All-optical orientation of linear molecules with combined linearly and elliptically polarized two-color laser fields

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Orientation of asymmetric molecules can be achieved with an intense nonresonant two-color laser field [1,2]. For linear molecules such as carbonyl sulfide (OCS), linearly polarized two-color laser field is used (Fig.1 a). Alignment is realized because molecules interact with the laser field through the anisotropic polarizability, which creates a symmetric potential. On the other hand, orientation takes place because the anisotropic hyperpolarizability interaction creates an asymmetric potential [3]. The magnitudes of the symmetric potential responsible for alignment and of the asymmetric potential responsible for orientation are determined by the polarizability and hyperpolarizability of the molecules, the intensities of the laser fields, and the relative phase difference between the two wavelengths. Since the polarizability is generally larger than the hyperpolarizability by a few orders of magnitude [4], simply increasing the laser intensities does not necessarily produce higher degrees of orientation. Although the asymmetry in the potential increases, a symmetric potential due to the polarizability interaction also becomes deeper, making the tunneling to a single direction more difficult [2,5]. To avoid this unfavorable situation, we propose using an elliptically polarized second-harmonic for the two-color laser fields as shown in Fig. 1(b). By tuning the ellipticity of the second-harmonic, it is possible to suppress the effect of the symmetric potential adiabatically created by the polarizability interaction while increasing the asymmetric potential. This strategy is useful for general molecules with smaller hyperpolarizability anisotropy and larger polarizability anisotropy.

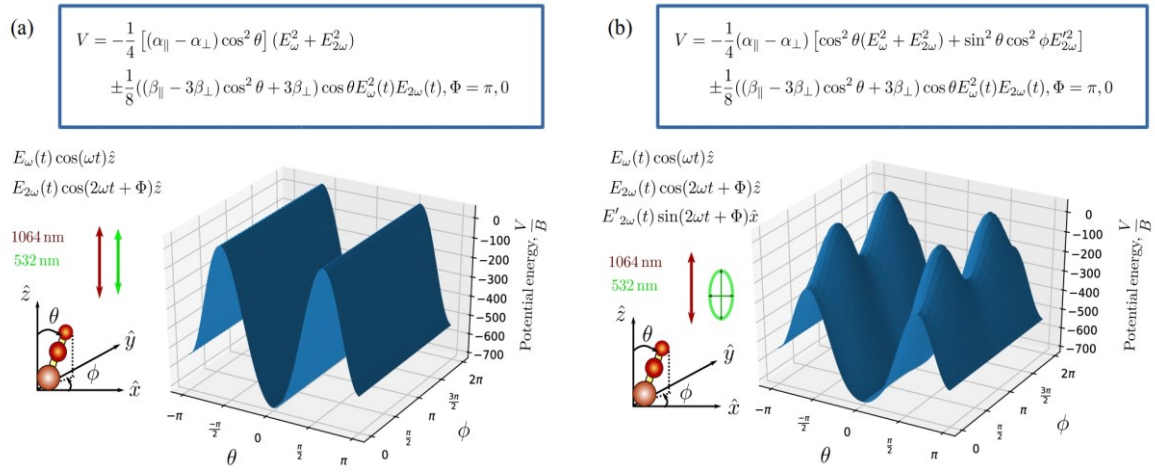

Fig. 1: Interaction potential for orientation of OCS molecules (a) when a linearly polarized two-color laser field is used, (b) when an elliptically polarized second harmonic for the two-color laser fields is used. The asymmetry between the two minima is rather exaggerated.

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