Coherent control in Atoms with ultrashort laser pulses

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Place du Capitole
Place du Capitole

Le Pont Neuf

Basilique Saint-Sernin
(1180 – 1478)
Control using shaped pulses

**Closed-loop**

- Pulse shaper
- System
- Optimization Algorithm
- GOAL

**Open-loop**

- Pulse shaper
- System
- Theory

- 2 ph. transitions: Silberberg, Nature 98
- Small molecules: Leone, JCP 98
- Quantum info: Bucksbaum, Nature 99
- 1 photon transition: Girard PRL 2002
- ....

In between...

Learning something on the interaction

- Molecules: Gerber, Science 98
- Clusters: Wöste, CP 2001
- Biological systems: Herek, Nature 2002
- Strong field: Levis, Science 2001
- ....

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2nd Sino-German workshop on Quantum Engineering, Reisensburg (22-26/9/2007)
Coherent control: opening the loop …

Open loop coherent control of simple systems

Many results:

- Raman transitions: Silberberg, Nature 02, Faucher 04, Motzkus 06…
- Small molecules: Leone et al., JCP 1998
- 1 photon transitions: Girard - Motzkus, PRL 2002
- Quantum information: Bucksbaum-Weinacht, Nature 99, PRL 98, Science 00 …
Many different ways to observe, study, control, use Quantum Interferences …

- External degree(s) of freedom: Matter waves (atoms, molecules, clusters, BEC …)
- Internal degree(s) of freedom: Ramsey fringes

In the Ultrashort world:
- Interferences of wave packets created by different laser pulses
- Interferences within the excitation process
Back to basics

Two level system ↔ laser pulses

$h\Omega(t) = \mu E(t)$

$\Omega \tau_0 > 1$

Strong field

$\Omega \tau_0 << 1$

Weak field

$\phi'' = 0$ FT limited

Rabi Oscillations

$\phi'' \neq 0$ Chirped

Adiabatic transfer

Coherent transients

In molecular potentials, the effect of chirp is opposite between weak and strong field

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Outline

I. Wave packet Interferences in atoms
II. Quantum state holography
III. Interferences within the excitation process
IV. Optics as an analog computer: Number factorization
Interferences between identical time-delayed quantum paths: Ramsey fringes - Theory

$\Psi(t) = |i\rangle + a e^{-i\omega t} |e\rangle + a e^{-i\omega (t-\tau)} |e\rangle$

*weak field regime:*

$a << 1$

$\Phi_{cc} = \omega \tau$

$\Phi_{cc} = 0 \ [2\pi]$ \ constructive interferences

$\Phi_{cc} = \pi \ [2\pi]$ \ destructive interferences
Interferences between identical time-delayed quantum paths: Ramsey fringes - Experiment

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Similar studies by
G. Fleming et al, JCP 93, 856 (1990); 95, 1487 (’91); 96, 4180 (92)
R. Jones, P. Buckbaum et al, PRL 71, 2575 (1993); 75, 1491 (95)
S.S. Vianna et al, PRA 74, 055402 (2006)
Interferences of free electron wavepackets

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(Received 24 April 2002; published 3 October 2002)

Interferences of free electron wave packets generated by a pair of identical, time-delayed, femtosecond laser pulses have been observed. Two different schemes have been used: (1) Rayleigh ionization with parallel laser polarization and (2) a two-photon transition with crossed polarizations. In both cases the intensity of light pulses is transferred to free electron wave packets. This demonstrates the potential for exciting experiments.


FIG. 1. Principle of the experiment. (a) Intensity of a pair of identical 30 fs Gaussian laser pulses separated by a time delay $\tau$. (b) $P_\alpha(\omega_\alpha)$ corresponding photoelectron spectrum for threshold electrons. Spatial distribution of the electron wave packets: (c) after creation by the pulse pair, (d) free motion of the wave packets leading to an overlap due to dispersion, and (e) long term evolution of the interference pattern.

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Wp interferences with a pulse shaper

- High Res. Pulse Shaper:
  - Phase-Amplitude control with 640 pixels.
  - Shaping window of 35 ps.
  - High complexity.
  - High amplitude dynamic (30 dB).
  - 75% power transmission.

Test of interferometric stability

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

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

1+1 = 0 to 4

Ramsey fringes
Outline

I. Wave packet Interferences in atoms
II. Quantum state holography
   Measuring the time evolution of the quantum state and not only the population
III. Interferences within the excitation process
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Application of wave packet interferences to Quantum State Holography

Excited state evolution through chirped pulse excitation

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

Degert et al, PRL 89, 203003(2002)

Cornu spiral

Re(\(a_e(t)\))

Im(\(a_e(t)\))

6d, 8s

Rb

6p-5s Fluorescence

\(\tau_c \approx 20\, \text{ps}\)

at resonance

\(\phi'' = -8 \times 10^5 \, \text{fs}^2\)
Quantum state holography I

- **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 driving field

- **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
Quantum state holography II

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 III

From CT to …

\[ CT_A(t) \propto |a_{e1}(t) + a_{e2}(t)|^2 \]

\[ CT_B(t) \propto |a_{e1}(t) + i.a_{e2}(t)|^2 \]

By combination of the two CT…
Quantum state holography IV

…the evolution of the atomic quantum state

By combination of the two CT...

Quantum state holography

- **Principle** (*Leichtle, Schleich, Averbukh, Shapiro* *PRL* 80 (1998)): Interference between a reference and an object quantum state. Several measurements at different delays provide reconstruction of the object state.

- **Two experiments**:

Our scheme is a direct measurement of the w.p. during the interaction.

- **See also** *Ohmori et al*, *PRL* 96, 093002 (2006)
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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
  - With or without intermediate states
  - 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
Two-photon transitions with chirped pulses

For $|\phi''| \gg \frac{1}{\delta^2}$: $a_e \propto \delta = \omega_{ig} - \omega_0$

$$\frac{i}{2\pi\delta} \int_{-\infty}^{+\infty} \tilde{E}(\omega_{eg} - \omega) \tilde{E}(\omega) d\omega + \frac{1}{2} \tilde{E}(\omega_{ei}) \tilde{E}(\omega_{ig})(-\text{sgn}(\phi'' \delta) + 1)$$
Two-photon transitions

\[ e^{i\phi''\delta^2} \]

\[ |a_e|^2 \]

\[ \phi'' < 0 \]

\[ 2\delta \]

\[ |g, +1\rangle \]

\[ |i, 0\rangle \]

\[ |e, -1\rangle \]

Dressed states

Excitation probability

\[ \frac{\approx 1}{\tau_c} \]

\[ \delta > 0 \]

\[ a_e^2 \]

\[ |\phi''(fs^2)| \]

\[ h\nu \]

Rb

5s → 5p → 5d

B. Noordam et al

PRL 92, PRA 94

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Ladder climbing in Rb


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Na ladder climbing with chirped pulses

\[
\begin{align*}
|3S, h\nu\rangle &\quad |3P_{3/2}\rangle \\
|3P_{1/2}\rangle &\quad |5S, -h\nu\rangle
\end{align*}
\]

Fluorescence observed as a function of chirp rate
Na (3s → 3p → 5s) at 605 nm

B. Chatel, et al.,
PRA, 70, 053414 (2004).
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From Coherent transients and the Fresnel integral …

... to the Gauss sum and the factorization of numbers!

(with Wolfgang P. Schleich (Ulm))

\[
a_e(t) \propto \int_{-\infty}^{t} e^{-2\ln 2 \left( \frac{t'}{\tau_c} \right)^2} e^{-i\frac{t'^2}{2\phi''}} dt'
\]
From 2 to many levels: numbers factorization

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.

Multipath interferences in atoms

But Atoms have not enough levels ...

... and Molecules have anharmonicities!

First successful attempts

- D. Bigourd, B. Chatel, B. Girard and W. P. Schleich, "Factorization of Numbers with the temporal Talbot effect: Optical implementation by a sequence of shaped ultrashort pulses", submitted.

Each term of the Gauss sum is « calculated » separately!

FIG. 1 (color online). Factorization interference pattern for \( N = 157573 = 13 \times 17 \times 23 \times 31 \) obtained from the Gauss...
Our experiment with shaped pulses

Sequence of shaped pulses with phases equal to the argument of the Gauss sum

\[ H(\omega) = \sum_{0}^{m} \exp\left[i\theta + im\tau(\omega - \omega_p)\right] \]

\[ \theta = \frac{2\pi m^2 N}{l} \]

Hence we have

\[ I(\omega_p) = \left| A^{(M)}_N(l) \right|^2 \]
First results

N=19043 = 137*139
with M=9 pulses

N=105 = 3*5*7
with M=4 pulses

X Theory

Exp.
Efficiency as a function of the number of pulses

N = 19043 = 137 * 139

Prediction (Schleich et al 2007):
To obtain all truncated Gauss sums of Ghost factors below 0.5, one should have

\[ M \geq 0.74^{\frac{4}{\sqrt{N}}} \approx 8 \]
Conclusion: Interferences, 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 $da_e/dt$):
  - 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!!

- Effect of chirp: hard to draw systematics!

- Factorization: Looking for direct schemes to generate the Gauss sum

... More interferences to come!
Interferences of colliding vibrational wave packets

- Two counter propagating vibrational wave packets in I₂ B state
- High resolution (1 pm – 50 fs) required to observe this transient standing wave (5 pm – 300 fs periodicity)
- Partial mapping of the wave packet pattern
- Possible « Applications »?
  - Create more complex structure
  - Probe decoherence (environment, induced perturbation)?
  - Extension to polyatomic molecules (probe vibrational energy transfer)