Non-Hermitian Floquet Theory

Application to Dynamical Interference

Hao Liang

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School of Physics, Peking University
Beijing, 100871 China
Email: haoliang@pku.edu.cn

What is Strong Field Physics?

Photoelectric effect: we learned from textbook

- Light with frequency below the threshold is unable to ionize electron
- Kinetic energy of photoelectron is independent of light intensity
- Electron doesn't require time to absorb energy
- Free electron cannot absorb a photon

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- Multi-photon ionization
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- Ac-Stark shift/U_p shift
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- Ionization time-delay $\sim 10^{-17}$ s
- Free electron cannot absorb a photon
- Above threshold ionization

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Ac-Stark Shift

For an atomic or a molecular system, the single-electron Hamiltionian in laser field (with dipole approximation) is

$$\begin{aligned} H &= \frac{1}{2}[\boldsymbol{p} + \boldsymbol{A}(t)]^2 + V(\boldsymbol{r}) \\ &= \underbrace{\frac{1}{2}\boldsymbol{p}^2 + V(\boldsymbol{r})}_{H_0} + \underbrace{\boldsymbol{p} \cdot \boldsymbol{A}(t)}_{V_I(t)} \underbrace{\cancel{+ \frac{1}{2}\boldsymbol{A}^2(t)}}_{\text{gauge free}} . \end{aligned}$$

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(At least far from atom,) plane wave is still eigenstate of the system with eigen energy unchange

$$\psi_{\text{Volkov}} = \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r} - \mathrm{i}E_k t)\exp(-\mathrm{i}\boldsymbol{k}\cdot\int \boldsymbol{A}(t)\,\mathrm{d}t),$$

But “average energy” of ground state changes from E_0 to $E_0 + \delta$.

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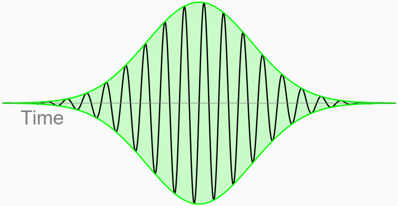
But “average energy” of ground state changes from E_0 to $E_0 + \delta$.

Thus, for n-photo ionization, the energy conservation law changes

$$E_k = E_0 + n\omega \quad \rightarrow \quad E_k = E_0 + \delta + n\omega$$

Dynamical Interference

In practical, we use laser pulse instead of monochromatic field.



Demekhin, P. V., *et al. Phys. Rev. Lett.*, **108**, 253001 (2012)

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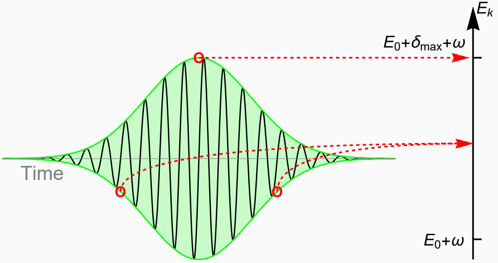

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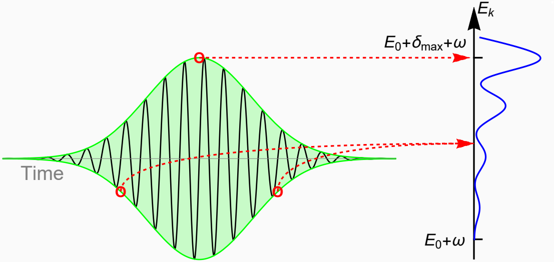

- E_k move forward and backward as instantaneous intensity change.
- Time domain double-slit in the rising and falling part.

Demekhin, P. V., *et al. Phys. Rev. Lett.*, **108**, 253001 (2012)

Essential Condition

Ground state amplitude

$$a_0(t) \equiv \langle 0|\Psi(t)\rangle \approx \exp\left\{-\mathrm{i}E_0 t - \mathrm{i}\int^t \delta[I(\tau)]\,\mathrm{d}\tau - \int^t \frac{\gamma[I(\tau)]}{2}\,\mathrm{d}\tau\right\}$$

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PMD according to modified perturbation theory

$$A(\boldsymbol{k}) = \left|\int \left\langle \boldsymbol{k} \,\middle|\, \mathrm{e}^{\mathrm{i}E_k t}\boldsymbol{p}\cdot\boldsymbol{A}_0(t)\mathrm{e}^{-\mathrm{i}\omega t}a_0(t) \,\middle|\, 0\right\rangle \,\mathrm{d}t\right|^2$$

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For $E_k = E_0 + \omega + \delta[I(t_1)] = E_0 + \omega + \delta[I(t_2)]$, we have

$$A \propto |a_0(t_1) + \mathrm{e}^{\mathrm{i}(E_k-\omega)(t_2-t_1)}a_0(t_2)|^2$$

$$\propto \left|1 + \exp\left\{\mathrm{i}\int_{t_1}^{t_2}(\delta[I(\tau)] - \delta_0)\,\mathrm{d}\tau - \int_{t_1}^{t_2}\frac{\gamma[I(\tau)]}{2}\,\mathrm{d}\tau\right\}\right|^2$$

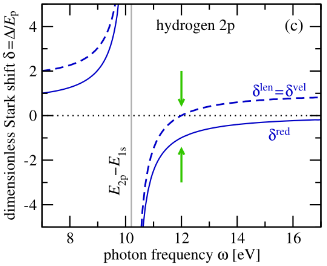
It requires $\delta > \sqrt{\pi}\gamma$ for linear-response region.

Baghery, M.,*et al. Phys. Rev. Lett.*, **118**, 143202 (2017)

Possible Solutions

Increase δ
Near-resonance ionization from excited state

$$\delta \sim \sum_i \frac{|d_{i0}|^2}{E_i - E_0 \pm \omega}$$



Phys. Rev. Lett., **118**, 143202 (2017)

Decrease γ

- Multi-photon ionization:
  $\gamma \propto I^n$ where $\delta \propto I$.

  Phys. Rev. A, **93**, 023419 (2016)
- Stabilization at high intensity:
  $\gamma \to 0$ when $I \gtrsim 10^{18}$ W/cm^2.

  OE **26**, 019921 (2018)
- Ionization suppression for diatomic molecule: would be discussed latter.

Question: How to compute δ and γ?

Floquet Theory

In the presence of a monochromatic field, the Hamiltonian of the system is expressed as

$$H(t) = H_0 + \frac{1}{2}(V_l^\dagger e^{i\omega t} + V_l e^{-i\omega t}),$$

one can remove the time-dependence by introducing photon field

$$H = H_0 \otimes I + \frac{1}{2}(V_l^\dagger \otimes a + V_l \otimes a^\dagger) + \hbar\omega\mathcal{N},$$

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- Decay indicates the imaginary part of energy $E = E_0 + \delta - i\gamma/2$.

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Quasi-energy

- Decay indicates the imaginary part of energy $E = E_0 + \delta - i\gamma/2$.
- $\mathrm{Re}\, i\sqrt{2E_k}r > 0$ for $\mathrm{Re}\, E_k > 0$ and $\mathrm{Im}\, E_k < 0$, non-square integrable.

Complex Eigenvalues

H_0 usually contains several discrete levels and a continue spectra

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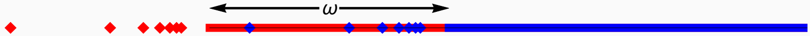
In the $V_0 \to 0$ limit, spectra for different photon number can be put together (only show for n and $n-1$ photon number state for clear)



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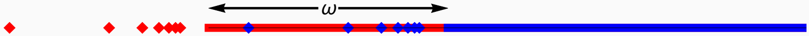
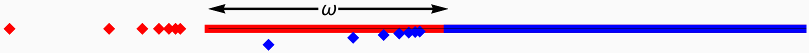


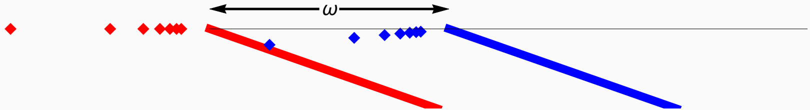
With non-zero interaction, due to the 'level repulsion', the discrete states have to be complex



$$E_i' = E_i + \delta - \mathrm{i}\gamma/2$$

Balslev-Combes theorem

Upon the transformation $\boldsymbol{r} \to \boldsymbol{r}e^{i\alpha}$, the discrete spectra of H will not change while its continue spectra rotate in complex plane with angle -2α.



Simon, B., *Ann. Math.*, **97**, 247 (1973)

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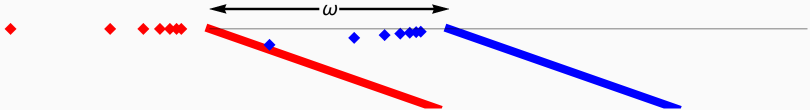
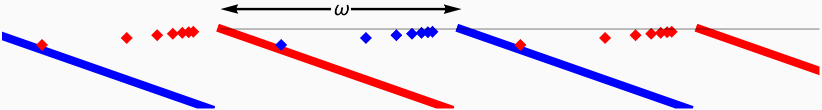


After considering all photon number states, we have these structures



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Arnoldi Propagator for Finding Eigenvalues

Since the spectra is periodic in energy domain with period of ω, we can remove such complexity by investigating the spectra of

$$U(2\pi/\omega) \equiv \exp(-2\pi \mathrm{i} H/\omega),$$

which is equivalent to original propagator

$$\mathcal{T} \exp\left[-\mathrm{i}\int_0^{2\pi/\omega} H(s)\,\mathrm{d}s\right],$$

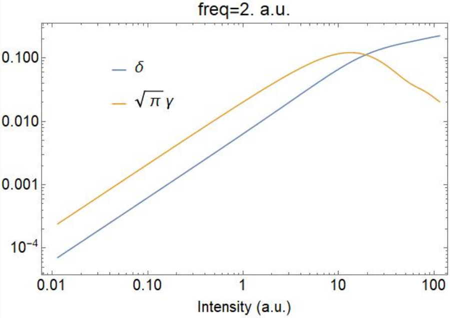
and can be solved with numerical method for usual time-dependent Shrödinger equation.

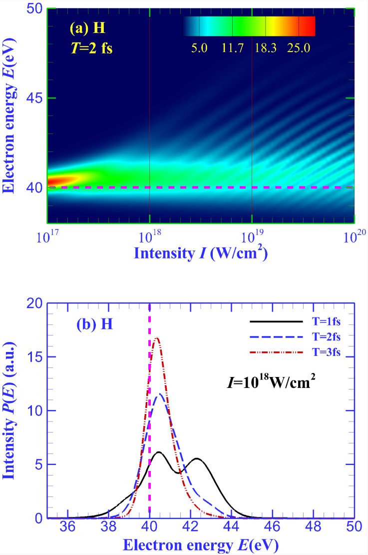
Thus one can diagonalize U in Krylov subspace $\mathrm{span}\{\psi, U\psi, \ldots, U^n\psi\}$.

Telnov, D. A. and Chu, S.-I. *J. Phys. B: At. Mol. Opt. Phys.* **37**, 1489 (2004).

Results for H Atom

For $\omega = 2\,\text{a.u.}$, $\delta > \sqrt{\pi}\gamma$ happens when $I \gtrsim 20\,\text{a.u.}$, which coincide with previous calculation on H atom.

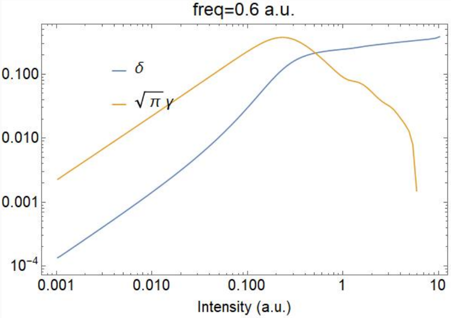


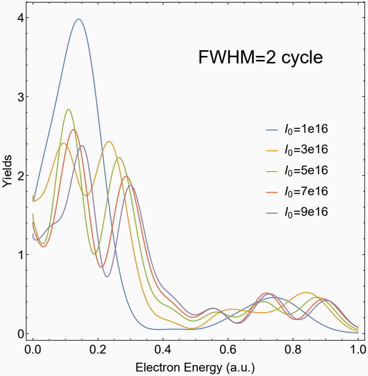


Jiang, W. C., *et al.* OE **26**, 019921 (2018)

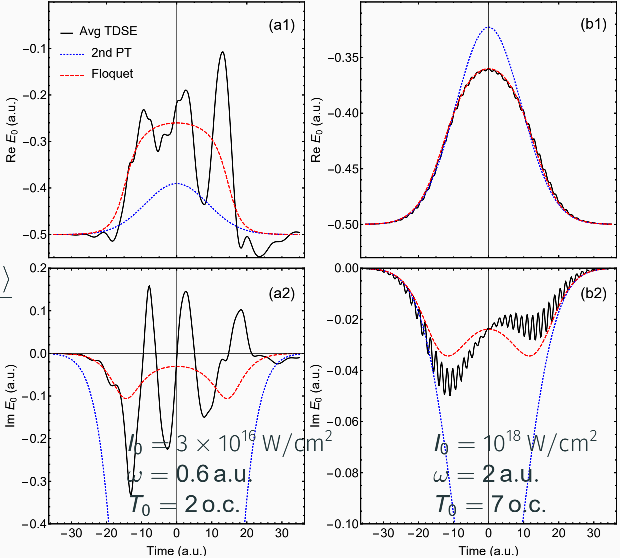
Results for H Atom - Cont.

For $\omega = 0.6\,\text{a.u.}$, stabilization (thus dynamics interference) happens at a much low intensity





$$E = \mathrm{i}\frac{\mathrm{d}\log\langle 0|\Psi(t)\rangle}{\mathrm{d}t}$$



$I_0 = 3 \times 10^{16}$ W/cm^2
$\omega = 0.6$ a.u.
$T_0 = 2$ o.c.

$I_0 = 10^{18}$ W/cm^2
$\omega = 2$ a.u.
$T_0 = 7$ o.c.

Why does atom stabilize in ultrahigh intensity laser field?

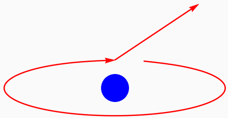
- Laser field is dominant than Coulomb potential.
- Electron oscillates following the vector potential

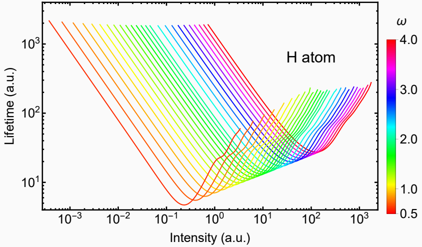
$$\boldsymbol{p} + \boldsymbol{A}(t) = \text{cosnt.}$$

- Ionization comes from Rutherford scattering

$$\sigma \propto 1/E^2$$

- Such mechanism is valid when $A_0^2/2 \sim E_0 + \omega$.

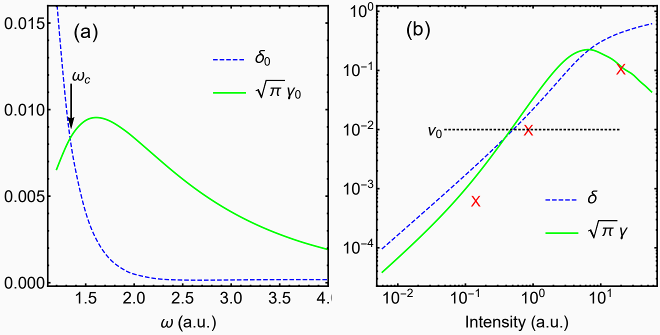




Pont, M. and Shakeshaft, R. *Phys. Rev. A* **44**, R4110 (1991).

Results for H_2^+ - Fixed nuclear

For fixed-nuclear H_2^+ at frequency close to threshold, $\delta > \sqrt{\pi}\gamma$ can be achieved without stabilization.



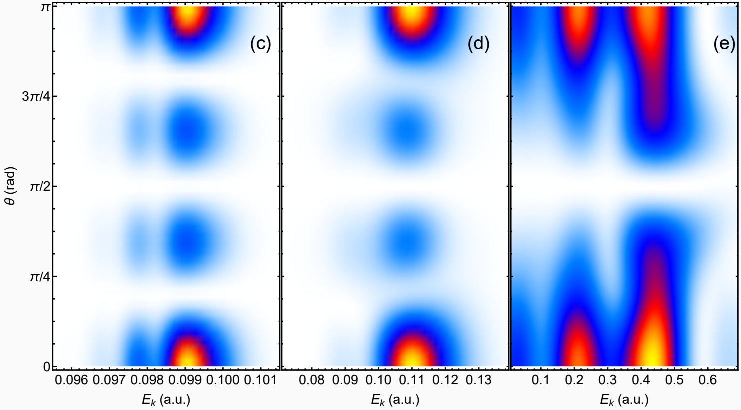
Results for H_2^+ - Fixed nuclear

Strong distortion in angular distribution at high intensity

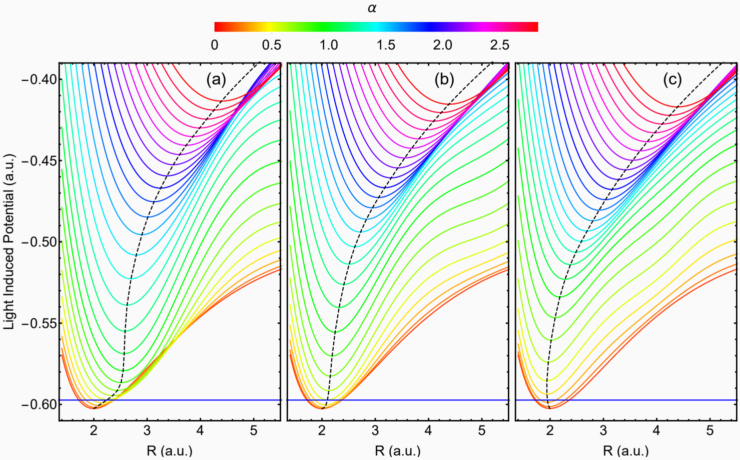
$I_0 = 5 \times 10^{15}\,\mathrm{W/cm^2}$ $T_0 = 256\,\mathrm{o.c.}$

$I_0 = 3 \times 10^{16}\,\mathrm{W/cm^2}$ $T_0 = 32\,\mathrm{o.c.}$

$I_0 = 7 \times 10^{17}\,\mathrm{W/cm^2}$ $T_0 = 3\,\mathrm{o.c.}$



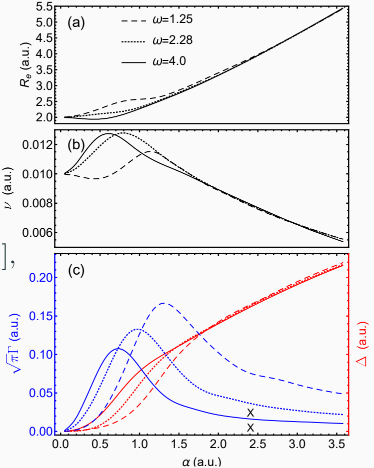
Potential Energy Curves



- Information of field-dressed vibrational state under BO approx. can be obtained by diagonalizing

$$H_{\text{BO}} = -\frac{1}{2M}\frac{\partial^2}{\partial R^2} + \frac{1}{R} + E_0[R; I(t), \omega],$$

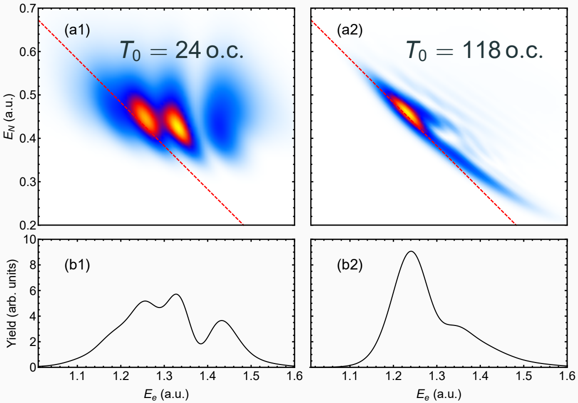
- Effects of the nuclear motion break down the interference condition in perturbation region.



Results for H_2^+ - BO Approx.

Solve nuclear motion on PEC to get Joint energy distribution

$$P_{\mathrm{ad}}(E_N, E_e) = \left| \int_{t_s}^{t_e} \mathrm{d}t\, e^{\mathrm{i}(E_N + E_e - \omega)t} \int_0^{\infty} \Phi_f^*(R; E_N) \sqrt{\frac{\gamma[R; I(t)]}{2\pi}}\, \Phi(R, t)\, \mathrm{d}R \right|^2,$$



Stretch R to a large value adiabatically is impossible since $\Gamma > \nu$.

Conclusion and Perspectives

- Developed a non-Hermitian Floquet program for evaluation of the ac-stark shift and decay rate of H atom and H_2^+.
- Identified the parameter region of dynamical interference.
- Electron-nuclear correlation is discussed under the framework of BO approx.
- Manuscript submitted to *Phys. Rev. A.*

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Thanks for attention!

High Frequency Floquet Theory

Under the gauge transformation

$$\Psi \to \exp\left[\mathrm{i}\boldsymbol{p} \cdot \int \boldsymbol{A}(t)\,\mathrm{d}t\right]\Psi,$$

we have Kramers-Henneberger Hamiltonian

$$H_{\mathrm{KH}}(t) = \frac{1}{2}\boldsymbol{p}^2 + U\left[\boldsymbol{r} + \int \boldsymbol{A}(t)\,\mathrm{d}t\right].$$

Expand $\boldsymbol{U}(\boldsymbol{r}, \boldsymbol{t})$ into Fourier series and treat high frequency terms as perturbation, we notice that energy shift in this scheme is only dependent on amplitude of $\int \boldsymbol{A}(t)\,\mathrm{d}t$, i.e., $\boldsymbol{\alpha}$.