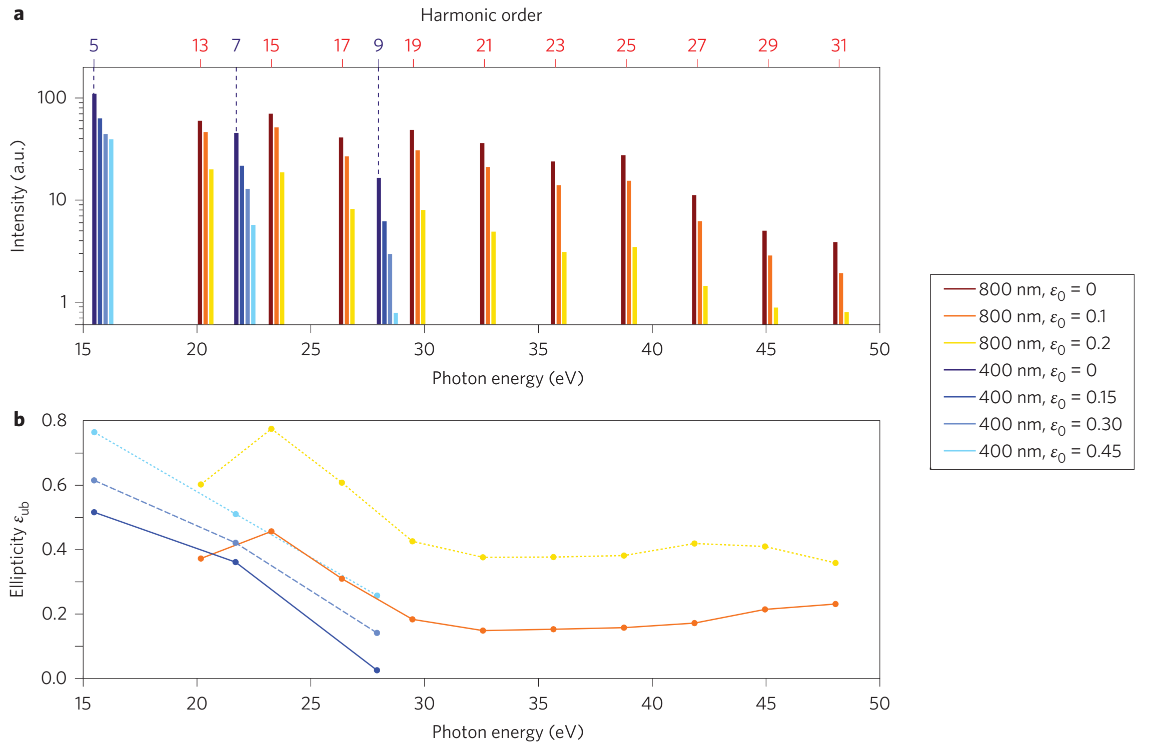

Figure 2 | Optical polarimetry measurements of HHG in SF_6. a, Harmonic signal produced by 800 nm, 1.3×10^{14} W cm^{-2} pulses (brown to yellow, harmonics 13 to 31), and by 400 nm, 1×10^{14} W cm^{-2} pulses (dark to light blue, harmonics 5 to 9). a.u., arbitrary units. **b**, Ellipticity of the harmonic radiation (upper limit) determined using an XUV polarizer. The error on ellipticity measurements, determined by comparing consecutive measurements in identical conditions, is ±0.03.

orthogonal component of the harmonic emission under the influence of the resonance. These results demonstrate the role of bound–bound resonant transitions in the generation of highly elliptical harmonics.

Resonant below-threshold HHG holds great potential for creating circular XUV sources. It is very easy to implement, and is bright enough for applications in the gas phase. Using a calibrated photodiode we measured that this source produces 7×10^6 photons per pulse for harmonic 5 when driven by 30% ellipticity; this can be extended to higher flux by increasing the generating pressure, without suffering from reabsorption like higher harmonics[29]. On the other hand, it is restricted to low photon energies, below the ionization potential of the gases conventionally used for intense HHG (argon, 15.76 eV; krypton, 14 eV; xenon, 12.13 eV). Can quasi-circular harmonics also be generated using resonances lying above the ionization threshold? To answer that question we modified the potential used in our calculations. We added a potential barrier that creates a shape resonance in the continuum, around the energy of harmonic 7. Such shape resonances are very common in ionization spectra of molecules in the 10–20 eV range above the ionization threshold. The modification of the potential also created another resonance, much broader in energy and higher in the continuum (from 27 eV to 36 eV). The dipole moments associated with these resonances are shown in the Supplementary Information. The results shown in Fig. 1c indicate that continuum resonances boost the orthogonal component of the harmonic emission, giving ellipticities of H7 and H9 of up to 0.75.

The simulations demonstrate the generality of quasi-circular HHG by different types of resonances, opening a path to the generation of highly elliptical XUV pulses over a broad spectral range, at higher photon energies. To validate this idea and design the ideal source for applications we chose SF_6 as a generating medium. We recently conducted high-harmonic spectroscopic studies of this molecule[28], which revealed that the harmonic emission in the 20–25 eV range was dominated by a shape resonance in the A ionization channel (associated with the first excited state of the cation)[27]. Figure 2 shows the harmonic intensity and ellipticity produced in SF_6 using 800 nm pulses. Ellipticity ε_{ub} reaches unprecedented values (between 60 and 80% for a fundamental ellipticity of only 20%) between harmonics 13 and 17 (20–27 eV). This is in agreement with the position of the shape resonance. A secondary maximum in the ellipticity around 45 eV could also be due to resonances in the A/B channel at 35 eV and 49 eV (ref. 27).

We also performed additional characterization of the elliptical XUV radiation. The beam shows low divergence (2–4 mrad) and excellent spatial coherence properties; creating two harmonic sources spatially shifted by ~100 μm results in a nice Youngs' slits inteference pattern with high contrast (~60% visibility). This ensures a good focusability of the elliptically polarized harmonics, which could be crucial for further applications. The spectral width of harmonics 13 to 17 is ~150 meV full-width at half-maximum. The ultrashort character of the harmonic emission was checked by initiating molecular vibrations in SF_6, which modulate the harmonic yield with a 43 fs period[31], indicating that the emission is shorter than this period.

The generation of a dense frequency comb with ~3 eV step (HHG at 800 nm) can be problematic in studying the photoionization of polyatomic molecules with congested spectra. This issue can be circumvented by increasing the spacing between consecutive harmonics, using a shorter-wavelength fundamental pulse. The results obtained using 400 nm fundamental pulses, which lead to a 6.2 eV step in the harmonic comb, are shown in Fig. 2. The decay of the harmonic efficiency with ellipticity is less important, in particular for harmonic 5, enabling the use of higher ε_0. The harmonic maximum ellipticities still reach very high values: almost 80% for harmonic 5 (15.5 eV) and 50% for harmonic 7 (21.7 eV) with