

Orientation of linear asymmetric molecules with combined linearly and elliptically polarized two-color laser fields

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Orientation of linear asymmetric molecules can be achieved by using a nonresonant two-color laser field [1,2]. The two-color laser field is given by $\mathbf{E}(t) = [E_\omega(t) \cos(\omega t) + E_{2\omega}(t) \cos(2\omega t + \Phi)] \hat{z}$ where $E_\omega(t)$ and $E_{2\omega}(t)$ are the time envelopes of the fundamental pulse and the second harmonic pulse, respectively. The phase difference between these two pulses is Φ . The time-dependent interaction Hamiltonian for a linear molecule is given by:

$$H(t) = B\mathbf{J}^2 - \frac{1}{4}[(\alpha_{\parallel} - \alpha_{\perp}) \cos^2 \theta] (E_\omega^2(t) + E_{2\omega}^2(t)) - \frac{1}{8}((\beta_{\parallel} - 3\beta_{\perp}) \cos^2 \theta + 3\beta_{\perp}) \cos \theta \cos \Phi E_\omega^2(t) E_{2\omega}(t). \quad (1)$$

Here, $\mathbf{J}^2$ is the squared angular momentum, B is the rotational constant of the molecule, θ is the angle between the molecular axis and $\hat{z}$, $\alpha_{\parallel}$, $\alpha_{\perp}$ and $\beta_{\parallel}$, $\beta_{\perp}$ are the polarizability and hyperpolarizability components parallel and perpendicular to the molecular axis, respectively. For OCS molecules, the potential part in Eq. (1) is plotted in Fig.1. The asymmetric part of the total potential shown in Fig. 1(b) is almost 100 times smaller than the total potential, making it difficult to orient general molecules like OCS [3]. On the other hand, Some molecules such as FCN have larger hyperpolarizability by an order of magnitude [4], allowing higher degrees of orientation.

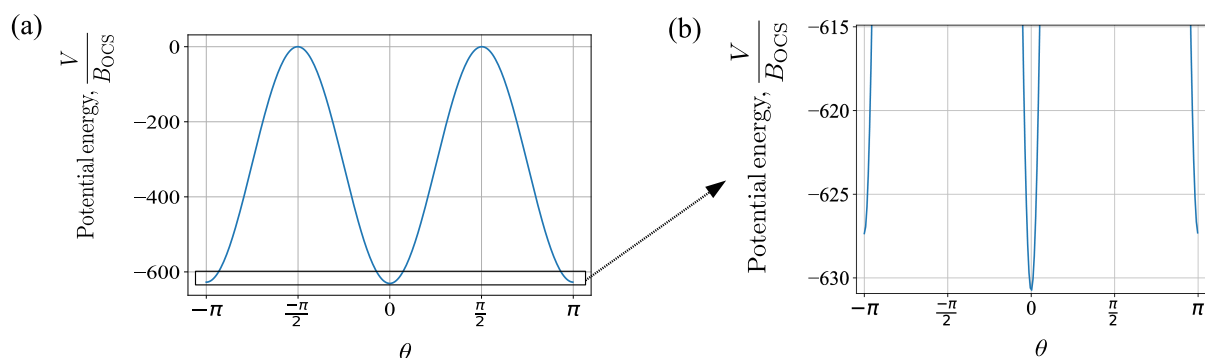


Fig. 1: (a) Interaction potential of a OCS molecule under a two-color linearly polarized laser field, (b) The enlarged asymmetric part of the potential responsible for orientation.

We introduce an elliptically polarized second harmonic pulse in the original two-color laser field as: $\mathbf{E}(t) = [E_\omega(t) \cos(\omega t) + E_{2\omega}(t) \cos(2\omega t + \Phi)] \hat{z} + E'_{2\omega}(t) \sin(2\omega t + \Phi) \hat{x}$, when the interaction potential, after taking an average over one laser oscillation, is given by:

$$H(t) = B\mathbf{J}^2 - \frac{1}{4}(\alpha_{\parallel} - \alpha_{\perp}) [\cos^2 \theta (E_\omega^2(t) + E_{2\omega}^2(t)) + \sin^2 \theta \cos^2 \phi E_{2\omega}'^2(t)] \pm \frac{1}{8}((\beta_{\parallel} - 3\beta_{\perp}) \cos^2 \theta + 3\beta_{\perp}) \cos \theta E_\omega^2(t) E_{2\omega}(t), \Phi = \pi, 0. \quad (2)$$

In Eq. (2), the $\sin^2 \theta \cos^2 \phi E_{2\omega}'^2(t)$ term serves to increase the tunnel transition probability from the shallower potential well to the deeper one thanks to the potential distortion along the azimuthal angle ϕ with the potential minima located at $\phi = 0$ and $\phi = \pi$. In this presentation, the application of this method on molecules with different orders of hyperpolarizability such as OCS and FCN will be discussed.

References: [1] T. Kanai and H. Sakai, J. Chem. Phys. **115**, 5492 (2001), [2] K. Oda et al., Phys. Rev. Lett. **104**, 213901 (2010), [3] G. Maroulis and M. Menadakis, Chem. Phys. Lett. **494**, 144 (2010), [4] G. Maroulis and C. Pouchan, Chemical Physics **215** (1997) 67-76.