Laser Induced Electron Diffraction and Electron Holography

Laser Induced Electron Diffraction

In this exercise, we will introduce the basic ideas behind the ultrafast laser-induced electron diffraction (LIED) and electron holography concepts. We will combine several concepts, previously introduced in this class, such as electron-ion elastic scattering combined with strong-laser-field ionization (SFI) and the semi-classical model of the electron propagation in a linearly polarized laser field. For the preparation for this class please read the following articles 1-3.


In the three-step model of strong-field light-matter interactions, the electron ionized slightly after the peak of the laser field can return to its parent ion with the maximum energy of $3.17U_p$, where $U_p$ stands for the ponderomotive potential of the laser field. Compared with the traditional electron diffraction (ED) concepts, used for retrieving atomic and molecular structure, the LIED can be seen as self-imaging tool with a potential for resolving molecular structural dynamics.

At the instant of the rescattering the attosecond electron wave packet can elastically scatter from a parent ion, with a certain differential cross section. Since the electrons are born in the continuum at different phases of the laser field and travel different classical trajectories, many of them miss the parent ion, while some of them elastically scatter off the parent ion. The electrons that scatter off the parent ion can be used for self-imaging of the molecular structure with the attosecond temporal an picometer spatial resolution. From left to right, Figure 1 shows a typical few-femtosecond laser electric field, calculated 2D electron momenta ionized from H atom by a strong laser filed, and the electron-ion differential cross section (DCS) calculated from the back-scattering rings (BRRs).

![Figure 1](image)

*Figure 1 Left: Laser electric field (E) and the corresponding vector potential (A) with a two-optical-cycle pulse duration. Middle: 2D electron momenta with two BRRs at $P_z=-1.8$ a.u. and $P_z=2$ a.u., and the electron holography structure in the range from $P_z=-1.2$ a.u. to $P_z=1.2$ a.u.. Right: Calculated electron diffraction (ED) differential cross-section (DCS) and the LIED DCS (dashed line).*

As labeled by the red and blue arrows in Fig. 1 a), when the electron tunnels to the right-hand side of the hydrogen atom, along the laser polarization direction (i.e. position “a” in the left panel of Fig. 1), at the instant of the rescattering (position “b” in the left panel of Fig. 1) the electron gets a kick back from laser field (i.e. the
vector potential is at its maximum at this point) in the same direction where the electron came from. On the other hand, at the position “a”, an electron is released on to the left-hand side of the atom, and it can backscatter to the left by getting the momentum kick from the laser field (i.e. vector potential at instant “b”).

1) Identify the two BRRs in Fig. 1 b) corresponding to the two situations described above (i.e. electron released at instant “a” vs electron released at instant “b”). Which ring has a larger displacement ($p_A$) from the center of the momenta distributions along the laser polarization $p_l$? Which has a larger radius ($p_B$)? Discuss why this is so. $p_A$ and $p_l$ are the drift momentum along the laser polarization and the recollission momentum, respectively. Approximate the ratios of the recollision and the drift momenta ($p_B/p_A$) for the case “a”. Note: the electrons on the single, cut-off trajectory have the highest $p_B/p_A$ ratio that cannot be larger than $p_B/p_A = 1.26$. The cut-off electrons are the ones coming back with the maxim energy of $3.17U_0$.

2) Use the calculated BRR momenta for the highest energy ring in Fig. 1b) to approximate the laser intensity used in this calculation. Hint: use the cut-off energy along the laser polarization from Fig. 1b).

3) Identify two smaller (ghost-like) BRRs in Fig. 1b). Where are they coming from? Discuss their origin in terms of the laser electric field and the vector potential as shown in Fig. 1a). How does the radius and the back-scattering displacement scale as the laser field ramps up and down? Hint: approximate and compare the $p_B/p_A$ ratios for the three situations in Fig. 1a) labeled as a, b and c.

**Electron Holography**

The complex structure in the electron-momentum distribution between the BRRs in Fig. 1 c), taken from reference 3, was considered to be too complex to interpret in detail, and explained in terms of direct ionization from the laser field. In this part of the exercise, we will show that the major features of these 2D electron momenta can be quantitatively reproduced using the concepts and models introduced in this class. In reference 1, it was shown that the electron momenta can be seen as 2D electron holography patterns, where the direct and rescattered electrons interfere on the electron detector. In reference 2, it was shown that the electron holographic patterns can be quantitatively reproduced by using a spherical and plane wave (SPW) interference model which assumes that the electron wave packet, freed from the ion by the strong laser field, can be seen as a plane wave, and the rescattered electrons can be seen as a spherical wave. The interference of the spherical and plane waves creates complex electron 2D momenta with several minima and maxima along the laser polarization. Such structure was named spider structure in ref. 3. The distance between the interference minima depends solely on the relative phases of the spherical and plane electron wave packets, and is given by the time the electron spends in the continuum before the instant of the rescattering.

![Image](image.png)

*Figure 2 a) The plot of the real part of the electron plane wave in the 2D momentum space (i.e. momenta given in atomic units parallel and perpendicular to the laser polarization. b) The plot of the real part of the electron spherical wave in the 2D momentum space. c) The superposition of the plane (a) and spherical (b) wave packets shows the spider-like structure. d) The experimental electron 2D momenta of an Xe atom ionized by a 1300 nm laser field with intensity of $5 \times 10^{13}$ W/cm$^2$. The white lines are the interference minima extracted from the SPW model shown in c). e) Classical trajectories of the electrons in the 1300 nm laser field of the intensity of $5 \times 10^{13}$ W/cm$^2$. We plot five different electron trajectories born at five different phases just after laser field peak (the black line). The phase of the plane wave is given by $\Psi_{\text{plane}} \propto e^{iP_{l}/a}$, and the phase of the spherical wave is given by...*
\[ \Psi_{\text{spherical}} \propto e^{ikP_{\text{total}}}, \]
where the total momentum (parallel and perpendicular) is given by
\[ P_{\text{total}} = \sqrt{P_0^2 + P_1^2}. \]
k is the modulation frequency.

Fig. 2 shows the plane and spherical waves plotted in the 2D momentum space (a-b), together with the SPW interference plot (c), and the corresponding experimental data (d). Fig. 2 (e) shows five classical trajectories of the electrons in the 1300 nm laser field born at five different phases close to the laser field peak. We see that all of the trajectories have the maximum excursion distance from the parent ion of ~5 nm before they get accelerated back towards the parent ion. The electron released in the continuum at the angle of 197° (i.e. 17° after the peak of the pulse) returns with the highest kinetic energy to the parent ion (i.e. 3.17U_p). These high-energy electrons revisit the parent ion roughly 2/3 of the optical laser cycle after the ionization. All this can be calculated from the classical trajectory equation:

\[ z(t) = \frac{eE_0(t)}{m_0\omega^2} \left[ \sin(\omega t_0)(\omega t - \omega t_0) + \cos(\omega t_0) - \cos(\omega t) \right]. \]

4) Use this equation to reproduce the trajectories of the electrons born at the 189° phase for the 266 nm, 400 nm, 800nm, 1300 nm, and 2000 nm. Calculate the maximum excursion distances of the electron from the parent ion for all the cases.

5) Assuming that at the instant of the rescattering the electron is back to its origin (i.e. \( z(t) = 0 \)), solve the above equation numerically and plot the initial versus returning phase for an 800 nm wavelength. Confirm that the electron born at 189° would return 2/3 of the optical-cycle later.

6) Assuming that the rescattering takes place 2/3 of the optical cycle after the electron is released in the continuum, how much time does the electron spend in the continuum before the rescattering takes place for an 800 nm laser field? Calculate these times for the laser beams of 266 nm, 400 nm, 800nm, 1300 nm and 2000 nm. What are the cut-off electron energies for these laser wavelengths considering the laser intensity of 5x10^{13} W/cm²?

7) How many times, on these cut-off trajectories, does the electron revisit the parent ion for the five different wavelengths assuming the pulses duration of 40 fs?

Figure 3, from ref. 3, shows the classical action \( S \) as a function of the momentum parallel to the laser polarization. The actions \( S \) is defined as follows:

\[ S(p, t, t_b) = \int_{t_b}^{t} \left( \frac{p(t')^2}{2m_e} + I_p \right) dt', \]
where \( I_p \) is the ionization potential of the atom at hand (i.e. Xe, Kr, Ar, etc.), \( p(t) \) is the momentum, \( t_b \) the instant of the ionization, and \( m_e \) the electron mass.

Figure 3 Action “S” vs the parallel momenta for three different laser wavelengths. The given slopes define the modulation frequencies that depend only on the laser wavelength for the given laser intensity. The phase of the electron in the laser field is given by \( e^{i\omega t}. \)
8) Reproduce Figs. 2 a)-c) by using the parameters given in the caption of Fig. 2. Hint: use the frequency modulation calculated by the slope of the action vs $p$-parallel for the 1300 nm laser wavelength as shown in figure 3.

9) Calculate the frequency modulation for the 266 nm and 400 nm laser wavelengths, as shown in Fig. 3, and plot the SPW electron holograms for an Xe atom for the 266 nm, 400 nm, 800 nm, 1300 nm, and 2000 nm laser wavelengths assuming the intensity of $5 \times 10^{13}$ W/cm$^2$. $I_p (\text{Xe}) = 12.1$ eV. Confirm that the angle of the spider minima depends only on the phase between the plane and spherical waves. For the phase calculations take into the account that the integration time takes place from the instant of the ionization to the instant of the first rescattering event (i.e. you could assume that this phase is $2/3$ of the optical cycle for all the given laser wavelengths).

Useful equations:

$$E_{\text{field}} = \frac{1}{4\pi\epsilon_0} \frac{e}{a_0^2} = 5.14 \cdot 10^9 \frac{V}{\text{cm}}$$

is 1 atomic unit of electric field.

An atomic unit of laser intensity is defined as: $I = \frac{1}{2} \epsilon_0 c E_{\text{field}}^2 = 3.51 \cdot 10^{16} \frac{W}{\text{cm}^2}$.

$F(t) = -\partial A(t)/\partial t$, the relation between the electric field the vector potential.

$$U_p = \frac{e^2 E_0^2}{4m_e \omega^2} = 9.33 \cdot 10^{-14} I \lambda^2 \text{ [eV]},$$

ponderomotive potential in eV for the intensity given in W/cm$^2$ and wavelength in micrometers. One atomic unit of energy 27.211 eV. In atomic units, the energy and momentum are related as $E=p^2/2$.

Also, the ponderomotive potential is related to the vector potential as: $U_p=A_r^2/2$, where $A_r$ is the magnitude of the vector potential at the instant of rescattering, as shown in fig. 1 a).

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