Quantum control of molecular process by using phase-controlled lights and its application to instrumentation frontier

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Intense phase-controlled two-color laser fields consisting of a fundamental light and a second harmonic light (hereafter the ω+2ω scheme) have been investigated to achieve quantum control of tunneling ionization (TI) in the space domain and the resultant selective ionization of oriented molecules (SIOM) in simple molecules [1-6]. The principle of quantum control of TI in the space domain and the resultant SIOM are described below. The total electric field of the linearly polarized ω+2ω pulse is given by $E(t) = E_1 \cos(\omega t) + E_2 \cos(2\omega t + \phi)$, where E_1 and E_2 are the amplitudes of the electric fields and ϕ is the relative phase difference between the fundamental and the second-harmonic light. The amplitude of the electric field in the positive (negative) direction is twice that in the negative (positive) direction when $\phi = 0$ ($\phi = \pi$) and $E_1 = 2E_2$. According to the molecular Ammosov-Delone-Krainov (ADK) model, ionized electrons are much more strongly extracted by the tunneling process from the high-density part of electronic clouds along the direction of an electric field. For monochromatic laser fields with a symmetric waveform, electrons are removed at the same rate from molecules in both the negative and positive directions along the laser polarization. In contrast, when an asymmetric two-color ω+2ω field induces TI of molecules with an asymmetric highest occupied molecular orbital (HOMO) structure, electrons are much more likely to be removed from the high-density part of the HOMO opposite to the direction of the electric field vector at its maximum. Consequently, molecules initially oriented in the direction of the field maximum are much more strongly ionized than randomly oriented molecules. We have experimentally demonstrated that, as a consequence of directionally asymmetric TI, SIOM induced by phase-controlled ω+2ω laser fields reflects the asymmetric geometry of the HOMO structure. SIOM is free of laser wavelength constraints and is observed over a wide range of pulse duration from the fs to the ns regime[5]. Furthermore, SIOM is free of the constraints of size, weight[6], and polarity of molecules[3,4], and this is an advantage compared to dynamic molecular orientation, since it is difficult to orient large heavy molecules that require large torques at practical laser intensities and it is impossible to orient nonpolar molecules with asymmetric structures.

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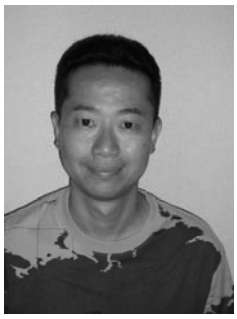

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