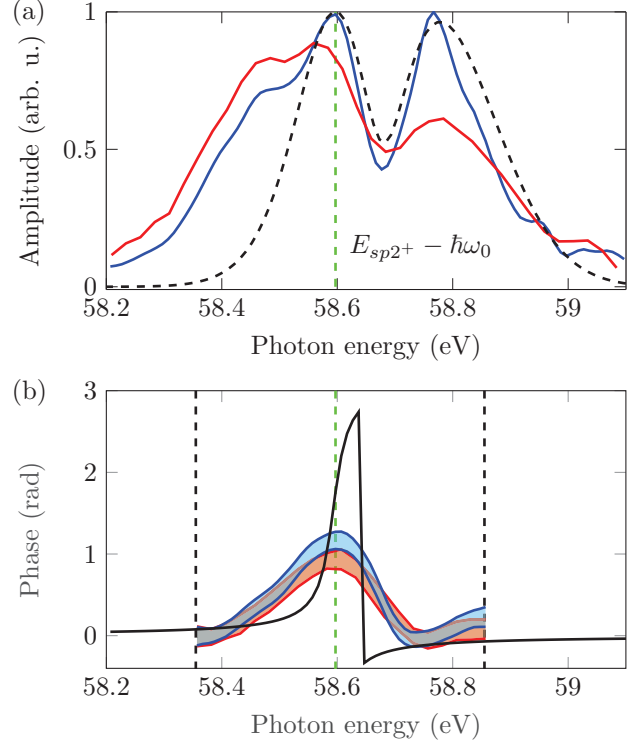


**Figure 3.** Deconvolution of the photoelectron spectrum. Profile of the resonant harmonic (H39) before (red) and after (blue) deconvolution. The green dashed line indicates the position of the resonance.

Figure 3 shows the photoelectron spectrum obtained by absorption of the 39th harmonic, close to the  $sp2^+$  resonance, before and after deconvolution (respectively red and blue curves). The deconvolution reduces the width of the harmonic and enhances the characteristic asymmetry of the Fano profile. The experimental deconvolved spectrum shows a minimum after the resonance and a second peak at higher energies which corresponds to the nonresonant spectrum of the harmonic [25].

We applied the deconvolution algorithm to the full RABBIT trace and extracted the amplitude and phase of the sidebands from the new spectrogram. In figure 4, we compare the amplitude (a)  $|A_{39-1}|$  calculated using equation 4, and phase (b) of the two-photon transition before (red) and after (blue) deconvolution, in conditions such that H39 is resonant with the  $sp2^+$  state. The deconvolution gives sharper features in figure 4(a), which agree well with theoretical results indicated by the dashed black line (see Section 2.4 and 4.1). We could not identify the spectral feature observed around 58.4 eV. For the phase variation, the deconvolution leads to a slightly sharper phase evolution around the resonance. However, the latter is still quite different from the resonant one-photon dipole phase shown as black line in figure 4(b). According to equation 2, this phase displays a smooth  $\pi$  variation characteristic of the arctan function around  $\epsilon = 0$ , followed by a sudden  $\pi$  phase jump at  $\epsilon = -q$ . The reason for the difference is thus not the convolution with the RF of the spectrometer, as was the case in [25], but finite pulse effects, as discussed below.



**Figure 4.** Amplitude and phase of the two-photon electron wave packet ( $A_{39-1}$ ) emitted when H39 is resonant with  $sp2^+$ . (a) Normalized amplitude before (red line) and after (blue line) deconvolution. The latter is in good agreement with the simulations (black dashed line). The green dashed line indicates the position of  $E_{sp2^+} - \hbar\omega_0$ . (b) Spectral phase before (red) and after (blue) deconvolution. We set a threshold of 30% of the maximum intensity of the sideband as a limit below which we consider that the intensity to noise ratio is too low to reliably extract the phase. The black dashed lines correspond to the limits of the energy region in which the intensity of the sideband is above the threshold. The shaded area represents the standard deviation given by the fit. The black solid line corresponds to the phase of the resonant one-photon transition amplitude (see equation 2), shifted down by one laser photon energy.

### 3.2. Finite pulse effects

It is often considered that in the RABBIT scheme, the IR field makes a perfect replica of the wave packet excited by the harmonic and that the amplitude and phase measured in the sideband correspond to that of the one-photon wave packet. While in the case of long pulses with narrow spectra, the correspondence between one- and two-photon wave packets is justified, this approximation breaks down when the bandwidths of the IR and XUV pulses become large. Indeed, in the presence of broad pulses, multiple combinations of frequencies can lead to the same final state, thus giving rise to a coherent mixing of the different frequencies of the one-photon wave packet. In [36], Jiménez-Galán *et al.* showed that this mixing of frequency components can induce a variety of effects, referred to as finite pulse