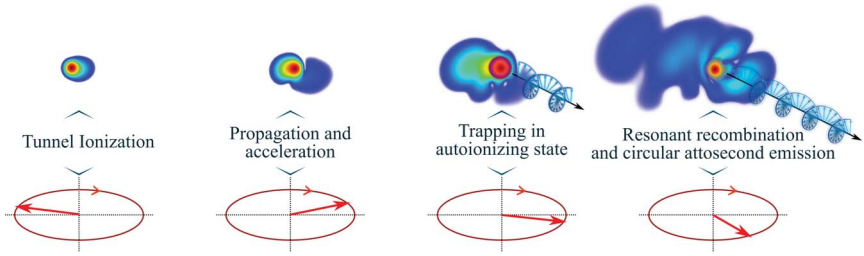

Fig. 3 Mechanism of elliptical resonant high-harmonic generation. The top color plot represents the electronic wavefunction from a 2D quantum calculation, evolving in an elliptical laser field whose instantaneous direction is depicted at the bottom.

orthogonal component to the harmonic electric field when driven by an elliptical laser. This results in the emission of quasi-circularly polarized light. This process is schematized in Fig. 3. An electron wavepacket tunnels out from the molecule and is driven by the elliptical strong field. When the wavepacket returns near its parent ion, it can get trapped into the potential well associated to the resonance before recombining. This adds an additional step to the standard 3-step model of HHG, as proposed by V. Strelkov.[38] Our simulations indicate that the shape resonance modifies the spatial profile of the electron wavepacket as it approaches the ionic core, as illustrated in Fig. 3.[39] Its perpendicular component increases, leading to a higher orthogonal harmonic field. Depending on the exact shape of the potential, this process can lead to the emission of quasi-circular high-order harmonics. This property was experimentally verified in the case of the shape resonance around 22–23 eV in SF_6 molecules, as well as for Rydberg resonances below the ionization threshold of argon.[39]

The PECD measurements were performed using the 1 kHz Aurore laser system at CELIA, which delivers 25 fs, 7 mJ pulses at 800 nm. The laser pulses were frequency doubled in a 200 μm thick BBO crystal and focused by a 50 cm lens in a 3 mm long cell filled with SF_6 molecules. Using 400 nm rather than 800 nm pulses for HHG increases the energy spacing between harmonics from 1.55 to 3.1 eV, easing the assignment of the photoelectron spectra. The laser ellipticity was controlled using a motorized zero order half waveplate in front of a fixed zero order quarter waveplate, in order to keep the orientation of the polarization ellipse fixed in space. The harmonics were directly sent into the interaction region of a Velocity Map Imaging Spectrometer (VMIS), without any focusing optics to avoid modifying the XUV polarization state and perturbing the ionization process with an additional intense 400 nm field. A motorized aperture was tuned to limit the diameter of the XUV beam and achieve a good resolution of the photoelectron images. A sample of enantiopure (+)-fenchone molecules (Sigma Aldrich) at room temperature was connected to a heated metallic nozzle (390 K) with 300 μm diameter located 7 cm away from the interaction zone of the VMIS. The pressure in the interaction region was 2×10^{-6} mbar (with a 5×10^{-8} mbar background pressure).

The velocity map imaging spectrometer records the bidimensional projection of a tridimensional photoelectron angular distribution $S_p(E, \theta)$, which can be decomposed as a sum of Legendre polynomials P_i:

