Orientation of linear molecules by two-color laser fields with orthogonal polarizations

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Molecular orientation methods using parallel polarized nonresonant two-color laser pulses have been proposed [1] and demonstrated [2]. In the method, the anisotropic hyperpolarizability interaction with the two-color fields is exploited. Recently, we have proposed some strategies to improve molecular orientation by optimizing relative delay and intensities of two-color laser pulses [3]. In the present work, we demonstrate that two-color fields with orthogonal polarizations can improve the adiabaticity of the orientation dynamics, leading to higher degrees of orientation compared to the parallel polarized two-color fields [4].

Figure 1 shows orientation dynamics of the OCS ground state pumped by the two-color laser fields with parallel (a) and orthogonal (b) polarizations. For the two polarization configurations, other conditions such as the intensities and the pulse widths are the same. As can be seen from the comparison between (a) and (b), the orthogonal polarization configuration provides higher degrees of molecular orientation $\langle \cos\theta \rangle$. This is due to the temporally synchronized formations of alignment and orientation potentials in the direction along the 2ω polarization, which reduces the nonadiabatic effects. The strongly oriented molecules in the presence of ns two-color laser pulses can be transferred to the completely field-free condition by employing the plasma shutter technique [5,6]. More details of the orientation dynamics and the future prospects will be given in the presentation.



Fig. 1. Degrees of orientation $\langle \cos\theta \rangle$ as a function of time for parallel (a) and orthogonal (b) polarization configurations. The peak intensities of the fundamental (ω) and the second harmonic (2ω) laser pulses are 3 $\times 10^{11}$ W/cm² and 5 $\times 10^{11}$ W/cm², respectively. Results from the time-dependent Schrödinger equation (TDSE), which naturally include the nonadiabatic effects, and ones from the adiabatic approximation (AA) are shown. In the orthogonal polarization configuration, the TDSE result is well reproduced by the AA one, meaning that the orientation dynamics is quasi-adiabatic. The higher degrees of orientation are achieved by using the orthogonally polarized two-color fields.

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