,t_i) = \mathbb{1}\, U(t,t_i)\, \mathbb{1} = \ldots$$

Eigenstate:

$\hat{\mathbb{p}}\,|\mathbb{k}\rangle = \mathbb{k}\,|\mathbb{k}\rangle$

real vector

$$\ldots = \int d^3k''\, |\mathbb{k}''\rangle\langle\mathbb{k}''| \left(\sum_{n=0}^{\infty} \frac{(-i/2)^n}{n!} \int_{t_i}^{t} dt' \left[\hat{\mathbb{p}} + \mathbb{A}(t')\right]^2 \right)^n \int d^3k'\, |\mathbb{k}'\rangle\langle\mathbb{k}'|$$

Insert identities (best trick ever!)

$$U(t,t_i) = \mathbb{1}\, U(t,t_i)\, \mathbb{1} = \dots$$

Eigenstate:

$\hat{\mathbb{p}}|\mathbb{k}\rangle = \mathbb{k}|\mathbb{k}\rangle$

real vector

$$\dots = \int d^3k'' \, |\mathbb{k}''\rangle\langle\mathbb{k}''| \left(\sum_{n=0}^{\infty} \frac{(-i/2)^n}{n!} \int_{t_i}^{t} dt' \left[\hat{\mathbb{p}} + \mathbb{A}(t') \right]^2 \right)^n \int d^3k' \, |\mathbb{k}'\rangle\langle\mathbb{k}'|$$

Orthogonality:

$\langle\mathbb{k}''|\mathbb{k}'\rangle = \delta(\mathbb{k}''-\mathbb{k}')$

$$\dots = \int d^3k'' \, |\mathbb{k}''\rangle\langle\mathbb{k}''| \left(\sum_{n=0}^{\infty} \frac{(-i/2)^n}{n!} \int_{t_i}^{t} dt' \underbrace{\left[\mathbb{k}' + \mathbb{A}(t') \right]^2}_{\text{scalar}} \right)^n \int d^3k' \, |\mathbb{k}'\rangle\langle\mathbb{k}'|$$

Insert identities (best trick ever!)

$$U(t,t_i) = \mathbb{1}\, U(t,t_i)\, \mathbb{1} = \ldots$$

Eigenstate:
$\hat{\mathbb{p}}|\mathbb{k}\rangle = \mathbb{k}|\mathbb{k}\rangle$
real vector

$$\ldots = \int d^3k'' \, |\mathbb{k}''\rangle\langle\mathbb{k}''| \left(\sum_{n=0}^{\infty} \frac{(-i/2)^n}{n!} \int_{t_i}^{t} dt' \left[\hat{\mathbb{p}} + \mathbb{A}(t')\right]^2 \right)^n \int d^3k' \, |\mathbb{k}'\rangle\langle\mathbb{k}'|$$

Orthogonality:
$\langle\mathbb{k}''|\mathbb{k}'\rangle = \delta(\mathbb{k}''-\mathbb{k}')$

$$\ldots = \int d^3k'' \, |\mathbb{k}''\rangle\langle\mathbb{k}''| \left(\sum_{n=0}^{\infty} \frac{(-i/2)^n}{n!} \int_{t_i}^{t} dt' \underbrace{\left[\mathbb{k}' + \mathbb{A}(t')\right]^2}_{\text{scalar}} \right)^n \int d^3k' \, |\mathbb{k}'\rangle\langle\mathbb{k}'|$$

$$\ldots = \int d^3k \, |\mathbb{k}\rangle\langle\mathbb{k}| \, e^{-\frac{i}{2}\int_{t_i}^{t} dt' \left[\mathbb{k} + \mathbb{A}(t)\right]^2}$$

Diagonal in canonical momentum

Kinetic momentum of electron in the field $\boxed{\pi(t) = \mathbb{k} + \mathbb{A}(t).}$

How to derive:

Strong-field approximation (SFA):

- Use DYSON expansion

Laser is perturbation

- First consider the laser as a perturbation : $H = H_A + V_A$

$$U(t,t_i) = U_A(t,t_i) - i\int_{t_i}^{t} dt'\, U(t,t')\, V_A(t')\, U_A(t',t_i)$$

How to derive:

Strong-field approximation (SFA):

- Use DYSON expansion

Laser is perturbation

- First consider the laser as a perturbation : $H = H_A + V_A$

$$U(t,t_i) = U_A(t,t_i) - i\int_{t_i}^{t} dt'\, U(t,t')\, V_A(t')\, U_A(t',t_i)$$

change "perturbations".

Atomic potential is perturbation

- Second consider the atomic potential as a perturbation : $H = H_V + V_V$

$$U(t,t_i) = U_A(t,t_i) - i\int_{t_i}^{t} dt'\, U_V(t,t')\, V_A(t')\, U_A(t',t_i)$$

$$\ldots + (-i)^2 \int_{t_i}^{t} dt'' \int_{t_i}^{t''} dt'\, U(t,t'')\, V_V(t'')\, U_V(t'',t')\, V_A(t')\, U_A(t',t_i)$$

Compare with derivation by: Lewenstein et al. PRA 49, 2117 (1994)

How to derive:

Strong-field approximation (SFA):

- Use DYSON expansion

Laser is perturbation

- First consider the laser as a perturbation : $H = H_A + V_A$

$$U(t,t_i) = U_A(t,t_i) - i\int_{t_i}^{t} dt'\, U(t,t')\, V_A(t')\, U_A(t',t_i)$$

change "perturbations".

Atomic potential is perturbation

- Second consider the atomic potential as a perturbation : $H = H_V + V_V$

$$U(t,t_i) = U_A(t,t_i) - i\int_{t_i}^{t} dt'\, U_V(t,t')\, V_A(t')\, U_A(t',t_i)$$

$$\ldots \cancel{+ (-i)^2 \int_{t_i}^{t} dt'' \int_{t_i}^{t''} dt'\, U(t,t'')\, V_V(t'')\, U_V(t'',t')\, V_A(t')\, U_A(t',t_i)}$$

Compare with derivation by: Lewenstein et al. PRA 49, 2117 (1994)

Modification of XUV photoionization by IR-dressed continuum.

Shifted photoelectron peak with dynamical interference (dressed continuum)

Single photoelectron peak (usual Einstein's photoelectric effect)

Ramsey interference when different envelopes are used.

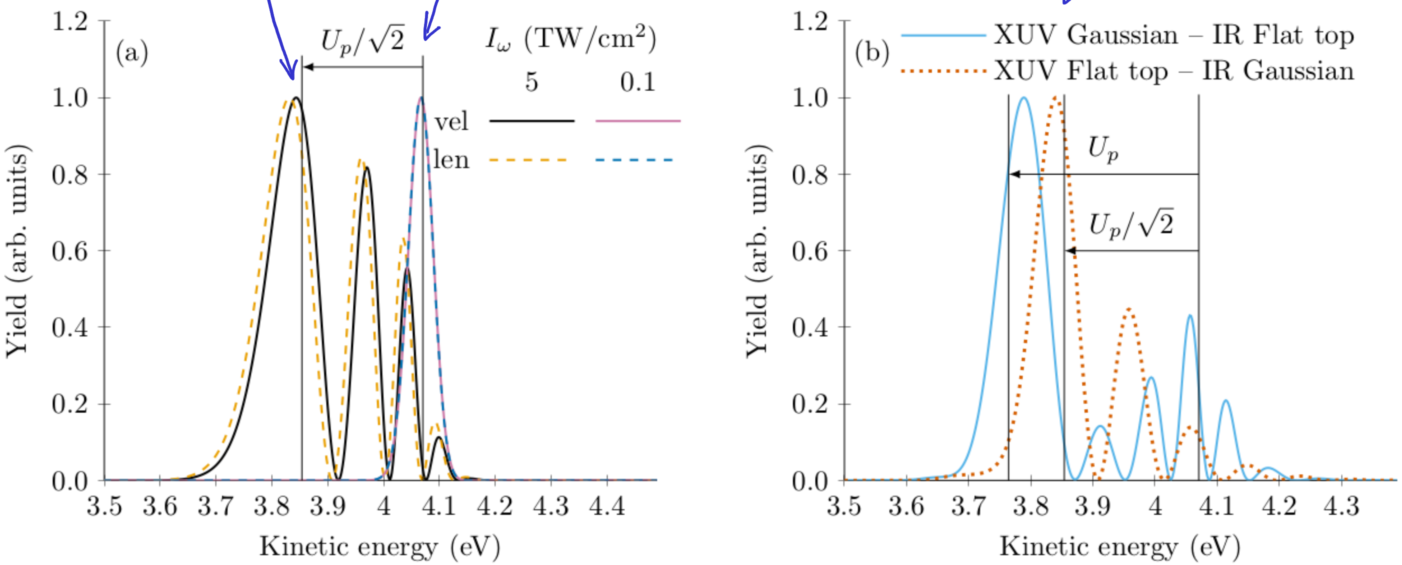


FIG. 5. Numerical demonstration of laser-assisted dynamical interference. (a) Single XUV pulse and IR pulse using truncated Gaussian envelopes. (b) Mixing flat-top and truncated Gaussian envelopes of the XUV and IR fields with $I_\omega = 5 \times 10^{12}$ W/cm^2. The expected classical energy shift is shown with an arrow and labeled U_p and $U_p/\sqrt{2}$ for a flat-top envelope and a Gaussian envelope, respectively.

Bertolino et al. Phys. Rev. A 106, 043108 (2022)

How to obtain LENGTH gauge Volkov states?

Electric field

Position operator

$$H_V^\ell(t) = H(t) = \frac{1}{2}\hat{\mathbb{p}}^2 + V(r) + \mathbb{E}(t)\cdot\mathbb{r}$$

PROBLEM : $H_V^\ell(t)$ is a CASE III Hamiltonian.

GOOD NEWS : We already know the propagator in velocity gauge.

$\rightarrow$ Gauge transformation.

$$\boxed{|\Psi^\ell(t)\rangle = e^{-i\chi}|\Psi^v(t)\rangle}$$

GAUGE TRANSFORMATIONS:

$$H(\mathbf{r},t) = \frac{1}{2}[\mathbf{p} + \mathbf{A}]^2 - A_0 + V(r)$$

vector potential

scalar potential

other potential

electron $q=-1$

FIRST KIND:

$$\begin{cases} \mathbf{A} \rightarrow \mathbf{A} + \nabla\chi \\ A_0 \rightarrow A_0 - \dfrac{\partial\chi}{\partial t} \end{cases}$$

SECOND KIND:

$$\begin{cases} \Psi(\mathbf{r},t) \rightarrow \Psi(\mathbf{r},t)\, e^{-i\chi(\mathbf{r},t)} \\ H \rightarrow e^{-i\chi} H e^{i\chi} + \dfrac{\partial\chi}{\partial t} \end{cases}$$

Recall unitary transformation

Transformation from velocity to length:

$$\chi = \chi_{\ell \leftarrow v} = -A(0,t)\cdot\mathbf{r} \longrightarrow \boxed{\Psi_\ell = \Psi_v\, e^{+iA(t)\cdot\mathbf{r}}}$$

(another minus.)

Details about the gauge transformation:

$$H^{\ell} = e^{-i\chi} H^{\nu} e^{+i\chi} + \left(\frac{\partial \chi}{\partial t}\right), \quad \text{where} \quad \chi = -\mathbb{A}(t)\cdot \mathbb{r} \Rightarrow \left(\frac{\partial \chi}{\partial t}\right) = -\left(\frac{\partial \mathbb{A}}{\partial t}\right)\cdot \mathbb{r} = \mathbb{E}\cdot \mathbb{r}$$

$$H^{\ell} = e^{i\mathbb{A}\cdot\mathbb{r}} \Big[\underbrace{\frac{\hat{\mathbb{P}}^2}{2}}_{1} + \underbrace{\mathbb{A}\cdot\mathbb{p}}_{2} + \underbrace{\frac{\mathbb{A}(t)^2}{2}}_{3} + V(r) \Big] e^{-i\mathbb{A}\cdot\mathbb{r}} + \mathbb{E}\cdot\mathbb{r}$$

Apply on test function:

$$\frac{\hat{\mathbb{p}}^2}{2}\left(e^{-i\mathbb{A}\cdot\mathbb{r}} f\right) = \frac{(-i)^2}{2} \nabla^2 \left(e^{-i\mathbb{A}\cdot\mathbb{r}} f\right) = -\frac{1}{2}\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}\right)\left(e^{-i\mathbb{A}\cdot\mathbb{r}} f\right)$$

$$= -\frac{1}{2}\frac{\partial}{\partial x}\left(-iA_x e^{-i\mathbb{A}\cdot\mathbb{r}} f + e^{-i\mathbb{A}\cdot\mathbb{r}} \frac{\partial f}{\partial x}\right) + \dots$$

$$= -\frac{1}{2}\Big(\overbrace{(-i)}^{-1} A_x^2 e^{-i\mathbb{A}\cdot\mathbb{r}} f - iA_x e^{-i\mathbb{A}\cdot\mathbb{r}} \frac{\partial f}{\partial x} - iA_x e^{-i\mathbb{A}\cdot\mathbb{r}} \frac{\partial f}{\partial x} + e^{-i\mathbb{A}\cdot\mathbb{r}} \frac{\partial^2 f}{\partial x^2} + \dots$$

$$= \frac{1}{2}\Big(\underbrace{\mathbb{A}^2 f}_{1'} + \underbrace{2i(\mathbb{A}\cdot\nabla f)}_{1''} - \underbrace{\nabla^2 f}_{1'''}\Big) e^{-i\mathbb{A}\cdot\mathbb{r}}$$

$$\mathbb{A}\cdot\mathbb{p}\, e^{-i\mathbb{A}\cdot\mathbb{r}} f = -i\mathbb{A}\cdot\nabla\left(e^{-i\mathbb{A}\cdot\mathbb{r}} f\right) = -i\mathbb{A}\cdot\left(-i\mathbb{A} e^{-i\mathbb{A}\cdot\mathbb{r}} f + e^{-i\mathbb{A}\cdot\mathbb{r}} \nabla f\right) = \dots$$

$$\dots = \Big(\underbrace{-\mathbb{A}^2 f}_{\text{Cancels with 3 and } 1'} \underbrace{- i\mathbb{A}\cdot\nabla f}_{\text{Cancels with } 1''}\Big) e^{-i\mathbb{A}\cdot\mathbb{r}} \Rightarrow \text{Only } 1''' \text{ remains. (pew!)}$$

$$\boxed{H^{\ell} = \frac{\hat{\mathbb{P}}^2}{2} + \mathbb{E}(t)\cdot\mathbb{r} + V(r)}$$

LENGTH GAUGE VOLKOV PROPAGATOR:

$$U_V^{\ell}(t,t_i) = e^{i\mathbb{A}(t)\cdot\mathbb{r}}\, U_V^{v}(t,t_i)\, e^{-i\mathbb{A}(t_i)\cdot\mathbb{r}} = \ldots$$

$$\ldots = e^{i\mathbb{A}(t)\cdot\mathbb{r}} \left(\int d^3k \; |\mathbb{k}\rangle \; e^{-\frac{i}{2}\int_{t_i}^{t} d\tau\,[\mathbb{k}+\mathbb{A}(\tau)]^2} \; \langle\mathbb{k}| \right) e^{-i\mathbb{A}(t_i)\cdot\mathbb{r}} = \ldots$$

$$\ldots = \int d^3k \; e^{-\frac{i}{2}\int_{t_i}^{t} d\tau\,[\mathbb{k}+\mathbb{A}(\tau)]^2} \; |\mathbb{k}+\mathbb{A}(t)\rangle \langle \mathbb{k}+\mathbb{A}(t_i)|$$

Instantaneous kinetic energy

Kinetic momentum out

Kinetic momentum in

NOTE: Not diagonal in kinetic momentum for general times during the laser pulse.

Conclusions:

- Simple problems with analytical propagators:
  - Two-level atom in laser field (Rabi model)
  - Free electron in laser field (Volkov states)
- Transformations were used to find propagators using standard exponential solutions: CASES I, II, III.
- Application of Dyson expansions with "different" perturbations is the basis for the strong-field approximation and other problems involving few-level models.

THANK YOU FOR YOUR ATTENTION!

(Also thanks to the Wallenberg Foundation, the Olle Engkvist Foundation and the Swedish Research Council.)