Generation of optical Schrödinger cat states in intense laser–matter interactions

M. Lewenstein1,2, M. F. Ciappina1,3,4,5, E. Pisanty1,6, J. Rivera-Dean1, P. Stammer1,6, Th. Lamprou7,8 and P. Tzallas1,7,9

The physics of intense laser–matter interactions1–4 is described by treating the light pulses classically, anticipating no need to access optical measurements beyond the classical limit. However, the quantum nature of the electromagnetic fields is always present5. Here we demonstrate that intense laser–atom interactions may lead to the generation of highly non-classical light states. This was achieved by using the process of high-harmonic generation in atoms6–8, in which the photons of a driving laser pulse of infrared frequency are upconverted into photons of higher frequencies in the extreme ultraviolet spectral range. The quantum state of the fundamental mode after the interaction, when conditioned on the high-harmonic generation, is a so-called Schrödinger cat state, which corresponds to a superposition of two distinct coherent states: the initial state of the laser and the coherent state reduced in amplitude that results from the interaction with atoms. The results open the path for investigations towards the control of the non-classical states, exploiting conditioning approaches on physical processes relevant to high-harmonic generation.

Decades after the invention of laser6, the development of high-power lasers7–9 enabled scientists to explore laser–matter interactions driven by laser fields with strengths comparable to or stronger than the atomic potential. This research domain, termed strong-laser-field physics, opened the way for studies ranging from relativistic optics10, to high-harmonic generation (HHG) and ultrafast optoelectronics11. A large amount of work was conducted in the past for the description of intense laser–atom interaction. In the early days, it was described perturbatively in a quantized manner using multiphoton processes12,13 and, later on, in the strong-field limit by semiclassical approaches such as the three-step model10–12. However, due to the high photon number of the driving field, the description of these interactions in the strong-field limit remains incomplete as it relies on semiclassical approximations treating the atomic system quantum mechanically but the electromagnetic field classically, that is, ignoring the quantum nature of the electromagnetic radiation, which is always present. Revealing the quantum nature of light in strongly laser-driven interactions, besides its fundamental physical interest, is important for applications in basic research and technology. This is because it bridges the gap between strong-laser-field physics1–4 and quantum optics15, providing access to the development of strong-field quantum electrodynamics, and of a new class of non-classical light sources (such as squeezed, Fock and Schrödinger ‘cat’ states, and so on)13–17, which are at the core of quantum technology18–20.

To unmask the quantum nature of light in strongly laser-driven interactions and show its impact on the aforementioned directions, we have used the HHG process in atoms14,15, which is one of the most fundamental processes in strong-laser-field physics. The understanding of the HHG process was boosted from the formulation of the semiclassical three-step model15. In this approach, HHG from a single atom/molecule/solid is initiated by an electron’s tunnelling to the continuum, its subsequent acceleration in the intense laser field and finally its recombinations to the ground state of the target. To provide a fully quantized description of the HHG process, we have to rigorously answer the following questions. (1) What is the quantum depletion of the coherent state of the fundamental laser mode? (2) What is the quantum state of the generated harmonics? Although several groups have attempted to study this problem theoretically21–25 and experimentally26–28, none of these efforts have provided a rigorous answer to the above questions. Here we show that if the initial state of a system of N atoms in their ground state is impinged by a laser in a coherent state of amplitude α, then the resulting quantum states of the fundamental and harmonic modes are coherent. However, due to coherences and correlations between the fundamental and the harmonics, the fundamental mode amplitude is shifted (that is α → α + δα), where δα is negative and reflects the energy conservation, which, in the context of the semiclassical three-step model15, represents the energy losses of the fundamental mode due to the recollision process. While at the single atom level δα is negligibly small, it may become quite important due to the cooperative effect in HHG and the phase matched contribution of many atoms. Hence, the final state of the fundamental mode, conditioned on harmonic generation, is a superposition of the two coherent states: the initial state and the shifted one. This is, in general, highly non-classical: it interpolates between a Schrödinger ‘kitten’ state, corresponding to a coherently shifted first Fock state (for |δα| ≪ 1), and a ‘cat’ state (for 0 < |δα| < 1). We exactly calculate δα, as well as the coherent amplitudes of the harmonics, and we confirm experimentally the generation of the ‘cat’ state, by characterizing the quantum state of the fundamental field exiting the HHG medium when conditioned to HHG. This was achieved by combining the quantum tomography (QT) approach23,28 with a photon-correlation-based method, namely the quantum spectrometer (QS)15,27.
Starting from the time-dependent Schrödinger equation (TDSE), describing both the atom and light quantum mechanically, it can be shown (Methods) that the evolution of the state of the laser and harmonic modes \( \{q\} \), conditioned on the atomic ground state and upon neglecting of the (small) continuum/excited part, is described by

\[
i\hbar \frac{\partial}{\partial t} \phi(t) = -\mathbf{E}_\text{L}(t) \cdot (\hat{d}_1(t)) \phi(t) . \tag{1}
\]

Here \( \phi(t) \) is the quantum state describing the photonic degrees of freedom, \( \mathbf{E}_\text{L} \) is the classical field, \( \hat{d}_1(t) \) describes the quantum fluctuating part of the laser electric field and \( \mathbf{d}_1(t) \) is the quantum averaged time-dependent dipole moment induced by the classical part of the laser pulse, which can be efficiently calculated solving the TDSE, or even easier using the strong-field approximation\(^\text{16}\). The solution is (Methods):

\[
|\phi(t)\rangle = \left( |\alpha_L + \delta \alpha_L \rangle e^{-\text{Im} t} , \beta_q e^{\text{Im} t} , \ldots , \beta_q e^{-\text{Im} t} , \ldots \right) , \tag{2}
\]

where \( \delta \alpha_L = -i \text{g}(\omega(t)) \cdot \mathbf{d}_\text{sa} \), \( \beta_q = -i \text{g}(\omega_L) \sqrt{\mathbf{a}} \cdot \mathbf{d}_\text{sa} \), \( \text{g}(\omega) \) is the frequency of the fundamental laser field, and \( \text{g}(\omega_L) \) is the effective coefficient entering into the expansion of the electric field into the quantized modes (Methods). Here \( \mathbf{d}_1 = \int_0^t \mathbf{E}(t)e^{i\omega t}(\mathbf{d}_1(t)) \) is a Fourier transform of the time averaged semiclassical dipole moment (Methods) weighted by the pulse envelope \( \mathbf{E}(t) \). Assuming that \( N \) atoms contribute to HHG coherently in a phase-matched way, we obtain that the final state of the fundamental and harmonic fields, after the pulse being coherent, is \( |\alpha_L + \delta \alpha_L \rangle \) and \( |\beta_q\rangle \) (Fig. 1) respectively, with

\[
\delta \alpha_L = -i \text{Ng}(\omega_L) \cdot \mathbf{d}_\text{sa} , \tag{3}
\]

\[
\beta_q = -i \text{Ng}(\omega_L) \sqrt{\mathbf{a}} \cdot \mathbf{d}_\text{sa} . \tag{4}
\]

The above, rigorous and analytic expressions constitute one of the main results of this work as they provide a direct solution to the problem concerning the quantum nature of light in strong-field laser–atom interaction, and the key parameters that can be used to control the properties of the light states exiting the atomic medium.

The consequences of this result towards the generation of non-classical light states is coming from the projection of the fundamental mode on its part corresponding to the HHG, which can be achieved experimentally by utilizing the QS approach\(^\text{26,27}\). To show the result of this action, we use the conditions of the experiment presented below, where the QT of the fundamental mode, after its projection to HHG, is performed in two steps (Fig. 1). The first consists in reducing the amplitude of this mode by a factor \( \cos(r) \). In this way, \( \alpha_L \rightarrow \cos(r)\alpha_L = \alpha \) and \( \delta \alpha_L \rightarrow \cos(r)\delta \alpha_L = \delta \alpha \). The second step corresponds to condition on HHG, that is, post-selection of the part of the state that includes at least one harmonic photon. Mathematically it means that \( |\alpha + \delta \alpha \rangle \rightarrow (1 - |\alpha \rangle \langle \alpha |) (\alpha + \delta \alpha) = |\alpha + \delta \alpha \rangle - (\alpha |\alpha \rangle + \delta \alpha |\alpha \rangle) \), that is, the HHG conditioning allows us to project the final coherent state onto the part of the fundamental field that has been affected by the HHG process, that is, everything that was not in the initial state \( |\alpha \rangle \). This is the action that shows the coupling of the initial coherent state with the shifted one (via the matrix element \( \xi = (\alpha |\alpha + \delta \alpha \rangle \) and creates the superposition of the shifted coherent state with the initial state, weighted by the coupling factor \( \xi \). It is elementary to see that the state is a superposition of the two coherent states, so it is a Schrödinger ‘cat’ state. However, if the two coherent states are very close to each other, this is a ‘kitten’ rather than a genuine ‘cat’. In fact, we obtain two limiting cases.

(1) If \( |\xi| \approx 1 \), then the post-selected state \( |\psi_{\text{post}}\rangle \), conditioned on HHG, is nevertheless quite non-classical, corresponding to a shifted Fock state, \( |\psi_{\text{post}}\rangle = (\alpha^2 - \alpha^2 |\alpha \rangle \) (where \( \alpha^2 \) is the creation operator affecting the fundamental mode and \( \alpha^2 \) is the complex conjugate of reduced in amplitude coherent state), with a Wigner function \( W(\beta) = (4|\beta - \alpha|^2 - 1)e^{-|\beta - \alpha|^2} \) (Fig. 2a,b).

(2) If \( 0 < |\xi| < 1 \), the state \( |\psi_{\text{post}}\rangle \) corresponds indeed to a ‘cat’ state, depicting a Wigner function \( W(\beta) = e^{-|\beta - \alpha|^2} + e^{-|\beta - \alpha|^2} - (e^{2|\beta - \alpha|^2} - e^{-2|\beta - \alpha|^2})e^{-2|\beta - \alpha|^2} \) (Fig. 2c,d) with negative regions\(^\text{29}\).

The experimental results presented here suggest the generation of a ‘cat’ state (that is, case 2). Since \( \alpha_L \approx 10^4 \), that would require that \( \text{Ng}(\omega_L) \cdot \mathbf{d}_\text{sa} \approx 10^8 \). We expect the atomic dipole moment to be of the order of \( e \) times the electron excursion amplitude, which is 5–50 Å, that is, 10–100 atomic units (a.u.). This would imply that the number of atoms participating in the coherent, phase-matched HHG is \( N \approx 10^{12} - 10^{15} \), which is consistent, within the large uncertainty, with the experimental estimations (Supplementary Sections 1 and 2). The experiment has been performed using the set-up shown in Fig. 3a (for more details, see Supplementary Section 1). The arrangement consists of an interaction area where the harmonics are generated, and the QT and QS approaches. The QS was used to project the fundamental mode on its part corresponding to the harmonic generation (Supplementary Section 3), and the QT to characterize the quantum state of the light field exiting the atomic medium (Supplementary Section 4). In the first branch of the interferometer, the linearly polarized \( \sim 35 \text{fs} \) infrared (IR) laser pulse, after passing through a beam separator (BS\(\)) was focused with an intensity \( \sim 8 \times 10^{13} \text{W cm}^{-2} \) into xenon atoms. It is here where the strong-field laser–atom interaction and HHG process takes place and high harmonics with \( q \leq 23 \) were generated (Supplementary Section 1). The mean photon number of the IR field exiting the medium was reduced to the level of few photons per pulse, with the QS selecting, for each laser shot, only the IR photons related to the HHG. The light field \( (E_L) \) after the QS was spatiotemporally overlapped in a beam splitter (BS\(\)) with the local oscillator laser field \( (E_L) \) coming from the second branch of the interferometer, which contains a translation stage that

---

**Fig. 1** Schematic representation of the generation of optical ‘cat’ states. The coherent laser state \( |\alpha_L\rangle \) (thick red lines) interacts with atoms and in consequence high harmonics (thick blue lines) are generated. The inset shows an intuitive picture of the electron recollision process that leads to HHG. The oscillating laser field \( E(t) \) is depicted with thick red curve and the electron paths with thin green curves. After the interaction, the harmonic modes are coherent \( |\beta_q\rangle \) and the fundamental is an amplitude-shifted coherent state \( |\alpha_L + \delta \alpha_L\rangle \). This state after an amplitude attenuation \( |\alpha + \delta \alpha\rangle \), is projected on its part corresponding to HHG and becomes a Schrödinger ‘cat’ state \( |\psi_{\text{post}}\rangle \) with \( \xi = (\alpha |\alpha + \delta \alpha \rangle \).
Fig. 2 | Calculated Wigner functions of a Schrödinger ‘kitten’ and a ‘cat’ state. The Wigner functions \(W(\beta)\) are plotted according to the terminology of ref. 29. \(\beta\) is a variable such that \(\text{Re}(\beta - \alpha) \equiv x\) and \(\text{Im}(\beta - \alpha) \equiv p\). \(x\) and \(p\) are the values of the quadrature field operators \(\hat{x} = (\hat{a} + \hat{a}^\dagger)/\sqrt{2}\) and \(\hat{p} = (\hat{a} - \hat{a}^\dagger)/\sqrt{2}\). \(\delta\alpha\) represents the amplitude shift of the initial coherent state \(|\alpha\rangle\) and \(\zeta = (\alpha|\alpha + \delta\alpha\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\rangle\ratio
introduces a phase shift $\varphi$ between the $E_i$ and $E_m$ fields. The power of the $E_i$ was about $\sim 10^7$ higher than the $E_m$. The interfering fields after BS, were recorded by a balanced detector, which provides at each value of $\varphi$ the photocurrent difference $i_m$. These values correspond to the measurement of the electric-field quadrature operator, and have been used for the reconstruction of the Wigner function according to the well-known methodology described in refs. 10,26 (for details, see Supplementary Section 4). As expected, when the HHG process was switched off, the state of the driving IR laser field is coherent, depicting a Wigner function with a Gaussian distribution (for details, see Supplementary Section 4). After that, we show that the quantum states of the fundamental and the harmonics oscillate at very different frequencies, with the fundamental and the harmonics in a large variety of intense laser–matter interactions31,32. The global coherent state is a product state, but only the effective harmonic modes after the interaction are coherent, with the fundamental to be shifted due to quantum effects. When the latter is conditioned onto HHG, it results in a superposition of two coherent states, which interpolates between a ‘kitten’ and a ‘cat’ state. This was experimentally confirmed by measuring the Wigner function of the ‘cat’ state. The method is applicable for the production of large optical ‘cat’ states and in a large variety of intense laser–matter interactions31.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-021-01317-w.

Received: 24 December 2020; Accepted: 1 July 2021; Published online: 19 August 2021

Fig. 4 | Measurement of the Wigner function of the genuine Schrödinger ‘cat’ state. a, Measured Wigner function $W(x,p)$ of the IR field (with $\langle n\rangle \approx 1.98 \pm 0.04$) when the HHG process and the conditioning to the HHG, were switched on (that is, xenon gas jet and QS switched on). $x$ and $p$ are the values of the quadrature field operators $\hat{x} = (\hat{a} + \hat{a}^\dagger)/\sqrt{2}$ and $\hat{p} = (\hat{a} - \hat{a}^\dagger)/\sqrt{2}$. The error of the amplitude of $W(x,p)$ is $\pm 0.002$. In agreement with the theoretical predictions, the function clearly shows a Schrödinger ‘cat’ state. It depicts a ring structure with a maximum $W_{\text{max}} \approx 0.21$ at $(x,p) \approx (0,-1.9)$ and a negative minimum $W_{\text{min}} \approx -0.004$ at $(x,p) \approx (0,-0.8)$. b, Contour colour plot of the projected Wigner function on the $(x,p)$ plane.
20. Deutsch, I. H. Harnessing the power of the second quantum revolution. *PRX Quantum* 1, 020101 (2020).
21. Diestler, D. J. Harmonic generation: quantum-electrodynamical theory of the harmonic photon-number spectrum. *Phys. Rev. A* 78, 033814 (2008).
22. Gonoskov, I. A., Tsatrafyllis, N., Kominis, I. K. & Tzallas, P. Quantum optical signatures in strong-field laser physics: infrared photon counting in high-order harmonic generation. *Sci. Rep.* 6, 32821 (2016).
23. Gombkötő, Á., Varró, S., Mati, P. & Földi, P. High-order harmonic generation as induced by a quantized field: phase-space picture. *Phys. Rev. A* 101, 013418 (2020).
24. Gorlach, A., Neufeld, O., Rivera, N., Cohen, O. & Kaminer, I. The quantum-optical nature of high harmonic generation. *Nat. Commun.* 11, 4598 (2020).
25. Yangaliev, D. N., Krainov, V. P. & Tolstikhin, O. I. Quantum theory of radiation by nonstationary systems with application to high-order harmonic generation. *Phys. Rev. A* 101, 013410 (2020).
26. Tsatrafyllis, N., Kominis, I. K., Gonoskov, I. A. & Tzallas, P. High-order harmonics measured by the photon statistics of the infrared driving-field exiting the atomic medium. *Nat. Commun.* 8, 15170 (2017).
27. Tsatrafyllis, N. et al. Quantum optical signatures in a strong laser pulse after interaction with semiconductors. *Phys. Rev. Lett.* 122, 193602 (2019).
28. Lvovsky, A. I. & Raymer, M. G. Continuous-variable optical quantum-state tomography. *Rev. Mod. Phys.* 81, 299–332 (2009).
29. Schleich, W. P. *Quantum Optics in Phase Space* (Wiley-VHC Verlag, 2001).
30. Breitenbach, G., Schiller, S. & Mlynek, J. Measurement of the quantum states of squeezed light. *Nature* 387, 471–475 (1997).
31. Krausz, F. & Ivanov, M. Y. Attosecond physics. *Rev. Mod. Phys.* 81, 163–234 (2009).
32. Nayak, A. et al. Saddle point approaches in strong field physics and generation of attosecond pulses. *Phys. Rep.* 833, 1–52 (2019).

**Publisher’s note** Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

© The Author(s), under exclusive licence to Springer Nature Limited 2021
Methods

Theoretical approach. Of course, the theoretical approach relies on several simplifying assumptions, but all of them can be avoided using less restrictive approximations. For instance, the reason why we can achieve these results is because we condition our analysis on the electrons being in or back in the ground state, ‘ignoring’ in a sense the electrons being ionized. Also, to describe a laser pulse with a given envelope, one needs a continuum of photon modes contributing to this wave packet. We avoid this difficulty by multiplying the atom-field interaction Hamiltonian by an envelope function $\hat{r}(t)$, in some analogy to scattering theory in many-body quantum field theories. Our starting point is the TDSE

$$i\hbar \frac{\partial}{\partial t} \langle \hat{\Psi}(t) \rangle = \hat{H} \langle \hat{\Psi}(t) \rangle,$$

where the Hamiltonian, $\hat{H}$, describes the laser-target system in the single-active-electron (SAE) approximation, and is the sum of three terms, that is, $\hat{H} = \hat{H}_0 + \hat{U} + \hat{V}$, where $\hat{H}_0 = \hat{H}_0^\text{sc} + \hat{V}(\hat{r})$ is the laser-free Hamiltonian of the atomic or molecular system with $\hat{V}(\hat{r})$ being the effective SAE atomic or molecular potential, $n$ is the electron mass; $\hat{U} = \hat{U}_c - \hat{U}_d$ is the dipole coupling, which describes the interaction of the atomic or molecular system with the laser radiation, written in the length gauge and under the dipole approximation; and, finally, $\hat{H}_0 = \int d\omega \hat{h}_0(\omega) \hat{a}_\omega^\dagger \hat{a}_\omega$ is the electromagnetic (E) field Hamiltonian containing all the frequency modes.

In principle, to describe laser/harmonic pulses of finite duration and spatial extension, the full continuum spectrum of the EM field must be considered, as stated in our original definition of $\hat{H}_0$. Here, we simplify the Hamiltonian to a sum of effective discrete modes. The free EM field Hamiltonian reduces in our case to

$$\hat{H}_0 = \hbar \omega_0 \hat{a}_0^\dagger \hat{a}_0 + \sum_\omega \hbar \omega_\omega \hat{a}_\omega^\dagger \hat{a}_\omega,$$

where $\hat{a}_0^\dagger (\hat{a}_0)$ and $\hat{b}_\omega^\dagger (\hat{b}_\omega)$ are creation (annihilation) operators of the laser and harmonic modes, respectively. To account for finite pulse duration, we model the electric field operator as

$$\hat{E} = -i\hbar g(\omega_{\text{eff}}) \int \frac{d\omega}{2\pi} \hat{a}_\omega \hat{d}_\omega^\dagger e^{i\mathbf{q} \cdot \mathbf{r}} + \text{c.c.},$$

where $g(\omega_{\text{eff}})$ is the quantum fluctuating part of the laser fields described in equation (9), and $\hat{d}_\omega$ is the quantum mechanical parameter entering into the expansion of the electric field into the modes by $\hat{E}(\mathbf{r}, t) \propto \sum_\omega \sqrt{\bar{\nu}_\omega} \hat{b}_\omega^\dagger$, where $\bar{\nu}_\omega$ is the effective quantization volume $\nu_\omega$ defined by $\nu_\omega = \int d\omega \sqrt{\nu_\omega} d\omega$, and for the typical frequencies used in strong-field physics is very small, on the order of $10^{-6}$ in a.u.

Equation (5) needs to be solved starting from the initial condition $\langle \hat{\Psi}(0) \rangle = |g, \alpha_1, \hat{\Theta}_0 \rangle$, that is for the electron initially being in its ground state $|g\rangle$, the laser in a coherent state $|\alpha_1\rangle$ and the harmonics in the vacuum state $|\Theta_0\rangle$. To this aim, we write $\langle \hat{\Psi}(t) \rangle = \exp(-i\hat{H}_0 t/\hbar) \langle \hat{\Theta}_0 \rangle$, where $\hat{\Theta}_0$ is the Glauber’s shift operator creating a coherent state $|\hat{\Theta}_0 \rangle$ from the vacuum of the laser mode, $|\hat{\Theta}_0 \rangle$. The second unitary operator transforms to the interaction picture with respect to the EM field. With these transformations, the initial state of the system is now described by $\langle \hat{\Psi}(t) \rangle = |g, \hat{H}_0(t), \hat{\Theta}_0 \rangle$ and the electric field part of the Hamiltonian shown in equation (5) becomes time dependent and gains an extra factor describing the behavior of the ‘classical’ field, that is, the mean value $\langle \alpha_1 \hat{E}(t) | \hat{a}_0 \rangle$. More explicitly, our Schrödinger equation now reads

$$i\hbar \frac{\partial}{\partial t} | \hat{\Psi}(t) \rangle = \left[ \hat{H}_0 - \hat{E}_Q(t) \cdot \hat{\eta} \right] | \hat{\Psi}(t) \rangle,$$

where $\hat{H}_0(t) = \hat{H}_0 - \hat{E}_Q(t) \cdot \hat{r}$ and $\hat{E}_Q(t) = -i\hbar g(\omega_{\text{eff}}) \int \frac{d\omega}{2\pi} \hat{a}_\omega^\dagger \hat{a}_\omega e^{i\mathbf{q} \cdot \mathbf{r}} - \alpha_1 e^{-i\mathbf{q} \cdot \mathbf{r}}$, the ‘classical’ electric field of the laser pulse. The quantum correction is

$$\hat{E}_Q(t) = -i\hbar g(\omega_{\text{eff}}) \int \frac{d\omega}{2\pi} \hat{a}_\omega^\dagger \hat{a}_\omega e^{i\mathbf{q} \cdot \mathbf{r}} - \alpha_1 e^{-i\mathbf{q} \cdot \mathbf{r}} + \sum_\omega \sqrt{\bar{\nu}_\omega} \hat{b}_\omega^\dagger e^{-i\omega t/\hbar}.$$

The next step is to go to the interaction picture with respect to $\hat{H}_0(t)$, something that we achieve with the following transformation

$$| \hat{\Psi}(t) \rangle = \mathcal{T} \exp \left[ -i \int_0^t \hat{H}_0(t') dt'/\hbar \right] | \hat{\Psi}(0) \rangle,$$

where $\mathcal{T}$ denotes the time-ordered product. Then we obtain:

$$i\hbar \frac{\partial}{\partial t} | \hat{\Psi}(t) \rangle = \hat{E}_Q(t) \cdot \hat{\eta} | \hat{\Psi}(t) \rangle.$$

The transformation depicted in equation (10) does not alter the initial condition for the system, but introduces in our Schrödinger equation the dynamics driven by the semiclassical term $\hat{H}_c(t)$ through the time-dependent dipole operator $\hat{r} \hat{H}(t)$, written now in the Heisenberg picture with respect to $\hat{H}_c(t)$. Due to this evolution, the electron might be ionized in the continuum (above threshold ionization process), or (hardly) remains in some bound excited state. Most of the physics relevant for HHG happens in the ground state; whatever remains there, remains there; whatever recombines to it, is related to harmonic emission. Therefore, it makes sense to condition equation (11) on the ground state, that is to consider $| \phi(t) \rangle = \langle g | \hat{\psi}(t) \rangle$, which fulfills equation (11), that is

$$i\hbar \frac{\partial}{\partial t} | \phi(t) \rangle = -\hat{E}_Q(t) \cdot \hat{\eta} | \phi(t) \rangle.$$
the Secretaria d’Universitats i Recerca del Departament d’Empresa i Coneixement de la Generalitat de Catalunya, as well as the European Social Fund (I’FSE invertí en el teu futur)–FEDER. P.T. group acknowledges LASERLABEUROPE (H2020-EU.1.4.1.2 grant ID 654148), FORTH Synergy Grant AgiiDA (grant no. 00133), the European Union’s Horizon 2020 framework programme for research and innovation under the NFPA-Europe-Pilot project (grant no. 101007417), the HELAS-CH (MIS grant no. 5002735) (which is implemented under the Action for Strengthening Research and Innovation Infrastructures, funded by the Operational Program Competitiveness, Entrepreneurship and Innovation (NSRF 2014-2020) and co-financed by Greece and the European Union (European Regional Development Fund)), and the European Union’s Horizon 2020 research. ELI-ALPS is supported by the European Union and co-financed by the European Regional Development Fund (GINOP grant no. 2.3.6-15-2015-00001).

Author contributions
M.L. supervised the theoretical part of the work; M.F.C., J.R.-D. and E.P. equally contributed to the manuscript preparation and the development of the theoretical approach; P.S. contributed to the theoretical calculations; Th.L. contributed in the experimental runs and data analysis; P.T. supervised the experimental part of the work.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41567-021-01317-w.
Correspondence and requests for materials should be addressed to M.L. or P.T.
Peer review information Nature Physics thanks the anonymous reviewers for their contribution to the peer review of this work.
Reprints and permissions information is available at www.nature.com/reprints.