Entanglement and Squeezing of the Optical Field Modes in High Harmonic Generation

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(Received 31 October 2023; revised 7 February 2024; accepted 29 February 2024; published 5 April 2024)

Squeezed optical fields are a powerful resource for a variety of investigations in basic research and technology. However, the generation of intense squeezed light is challenging. Here, we show that intense squeezed light can be produced using strongly laser driven atoms and the so far unrelated process of high harmonic generation. We demonstrate that when the intensity of the driving field significantly depletes the ground state of the atoms, leading to dipole moment correlations, the quantum state of the driving field and the generated high harmonics are entangled and squeezed. Furthermore, we analyze how the resulting quadrature squeezing of the fundamental laser mode after the interaction can be controlled. The findings open the way for the generation of high intensity squeezed light states for a wide range of applications.

DOI: 10.1103/PhysRevLett.132.143603

Introduction.—The field of attosecond science [1] has recently established connections to the field of quantum optics and quantum information science [2,3]. This is mainly due to the efforts in describing systems driven by strong light fields with fully quantized approaches [4], going beyond the extremely successful (semi)classical methods [5]. In particular, the process of high harmonic generation (HHG) [6,7] has been the subject of various investigations to gain novel insights into the radiation properties of the scattered field. These insights were elusive from a semiclassical perspective without the quantization of the electromagnetic field [8–10]. It was shown that the final quantum state of the field modes are given by product coherent states [9,10], which holds when assuming small depletion of the ground state and is related to vanishing dipole moment correlations [11] (note that a coherent state driving field needs to be assumed as well [12]). However, further analysis has shown that the field state generated in the HHG process is entangled [13,14]. In this work, we show how the description of the final quantum optical state after HHG by means of product coherent states is a consequence of the aforementioned approximations [15]. In particular, we show that all field modes are naturally entangled, and further show that each mode exhibits squeezing.

We illustrate the onset of mode squeezing for increasing driving field strength, due to the increased ground state depletion leading to dipole moment correlations. As a proof of principle we further show that the quadrature squeezing of the fundamental driving mode can be tuned by varying the carrier-envelope phase (CEP) of the driving laser field. This allows one to rotate the angle of the squeezed field quadrature. This is particularly interesting since the squeezing already occurs in the simplest scenario when driving uncorrelated atoms with classical laser light. Thus far, squeezed light has been considered to drive the HHG process [16,17], showing an extension of the generated spectrum. However, the other field properties besides the spectrum, such as squeezing [8] or quantum coherence [12], only received very limited attention in HHG. Here, we will observe two-mode squeezing between all field modes participating in the process, which naturally leads to an entangled state between all modes [18].

Generating such squeezed and massive entangled states is of importance for modern quantum technologies [19–21]. This further highlights the use of strong-laser driven systems for quantum state engineering of light [4,14], which has already shown the ability to generate non-classical states of light by means of optical Schrödinger cat states [9] having sufficient photon numbers to induce nonlinear phenomena [22]. This work further manifests the intrinsic properties in the HHG process beyond the semiclassical framework by showing that entanglement and squeezing naturally occurs even in simple gas targets.

Quantum optical HHG without dipole correlations.—In the quantum optical description of HHG the interaction of an intense laser field, described by the coherent state $|\alpha\rangle$, with an atom in the ground state $|g\rangle$ is given by the following interaction Hamiltonian (a detailed derivation can be found in [4]):

$$H_I(t) = -d(t)E_Q(t), \tag{1}$$

where the electric field operator is given by

$$E_Q(t) = -igf(t)\sum_{q=1}^{N}\sqrt{q}\,(a_q^\dagger e^{i\omega_q t} - a_q e^{-i\omega_q t}), \tag{2}$$

with $q \geq 2$ being the harmonic field modes and the dimensionless function $f(t)$ is introduced for using a discrete set of modes and takes into account the finite duration of the driving pulse. The electric field operator is coupled to the time-dependent dipole moment operator

$$d(t) = U_{sc}^\dagger(t) d U_{sc}(t). \tag{3}$$

The dipole moment is in the interaction picture of the reference frame $U_{sc}(t) = \mathcal{T}\exp[-i\int_0^t d\tau H_{sc}(\tau)]$, with respect to the semiclassical Hamiltonian of the electron $H_{sc}(t) = H_A - dE_{cl}(t)$. This semiclassical Hamiltonian is the same as the one traditionally considered in semi-classical HHG theory [6], where $H_A = p^2/2 + V(r)$ is the pure electronic Hamiltonian, and

$$E_{cl}(t) = \mathrm{Tr}[E_Q(t)|\alpha\rangle\langle\alpha|] = igf(t)(\alpha e^{-i\omega t} - \alpha^* e^{i\omega t}), \tag{4}$$

is the classical part of the driving laser electric field. Solving the dynamics for the parametric process of HHG, in which the electron is found in the ground state after the end of the pulse, the dynamical equation for the field state conditioned on the electronic state $|g\rangle$ is given by

$$i\partial_t|\Phi(t)\rangle = -\langle g|d(t)E_Q(t)|\Psi(t)\rangle, \tag{5}$$

where $|\Phi(t)\rangle = \langle g|\Psi(t)\rangle$, with $|\Psi(t)\rangle$ the state of the total system. The general solution for the field state conditioned on the electronic ground state is given by [14]

$$|\Phi(t)\rangle = \langle g|U(t)|g\rangle|\Phi_i\rangle = K_{HHG}|\Phi_i\rangle, \tag{6}$$

with the conditioned evolution operator

$$K_{HHG} = \langle g|\mathcal{T}\exp\left[i\int_0^t dt' d(t')E_Q(t')\right]|g\rangle, \tag{7}$$

which solely acts on the initial field state $|\Phi_i\rangle = |\{0_q\}\rangle$. To obtain the exact solution for the field we introduce the identity on the electronic subspace $\mathbb{1} = |g\rangle\langle g| + \int dv|v\rangle\langle v|$ (for the sake of simplicity we use a $1d$ momentum representation) [4,9,10], and in the spirit of the strong field approximation (SFA) [5] we neglect continuum-continuum transitions such that we have to solve the following set of coupled equations:

$$\partial_t|\Phi(t)\rangle = iE_Q(t)\langle g|d(t)|g\rangle|\Phi(t)\rangle + i\int dv E_Q(t)\langle g|d(t)|v\rangle|\Phi(v,t)\rangle, \tag{8}$$

$$\partial_t|\Phi(v,t)\rangle = iE_Q(t)\langle v|d(t)|g\rangle|\Phi(t)\rangle, \tag{9}$$

where we have defined $|\Phi(v,t)\rangle = \langle v|\Psi(t)\rangle$ for the continuum state $|v\rangle$ of the electron with velocity v. Solving the exact dynamics for $|\Phi(t)\rangle$ in (7) is a tedious task, and approximations based on the physical situation under consideration are necessary. For instance, we can assume that the depletion of the electronic ground state is small [6], such that the second term in (8) can be neglected since it is proportional to the total continuum state amplitude. The remaining equation can then be solved and is given by [14]

$$K_{HHG} \simeq \mathcal{T}\exp\left[i\int_0^t dt'\langle d(t')\rangle E_Q(t')\right] = \mathbf{D}[\chi] \tag{10}$$

leading to a displacement operation for each mode $\mathbf{D}[\chi] = \prod_q D[\chi_q]$, with the coherent state displacements $\chi_q \propto \int_0^t dt' f(t')\langle d(t')\rangle e^{i\omega_q t'}$, which are proportional to the Fourier transform of the dipole moment expectation value $\langle g|d(t)|g\rangle = \langle d(t)\rangle$. Note that the approximation to obtain Eq. (10) is equivalent to neglecting dipole moment correlations of the electron [11,13,14], such that the final field state

$$|\Phi(t)\rangle = K_{HHG}|\{0_q\}\rangle = |\{\chi_q\}\rangle \tag{11}$$

is given by product coherent states. We shall now show that the actual field state in the process of HHG is not given by product coherent states when taking into account the dipole moment correlations, and we will see that the proper final field state is entangled and squeezed.

Including dipole moment correlations.—The crucial approximation to find the expression for the final field state in (11) is based on the assumption of negligible depletion of the ground state amplitude, equivalent to neglecting dipole moment correlations of the electron. That is, the approximation from (7) to (10) leading to the product coherent states of all field modes (11), due to a linear expression in the field operators $a_q^{(\dagger)}$ in (10). In typical experiments with moderate laser intensities ($\leq 1\times 10^{14}$ W/cm^2), this assumption is reasonable, but if we increase the intensity to higher values, the ground state depletion can start to play a role. Formally, the dipole moment correlations imply that the exact interaction Hamiltonian in the propagator (7) does not commute at different times and remains an operator in the total Hilbert space ($\mathcal{H}_A \otimes \mathcal{H}_F$) of the electron plus the field

$$[H_I(t_1), H_I(t_2)] \in \mathcal{H}_A \otimes \mathcal{H}_F. \tag{12}$$

In contrast, the commutator of the approximate interaction Hamiltonian $H_I'(t) = -\langle d(t)\rangle E_Q(t)$ is just a complex number, i.e., $[H_I'(t_1), H_I'(t_2)] \in \mathbb{C}$, and thus, when solving (7), the modes do not mix. Going beyond the linear term of the field operator $E_Q(t)$ would lead, for instance, to squeezing in the field modes. Furthermore, all field modes will become entangled due to the mixing of the field operators $a_q^{(\dagger)}$ of the different modes. We therefore, anticipate signatures of squeezing and entanglement between the modes when going beyond the approximation of neglecting dipole moment correlations. This is done by solving (8) without the assumption of negligible ground state depletion of the electron, i.e., taking into account the second term of the continuum states. We thus solve the dynamics for higher orders of the exact interaction Hamiltonian $H_I(t) = -d(t)E_Q(t)$ instead of $H_I' = -\langle d(t)\rangle E_Q(t)$ as done in (10). Especially, we want to find a solution of (8), and have

$$\partial_t|\Phi(t)\rangle = iE_Q(t)\langle d(t)\rangle|\Phi(t)\rangle - E_Q(t)\int_0^t dt' \int dv\, d_{vg}^*(t) d_{vg}(t') E_Q(t')|\Phi(t')\rangle, \tag{13}$$

where $d_{vg}(t) = \langle v|d(t)|g\rangle$. In order to find the evolution operator for the state $|\Phi(t)\rangle$, we perform a Markov type approximation $|\Phi(t')\rangle \to |\Phi(t)\rangle$, and make use of the identity on the electronic subspace $\int dv|v\rangle\langle v| = \mathbb{1} - |g\rangle\langle g|$, such that

$$\partial_t|\Phi(t)\rangle = [iE_Q(t)\langle d(t)\rangle - \langle \dot{Q}(t)Q(t)\rangle]|\Phi(t)\rangle, \tag{14}$$

where we have defined

$$Q(t) = \int_0^t dt' E_Q(t')[d(t') - \langle d(t')\rangle]. \tag{15}$$

Neglecting the commutators $[Q(t), Q(t')]$ we find [23]

$$\langle \dot{Q}(t)Q(t)\rangle = \frac{1}{2}\frac{\partial}{\partial t}\langle Q^2(t)\rangle, \tag{16}$$

and thus, the evolution of the initial state reads

$$|\Phi(t)\rangle = \mathbf{D}[\chi]e^{-\frac{1}{2}\langle Q^2(t)\rangle}|\{0_q\}\rangle. \tag{17}$$

Here, we have used that $[E_Q(t), Q(t')]$ is a complex number such that the two operations in (17) can be factorized. The additional contribution is quadratic in $Q(t)$ and thus in the electric field operator and therefore leads to squeezing and mixing between the field modes. Evaluating this term we find

$$\langle Q^2(t)\rangle = \int_0^t \int_0^t dt' dt'' E_Q(t')E_Q(t'') \times \langle [d(t') - \langle d(t')\rangle][d(t'') - \langle d(t'')\rangle]\rangle. \tag{18}$$

As done in previous works [9,10], we will focus on the fundamental mode of the driving field (with operators $a_1^{(\dagger)} = a^{(\dagger)}$) since the amplitudes of the harmonics are much smaller. We can thus write

$$\langle Q^2\rangle = -g^2[(a^\dagger)^2\langle \mathcal{D}(\omega)\mathcal{D}(\omega)\rangle + a^2\langle \mathcal{D}^\dagger(\omega)\mathcal{D}^\dagger(\omega)\rangle - a^\dagger a\langle \mathcal{D}(\omega)\mathcal{D}^\dagger(\omega)\rangle - aa^\dagger\langle \mathcal{D}^\dagger(\omega)\mathcal{D}(\omega)\rangle], \tag{19}$$

where we have considered $t \to \infty$ and defined

$$\mathcal{D}(\omega) = \int_0^\infty dt' f(t')e^{i\omega t'}[d(t') - \langle d(t')\rangle]. \tag{20}$$

We expect all the averages to be comparable, i.e., $|\langle \mathcal{D}(\omega)\mathcal{D}(\omega)\rangle| = \langle \mathcal{D}^\dagger(\omega)\mathcal{D}(\omega)\rangle = B$, where we have used that $\mathcal{D}^\dagger(\omega)\mathcal{D}(\omega)$ is self-adjoint and defined $\langle \mathcal{D}(\omega)\mathcal{D}(\omega)\rangle^* = \langle \mathcal{D}^\dagger(\omega)\mathcal{D}^\dagger(\omega)\rangle \equiv Be^{-2i\psi}$, such that we can write

$$\langle Q^2\rangle = 2g^2B\left[\frac{i}{\sqrt{2}}(a^\dagger e^{i\psi} - ae^{-i\psi})\right]^2. \tag{21}$$

Interestingly, this includes a quadrature operator, leading to squeezing along the momentum quadrature

$$P_\psi = \frac{i}{\sqrt{2}}(a^\dagger e^{i\psi} - ae^{-i\psi}). \tag{22}$$

We can now write down the final field state of the fundamental mode after the process of HHG, which is given by (in the laboratory frame)

$$|\Phi_1\rangle = D[\alpha]D[\chi_1]S(\psi)|0\rangle, \tag{23}$$

which is a displaced and squeezed vacuum state, i.e., a high photon number squeezed coherent state, with the squeezing operator given by

$$S(\psi) = \exp[-g^2BP_\psi^2]. \tag{24}$$

We note that varying the squeezing phase ψ in (22) allows us to consider squeezing along different directions given by the quadrature operator P_ψ.

Bright squeezed states in HHG.—In order to quantify the squeezing in the fundamental mode, we need the correlations of the dipole moment fluctuations around its mean $\mathcal{D}(\omega)$ in (20). We therefore numerically solve [24]

$$\langle \mathcal{D}(\omega)\mathcal{D}(\omega)\rangle = \int_0^\infty dt' \int_0^\infty dt'' f(t')f(t'')e^{i\omega t'}e^{i\omega t''} \times [\langle d(t')d(t'')\rangle - \langle d(t')\rangle\langle d(t'')\rangle], \tag{25}$$

from which we obtain the single atom contribution to the squeezing $|\langle\mathcal{D}(\omega)\mathcal{D}(\omega)\rangle| = B$. Taking into account that in the interaction region we have N_{at} atoms independently contributing to the HHG process we can write the total squeezing power via the squeezing parameter $r \equiv -g^2 B N_{\text{at}}$ in units of $[\text{db}] = 10\log_{10}(e^{2|r|^2})$. In the derivation of the final field state we have seen that including dipole moment correlations can originate from the depletion of the ground state. Since this depletion increases for increasing field strength, we first demonstrate the onset of the field squeezing in HHG. In Fig. 1 we show the squeezing of the fundamental mode for increasing field strength for two different pulse durations of the driving laser [29]. We can clearly see that the squeezing increases for increasing field strength due to the depletion of the ground state for fields of higher intensity. On the other hand, shorter pulses induce less depletion [5,30], and hence lead to smaller field squeezing. In summary, we find that the more depletion of the ground state, the squeezing increases. Thus, the approximation of neglecting the continuum population in (13), as done in previous works [9,10], does not hold anymore. Furthermore, we observe that the imaginary part of the squeezing parameter only contributes for relatively large field strength compared to the onset of overall squeezing [see Fig. 1(b)]. The appearance of imaginary squeezing comes from breaking the time inversion symmetry of the dipole moment correlations when taking into account depletion effects. It happens that the dipole correlations (25) are real within standard SFA theory without depletion, while the depletion effects break the time inversion symmetry leading to a complex valued squeezing parameter (see Supplemental Material [24]). Further, we note that for very short pulses the symmetry can also be broken for specific CEP values, even for relatively small depletion.

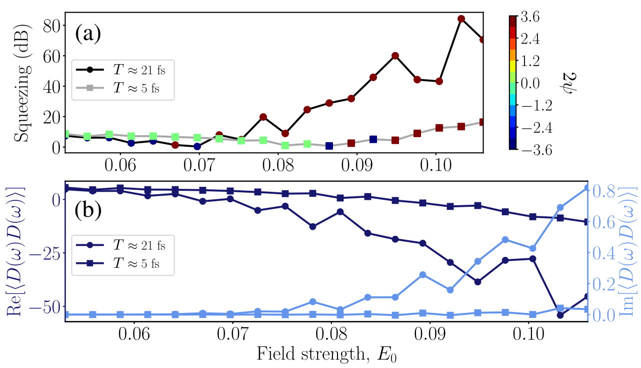

FIG. 1. (a) Squeezing parameter of the fundamental mode is shown as a function of the field strength for pulses with different duration. To convert to dB units, we employed the relation $[\text{db}] = 10\log_{10}(e^{2|r|^2})$, with $r \equiv -g^2 B N_{\text{at}}$ denoting the squeezing parameter. The target is an H atom in its 1s ground state. Here, we have considered that $N_{\text{at}} = 5 \times 10^{13}$ atoms independently contribute to HHG in the interaction region [9]. Specifically, $T \approx$ 21 fs (eight optical cycles) for the black curve with circular markers, whereas $T \approx 5$ fs (two optical cycles) for the gray curve with squared markers. (b) The dark blue curve represents the real component (leftmost axis) of Eq. (25) for the aforementioned pulse duration, while the lighter blue curve (rightmost axis) represents its imaginary component. In both cases, the intensity values range from 1×10^{14} W/cm^2 to 4×10^{14} W/cm^2 while the central wavelength has been fixed to $\lambda = 800$ nm. Depletion was taken into account using the TBI model described in Ref. [31] (see Supplemental Material [24]).

We have just seen that field squeezing occurs when the ground state depletion becomes relevant and that breaking the time inversion symmetry of the dipole correlations leads to imaginary squeezing. Therefore, we shall now consider explicit control schemes of the squeezing phase by tuning the driving laser field properties. In Fig. 2 we show the squeezing power in the fundamental mode after the HHG process for varying the carrier-envelope phase of the driving laser field. The squeezing phase $\psi = \frac{1}{2}\arg[\langle\mathcal{D}(\omega)\mathcal{D}(\omega)\rangle]$ is indicated with the color coding for each CEP value. We observe that by varying the CEP we can control the squeezing power and the squeezing phase, which allows one to rotate the angle of the quadrature operator along which the squeezing occurs. The arrows at different CEP values in Fig. 2 show different shapes of the electric field of the driving pulse. To further illustrate the effect of the squeezing on the fundamental mode, and to show the rotation of the quadrature operator, the Wigner function [32] of the state in (23) is shown in Fig. 3 for the CEP values indicated by the arrows in Fig. 2. We can see significant squeezing along the field quadratures and the ability to rotate the squeezing ellipse by varying the CEP. Further, we can see how the symmetry between the pulse shapes (see Fig. 2) is reflected in the orientation of the

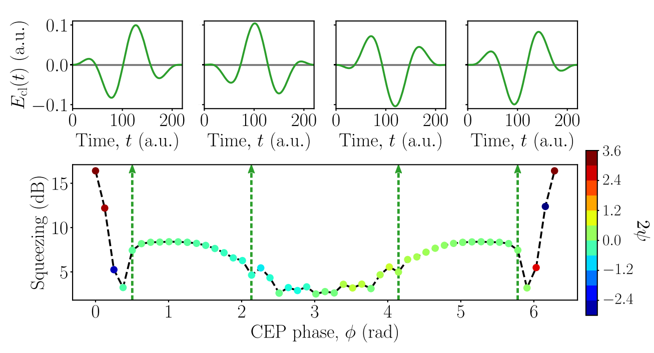

FIG. 2. Squeezing parameter of the fundamental mode $\langle\mathcal{D}(\omega)\mathcal{D}(\omega)\rangle$ versus the carrier-envelope phase in dB units. The driving laser field used in the calculations has a field strength of $E_0 = 0.106$ a.u. (corresponding to an intensity of 4×10^{14} W/cm^2) with a central frequency $\omega = 0.057$ a.u. (corresponding to a wavelength $\lambda = 800$ nm) and a $\sin^2$-shaped envelope with two cycles of total duration (approximately 5 fs). The laser field is plotted in the upper panels for each of the CEP values indicated. The arrows in this plot correspond to the values for which the Wigner function is shown in Fig. 3.

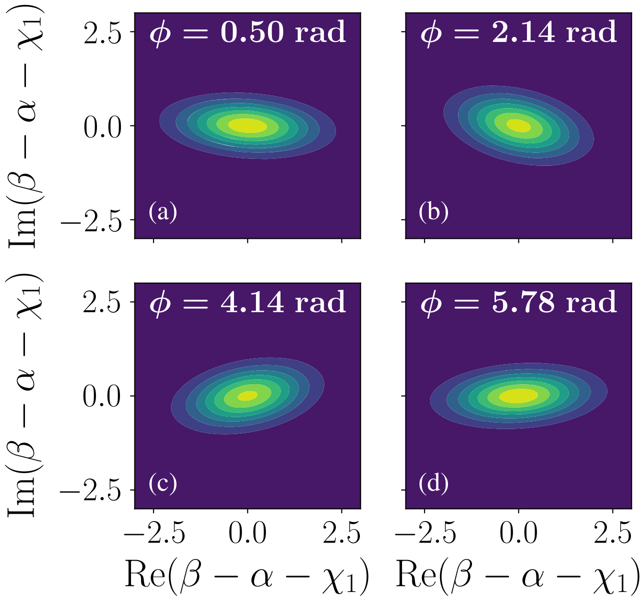

FIG. 3. Wigner function $W(\beta)$ of the state of the fundamental mode (23) for different values of the CEP ϕ, with the same laser parameters as specified in the caption of Fig. 2. The cases (a)–(d) correspond to the arrows indicated in Fig. 2, illustrating the ability to control the rotation of the quadrature squeezing by varying the CEP.

squeezing quadrature showing a rotation of the Wigner function in Fig. 3 between (a) and (d) of π. The same holds between (b) and (c) in Fig. 3, which emphasizes that there exists a relation between the CEP phase ϕ and the squeezing phase ψ, such that when applying a sign change and time-reversal symmetry operation on the driving pulse, the squeezing phase changes according to $\psi \to \pi - \psi$.

Entanglement in HHG.—Now, we shall take into account all harmonic field modes, to show that the final state of the field is in general entangled. Starting from (18) by considering all modes in the field operator, we can write

$$\langle Q^2 \rangle = -g^2 \sum_{q,p} \sqrt{qp} [a_q^\dagger a_p^\dagger \langle \mathcal{D}(\omega_q) \mathcal{D}(\omega_p) \rangle - a_q^\dagger a_p \langle \mathcal{D}(\omega_q) \mathcal{D}^\dagger(\omega_p) \rangle + \mathrm{H.c.}]. \tag{26}$$

This is a generic bilinear Hermitian operator for all the bosonic field modes participating in the process of HHG. The terms in which $q = p$, i.e., the pairs of identical harmonic photons created or annihilated, correspond to a single mode squeezing, and the terms with $q \neq p$ correspond to two-mode squeezing. It is known that those two-mode squeezing terms are responsible for entanglement between the two modes [18]. This further manifests that the actual field state in the process of HHG is in general entangled. In the particular case of HHG, where many different field modes participate in the process, this two-mode squeezing leads to massive entangled states. In view of quantum information processing tasks with continuous variable systems [19,20] the detection and characterization of entanglement in continuous variable systems is of great importance [33,34]. Taking into account that N modes participate in the process of HHG, we expect the final field state to exhibit genuine multipartite continuous variable entanglement [35], and its detection is the subject of future investigation.

Conclusions.—We have shown that the final field state in the process of HHG is an entangled state between all field modes and that each mode is squeezed. The onset of the squeezing and its physical interpretation based on dipole moment correlations is provided, as well as the explanation of the complex valued squeezing parameter. It is shown that the squeezing can be controlled by varying the CEP of the driving field, which allows one to rotate the quadrature operator along which the squeezing occurs. This establishes the process of HHG as a new light source for bright squeezed radiation. Showing that the field is entangled provides further usefulness of the recently emerging connection of attosecond physics with quantum information science [2,3]. Especially the generation of a genuine multipartite entangled continuous variable system is of fundamental and technological importance. In conclusion, we have uncovered previously unknown field properties in the HHG process, and we anticipate that driving more complex systems [36–38] or using nonclassical driving fields [16,17,39] could lead to further insights into the entanglement and squeezing properties of the generated harmonic light. Finally, we note that the derivation of the field state is generic, and that the dipole moment correlations are the constitutive components for the field entanglement and squeezing. Going beyond strong field dynamics in simple targets may provide novel light sources with unexpected properties.

P. S. acknowledges funding from the European Union's Horizon 2020 research and innovation program under the Marie Skłodowska-Curie Grant Agreement No. 847517. J. R.-D. acknowledges funding from the Government of Spain (Severo Ochoa CEX2019-000910-S and TRANQI), Fundació Cellex, Fundació Mir-Puig, Generalitat de Catalunya (CERCA program) and the ERC AdG CERQUTE. ICFO group acknowledges support from: Europea Research Council AdG NOQIA; MCIN/AEI (PGC2018-0910.13039/501100011033, CEX2019-000910-S/10.13039/501100011033, Plan National FIDEUA PID2019-106901GB-I00, Plan National STAMEENA PID2022-139099NB, I00, project funded by MCIN/AEI/10.13039/501100011033 and by the "European Union NextGenerationEU/PRTR" (PRTR-C17.I1), FPI); QUANTERA MAQS PCI2019-111828-2); QUANTERA DYNAMITE PCI2022-132919, QuantERA II Programme co-funded by European Union's Horizon 2020 program under Grant Agreement No 101017733); Ministry for Digital Transformation and of Civil Service of the Spanish Government through the QUANTUM ENIA project call—Quantum Spain project, and by the European Union through the Recovery, Transformation and Resilience Plan—NextGenerationEU within the framework of the Digital Spain 2026 Agenda; Fundació Cellex; Fundació Mir-Puig; Generalitat de Catalunya (European Social Fund FEDER and CERCA program, AGAUR Grant No. 2021 SGR 01452, QuantumCAT\U16-011424, co-funded by ERDF Operational Program of Catalonia 2014-2020); Barcelona Supercomputing Center MareNostrum (FI-2023-1-0013); Funded by the European Union. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union, European Commission, European Climate, Infrastructure and Environment Executive Agency (CINEA), or any other granting authority. Neither the European Union nor any granting authority can be held responsible for them (EU Quantum Flagship PASQuanS2.1, 101113690, EU Horizon 2020 FET-OPEN OPTOlogic, Grant No. 899794), EU Horizon Europe Program (This project has received funding from the European Union's Horizon Europe research and innovation program under Grant Agreement No. 101080086 NeQST Grant Agreement No. 101080086—NeQST); ICFO Internal "QuantumGaudi" project; European Union's Horizon 2020 program under the Marie Sklodowska-Curie Grant Agreement No. 847648; "La Caixa" Junior Leaders fellowships, La Caixa" Foundation (ID 100010434): CF/BQ/PR23/11980043.

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