The recent developments in strong-field physics make it possible to probe the molecular structure and electron dynamics with attosecond and Ångström resolutions [1–4]. As a potential tool of imaging the molecular electron dynamics, molecular orbital tomography (MOT) based on high-order harmonic generation (HHG) has attracted a great deal of attention in the past decade [5–9]. This tomographic method was first proposed and successfully carried out by J. Itatani et al. [5–9] to reconstruct the highest occupied molecular orbital (HOMO) of N2 in experiment. Since then, much effort has been expended to extend the MOT method to molecules other than N2 [8,9]. It has been shown that the MOT method can be directly applied to the symmetric molecular orbitals. While for asymmetric molecular orbitals, to reconstruct the orbitals from HHG, the returning wave-packets should be controlled to recollide with the parent ion from only one direction [9].

It was proposed that the unidirectional recollisions can be achieved by using an extremely short tailored laser pulse [9]. In this scheme, by stabilization and control of the carrier-envelope phase (CEP), the returning wave-packet can be controlled. However, this scheme requires a single-cycle pulse with a stabilized and controllable carrier-envelope phase, which is a rather stringent requirement. This kind of laser pulse with a sufficient intensity is still not available for many laboratories. Therefore, a method with less stringent experimental conditions to achieve the unidirectional recollisions is preferred for tomographic reconstruction of asymmetric molecular orbitals. It has been shown that the two-color laser field is an efficient tool to control the electron dynamics. For example, a tailored two-color laser field has been used to control the electron dynamics for producing attosecond pulses [10,11]. In this Letter, we theoretically demonstrate a method for tomographic imaging of asymmetric molecular orbitals with a multicycle two-color laser pulse. By adjusting the relative phase of the two fields, the returning wave-packets are forced to recollide from one direction for all the orientations of the molecule. Thus the reconstruction of the asymmetric orbitals can be carried out with multicycle laser field. This releases the stringent requirement of a single-cycle pulse with a stabilized and controllable carrier-envelope phase for the tomographic imaging of asymmetric molecular orbitals. © 2013 Optical Society of America

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Fig. 1. Ionization rate as a function of the ionization time (blue solid line) for the two-color field with $\phi = 0.0\pi$ (panel a) and $\phi = 0.9\pi$ (panel c). The corresponding electric fields are also depicted by the red dashed line. The probability that an electron returns with momentum $k$ for $\phi = 0.0\pi$ and $\phi = 0.9\pi$ are presented in panel b and panel d, respectively.

is normalized to 1. The CO molecule is oriented at 0°. The corresponding electric fields are also displayed by the red dashed lines in this figure. As shown in the first row for $\phi = 0.0\pi$, the tunnel ionization of the electron is confined around the peaks of the electric field with positive amplitude. Correspondingly, the electrons recollide with positive momentum, as shown in panel b. In second row for $\phi = 0.9\pi$, the electron is ionized around the peaks with negative amplitude and then all the electrons return with negative momentum. For both cases, the returning electrons are controlled to approach the parent ion from one side, which is also suggested in ref. [1].

To further demonstrate the efficient control of the returning electron by the two-color fields, the fraction of the electrons with negative momentum as a function of the relative phase $\phi$ is presented in Fig. 2. The wavefunction of CO from the Gaussian 03 $ab$ initio code [14] is also depicted in Fig. 2a, where the orientation angle is 0°. According to the MOT theory, the unidirectional recollision is required for all the orientations of the molecule. Therefore, the momentum distribution of the returning electron for other orientations are also investigated. The results for molecule CO oriented at 0° (blue line), 90° (black line) and 180° (red line) are presented in Fig. 2(b). As shown in Fig. 2(b), by adjusting the relative phase of the two fields, the two-color multicycle laser pulse can provide an efficient control of the returning wave-packet. For the cases $0.7\pi \leq \phi \leq 1.0\pi$, the electron returns are well confined to only one direction for all the orientations. This offers the possibility of the reconstruction of asymmetric molecular orbitals with multicycle laser pulse.

Fig. 2. (a) A normalized two-dimensional projection of the highest occupied molecular orbital of CO obtained from the Gaussian 03 $ab$ initio code. The orientation angle is at 0°. (b) Fraction of the electron returning with negative momentum as a function of the relative phase $\phi$ for three different orientations of CO molecule. The blue, black and red lines correspond to the orientation angle 0°, 90° and 180°, respectively.

To reconstruct the HOMO of CO, the odd harmonics 11 to 101 at 19 different angles between 0 and 180°

Fig. 3. (a) The harmonic spectrum of CO oriented at the angle 0° for the two-color field with $\phi = 0.9\pi$. The double arrow indicates the spectral range (harmonics 11 to 101) sampled for orbital reconstruction. (b) The corresponding time-frequency distribution of HHG.

In the following, we choose the two-color multicycle laser fields with $\phi = 0.9\pi$ to carry out the reconstruction of the HOMO of CO. The reference Kr atom with the same ionization potential (0.519 a.u.) as that of the HOMO of CO is used [13]. The complex amplitudes of the high-order harmonics are calculated with the SFA model [15]. In Fig. 3, the high-order harmonic spectrum (panel a) and the corresponding time-frequency distribution of HHG (panel b) are presented for the orientation angle 0°. From Fig. 3(b), one can see that the emission of the high-order harmonics are well confined to the time when the direction of the electric field is positive. Therefore, the wave-packets approach the core mainly from one side, which is consistent with the result obtained within semi-classical model as shown in the second row of Fig. 1.

To reconstruct the HOMO of CO, the odd harmonics 11 to 101 at 19 different angles between 0 and 180°
are sampled. Following the reconstruction procedure in the velocity form based on the plane wave approximation [1], the HOMO of CO is reconstructed by using the dipole matrix elements projected perpendicular to the internuclear axis. In Fig. 4(a), the normalized real part of the reconstructed orbital is presented with the same color scale as that of Fig. 2(a). As shown in Fig. 2(a) and Fig. 4(a), the structure of the HOMO of CO is well reproduced with the multicycle two-color laser field. In detail, the reconstructed orbital possesses three main lobes with alternating signs, separated by two nodal surfaces. For clarity, cuts along the internuclear axis for the reconstructed orbital (red line) and the ab initio orbital (blue line) are depicted in Fig. 4(b). As shown in this figure, the two positive maxima of the reconstructed and ab initio orbital have the same values, as indicated by the two black arrows. Whereas some distortions are also observed in the reconstructed orbital. For example, the detailed structure near the left nucleus is lost and the amplitude at the negative maximum is lower than that of the exact wavefunction. To find the origin of these distortions, we calculate the Fourier-filtered ab initio orbital with the k-range corresponding to the sampled spectral range. A cut along the internuclear axis for the Fourier-filtered ab initio orbital is depicted by the dashed green line in Fig. 4b. One can see that the same deviations are observed for the Fourier-filtered ab initio orbital. Therefore, these distortions mainly originate from the limited spectral range, of which the effect on the reconstruction has been discussed in detail in [1, 8]. Besides, there is a slight shift of the reconstructed wavefunction relative to the exact wavefunction and the origin of the shift is still an open question. Although only the HOMO of CO is considered in our simulation, our scheme can be extended to other asymmetric molecular orbitals by properly choosing the relative phase and intensity of the two-color fields. Experimentally, there still exist some challenges such as the orientation of the molecules [16] and the measurements of the harmonic phase and polarization, which is systematically discussed in [1, 2].

In summary, we theoretically demonstrate a method for tomographic reconstruction of asymmetric molecular orbitals with multicycle two-color laser pulses. The unidirectional recollision of the electron wave-packet is achieved for all the orientations of the molecules by adjusting the relative phase of the two fields, and then the asymmetric molecular orbital is satisfactorily reconstructed with multicycle laser pulses. This releases the stringent requirement of a single-cycle pulse with a stabilized and controllable carrier-envelop phase for the tomographic imaging of asymmetric molecular orbitals.

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