All-optical three-dimensional molecular orientation 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 linearly polarized two-color laser field [1,2]. All-optical three-dimensional molecular alignment and orientation can also be realized by applying linearly polarized two-color laser fields, where the polarizations of the fundamental pulse and the second harmonic pulse are crossed [3,4]. Here, we propose a novel approach of realizing all-optical three-dimensional molecular orientation, where a linearly polarized fundamental pulse and an elliptically polarized second harmonic pulse are applied: $\mathbf{E}(t) = [E_\omega(t)\cos(\omega t) + E_{2\omega}(t)\cos(2\omega t + \Phi)]\,\hat{z} + E'_{2\omega}(t)\sin(2\omega t + \Phi)\hat{x}$ [5]. Here, $E_\omega(t)$ is the time profile of the fundamental pulse. $E_{2\omega}(t)$ and $E'_{2\omega}(t)$ are the time envelopes along the major and minor axes of the elliptically polarized second harmonic pulse, respectively. The interaction potential, after taking an average over one ω-laser oscillation, is given by:

$$\begin{aligned}\hat{H}_i = &-\frac{1}{4}\alpha_{zy}\left[\cos^2\phi\sin^2\theta E'^2_{2\omega}(t) + \cos^2\theta\left(E^2_\omega(t) + E^2_{2\omega}(t)\right)\right] \\ &-\frac{1}{4}\alpha_{xy}\left[(\cos\phi\cos\theta\cos\chi - \sin\phi\sin\chi)^2 E'^2_{2\omega}(t) + \cos^2\chi\sin^2\theta\left(E^2_\omega(t) + E^2_{2\omega}(t)\right)\right] \\ &\pm\frac{3}{8}\left(\beta_{zxx}\cos^2\chi + \beta_{zyy}\sin^2\chi\right)\cos\theta\sin^2\theta E^2_\omega(t)E_{2\omega}(t) \\ &\pm\frac{1}{8}\beta_{zzz}\cos^3\theta E^2_\omega(t)E_{2\omega}(t),\end{aligned} \tag{1}$$

where the phase differences between the two wavelengths are $\Phi = \pi$ or 0 for a plus or minus sign, respectively, $\alpha_{zy} = \alpha_{zz} - \alpha_{yy}$, and $\alpha_{xy} = \alpha_{xx} - \alpha_{yy}$. The simulated degrees of alignment $\langle\langle\cos^2\theta\rangle\rangle$ and orientation $\langle\langle\cos\theta\rangle\rangle$ for iodobenzene molecules as a function of the peak intensity of the fundamental pulse I_ω, under the assumption of adiabatic approximation, are shown in Figs. 1 (a) and (b), respectively. In the calculations, the peak intensities along the major and minor axes of the elliptically polarized second harmonic pulse are also increased as a function of the peak intensity of the fundamental pulse: $I_{2\omega} = 1.1I_\omega$ and $I'_{2\omega} = 0.98I_\omega$.

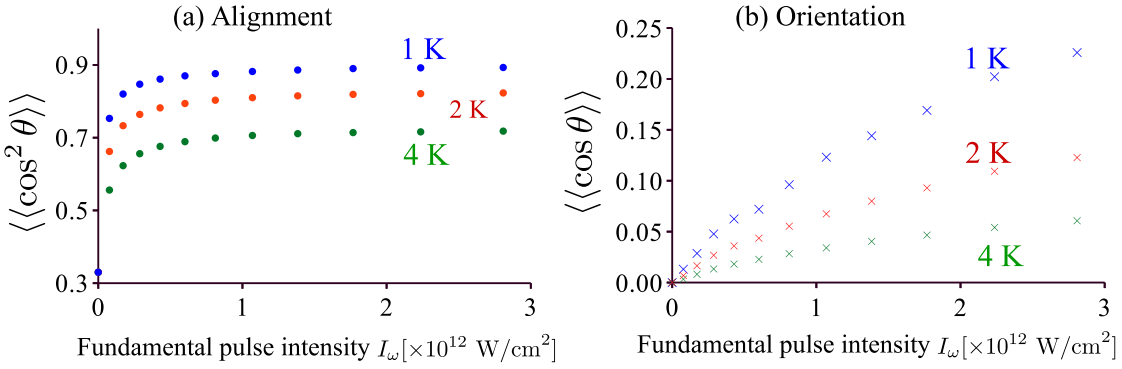

Fig. 1: Degrees of (a) alignment $\langle\langle\cos^2\theta\rangle\rangle$ and (b) orientation $\langle\langle\cos\theta\rangle\rangle$ of iodobenzene molecules as a function of the peak intensity of the fundamental pulse.

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] N. Takemoto and K. Yamanouchi, Chem. Phys. Lett. **451**, 1 (2008), [4] K. Lin *et al.,* Nat. Commun. **9**, 5134 (2018), [5] M. M. Hossain, PhD thesis (The Univ. of Tokyo) (2019).