

using Fresnel-Kirchhoff diffraction formula [13] for each harmonic.

We apply this principle to our case, when the wavefront structure of each harmonic is taken from our measurements, but we arbitrary assume that the attosecond pulses within the train are transform limited at the center of the focusing mirror (Fig. 3(d)) i.e. $\varphi_N = N\omega_0 t_e$, where t_e is harmonic emission time. As we move along or across the beam, the relative spectral amplitude and phase of the harmonics changes. Consequently the temporal structure of each attosecond pulse in the pulse train depends on where the pulse is measured. Using the measured spectral wavefronts in our experiment, the temporal profiles is shown for selected points along the beam in Fig. 3(a)–3(c). Even in cases where the atto-chirp [14, 15] has been corrected, it can only be corrected strictly speaking at one point.

There are at least two approaches for bringing the harmonics to a common focus and generating Fourier limited attosecond pulses and pulse trains. (1)The phase of any harmonic depends on both the intensity and the phase of generating beam. Optimizing the spatial profile (both phases and amplitudes) of the fundamental beam will allow us to minimize the chromatic wavefront variations between different harmonics. (2)The chromatic aberrations that we measure are reproducible. They can, in principle, be corrected with specially designed linear optics.

4. Conclusion

In conclusion, within the signal-to-noise limits of our experiment, we have obtained complete (phase and amplitude) information on wavefront profile of each harmonic. We have shown that spectrally resolved wavefronts allow the field distribution to be found anywhere. If we know the position of the generating medium with sufficient accuracy, this includes the position at the generating medium itself. The wave front amplitude and phase in the generating medium isolate an imprint of the underlying single atom or molecule response. This will solve the long standing problem of intensity and phase focal volume averaging in attosecond science experiments.

Within a 3-step model [16], for a thin, low density gas, the measured wavefront phase $\phi_N(\vec{r}, z_{jet})$ is given by the sum of four contributions:

$$\phi_N(\vec{r}, z_{jet}) = \phi_t(I(\vec{r}, z_{jet}), \omega_N) - |\alpha_N|I(\vec{r}, z_{jet}) + \phi_r(\omega_N, z_{jet}) + N\phi_{fund}(\vec{r}, z_{jet}) \quad (2)$$

Where $\phi_{fund}(\vec{r}, z_{jet})$ is the wavefront phase of the fundamental beam at the jet position and can be measured. $\phi_r(\omega_N, z_{jet})$ is the phase contribution of the transition moment [17]. $\phi_r(\omega_N, z_{jet})$ is intensity independent for a single orbital, but intensity dependent for multiple orbitals. The phase that the electron acquires in the continuum is $-|\alpha_N|I(\vec{r}, z_{jet})$. This phase can be calculated within the strong field approximation and its corrections [5, 6]. $\phi_t(I(\vec{r}, z_{jet}), \omega_N)$ is the yet not fully understood tunneling phase [18, 19]. It might be intensity dependent. Therefore, wavefront structure will serve as a sensitive measurement of the intensity dependence of these parameters.

Once the relative phase of each harmonic is determined and spectrally resolved wavefronts are measured, then we have shown that we can resolve the temporal profile of an attosecond pulse anywhere in space. No matter what the pulse temporal structure should prove to be, we have already determined that there will be significantly different temporal profile at the center and at the edges of a beam and the temporal profile must change as the beam passes through the respective foci of the harmonics. Therefore, any spatially extended measurement with attosecond pulses will be affected and any measurement of the pulse itself is also affected. The result of any experiment will be effectively averaged over the different temporal profiles at different positions in the interaction region of the focal volume.

Finally, attosecond science has been restricted to spatially averaged measurements to image orbitals [10], tracing molecular dynamics [20], identify and time resolve tunneling wave packets [18] and to follow Auger decay [21]. Spectrally resolved wavefronts and the complete spatio-temporal characterization of attosecond pulses that they facilitate will allow us

much greater experimental precision in all of these experiments, facilitating access to a single molecule response in the strong field.

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