Wavepacket dynamics and interferences in atoms and molecules

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Thanks to:
Damien Bigourd (post-doc)
Antoine Monmayrant (ph D)
Sébastien Weber (ph D)
Béatrice Chatel (CNRS)
Wave packets and interferences

A wave packet is a Coherent superposition of several states:
- Vibrational wave packet (*many levels*)
- Rydberg electron wave packet (*many levels*)
- Free electron wave packet (*continuum of levels*)
- Spin-orbit wave packet (*2 levels*)
  … Single level

Interferences:
- between wave packets created by different laser pulses
- within the excitation process
Principle of experiments

Quantum system (atom, molecule)

- single ultrashort pulse
- sequence of ultrashort pulses
- complex shaped pulse

Observation of the temporal evolution
with a probe pulse
of the final state
without time resolution

$I(\tau)$
$I(\text{shape})$

Principle of experiments
Invited talks

Wave Packets Dynamics
(Magpie A)

12:00 Helen Fielding, University College, London (UK)
   “Setting the quantum clock: Localisation of Rydberg wave packets in H₂”

12:20 Kenji Ohmori, Institute for Molecular Sciences, Okazaki (Japan)
   “Tailoring Picometric Quantum Carpets by Controlling Ultrafast Wave-Packet Interference”

12:40 Terry Mullins, Albert-Ludwigs-Universität Freiburg (Germany)
   “Coherent transients in the photoassociation of ultracold atoms by femtosecond pulses”
Invited talks

Wave Packets Dynamics (Magpie A)

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“Setting the quantum clock: Localisation of Rydberg wave packets in H2”

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“Coherent transients in the photoassociation of ultracold atoms by femtosecond pulses”
Outline

I. Wave packet Interferences in atoms

II. Quantum state holography

III. Control of wave packet dynamics

IV. Interferences within the excitation of a multiphoton transition

\[ \tau \]

\[ 6d, 8s \]

\[ 5p^2P_{1/2} \]

\[ 5s \]

Rb

Fluorescence (420 nm)
Interferences between identical time-delayed quantum paths: Ramsey fringes - Theory

\[ |\Psi(t)\rangle = |i\rangle + a e^{-i\omega t} |e\rangle + a e^{-i\omega (t-T)} |e\rangle \]

**weak field regime:** \[ |a|^2 \ll 1 \]

\[ \text{Period} = \frac{2\pi}{\omega} \]

Excited state population

Time delay \( T \)

**interference phase oscillates rapidly with** \( T \)

\[ \phi_{cc} = \omega T \]

\[ \phi_{cc} = 0 \ [2\pi] \quad \text{constructive interferences} \]

\[ \phi_{cc} = \pi \ [2\pi] \quad \text{destructive interferences} \]
Ramsey interferences with a pulse shaper (1)

High Res. Pulse Shaper:
- Phase - Amplitude control with 640 pixels.
- Shaping window of 35 ps.
- High complexity.
- 75% power transmission.

\[ E_S(\omega) = H(\omega) \tilde{E}_E(\omega) \]

\[ H(\omega) = M(f \gamma \omega) \]


Ramsey interferences with a pulse shaper (2)

A sequence of 2 FT limited pulses with a relative phase $\theta$…

$$H(\omega) = \left\{ 1 + \exp \left[ i\theta + iT(\omega - \omega_p) \right] \right\} / 2$$
Outline

I. Wave packet Interferences in atoms

II. Application of wave packet interferences to Quantum state holography

Measuring the time evolution of the quantum state and not only the population

III. Control of wave packet dynamics

IV. Interferences within the excitation multiphoton transition
Excited state evolution through chirped pulse excitation.

\[ \tau_c \approx 20 \text{ ps} \]

\[ \phi'' = -8 \times 10^5 \text{ fs}^2 \]

T. Mullins’ talk (parallel session)

Coherent transients produced by a chirped pulse: Interferences within a single pulse

\[ a_e(t) \propto \int_{-\infty}^{t} e^{-\ln 2 \left( \frac{t'}{\tau_c} \right)^2 - i \frac{t'^2}{2\phi''}} dt' \]

Degert et al, PRL 89, 203003 (2002)
Quantum state holography (1)

**Principle:**
- Use a first pulse to create a local oscillator in the atom
- The population in 5p (probed in real time) reflects the beating between this local oscillator and the wave function excited by the chirped pulse

**Two level system: 5s-5p²P1/2 in Rb**
- Red pump: resonant and chirped
- Yellow probe: shorter than pump and CT dynamic
- Fluorescence is monitored as function of the pump-probe delay
Two level system: 5s-5p²P1/2 in Rb

- Red pump: resonant and chirped
- Yellow probe: shorter than pump and CT dynamic
- Fluorescence is monitored as function of the pump-probe delay

\[ E_O(\omega) = H(\omega)E_I(\omega) \]

\[ H(\omega) = \left[ 1 + \exp \left( i\theta + i\phi^{(1)}(\omega - \omega_p) + i\frac{\phi^{(2)}}{2}(\omega - \omega_p)^2 \right) \right] / 2 \]
Quantum state holography (3)

From CT to …

A) $CT_A(t) \propto |a_{e1}(t) + a_{e2}(t)|^2$

B) $CT_B(t) \propto |a_{e1}(t) + ia_{e2}(t)|^2$

By combination of the two CT…
…the evolution of the atomic quantum state

By combination of the two CT...

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Coupled – Uncoupled state

\[ |\Psi_c\rangle = \Omega_1 |\phi_1\rangle + \Omega_2 |\phi_2\rangle \]

\[ |\Psi_{uc}\rangle = \Omega_2 |\phi_1\rangle - \Omega_1 |\phi_2\rangle \]

Interference phase

Condition to observe interferences:  
The coupled and uncoupled states should have different probe probabilities.

The wave packet evolves freely back and forth between the coupled and uncoupled states.
Spin-orbit precession in Rb

Fine structure: The coupled and uncoupled states belong to the uncoupled basis set. The free evolution corresponds to the spin precession around the total angular momentum. The spin of the electron is spectator during the pump step.
Spin-orbit precession in Rb

First observed in Potassium then in Rubidium


Is it possible to populate the uncoupled state first?
Can we populate directly the uncoupled state?

Use a shaped pulse with a phase shift of $\pi$ between the two resonances.
Spin orbit precession and control

- apply a $\pi$ phase step between the two excitation frequencies

The uncoupled state is first populated after the pulse.

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<tr>
<th>Without $\pi$-step</th>
<th>Coupled Uncoupled</th>
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Experimental results

The oscillations produced by the shaped pulse are phase shifted by $\pi$ with respect to the reference case.

No violation of first principles: The shaped pulse is longer than the spin-orbit precession!
This demonstration on the fine structure could be extended to molecular vibrational levels.

Can we make a non-Franck-Condon transition?

\[ \sum v a_v |v\rangle \]
Ramsey fringes with several excited states: wave packet interferences

Excitation of a superposition of the two states

Excited state population

Period $= \frac{2\pi}{\omega}$

Time delay $\tau$
Interferences between identical time-delayed quantum paths: Ramsey fringes - Experiment


- 3 0369 1 2 1 5

delay / ps

1.6 ps

Cs+/ a.u

35040 35060

Beats due to spin-orbit precession

Similar studies by
G. Fleming et al, JCP 93, 856 (1990); 95, 1487 (1991); 96, 4180 (1992)
W. J. van der Zande et al, PRL 72, 3783 (1994)

Femto group - LCAR - Toulouse
In Molecules: Wave packet interferences

In bound states

2nd wave packet

Interferences in the total cross-section (Ramsey fringes)


K. Ohmori’s talk (parallel session)
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Interferences in Ladder climbing

Quantum ladders:
- Vibration in molecules
- Rotation (within Raman process)
- Rydberg levels

Vibration in molecules
Rotation (within Raman process)
Rydberg levels
Interferences within the excitation process

- Takes advantage of the broad spectrum of ultrashort pulses:
  - Interferences between several quantum paths associated to different frequency components in multiphoton transitions
  - Control of interferences by changing the pulse shape (spectral phase)

- Many examples
  - Chirped pulses
    - Noordam et al, Chatel-Girard et al,
  - Shaped pulses
    - Silberberg et al, Dantus et al, Faucher et al

1 pulse – Two-photon transition
Na ladder climbing with **chirped pulses**

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**Na cell**

- **5S**
- **3P\(_{3/2}\)**
- **3P\(_{1/2}\)**

**Fluorescence**

- 330 nm
- 602 nm

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**CPA**

- 800 nm
- 1 kHz
- 1 mJ
- 130 fs

**NcOPA**

- 605 nm, 30 fs
- Width 25 nm
- 10 µJ

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**Stretcher**

**Fixed chirp**

**Variable chirp**

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**Fluorescence observed as a function of chirp rate**

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**Dressed states**

- \(|3S, h\nu\rangle\)
- \(|3P_{3/2}\rangle\)
- \(|3P_{1/2}\rangle\)
- \(|5S, -h\nu\rangle\)
Na (3s $\rightarrow$ 3p $\rightarrow$ 5s) at 605 nm

Contrast of 97%

$e^{i\phi''\delta_k^2}$

PRA, 70, 053414 (2004).
From 2 to many levels: numbers factorization with Gauss sums

Goal: Implement the Gauss sum in a physics experiment to factorize numbers

If $l$ is not a factor of $N$ then the phase oscillates rapidly and the sum is small.
If $l$ is a factor of $N$ then the phase is a multiple of $2\pi$ and the sum is equal to $1$.

\[
A_N^{(M)}(\ell) = \frac{1}{M+1} \sum_{m=0}^{M} \exp \left[ -2\pi i \frac{m^2 N}{\ell} \right],
\]

From 2 to many levels: numbers factorization with Gauss sums


Goal: Implement the Gauss sum in a physics experiment to factorize numbers

Number to factorize

If $l$ is not a factor of $N$, then $A^{(M)}_N(l)$ is small
If $l$ is a factor of $N$, then $A^{(M)}_N(l)$ is $1$ to $1$

...and Molecules have anharmonicities!

Femto group - LCAR - Toulouse
Wednesday Morning, January 9 2008

Plenary Session, Hans Frauenfelder, Chair

7:30 Wolfgang Schleich, Universität Ulm, “Factorization of numbers with classical and quantum interference”

Number Theory and Quantum Mechanics
Wolfgang Schleich, Chair

9:10 Ernst Rasel, Universität Hannover, “Gauss sum factorization with cold atoms”

9:30 Dieter Suter, Universität Dortmund, “Factorizing numbers with the Gauss sum technique: NMR implementations”

9:50 Béatrice Chatel, CNRS-Université Paul Sabatier-Toulouse III, “Factoring numbers with ultrashort laser pulses”
Conclusion: Interferences !!!

- Observation of the atomic quantum state during the light interaction
  - Measurement with a reference, but no spectral condition for the reference (unlike conventional interferometric methods)
  - Extension to multiple states (molecular nuclear wave packets)

- Pulse shape measurements (from $d\alpha_e/dt$):
  - Feasibility of reconstructing the phase and the amplitude using coherent transients
  - No intrinsic limitation to measure complex shapes
  - The only temporal limit is the linewidth of the atomic transition!
  - Requires a resonant bound state as a local oscillator!!

- Interferences in ladder climbing

- Towards « non Franck-Condon » transitions

  … More interferences to come !
FASTQUAST European network (starting Spring 2008)
« Ultrafast control of quantum systems by strong laser fields »

(Dijon, UC London, Imperial College, Oxford, Kassel, Aarhus, Sofia, Heraklion, Madrid, Weizmann, Fastlite, Femtolasers, APE)

☐ PhD (and postdoc) positions available soon
☐ Summer school in Cargese (Corsica, France)
  ■ 17-23 Aug 2008 – (M. Shapiro, B. Chatel, B. Girard)

« Basic concepts of femtosecond physics »