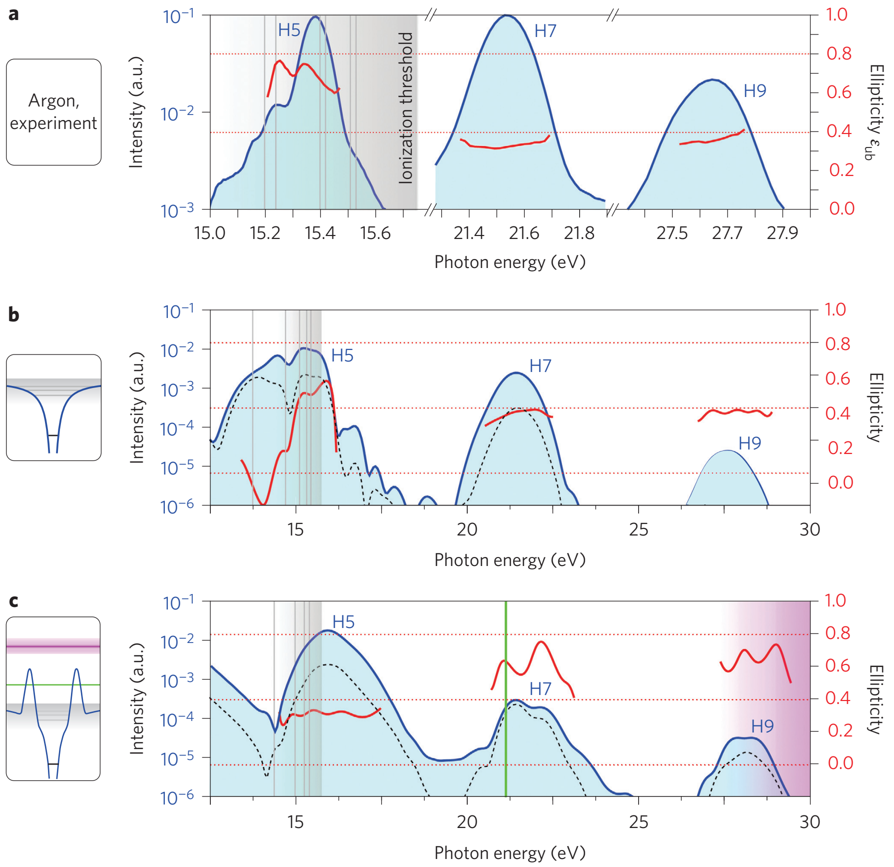

Figure 1 | Elliptical resonant HHG in argon. a, Experimental harmonic intensity (blue area, log-scale) and upper-bound ellipticity (red continuous line) generated using 404 nm, ~1×10^{14} W cm^{-2} pulses with $\varepsilon_0 = 0.4$. Thin grey vertical lines indicate positions of resonances. a.u., arbitrary units. **b**, Theoretical results at 400 nm, ~1×10^{14} W cm^{-2}, $\varepsilon_0 = 0.4$, using a soft-core Coulomb potential. The thin dashed black line shows the orthogonal component of the harmonic emission. **c**, Theoretical results using a modified potential with a shape resonance around 21 eV (green vertical line) and a broad resonance in the continuum from 27 to 36 eV (purple area).

404 nm pulse with $\varepsilon_0 = 40\%$ ellipticity reaches 75% ellipticity. A theoretical study shows that this effect is due to the presence of resonances below the ionization threshold. This study also indicates that continuum resonances, above the ionization threshold, also lead to highly elliptical harmonics. To extend the spectral range of elliptical XUV emission, we changed the generation gas for SF_6, a molecular system with a high ionization potential (15.7 eV) and a rich ionization continuum involving several resonances[27,28]. Optical polarimetry confirmed the existence of high ellipticity in a broad spectral region. The quality of this elliptical femtosecond XUV source enabled us to induce macroscopic asymmetries in the one-photon ionization of pure enantiomers of randomly oriented fenchone molecules, via the so-called photo-electron circular dichroism (PECD) process[2].

To study resonant HHG by elliptical laser fields we generated harmonics of 404 nm pulses in argon. Details about the experiment are provided in the Methods. Harmonic 5 is centred at 15.3 eV, just below the ionization potential of Ar (15.76 eV). Figure 1a shows the harmonic spectrum obtained using fundamental pulses with $\varepsilon_0 = 0.4$ ellipticity. Although the harmonics generated above the ionization threshold have a smooth Gaussian-like spectral profile, harmonic 5 is spectrally narrower and shows additional structures. Such spectral structuring of below-threshold harmonics has recently been identified as the signature of resonant bound–bound transitions in HHG[29].

Measuring light polarization requires a rotating polarizer, which modulates the intensity following the Malus' law[30]. This enables the polarization direction of the linear component of the radiation to be determined, but is not enough to measure the circular polarization rate S_3 because of the possible presence of unpolarized radiation (S_4), where S_i are the conventional Stokes parameters. As such, the Malus' law provides an upper bound of the ellipticity, $\varepsilon_{ub} = \sqrt{[1-(S_1^2+S_2^2)]}$. In our experiment, a fixed polarizer was used and the harmonic spectrum was recorded as a function of the rotation of the laser's main axis of polarization. ε_{ub} was extracted by Fourier transform of the XUV yield oscillations, for each component of the harmonic spectrum. The results are shown in Fig. 1a. Above the ionization threshold the ellipticity is slightly lower than that of the driving laser, and rather uniform over the spectral width of each harmonic. By contrast, harmonic 5 shows a much higher ellipticity, reaching 0.77, with an oscillating behaviour.

To investigate the influence of bound–bound resonances on harmonic ellipticity, we performed a theoretical study based on the resolution of the time-dependent Schrödinger equation in two dimensions. Details for the calculations are provided in the Supplementary Information. Figure 1b shows the harmonic spectrum and ellipticity obtained using a soft-core Coulomb potential mimicking the ionization potential of Ar. Similar to the experiment, above-threshold harmonics have a regular spectral profile and an ellipticity slightly lower than the fundamental pulse, while harmonic 5 shows structures both in the spectrum and ellipticity. The ellipticity is higher than 0.4 above 15 eV and becomes negative at 14 eV. Calculations show that the values of the ellipticity in the resonant area strongly depend on the exact structure of the potential, so the agreement with the experiment can only be qualitative. The high values of the ellipticity are associated with an increase of the