Supplementary Material: Complete photoionization experiments via ultra-fast coherent control with polarization-multiplexing

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Here we provide an extended presentation and discussion of the results for a polarization-shaped laser pulse, as shown in brief in figure 4 of the main article. In order to provide more insight into the formation of the PADs in these cases, figure 1 shows the calculated PADs, along with the population dynamics and laser field, for a few examples. Columns (a) & (b) show the same results as figure 4 in the main article. Of the results presented, (a) & (d) are examples of cases where there is a large difference in the $p_{+1}$ and $p_{-1}$ state populations, while (b) & (c) show cases where the population dynamics for $p_{+1}$ and $p_{-1}$ show similar (time-integrated) amplitudes, although distinct temporal features in response to the details of the laser field.

In case (a), the dominance of the $p_{+1}$ state at the 1-photon level means that the resultant PAD shares some gross structure with the result for a pure circularly polarized pulse - the banded equatorial distribution shown in figure 2(d) in the main article - but with a 6-fold equatorial lobe structure. This additional structure results from the coherent addition of ionization via the $p_{-1}$, which also has significant temporal structure. Case (d) shows the results of an opposite phase mask, $\phi_y = +\pi/2$, which results in a flip in the amplitudes of the L and R field components, and in the temporal profiles. In this case, the gross structure of the distribution is still somewhat similar, with the equatorial part of the distribution dominant, but the net effect of the coherent addition of the photoelectron waves (including the phase of the laser field, as shown in eqn. 2 in the main text) results in a more asymmetric structure, most clear in the $(X,Y)$ projection. The loss of sensitivity to the fine details of the full PAD in the 2D projections is clear here, with reflection symmetry recovered in the $(X,Z)$ and $(Y,Z)$ projections.

For examples (b) & (c), which both have similar $p_{+1}$ and $p_{-1}$ state populations overall, the gross shape of the PAD is much closer to the case of a linearly polarized pulse - the $L = 3$-type distribution shown in figure 2(a) in the main article - but with cylindrical symmetry breaking and frame rotation. These characteristics are most clearly observed by considering the major axis of the distribution (defined as a line through the maxima), which is rotated away from the $y$-axis by around $\pi/4$ radians, and has broken reflection symmetry due to the loss of cylindrical symmetry in the full PAD as compared to the linearly polarized case. The 2D projections may show even greater sensitivity to the pulse shape than the full $I(\theta,\phi)$ distribution, due to the enhanced effect of rotations of the distribution on the image plane projections; in this manner pulse shaping with imaging detection can lead to a wide range of 2D projections, even in the case where only a few partial wave channels are accessed in the ionization continuum. As discussed in the main text, the PADs are very sensitive to the exact pulse shape, with small changes in the spectral mask phase and extent significantly affecting the PADs. This is emphasized by cases (b) and (c), which show the effect of a small change to the extent of the phase mask. In (b) the phase mask is applied to the red half of the spectrum, while in case (c) the mask is applied to around 60% of the spectrum, thus crossing over into the blue half of the spectrum. This change shifts the peaks of the L and R components of the field slightly and broadens the wings of the pulse. The effect of these changes are clear in the population dynamics, with the $p_{+1}$ and $p_{-1}$ traces reflecting the change of the pulse shape. In the PADs, this results in a slight change to the overall symmetry of the full (3D) PAD, particularly evident in the $(X,Y)$ plane projection as unequal lobes in case (b), and also a rotation of the poles of the PAD of around $\pi/4$ radians in the $(X,Y)$ plane between the two cases. As discussed in the main text, the effect of frame rotations can be very significant in the 2D projections, and this is evident here in the very different $(X,Z)$ and $(Y,Z)$ projections for case (b) and (c).

While these examples only begin to show the details and range of this 3-photon ionization scheme, they do serve to illustrate further the interplay of the population and ionization dynamics inherent in this process (as shown in equations 2 & 3), including the effects of the coherent nature of the multiplexed pathways.
Figure 1. Calculated PADs for $E_{X,Y}(t)$ obtained by application of various spectral phase masks, population dynamics (2nd row) and laser field components (3rd row). Columns (a) & (b) show the same results as in the main article (figure 4), where masks of $\phi_y = -\pi/2$ and (b) $\phi_y = -0.995\pi$ were applied to the red half of the spectrum. Column (c) shows the same phase mask as (b), i.e. $\phi_y = -0.995\pi$, but extended slightly into the blue half of the spectrum (in this case applied to around 60% of the spectrum); column (d) shows the case of a $\phi_y = +\pi/2$. For the PADs, the full $I(\theta, \phi)$ distribution is shown in spherical projection, along with 2D projections onto different Cartesian image planes. The population dynamics are shown for the bound-states coupled at the 1-photon level (see figure 1 in the main text). The laser field components are expressed in a spherical basis, and the field envelope is also shown.