3 - Examples of strong-field approaches

3.(a) - Numerical solution of the time-dependent Schrödinger equation.

$i\partial_t \Psi(\vec{r},t) = H(t)\Psi(\vec{r},t)$ solved numerically

Advantage: no physical approximation
(all effects are present)

Disadvantage: difficult physical interpretation
(sometimes effects are difficult to disentangle)

- Easy to solve in 1D, more involved in 3D
- Problematic for many-electron systems
  => only recently this has been achieved for Helium by Ken Taylor's group in Belfast
  (context: laser-induced nonsequential double ionization

3.(b) - Classical methods

We have seen that, in general, strong-field phenomena can be described as laser-assisted rescattering or recombination processes.
Easiest description: classical models

Classical methods

Classical electron in a laser field

- **Electron propagation in the continuum:**

$$\ddot{\mathbf{r}}(t) = \underbrace{-\nabla V}_{neglected} - \mathbf{E}(t)$$

- **Initial conditions:**
  - $\mathbf{v}(t_0) \equiv 0$ (**vanishing drift velocity**)
  - $\mathbf{r}(t_0) \equiv 0$ (**electron released from the origin**)

$$\mathbf{v}(t) \equiv \mathbf{A}(t) - \mathbf{A}(t_0)$$

$$\mathbf{r}(t) \equiv \int_{t_0}^{t} \mathbf{A}(\tau)d\tau - (t - t_0)\mathbf{A}(t_0)$$

- **return time** t_1: $\mathbf{r}(t_1) \equiv 0$

- High-harmonic generation: $\Omega = |\varepsilon_0| + E_{kin}(t_1, t_0)$
- High-order above-threshold ionization: $\frac{1}{2}\left[\mathbf{p} - \mathbf{A}(t_1)\right]^2 = E_{kin}(t_1, t_0)$
- Nonsequential double ionization:

$$\sum_{i=1}^{2} \left[\mathbf{p}_i - \mathbf{A}(t_1)\right]^2 = E_{kin}(t_1, t_0) - \underbrace{|\varepsilon_{02}|}_{2^{nd}\text{ ionization potential}}$$

with $E_{kin}(t_1, t_0) = \frac{1}{2}[A(t_1) - A(t_0)]^2$ (kinetic energy upon return)

With this simple approach we can predict:

(a) The times in which the electron leaves and returns to its parent ion

(b) The kinetic energies of the electron upon return

(c) The cutoff energies for Above-threshold ionization / High-order harmonic generation

(d) Electron momentum distributions for laser-induced nonsequential double ionization

Disadvantage:

We cannot use this to compute spectra, as this does not account for quantum-interference effects.

Does not account for the internal structure of the atom

2- Semi-analytic approaches - strong-field approximation

L.V. Keldysh, Sov. Phys. JETP 20, 1307 (1965); F.H.M. Faisal, J. Phys. B 6, 289 (1973); H.R. Reiss, PRA 22, 1786 (1930).

(*) key idea

- The residual binding potentials are neglected when the electron is in the continuum
  => Volkov states (field-dressed plane waves) → analytical solution
- The laser field is neglected when the electron is bound (field-free states) → analytical solution

⊛ Advantages

- Clear, physical picture of the phenomenon in question (easily related to classical models)
- Computationally inexpensive

⊛ Drawbacks

- The SFA is not gauge invariant (especially problematic for molecules)
- Breaks down if the residual potentials become important (for instance, in extended systems)
- There is no proper justification for the approaches performed in the SFA.
- The continuum and bound states are not orthogonal

2.(a) – Derivation

⊛ Key definitions

Let us consider the time-evolution operator

$U(t,t')$ related to the Hamiltonian

$$H(t) = \underbrace{\frac{p^2}{2} + V}_{\text{atomic Hamiltonian } H_0} + \underbrace{H_{int}(t)}_{\text{interaction with the field}}$$

Velocity gauge: $H_{int}(t) = -\vec{P}\cdot\vec{A}(t) + \frac{1}{2}A^2(t)$

Length gauge: $H_{int}(t) = -\vec{r}\cdot\vec{E}(t)$

Reminder: 4th year quantum mechanics

The time-evolution operator $U(t,t')$ makes a physical

System evolves from a time t' to a time t

$$|\psi(t)\rangle = U(t,t')\,|\psi(t')\rangle$$

- Properties:

$$U(t,t') = U^{-1}(t',t) \;;\; U(t,t) = I \;;\; U(t,t')\,U(t',t'') = U(t,t'')$$

It also obeys the time-dependent Schrödinger equation

Let us now consider the Hamiltonians

H_0 (atomic Hamiltonian) and H^V (Gordon-Volkov Hamiltonian)

Time evolution operator

$U_0(t,t')$

Time-dep. Schrödinger equation

$$i\,\partial_t U_0(t,t') = \underbrace{\left[\frac{p^2}{2} + V\right]}_{H_0} U_0(t,t')$$

H_0 time independent

$$\Rightarrow U_0(t,t') = \exp\left[-i H_0 (t-t')\right]$$

Time evolution operator

$U^V(t,t')$

Time-dep. Schrödinger equation

$$i\,\partial_t U^V(t,t') = \left[\frac{p^2}{2} + H_{int}(t)\right] U^V(t,t')$$

(*) Starting point: Dyson equation (DuHamel equation)

$U(t,t')$ may be written as

$$U(t,t') = U_0(t,t') - i\int_{t'}^{t} U_0(t,\tau)\,H_{Int}(\tau)\,U(\tau,t')\,d\tau \quad (*)$$

Iteration: weak-field perturbation theory

$$U(t,t') = U_0(t,t') - i\int_{t'}^{t} U_0(t,\tau)\,H_{Int}(\tau)\,U_0(\tau,t')\,d\tau$$

$$- \int_{t'}^{t} d\tau \int_{t'}^{\tau} d\tau_2\; U_0(t,\tau)\,H_{Int}(\tau)\,U_0(\tau,\tau_2)\,H_{int}(\tau_2)\,U_0(\tau_2,t') + \dots$$

$H_{int}(t) \propto$ external field

$\Rightarrow$ The above-stated series converges for weak fields and diverges for strong fields

On the other hand, one may also write $U(t,t')$ as

$$U(t,t') = U_V(t,t') - i\int_{t'}^{t} U_V(t,\tau)\, V\, U(\tau,t')\, d\tau \qquad (**)$$

U_V: Gordon-Volkov time-evolution operator

Volkov, Zeit für Physik 94, 250 (1935); Gordon, ibid. 40, 117 (1926)

Iteration: Gordon-Volkov series (binding potential is treated as a perturbation)

If we consider the zero-order term in (**) and insert it in (*), we have

$$U^{SFA}(t,t') = U_0(t,t') - i\int_{t'}^{t} U_0(t,\tau)\, H_{int}(\tau)\, U^{V}(\tau,t')\, d\tau^{*}$$

Formally,

(a) Both series are mixed

(b) Keldysh: perturbation theory with a modified basis

(*) Please note: in the mainstream literature, the SFA is sometimes referred to as "non-perturbative". It is, however, perturbation theory with a modified basis.