Quantum Interference and Imaging

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Outline of Quantum Battle 2

• B1 – Interference is a mathematical trick (it is not real). All that can be measured is intensity or $|\psi|^2$.

• B2 – How are LIED and holography useful to measure molecular structure?

• B3 – All can be learned by just solving the TDSE. Why do experiments?

• B4 – No one should ever solve the TDSE.

• Open discussion - Decoherence is a symptom of an incomplete measurement and has no physical reality.
B1 – Benjamin
Interference is a mathematical trick (it is not real). All that can be measured is intensity or $|\psi|^2$. 
Quantisation in strong fields: ATI in Xe - Benjamin

\[ \gamma = \sqrt{\frac{I_p}{2U_p}} \]

\[ U_p \propto I_0 \lambda^2 \]

Photon picture is a successful model for the fundamental strong-field effects

\[ E_{\text{kin}} = n\hbar\omega_{IR} - I_P - U_P \]

\[ \hbar\omega_{XUV} = 19 \cdot \hbar\omega_{IR} \]

\[ \lambda = 56 \text{ nm} \]

L. V. Keldysh et al., Sov. Phys. JETP 20, 1307 (1965)
Figure adapted from K. Amini, D.Phil thesis, University of Oxford (2017)
Hypothesis: We don’t need no interference - Matthias

If somewhere is written “interference” this simply indicates that the author was incapable of identifying the conserved quantity of the system.

Example:
Angular positions of the interference maxima for the double slit:

$$\theta_n = n \cdot \frac{\lambda}{d}$$

→ The conserved quantity for the double slit

is one “doubleslititron” and has the value $$\frac{\lambda}{d}$$
ATI arises from interferences - Sebastian

Intensities:
- 780 nm: $4.8 \times 10^{13}$ W/cm$^2$
- 390 nm: $0.6 \times 10^{13}$ W/cm$^2$

the 2D TDSE code is based on a code by Uwe Thumm
Premises or propositions:

1. The ground state (\( GS \)) is the e-wave source.
2. The laser-GS interaction creates multiple time-space slits \( (S) \).
3. The slits are located about zero of the electric field at \( t_n \), let’s assume an approximated time-window of \( T_0/5 \).
4. By symmetry argument, these slits are separated by a laser-cycle \( T_0 \).
5. The final energy distribution will be given by the squared of the sum of the individual EWP's produced by each slit \( S_n \):

\[
P(\epsilon_k, t_F) = \left| \sum_n a_n(\epsilon_k, t_n, t_F) \right|^2 \text{ with } a_n(\epsilon_k, t_n, t_F) = A_n(\cdot) e^{-iS(\epsilon_k, t_n, t_F)}
\]
ATI peaks can be explained as an inter-cycle interference or by using energy conservation.

What about other types of interference?

Matthias
Sub-cycle electron trajectories - Kasra

Shape of atomic potential under zero field

Energy

t = 0

t = T/2

V = 0

\(-e^2/|z|\)

\(-eF(t)z\)

Back-scattering

Forward-scattering

Direct tunnelling

Shape of potential at maximum forward and reverse field

Detector plane

Energy

Displacement (z)

E(t)

Return energy [U,]

Time [cycles]

Types of interferences & holography - Matthias

- ATI
- "Temporal Double Slit"
- Strong-field PE holography

Scattering-free Interference
Intracycle Interference

Propagation in ionic field
Emission time difference

Intra-cycle interference cannot be modeled by photons - Sebastian

How can position–information or time–information be retrieved from measured electron momentum distribution without recollisions?

First, Intra-cycle interference cannot be modeled by photons, because the time scale needed to explain intra-cycle interference $T_{\text{intra}}$ is below one cycle of the light field.

→ The concept “photon” does not exist on sub-cycle time-scales.

Second, this also indicates that the wave-particle duality is nonsense on sub-cycle time-scales.

Poll:
- A) Photons do not exist at all
- B) Photons cannot be used to model physics on sub-cycle time scales ($t<1/E_{\text{ph}}$)
- C) Photons are just as good as time-dependent fields to model physics (wave particle duality)

Attosecond electron wave packet (EWP) - Alexis

- Single Attosecond EWP

\[ a_p(\tau_1) = A(p, \tau_1) e^{i\phi(p, \tau_1, t_f)}. \]

- How to recover the phase, if the measurement \( |a_p|^2 \) lost it 😣?

A. Chacon et al., PRA 87, 023408 (2013)

A. Chacon et al., PRA 87, 023408 (2013)
Extracting electron wavefunction if the phase is lost by measuring - Alex

💡 what about analogy?
Light wave-interferometry techniques, i.e. S

💡 But now with EWPs?

https://www2.physics.ox.ac.uk/research/ultrafast-quantum-optics-and-optical-metrology/

But but with the aim to the wavefunction still from EWPs interfering and dipoles are missing?


Quantum Battles 2020

Battle 2 - Quantum Interference and Imaging
How to extract dipole information?

Primises: 

\[ |a_p(\tau_1, \tau_2)|^2 = |A_1(p, \tau_1)e^{-i\phi(p, \tau_1, t_f)} + A_2(p, \tau_2)e^{-i\phi(p, \tau_2, t_f)}|^2 \]

\[ = A_1(p, \tau_1)^2 + A_2(p, \tau_2)^2 + 2A_1(p, \tau_1)A_2(p, \tau_2)\cos(\Delta \phi_{21}). \]
Applications of qspider to retrieve EWPs And Dipole “phases”:

SFA- ionisation from 2pz states of He+

EWP–retrieval  

Dipole Matrix Elem.
B2 – How are LIED and holography useful to measure molecular structure? - Benjamin

Laser-induced electron diffraction (LIED)

Atomic

Molecular

Strong-field holography

Reference

Signal

$\rho_r$ (a.u.)
LIED: quantum picture - Kasra


B. Wolter et al., PRX 5, 021034 (2015)
M. Pullen et al., Nature Commun. 6, 7262 (2015)

E. T. Karamatskos et al., JCP 150, 244301 (2019)


Amini et al., PNAS 116, 8173 (2019)
Holography measures the electron wave packet - Matthias

Conclusion: "In our experiment, electron holography provides information about the continuum electron wave packet rather than the scattering object"

M. Meckel et al., Nat. Phys. 10, 592 (2014)
Image orbitals without rescattering - Matthias


LIED works! - Kasra

\[ p_r = p_\parallel - A_r \]


Conditions for LIED - Kasra

MIR-LIED: Molecular structure retrieval
- Large $U_p$ – avoid influence/blurring of valence electron cloud
- Large momentum transfer range and small $\lambda_{dB}$ – spatial resolution $\rightarrow$ comparable to CED/UED
- Deep into quasistatic (tunneling) regime – you can use classical models and map classical trajectories to exp. data
- Avoid ionization saturation
- Large EWP extent $(x)$
- Coincidence single molecule imaging

NIR-Holography: Orbital structure retrieval
- Sensitive to valence electron cloud
- Too low $U_p$ and narrow momentum transfer range for molecular structure retrieval
- Boundary of quasistatic (tunnelling) regime
- Ionization saturation; easily ionize and fragment large molecules with small $I_p$

B. Wolter et al., PRX 5, 021034 (2015)
Sebastian

B3 – All can be learned by just solving the TDSE.

Why do experiments?
A short story of attoseconds - Matthias

\[ i \frac{\partial}{\partial t} \Psi = - \left[ \frac{1}{2} \Delta + \frac{1}{r} + \vec{E} \cdot \vec{r} \right] \Psi \]

The end.
Example: Attoclock in atomic hydrogen - Matthias

See also (original work in He, $\Delta t_D < 34$ as):


Hamiltonian of any molecule - Alexis

\[ H_0 = \sum_{i=1}^{N} \left( \frac{p_i^2}{2m} \right) + \sum_{k=1}^{K} \frac{p_k^2}{2M_k} - \sum_{i=1}^{N} \sum_{k=1}^{K} \frac{Z_k}{|r_i - R_k|} \]

- Kin. energy of the electrons
- Kin. energy of the nuclei
- Nucleus-electron attraction
- Electron-electron repulsion
- Repulsion between nuclei

Solutions for N>2, K>2 is very challenging to find

Alternative: DFT and TD-DFT

We can measure this! - Kasra
TDSE vs (semi-) classical - Benjamin

\[ \Psi(t = 0) \quad \text{TDSE} \quad \Psi(t \to \infty) \]

\[ E(t) \]

(Semi-) classical model
No one should ever solve the TDSE

CHANGE MY MIND
Example: Attoclock in atomic hydrogen - Sebastian

See also (original work in He, $\Delta t_D < 34$ as):


Reality is described by classical observables - Kasra

What we measure are intensities or $|\psi|^2$

- Electrons
- Ions
- Photons

Alternative interpretations of QM?

Poll:
- A) Phases can be measured in the lab
- B) Only $|\psi|^2$ can be measured in the lab
- C) Quantum effects can be measured directly by looking at classical observables
Extracting PES from time-dependent wave function - Benjamin

- Time-dependent Schrödinger equation:

\[ i \frac{\partial \Psi(r, t)}{\partial t} = [H_0 + V_I(t)] \Psi(r, t), \]

At the end of laser pulse \( t = T_p \) we get \( \Psi(r, T_p) \).

- How do we do we extract photoelectron spectra from the \( |\Psi(T_p)\rangle \)?

- The probability of finding the electron at the end of the laser pulse in a continuum state with the momentum \( k = (k, \Omega_k) \) is given by

\[
P(k, \Omega_k) = \frac{d^3P}{k^2 dk d\Omega_k} = |\langle \Phi_k | \Psi(T_p) \rangle|^2.
\]

- Popular approximate methods used for extracting PES:
  window-operator method (WO), tSURFF method, iSURV method, etc.
The differential ionization probabilities of H atoms for emission of electrons in the directions $\theta_k = 0^\circ$, $90^\circ$, and $180^\circ$, as functions of the photoelectron energy in units of the ponderomotive energy $U_p$, for the following laser-field parameters: $I = 10^{14}$ W/cm$^2$, $\lambda = 800$ nm, and pulse duration is six optical cycles [3].

PCS vs WO for Argon atom - Benjamin

Figure: Full PADs for the Ar atom.
In order to simulate an ionization experiment, one has to use appropriate continuum state $\Phi_k^(-)(r)$ which satisfy so-called incoming boundary condition.

The problem with the window-operator method is that it does not single out the contribution of the solution $\Phi_k^(-)$, but it includes an unknown linear superposition of the states $\Phi_k^(-)$ and $\Phi_k^+$. Therefore, it may lead and does lead to unphysical results, depending on the considered region of the spectrum.

Approximate method used for extraction of the PES from the time-dependent wave function should always be checked for the consistency with the PCS method.
Decoherence is a symptom of an incomplete measurement and has no physical reality.

Poll:
- A) Agree
- B) Disagree
- C) I am not sure (I am a cat - maybe)
Is the neon dimer decoherent? - Sebastian

Experiment:
- dissociation of a neon dimer in a circularly polarized strong laser field
- detect electron and ion in coincidence

Is the neon dimer decoherent? - Sebastian

Option B: Interference in momentum space can be used to infer position-information regarding the molecule

- Interference is recovered selecting only one channel (knowing if an additional photon is absorbed or not)
- Decoherence is a symptom of an incomplete measurement and has no physical reality

Some processes are incoherent by nature - Matthias

- Quantum physics is *microscopic* and *coherent*
- Experiments integrate over the *macroscopic* focal volume

Focal intensity distribution $I(\vec{r})$

Focal phase distribution $\phi_0(\vec{r})$

Poll: In HHG all atoms in the focal volume radiate coherently. On the other hand in ATI, electron emission from different atoms...

- A) Is inherently incoherent
- B) Is coherent, but we cannot observe the coherence
- C) I am not sure (I am a cat - maybe)

\[ Y_{\text{HHG}} = \left| \iiint_V \sqrt{Y_{\text{atom}}^{\text{HHG}}} e^{-i\phi(\vec{r})} d^3\vec{r} \right|^2 \]

\[ Y_{\text{ATI}} = \iiint_V Y_{\text{ATI}}^{\text{atom}} d^3\vec{r} \]
Decoherence - Sebastian

Decoherence is the loss of coherence of a quantum system caused by ignoring (i.e., tracing over) degrees of freedom to which the system is coupled and which are regarded as of no interest. The latter is termed the bath, although it need not be larger than one degree of freedom.


Future perspectives: dynamics in trivial and topological materials - Alexis

Extending ultrafast science for electron structural and dynamics in condensed matter phases:

Transition metal dicholoeidal
Topological Chern insulators
Topological insulators
Weyl semimental
Strongly-correlated materials ...

R. Silva et al Nat. Photonics (2019) HHG in Haldane Model
High harmonic in 3D topological insulators Bi$_2$Se$_3$

3D TIs:
- Insulating bulk bands
- Conducting at the surface bands
  (protected by Time-reversal symm.)

Topological anomalous
Harmonic vs the ellip. Of the 7.5 um driven–sources

D. Baykusheva, A. Chacon et al. (in revision process)
Towards the ultrafast e–h interferometry spectroscopy in trivial and topological materials?
Acknowledgements

CANTON SARAJEVO
Ministry for Education, Science and Youth
Thank you for your attention
and don’t forget to see the world from other point of view

– Quantum Interference Imaging Team
Extra slides
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Acknowledgements: Everyone
Comparison with conventional electron diffraction - Kas

Electron diffraction

Energies of 10 keV - 100 keV

Momentum transfer

\[ q = 2k_r \sin \frac{\theta_r}{2} \]

Modulated distribution

Spectrum gives internuclear distances

... temporal resolution limited to ~ 150-350 fs


K. Amini, J. Biegert, book chapter on UED/LIED in AAMOP vol. 69, Elsevier (2020). Free download until 5 August:

Elastic electron scattering - Kasra

\[ I_T(q) = I_A(q) + I_M(q) \]

Atomic

\[ q = 2k_r \sin \frac{\theta_r}{2} \]

Molecular

Atomic (incoherent) sum of scatterings

Molecular (coherent) interference

Quantisation in strong fields: ATI in Xe - Matthias

\[ E_{\text{kin}} = n\hbar \omega_{IR} - I_P - U_P \]

\[ U_P \propto I_0 \lambda^2 \]

\[ \hbar \omega_{XUV} = 19 \cdot \hbar \omega_{IR} \]

\[ \lambda = 56 \text{ nm} \]

Photon picture is a successful model for the fundamental strong-field effects.

L. V. Keldysh et al., Sov. Phys. JETP 20, 1307 (1965)
Figure adapted from K. Amini, D.Phil thesis, University of Oxford (2017)

\[ E_{\text{kin}} = 1.16\text{eV} \]

\[ I_P(Xe) \]
Quantisation in strong laser fields: Keldysh parameter - Kasra

\[ \gamma = \sqrt{\frac{I_p}{2U_p}} \]

\( \gamma \) - Keldysh parameter
Identifies the dominant photoionization regime

\( I_p \) - Ionization potential
Energy required to remove an electron from an atom or molecule

\( U_p \) - Pondereomotive energy
Average kinetic energy of a free electron in an oscillating electric field (e.g. laser field)

L. V. Keldysh et al., Sov. Phys. JETP 20, 1307 (1965)
Figure adapted from K. Amini, D.Phil thesis, University of Oxford (2017)
Quantisation in strong fields: ATI in Xe - Kasra

\[ E_{\text{kin}} = n\hbar\omega_{IR} - I_P - U_P \]

Photon picture is a successful model for modeling inter-cycle interference.

Two-color photoelectron holography - Kasra