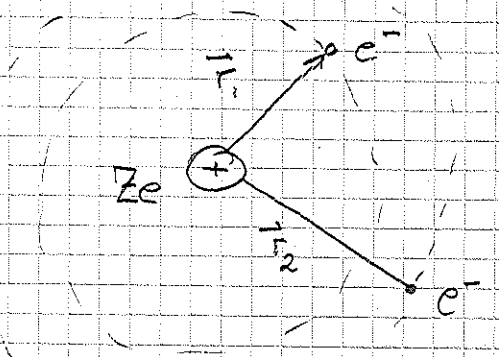
E.g.: negative interference of resonance states with comparable widths

cf. Tikhonova, Fedorov, Laser Physics 7, 574 (1997).

B. Two-electron atoms – basic electronic structure



$$H = H_0 + H_1$$

$$H_0 = \frac{p_1^2}{2} + \frac{p_2^2}{2} + V(r_1) + V(r_2)$$:

approximate, separable, includes "averaged" electronic correlation through central potential V

$$r_{12} = |\vec{r}_2 - \vec{r}_1| \qquad H_1 = -\frac{Z}{r_1} - V(r_1) - \frac{Z}{r_2} - V(r_2) + \frac{1}{r_{12}}$$

Neglect spin-dependent interactions (no relativistic) effects : good for low Z

$$\underset{\text{total}}{\psi}(\vec{r}_1, \vec{r}_2) = \psi(\vec{r}_1, \vec{r}_2) \cdot \chi(1,2)$$

spatial wf spin wf

$$\psi(\vec{r}_1, \vec{r}_2) = \pm \psi_\pm(\vec{r}_2, \vec{r}_1) \qquad \begin{cases} \chi(1,2) = -\chi(2,1) \\ \chi(1,2) = +\chi(2,1) \end{cases}$$

(Antisymmetrization postulate)

Ground state : $\vec{S} = \vec{s}_1 + \vec{s}_2$ (net spin)

$$S = \begin{cases} 0 & \text{singlet : "para He"} \\ 1 & \text{triplet : "ortho He"} \end{cases}$$

- Eigenstates of H_0 (optimize V to make H_1 als unimportant as possible) :

$$H_0 \psi_\pm^{(0)} = E^{(0)} \psi_\pm$$

$$\Rightarrow \quad \psi_{\pm}^{(0)} = \frac{1}{\sqrt{2}} \left\{ \varphi_{n_1 \ell_1 m_1}(\vec{r}_1)\, \varphi_{n_2 \ell_2 m_2}(\vec{r}_2) \pm \varphi_{n_1 \ell_1 m_1}(\vec{r}_2)\, \varphi_{n_2 \ell_2 m_2}(\vec{r}_1) \right\}$$

"configuration"

$$E^{(0)} = \varepsilon_{n_1 \ell_1} + \varepsilon_{n_2 \ell_2}$$

(φ, ε) : "(orbitals, orbital energies)"

Good quantum numbers : $n_1, \ell_1, m_1, n_2, \ell_2, m_2, L, \Pi_L, S, \Pi_S, \Pi$

$\vec{L} = \vec{\ell}_1 + \vec{\ell}_2$; $\Pi_{L,S}$: magn. quant. nos

$\Pi := (-1)^{\ell_1 + \ell_2}$: parity

- Include H_1 (non separable)

$\Rightarrow$ configuration interaction

$$|\alpha\rangle := \psi_{\pm}^{(0)}(n_1, \ell_1, m_1, n_2, \ell_2, m_2)$$

expand

$$|\psi\rangle = \sum_{\alpha} C_{\alpha} |\alpha\rangle$$

to solve

$$H |\psi\rangle = E |\psi\rangle$$

by projections on $\{\langle \alpha' |\}$.

$\Rightarrow$ Coupled equations:

$$(E - \varepsilon_{\alpha'}) C_{\alpha'} = \sum_{\alpha} \langle \alpha' | H_1 | \alpha \rangle C_{\alpha}$$

includes $\langle \alpha' | \frac{1}{r_{12}} | \alpha \rangle$: configuration coupling/mixing"

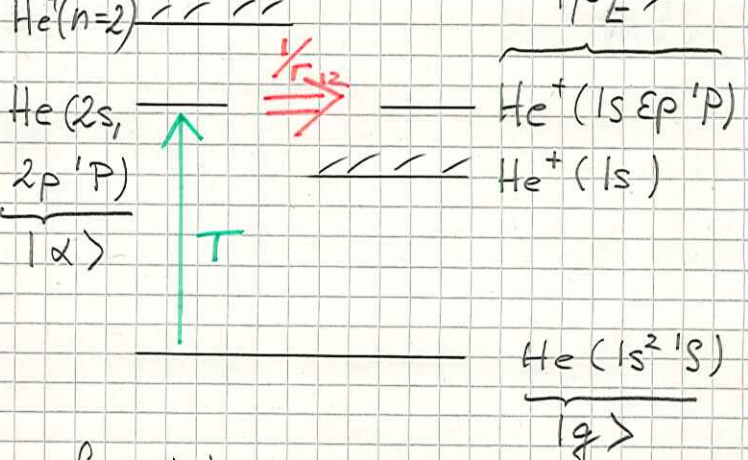
Good quantum numbers : L, Π_L, S, Π_S, Π

- Configurations may include all combinations of bound and continuous (= unbound orbitals).

– Doubly excited states (of He)

are unbound.

Decay by configuration interaction ("auto-ionization").



T: transition operator (e.g., opt. excitation):

$$|g\rangle \xrightarrow{T} \underbrace{a_E|\alpha\rangle + \int dE'\, b_{E'}|\beta_{E'}\rangle}_{|\psi_{E_{res}}\rangle}$$

$H|\psi_{E_{res}}\rangle = E|\psi_{E_{res}}\rangle$, project onto $|\alpha\rangle, \{|\beta_{E'}\rangle\}$,

define $\langle\alpha|H|\alpha\rangle = E_{res}$ (resonance energy)

$\langle\beta_E|H|\alpha\rangle = V_E$ (assumed constant near E_{res}),

use $\langle\beta_{E'}|H|\beta_E\rangle = E\,\delta(E-E')$

and solve coupled equations for $a_E, \{|\beta_{E'}\rangle\}$

$\Rightarrow$ Excitation cross section near $E = E_{res}$:

$$\sigma_{g\to|\psi_{E_{res}}\rangle} = \underbrace{|\langle\beta_{E_{res}}|T|g\rangle|^2}_{\sigma_0} \cdot \frac{(\varepsilon+q)^2}{1+\varepsilon^2}$$

$$\varepsilon := \frac{E-E_{res}}{\Gamma/2}$$

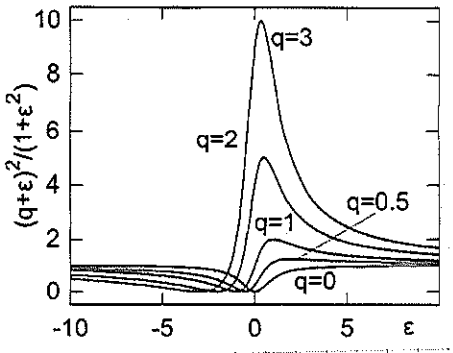
$$q := \frac{\langle\alpha|T|g\rangle}{\pi V_{E_{res}}\langle\beta_{E_{res}}|T|g\rangle}$$

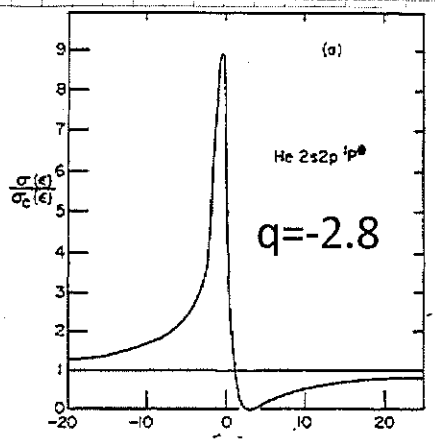
} "Fano resonance parameters"

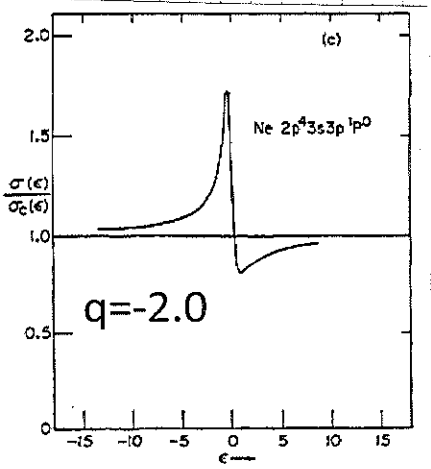
$$\Gamma := 2\pi V_{E_{res}}^2$$

U. Fano, Phys. Rev. A. 124, 1866 (1961)

C.D. Lin, Adv. At. Mol. Phys. 22, 77 (1986)

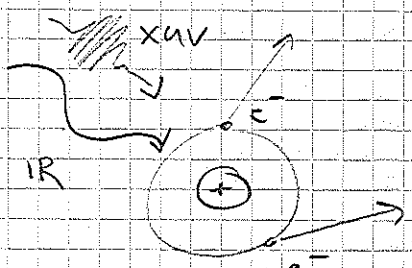






Examples for Fano (Feshbach) shape resonances
(U. Fano & J.W. Cooper, Rev. Mod. Phys. 40, 441 (1968),
≙ C.D. Lin et al. Attosec. & strong-field physics) → handout

Helium interacting with external IR + XUV fields



- single / double excitation / ionization
- excitation & ionization

$$H = H_0 + V_{int}$$

$$H_0 = \frac{p_1^2}{2} + \frac{p_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2}$$

$$V_{int} = \frac{1}{r_{12}} + \left[\vec{E}_{XUV}(t) + \vec{E}_{IR}(t)\right] \cdot (\vec{r}_1 + \vec{r}_2)$$

$$E_a(t) \underset{e.g.}{=} E_{a,0} \cos^2\left(\frac{\pi}{2}\frac{t}{\tau_a}\right) \cos(\omega_a t + \varphi_a)$$

for $|t| < \tau_a$ (0 else)

a = IR / XUV

Solve $H|\psi(t)\rangle = i\frac{\partial}{\partial t}|\psi(t)\rangle$ by expanding in spherical harmonics :

$$\Psi(\vec{r}_1,\vec{r}_2;t) = \sum_{L M} \sum_{\ell_1 \ell_2} \frac{\Psi^{(L M)}_{\ell_1 \ell_2}(r_1,r_2;t)}{r_1 r_2} \mathcal{Y}^{L,M}_{\ell_1 \ell_2}(\Omega_1,\Omega_2)$$

$$\mathcal{Y}^{L,M}_{\ell_1 \ell_2}(\Omega_1,\Omega_2) = \sum_{m_1,m_2} C^{L M}_{\ell_1 m_1 \ell_2 m_2} Y_{\ell_1 m_1}(\Omega_1) Y_{\ell_2 m_2}(\Omega_2)$$

$C^{LM}_{\ell_1 m_1 \ell_2 m_2}$: angular mom. coupling (Clebsch-Gordon) coefficients

$Y_{\ell m}$: spherical harmonics

$\rightsquigarrow$ coupled diff. equations for radial wavefunctions

$$i\frac{\partial}{\partial t} \Psi^{(L,0)}_{\ell_1 \ell_2} = \sum_{L,\ell_1',\ell_2'} \int d\Omega_1 \int d\Omega_2 \ldots \langle \ell_1 \ell_2, L, 0 | H | \ell_1' \ell_2' L', 0 \rangle \Psi^{L' 0}_{\ell_1' \ell_2'}$$

$M=0$ for assumed 'S initial (ground state)

coupled spher. harmonics $\mathcal{Y}^{LM}_{\ell_1 \ell_2}$

Solve coupled equations numerically, by wavefunction propagation on a $r_1 - r_2$ numerical grid, using, e.g., a finite-element discrete-variable representation (FE-DVR) scheme

cf.: A. Liu & U. Thumm, Phys Rev. A 89, 063423 (2014)

Observables

Numerical solution of TDSE $\Rightarrow$ $|\Psi(t)\rangle$

Project onto final state $\langle f|$ at sufficiently late time t_f

$\Rightarrow$ Prob. for transition to $|f\rangle$: $P = |\langle f | \Psi(t_f)\rangle|^2$

B.2 Many-electron atoms

- Direct & exchange interactions (between electrons)

N-electron state (Fock representation)

$$|\psi^N\rangle = |n_1, n_2, \ldots n_e \ldots\rangle$$

n_e : no. of particles in single-particle state $|e\rangle$

- occupation number operator:

$$N = a_e^+ a_e$$

with

$$\left.\begin{aligned} a_\mu^+ a_\nu^+ + a_\nu^+ a_\mu^+ &= 0 \\ a_\mu a_\nu + a_\nu a_\mu &= 0 \\ a_\mu a_\nu^+ + a_\nu a_\mu^+ &= \delta_{\mu\nu} \end{aligned}\right\} \text{Fermi-Dirac statistics}$$

$$\rightarrow a_\mu^+ a_\mu^+ = 0 \quad \text{(Pauli exclusion principle)}$$

$$N_e |\psi^N\rangle = n_e |\psi^N\rangle$$

a_e^+, a_e : create/destroy one particle in $|e\rangle$

Hartree-Fock method

Describe system of interacting fermions as an effective single-particle problem, introducing a mean-field single-particle potential $V^{HF}(\vec{r})$.

$V^{HF}(\vec{r}) \hat{=}$ averaged effect of $(N-1)$ particle & (time-dependent) external fields on any given electron.

- Exact Hamiltonian (coordinate representation, pair interactions only)

Examples: i) $He(1s^2\,{}^1S) \rightarrow He^+(n\ell=0) + e^-(\varepsilon\ell'=0)$:

$$P = |\langle n\ell m=0,\ \varepsilon\ell' m'=0^{(-)} | \Psi(t_f)\rangle|^2$$

$n = 1$ direct single ionization

$n > 1$ "shake up" — " —

ii) $He(1s^2\,{}^1S) \rightarrow He^{++} + e^-(\vec{k}_1) + e^-(\vec{k}_2)$

$$P(\vec{k}_1, \vec{k}_2) = |\langle \vec{k}_1^{(-)}, \vec{k}_2^{(-)} | \Psi(t_f)\rangle|^2$$

$P(\vec{k}_1, \vec{k}_2)\, d^3k_1\, d^3k_2$ double ionization probability

Note – "(–)" denotes incoming-wave Coulomb boundary conditions

$$- \langle \vec{k}_1^{(-)}, \vec{k}_2^{(-)} | \approx \mathcal{A}\{\langle \Psi_{\vec{k}_1}^{(Coul.-)} | \langle \Psi_{\vec{k}_2}^{(Coul.-)} |\}$$

three-body continuum state (not known analytically)

incoming Coulomb waves

poor approximation for strong final state interaction

Numerical example: IR laser assisted XUV ionization of He

handout ← A. Liu & U. Thumm, Phys. Rev. A 89, 063423 (2014)

Note: ◦ Sequential vs. non sequential double ionization by two photon absorption for $\hbar\omega \gtrsim 54.4$ eV

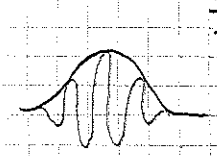
(See also A.L. & U.T., Phys. Rev. Lett. 115, 183002 (2015).

◦ Dipole selection rules → nodal structure in joint angular distributions

◦ Energy distributions depend on XUV spectral width & no. of photons

◦ "Conditional angular distribution": one emission angle is kept fixed (θ_1)

◦ "In-plane geometry": XUV, IR polarization, $\vec{k}_1$ & $\vec{k}_2$ in one plane



PHYSICAL REVIEW A 89, 063423 (2014)

Laser-assisted XUV few-photon double ionization of helium: Joint angular distributions

Aihua Liu and Uwe Thumm

We investigate few-photon extreme ultraviolet (XUV) double ionization of helium atoms without and in the presence of an assisting infrared (IR) laser field by numerically solving the time-dependent Schrödinger equation in full dimensionality within a finite-element discrete-variable-representation scheme. We discuss joint energy distributions for coplanar emission where the emitted electron momenta and polarization axis of the linearly polarized XUV and IR pulses lie in a plane. Our analysis focuses on joint angular distributions for highly correlated equal-energy-sharing double ionization by absorption of one, two, or three XUV photons and IR-laser-assisted single-photon XUV double ionization.

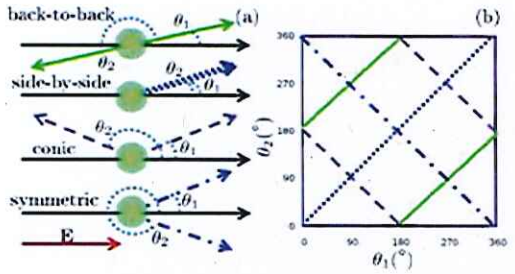


FIG. 1. (Color online) (a) Schematics for four different types of photoelectron emission patterns in the double ionization of helium. From top to bottom: back-to-back emission, side-by-side emission, conic emission, and symmetric emission. (b) Schematics for the identification of the corresponding emission patterns (with matching line types and colors) in joint angular distributions (see text).

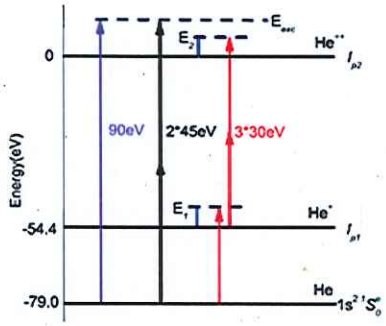


FIG. 2. (Color online) Energetics for nonsequential double ionization of helium by two 45-eV or one 90-eV photons and sequential double ionization by three 30-eV photons. I_{p1} and I_{p2} designate the first and second ionization threshold, respectively. For the examples considered in this section, the combined excess energy of the emitted electrons is $E_{exc} = E_1 + E_2 = 11$ eV.

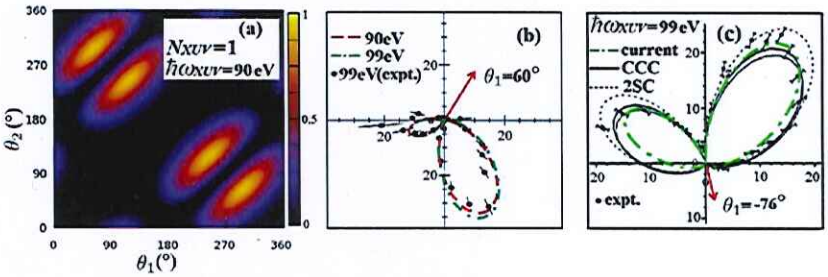


FIG. 4. (Color online) (a) Calculated normalized joint angular distribution for the double ionization of helium by one $\hbar\omega_{XUV} = 90$-eV XUV photon at equal energy sharing. The XUV pulse has a peak intensity of 10^{14} W/cm^2 and a pulse lengths of 1 fs. (b) Conditional angular distribution for $\theta_1 = 60°$. The red dashed line shows the calculated angular distribution for $\hbar\omega_{XUV} = 90$ eV and is scaled by the factor 0.55 relative to the distribution calculated for $\hbar\omega_{XUV} = 99$ eV (green dash-dotted line). The black dots with error bars show the distribution measured by Bräuning *et al.* [3] for $\hbar\omega_{XUV} = 99$ eV in units of b eV^{-1} sr^{-2}. Adapted from [3]. (c) Conditional angular distribution for $\theta_1 = -76°$ and $\hbar\omega_{XUV} = 99$ eV. The green dashed-dotted line shows our result, the solid black lines are convergent close-coupling calculations for different gauges by Kheifets and Bray [36], and the black dotted line shows a calculation with screened final-state Coulomb wave functions by Pont and Shakeshaft [37]. The black dots with error bars show the measured relative cross sections of Schwarzkopf *et al.* [35]. Adapted from [36].

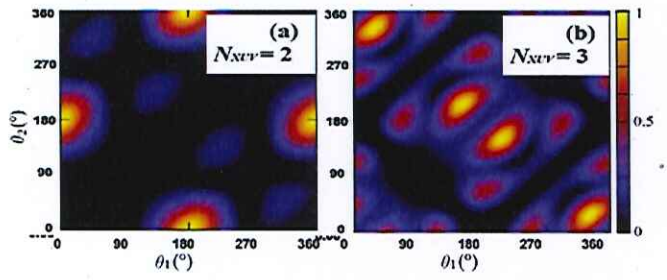


FIG. 5. (Color online) Calculated normalized (a), (b) joint angular distributions and (c), (d) conditional distributions for $\theta_2 = 0°$ for the double ionization of helium at equal energy sharing by (a), (c) two 45-eV photons and (b), (d) three 30-eV photons in XUV pulses with a peak intensity of 10^{14} W/cm^2 and a pulse lengths of 1 fs (see text).

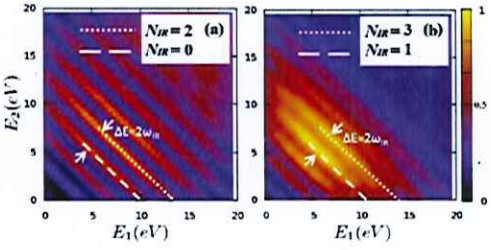


FIG. 6. (Color online) Normalized joint photoelectron energy distributions for IR-laser-assisted single-photon double ionization of helium at equal energy sharing in a 10-cycle XUV pulse of peak intensity 10^{14} W/cm^2, central photon energy $\hbar\omega_{XUV} = 89$ eV, and pulse length 0.46 fs. The assisting single-cycle laser pulse has a peak intensity of 3×10^{12} W/cm^2, photon energy of $\hbar\omega_{IR} = 1.61$ eV, and a pulse length of 2.6 fs. (a) Even effective numbers of IR photons. (b) Odd effective numbers of IR photons.

$$H_N = \sum_{i=1}^{N} \underbrace{\left\{ \underbrace{-\frac{\hbar^2}{2m}\Delta_i}_{t_i} + V(\vec{r}_i) \right\}}_{h_i} + \frac{1}{2}\sum_{\substack{i,j=1 \\ i\neq j}}^{N} V(\vec{r}_i, \vec{r}_j)$$

$V(\vec{r}_i)$: can include time-dep. external fields

- approx. Hamiltonian

$$H_N^{HF} = \sum_{i=1}^{N} \underbrace{\left(t_i + V^{HF}(\vec{r}_i) \right)}_{h_i^{HF}}$$

V^{HF}: HF potential, to be constructed

$$H_N^{HF}\,\Phi_N = E_N\,\Phi_N \,, \quad \Phi_N(\vec{r}_1 \ldots \vec{r}_N) = \frac{1}{\sqrt{N!}} \det\left(\varphi_i(\vec{r}_j) \right)$$

φ_i: orbitals

"Slater determinant for antisymmetrization

$$h^{HF}\,\varphi_i = \tilde{\varepsilon}_i\,\varphi_i$$

- Ritz' variational method

Determine Φ_N with minimal E_N by variation: $\varphi_\alpha \to \varphi_\alpha + \delta\varphi_\alpha$

$$\frac{\partial}{\partial \varphi_\alpha^*} \left\{ \langle \Phi_N | H_N | \Phi_N \rangle - \sum_{i=1}^{N} \varepsilon_i \int d\vec{r}\, |\varphi_i(\vec{r})|^2 \right\} \overset{!}{=} 0$$

↑ variational derivative

↑ Lagrange parameters to guarantee $\|\varphi_i\| = 1$

$\Rightarrow$ HF equations

$$\left\{ -\frac{\hbar^2}{2m} + \underbrace{V(\vec{r}) + \int d\vec{r}\,' \rho(\vec{r}\,')\, V(\vec{r}, \vec{r}\,')}_{\text{local}} \right\} \varphi_\alpha(\vec{r})$$

$$- \underbrace{\int d\vec{r}\,' \rho(\vec{r}, \vec{r}\,')\, V(\vec{r}, \vec{r}\,')\, \varphi_\alpha(\vec{r}\,')}_{\text{non local, due to exchange}} = \varepsilon_\alpha \varphi_\alpha(\vec{r})$$

$$\rho(\vec{r}) := \sum_{\substack{i=1 \\ \text{occ}}}^{N} \varphi_i^*(\vec{r})\,\varphi_i(\vec{r})$$

$$\rho(\vec{r},\vec{r}') := \sum_{\substack{i=1 \\ \text{occ}}}^{N} \varphi_i^*(\vec{r}')\,\varphi_i(\vec{r})$$

$$\Leftrightarrow \quad \left\{ -\frac{\hbar^2}{2m}\Delta + V^{HF} \right\} \varphi_\alpha = \varepsilon_\alpha \varphi_\alpha \,, \quad \varepsilon_\alpha \in \mathbb{R}$$

/ Self consistent solution of HF equations

Step 0 : approximate orbitals : $\{\varphi_i^{(0)}\}_{i=1,\dots N}$

calculate $V^{HF(0)} = V^{HF}[\{\varphi_i^{(0)}\}]$

Step 1 : solve eigenvalue problem

$$\left(T + V^{HF(0)}\right)\varphi_\alpha^{(1)} = \varepsilon_\alpha^{(1)}\varphi_\alpha^{(1)}$$

$\Rightarrow$ In general ∞ many solutions

Select $\{\varphi_1^{(1)},\dots,\varphi_N^{(1)}\}$ corresponding to N lowest energies $\varepsilon_\alpha^{(1)}$ (no degeneracies are assumed).

Only these orbitals are occupied (for the N particle ground state)

Step n : solve

$$\left(T + V^{HF(n-1)}\right)\varphi_\alpha^{(n)} = \varepsilon_\alpha^{(n)}\varphi_\alpha^{(n)}$$