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二色位相制御強レーザーパルスによる CO_2 分子の選択的結合切断

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Asymmetric Coulomb explosion of CO_2 in phase-locked two-color intense laser fields

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Laser pulse shaping has provided a unique approach to control reaction dynamics in the strong field regime. Compared with complex shaping schemes using a spatial light modulator [1], ω-2ω two-color mixing offers a simple method to control chemical reactions through spatial asymmetry of laser electric fields, as demonstrated in orientation-selective ionization of asymmetric molecules [2] and directional ejection of D^+ from D_2^+ [3]. Here we apply the two-color asymmetric laser pulses to selective breaking of one of the two equivalent C–O bonds of CO_2^{2+} in the Coulomb explosion process, $CO_2^{2+} \rightarrow CO^+ + O^+$, and discuss the dependence of the two-color relative phase and the laser field intensity.

The output of a femtosecond Ti:sapphire laser system (800 nm, 45 fs) was introduced into a phase-locked two-color intense laser pulse generator. The two-color laser electic field can be expressed as $E_{\omega+2\omega} = E_\omega\cos(\omega t) + E_{2\omega}\cos(2\omega t + \phi)$, where E_ω and $E_{2\omega}$ are the amplitudes of 800 nm and 400 nm laser electric fields, respectively, and ϕ is the relative phase which alters the laser pulse shape. The ω–2ω generator is an inline optical setup consisting a β-BBO crystal to generate the second harmonic light (400 nm, 110 fs), α-BBO crystals to compensate the relative phase between the two lights, and a pair of silica wedge plates to alter ϕ, which was locked by a feedback loop utilizing 2ω–2ω spectral interference [4]. The two-color laser pulse was focused onto the CO_2 introduced as a molecular beam, with a concave mirror (f = 75 mm) in a coincidence momentum imaging chamber. Fragment ions produced from CO_2 in the interaction region were accelerated to a position sensitive detector by four electrodes in the velocity mapping configuration. The three-dimensional momentum $\boldsymbol{p}(p_x, p_y, p_z)$ was obtained for each ion from the flight time (t) and position (x, y). For the coincidence detection, momentum matching condition $|\boldsymbol{p}_{CO^+} + \boldsymbol{p}_{O^+}| < 20$ a.u. was used.

The momentum distribution of O^+ fragment ions, detected in coincidence with CO^+, in a one-color laser field (800 nm, 5.1×10^{14} W/cm^2) is shown in Fig. 1(a). The corresponding images for two-color laser fields (800 + 400 nm, 5.2×10^{14} W/cm^2) are shown in Figs. 1(b), and (c), where the differences from the momentum images averaged over ϕ ($0 \le \phi < 2\pi$) are plotted. It is clearly seen that positive signals are observed on the larger electric field amplitude side, as depicted below in the figure, indicating that C–O bonds are selectively broken on the larger electric field amplitude side. For quantitative discussions, the asymmetric distribution of the O^+ fragments was evaluated by the asymmetric parameter, $A(\phi) = (Y_+(\phi) - Y_-(\phi))/(Y_+(\phi) + Y_-(\phi))$, where $Y_+(\phi)$ and $Y_-(\phi)$ are the yields of O^+ fragment ions, observed with positive and negative momenta as indicated in Fig. 1(a). The asymmetric parameter of O^+ fragment ions as a function of the relative phase ϕ is shown in Fig. 1(d).

The obtained results show a clear dependence on ϕ, indicating that bond scission can be controlled by the relative phase between the ω and 2ω laser fields [4]. This is in good agreement with a theoretical prediction based on the time-dependent adiabatic state approach [5], showing that the deformation of the CO_2^{2+} potential by the asymmetric laser field is responsible for the selective bond scission. However, at lower laser intensities, an increase in amplitude of the asymmetric parameter and a positive phase shift were identified, suggesting contributions from a different control mechanism.

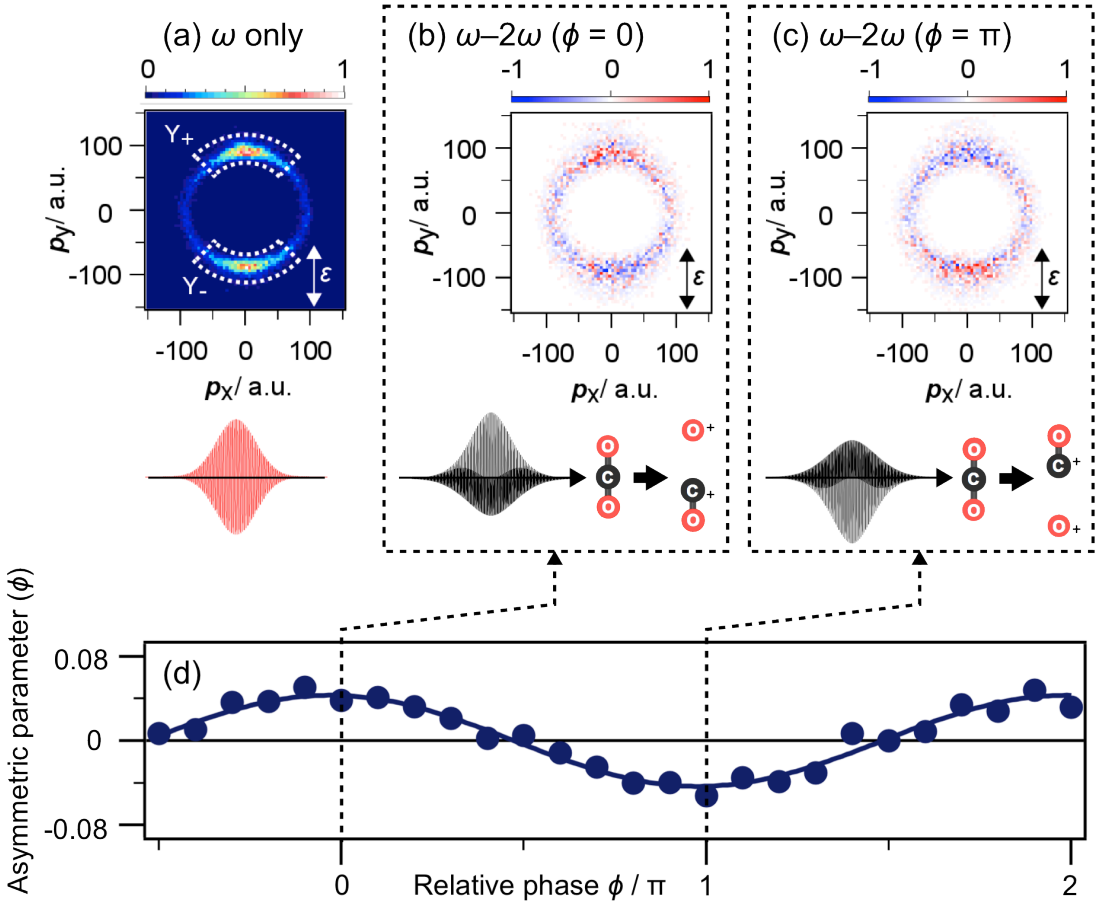

Figure 1: (a) Momentum image of the O^+ fragments produced in a one-color laser field (800 nm, 5.1×10^{14} W/cm^2). (b), (c) The difference momentum images, for $\phi = 0$ and π, respectively, in two-color laser fields (800 + 400 nm, 5.2×10^{14} W/cm^2). The laser polarization direction is noted with the double-headed arrow for all momentum images. The corresponding laser electric fields are shown below. (d) Asymmetric parameter plotted as a function of the relative phase. The solid line is a numerical fit by a sine function.

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