Coherent Control in Small Systems: Spin Dynamics, Cold Molecules....

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Coherent control:

- To take advantage of several quantum paths during the interaction.
- To play with interferences...
- How? By controlling laser parameters (energy, wavelength, spectral phases, spectral amplitude, polarisation...).
Our tools:
ultrashort pulses and pulse shaper

\[ E(t) = |E(t)| e^{i\phi(t)} \quad \xrightarrow{\mathcal{F}} \quad \tilde{E}(\omega) = |\tilde{E}(\omega)| e^{i\phi(\omega)} \]

Linear passive Filter
\[ \tilde{E}_S(\omega) = H(\omega) \tilde{E}_E(\omega) \]
with
\[ H(\omega) = A(\omega) e^{i\phi(\omega)} \]

\[ \tilde{E}_S(\omega) = M(f \gamma \omega) \tilde{E}_E(\omega) \]

Our high resolution pulse shaper

- Phase/Amplitude control over 640 pixels.
- Shaping window of 35 ps.
- 0.06 nm/pixel
- High amplitude dynamic (30 dB).
- 75% power transmission.

Three examples

- Coherent control to cool molecules

- Coherent control to manipulate spin precession in atoms

- Coherent transient revisited: Shape the pump and the probe
Cold molecules, Why?

Precise measurement

Fundamental test (e- dipole, chirality, constant variation)

Quantum information, computation, logic...

Quantum properties (dipole), BEC, BCS

Control of (Reactive) collisions: quantum chemistry
Photochemistry (photoassociation), Superchemistry (Bose-Enhancement)


Need ultra-cold molecules (T~0K) and in v=J=0
To cool molecules

From molecules

- Cryogenic cooling
- Slowing of supersonic beam
- Velocity filtering

From atoms

- External field (Feshbach) @ 1µK, 10^{12} at/cm^3
- Collision
- Photon-association @ 100µK, 10^{10} at/cm^3

GOAL

Translation cold + Vibration cold

Translation cold + Vibration Hot (not v=0)
Two approaches to cool molecules based on photo-association

- Use coherent control scheme as pump-dump: preliminary works in coll with I. Walmsley (Oxford)

- Optical pumping in coll with D. Comparat/P. Pillet (LAC-France): M. Viteau, A. Fioretti...
  - Incoherent approach
Incoherent approach

Makes cold molecules: Long Range Molecules (high $v$, but low $J$).

Several vibrational states populated

Then optical pumping to cool the vibration
Optical pumping and vibrational cooling

Viteau, Comparat, Pillet et al

To use the laser to excite all populated vibrational levels but frequency-limited in such a way to eliminate transitions from $v = 0$ level, in which molecules accumulate (creation of a dark state)
Choosing the dark state

To extend this incoherent optical pumping in order to accumulate molecules into other single selected vibrational level than the sole $v = 0$ one.

Very fast (<100 µs), $10^4$ pulses
Only few cycles to transfer 60% of the molecules → Almost no heating

Sofikitis, Chatel et al.

Using a comb to favour high FC factor
Three examples

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Quantum control using pathways interferences

Here we consider a pump probe experiment which allows to observe the dynamics of the superposition of the two intermediate states.
This dynamic can also be understood as interferences between different quantum paths.

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

The coupled state evolves freely back and forth towards the uncoupled state.
Spin-orbit precession in Rb

First observed in Potassium then in Rubidium

Spin-orbit precession in Rb

Fine structure: The coupled and uncoupled states belong to the uncoupled basis set.

The spin of the electron is spectator during the pump duration. The free evolution corresponds to the spin precession around the total angular momentum.
Changing the sign of the electric field by applying a $\pi$-step

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

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

Without $\pi$-step

With $\pi$-step

The oscillations of the coupled state are phase shifted by $\pi$ with respect to the reference.

Chatel et al, J Phys B, 41 (2008), 074023
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Coherent transients: principle

Fourier limited pulse

\[ \phi(\omega) = 0 \]

Chirped pulse

\[ \phi^{(2)} \gg \tau^2_0 \]

\[ \phi(\omega) = \frac{\phi^{(2)}}{2} (\omega - \omega_{eg})^2 \]

\[ P_e(t) = |a_e(t)|^2 \]

\[ |E(\omega_{eg})|^2 \]

\[ |E(t)|^2 \]

\[ t \text{ (fs)} \]

\[ \tau_0 \]

\[ \tau_c \]

\[ a_e(\tau) \propto \int_{-\infty}^{\tau} E(t)e^{i\omega_{eg}t} \, dt \]
Quantum holography: to follow the wave function in real time

To implement Temporal Fresnel lens


To use the coherence of the atom to reconstruct the electric field.

Monmayrant et al OL 31, 410 (2006)
Coherent Transients in the Femtosecond Photoassociation of Ultracold Molecules

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Quantum transients

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Experimental Set-up

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

Grating’s pair to chirp the probe.
Pump strongly chirped/probe TF limited

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

Pump TF limited
Probe strongly chirped
\( \text{chirp} = -1.4 \times 10^5 \text{ fs}^2 \)
\[
\begin{align*}
b_f(\tau) &= -\frac{\mu_{fe}\mu_{eg}}{4\hbar^2} \int_{-\infty}^{\infty} dt' E_s(t') e^{i\omega_{fe}t'} \int_{-\infty}^{t' + \tau} dt E_p(t) e^{i\omega_{eg}t} \\
&\quad + \int_{t' + \tau}^{+\infty} dt E_p(t) e^{i\omega_{eg}t}.
\end{align*}
\]
If now we apply an opposite chirp on the pump we are able to suppress the coherent transients

Pump strongly chirped
\( \text{Phisec}=1.4 \times 10^5 \text{ fs}^2 \)

Probe strongly chirped
\( \text{Phisec}=-1.4 \times 10^5 \text{ fs}^2 \)

Another way to characterize pulse in an unusual spectral range
Short dynamic even pulses are long

$\delta = 5\omega_0 s$

$\delta = 0$

Signal (a. u.)

Time (fs)

Case at resonance or without any intermediate state.
Preliminary work Using sum frequency mixing

\[ \chi^{(2)} \]

\[ \omega_{SFG} = \omega_p + \omega_s \]

Case of a quadratic phase

Shaped pump
Short probe

Shaped pump
Shaped probe
The final state acts as a spectral filter
Case of a cubic phase

A sort of focus point
Both in time and spectral domain!
- Experiment in progress
- A new spectroscopic approach
- Investigation to use HHG as either the pump or the probe (in coll P. Salières -CEA)
Conclusion

- Coherent control: a way to cool molecules
- Coherent control: To manipulate the spin by using shaped pulses
- To shape pump and probe could lead to a new spectroscopic approach: work under study
- FASTQUAST European network (PhD and postdoc positions)
  [http://icb.u-bourgogne.fr/fastquast/](http://icb.u-bourgogne.fr/fastquast/)