

Molecular alignment by two-color laser fields

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Outline

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3 Results

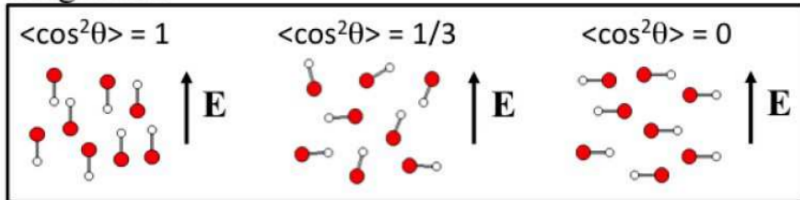
- One-color pulse
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Introduction: Molecules alignment

- Alignment has crucial applications in chemical reaction dynamics and stereochemistry related to molecular structures¹. Strongly aligned structures play an important role in quantum computing².
- Aligned molecules have been used to generate high harmonic generations (HHGs)³. The high-harmonic generation (HHG) process describe atoms in intense laser field, which show non linear behaviour and emit coherent radiations of multiple frequencies.

Alignment



¹Itatani, J. *et al.* English. *Nature* **432**, 867–871 (2004), Torres, R. *et al.* English. *Physical review letters* **98**, 203007 (2007).

²Shapiro, E. *et al.* English. *Physical Review A* **67**, 013406 (2003).

³Hay, N. *et al.* English. *Physical Review A* **65**, 053805 (2002).

Introduction: Molecules alignment

- Ionization of polar molecules are also crucially dependent on aligned molecules⁴. Multiphoton ionizations in the strong field strongly depend on the angle between the molecular axis and field polarizations⁵. When ICl molecule exposed to a 100 fs laser pulse and it is polarized parallel to alignment, up to six electrons can be easily removed due to the ionizing field, however, if the molecule is polarized perpendicular to field, it is not possible to remove more than two electrons.
- The investigations of a diatomic molecules interaction with external laser fields are highly relevant due to prospects for one of the possible realisation of a qubit and application to the quantum computing schemes⁶.

⁴ Petretti, S. *et al.* English. *Physical review letters* **104**, 223001 (2010).

⁵ Zuo, T. & Bandrauk, A. D. English. *Physical Review A* **52**, R2511 (1995).

⁶ DeMille, D. English. *Physical Review Letters* **88**, 067901 (2002), Ni, K.-K. *et al.* English. *Chemical science* **9**, 6830–6838 (2018).

Introduction: Molecules alignment

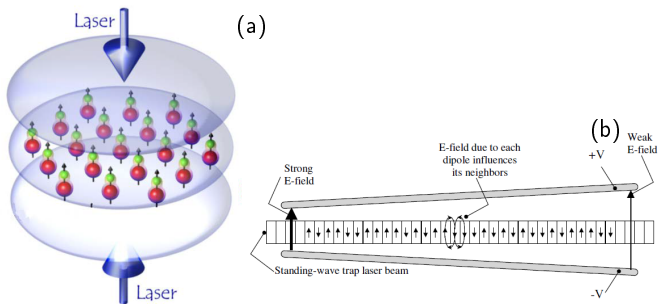


Fig.1 The schematic representation of the experimental molecule alignment methods: laser field (a) or/and static electric field (b).

Rigid rotor model

We consider a linear rigid rotor with a Hamiltonian of the form

$$\tilde{H} = B J^2 + \tilde{V}(\theta, t) \quad (1)$$

where $B = \hbar^2/2I$ is the rotational constant (with I the moment of inertia),

$$J^2 = -\frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial}{\partial \theta} \right) - \frac{1}{\sin^2 \theta} \frac{\partial^2}{\partial \phi^2} \quad (2)$$

is the operator of the angular momentum squared, and $\theta \in [0, \pi]$ and $\phi \in [0, 2\pi]$ are the polar and azimuthal angles⁷.

⁷Arfken, G. & Weber, H. *Mathematical Methods For Physicists International Student Edition*. English (Elsevier Science, 2005).

TDSE and Electric field

For a dimensionless Hamiltonian

$$H \equiv \tilde{H}/B = J^2 + V(\theta, \tau) \quad (3)$$

and dimensionless time $t \equiv B\tilde{t}/\hbar$, the corresponding time-dependent Schrodinger equation (TDSE) takes the form

$$i \frac{\partial}{\partial t} \psi(\theta, \phi, t) = H \psi(\theta, \phi, t). \quad (4)$$

The corresponding driving potential $V \equiv \tilde{V}/B$ is given by

$$V(\theta, \tau) = -\mu \cos \theta F(t) - \frac{1}{2} (\Delta\alpha \cos^2 \theta + \alpha_{\perp}) F^2(t) \quad (5)$$

where θ is the Euler angle between the internuclear molecular axis and the LFF Z-axis, μ the permanent electric dipole moment, $\Delta\alpha = \alpha_{\parallel} - \alpha_{\perp}$ the polarizability anisotropy, with α_{\perp} and α_{\parallel} being its perpendicular and parallel components.

TDSE and Electric field

One or two-color laser field linearly polarized along the laboratory fixed frame Z-axis is considered. The corresponding electric field $F(t) = F(t)Z$ is given by the biharmonic function

$$F(t) = F_{\omega}(t) \cos(\omega(t - t_1)) + F_{2\omega}(t) \cos(2\omega(t - t_2)), \quad (6)$$

with ω and 2ω , $F_{\omega}(t)$ and $F_{2\omega}(t)$, t_1 and t_2 being the laser frequency, electric field strength (laser pulse envelope) and laser pulse time delay of corresponding harmonic.

These parameters allow for comparing the dynamics that results from kicks of different shapes, lengths, and strengths.

Note that the energies are expressed in terms of the rotational constant B and that a rotational period amounts to $\tau_r = \pi$.

Rigid rotor model

- The time-dependent Schrödinger equation is solved by the combining the Strang-Marchuk split-operator method⁸ for the time variable, and the Discrete Variable Representation approach to the solving the PDE on each time step.
- The time-dependent Schrödinger equation is solved assuming that at $t = 0$ the molecule is in a field-free eigenstate $\psi(\Omega, t = 0) = Y_{J,M}(\Omega)$.
- In this work, the field-dressed rotational dynamics is analyzed in terms of the alignment expectation values

$$\langle \cos^2 \theta \rangle = \int \psi^*(\Omega, t) \cos^2 \theta \psi(\Omega, t) d\Omega, \quad (7)$$

with $\psi(\Omega, t)$ being the time-dependent wave function. The wave function $\psi(\Omega, t)$ and the expectation values depend on the laser field parameters (6).

⁸Marchuk, G. I. English. *Handbook of numerical analysis* **1**, 197–462 (1990).

One-color gaussian pulses: different pulse duration (FWHM)

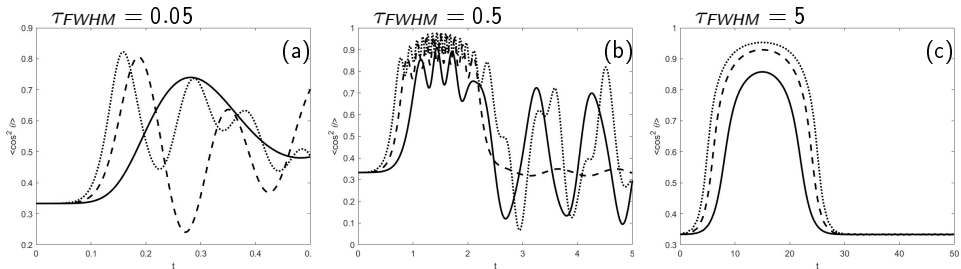


Fig.2. The alignment expectation value $\langle \cos^2(\theta) \rangle$ for the one-color pulse for different pulse duration. $F_0 = \max(F_\omega(t)) = \max(F_{2\omega}(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line.

One-color gaussian pulses: different pulse duration (FWHM)

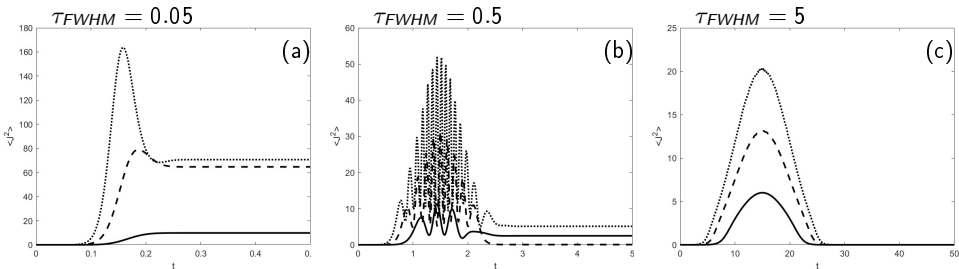


Fig.3. The angular momentum J^2 for the one-color pulse for different pulse duration. $F_0 = \max(F_\omega(t)) = \max(F_{2\omega}(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line.

Two-color gaussian pulses: different pulse duration (FWHM)

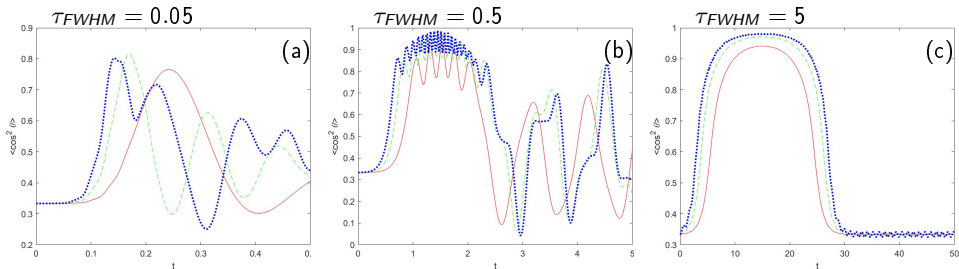


Fig.4. The alignment expectation value $\langle \cos^2(\theta) \rangle$ for the two-color pulse for different pulse duration. $F_0 = \max(F_\omega(t)) = \max(F_{2\omega}(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line.

Two-color gaussian pulses: different pulse duration (FWHM)

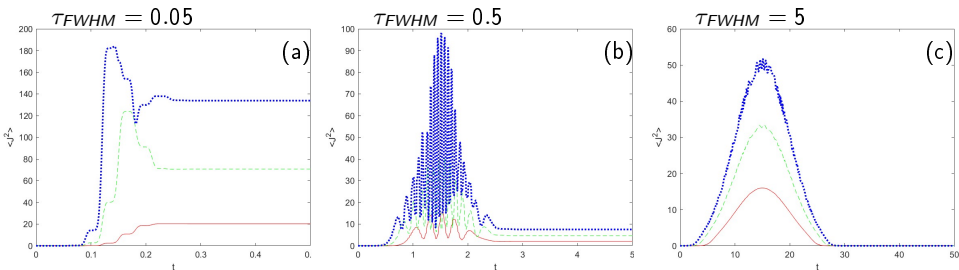


Fig.5. The angular momentum J^2 for the two-color pulse for different pulse durations. $F_0 = \max(F_\omega(t)) = \max(F_{2\omega}(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line.

Two-color gaussian pulses: different harmonics amplitudes ratio

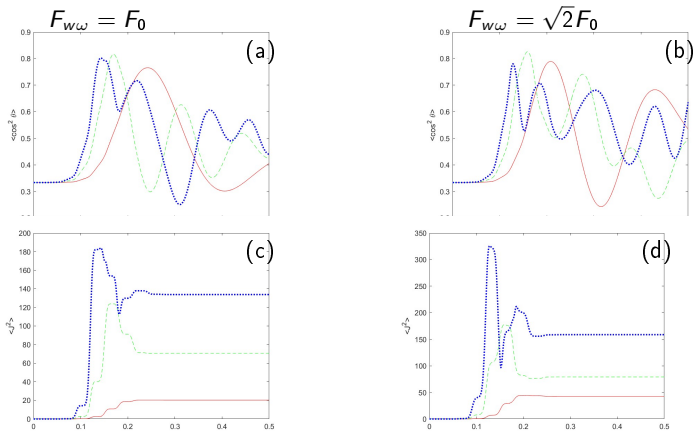


Fig.6. The alignment $\langle \cos^2(\theta) \rangle$ and the angular momentum J^2 expectation values for the two-color pulse for different harmonics amplitudes ratio. $F_0 = \max(F_\omega(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line. The FWHM of the pulses is fixed: $\tau_{FWHM} = 0.05$.

Two-color gaussian pulses: delay of the second harmonic pulse

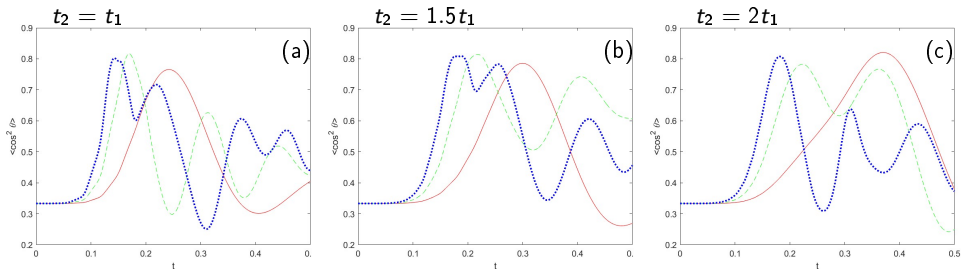


Fig.7. The alignment expectation value $\langle \cos^2(\theta) \rangle$ for the two-color pulse for different second harmonic pulse delay. $F_0 = \max(F_\omega(t)) = \max(F_{2\omega}(t)) = 100$ is marked by the solid line, $F_0 = 400$ — by the dashed line, $F_0 = 900$ is marked by the dot-dashed line. The FWHM of the pulses is fixed: $\tau_{FWHM} = 0.05$.

Conclusions

- The good agreement with results of the Ref^a was obtained for molecules alignment with one-color pulse. The influence of pulses with longer duration show almost adiabatic response of the rigid rotor. While shorter pulses lead to the strongly nonadiabatic behaviour with oscillating alignment even after pulse switch-off. The stronger laser field amplitudes allow to get higher molecules alignment during the pulse. The nonadiabatic behaviour also shows revivals of the alignment after short pulses.
- For two-color parallel laser fields the number of alignment cosine oscillations is increased comparing to the one-color case. While shapes of the dependences of the alignment cosine on the pulses duration are close to the one-color case, the peaks of the alignment cosine dependencies are higher.
- Obtained results show that increasing of the intensity of the second harmonic pulse leads to the higher alignment cosine during the pulses for short pulse durations.
- The variation of the second harmonic pulse delay (relative to the first harmonic pulse) allows to change interfering pattern of the pulses on the wave function, in order to increase the duration of the alignment for short pulses.

^aOrtigoso, J. et al. English. *The Journal of chemical physics* **110**, 3870–3875 (1999).

Thank you for your attention!