

Observation of a displaced squeezed state in high-harmonic generation

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High-harmonic generation (HHG) and quantum optics are historically two distant fields of science. HHG as a central technique to generate attosecond pulses via atoms, molecules, or solids has been mostly treated as a classical source of light. Entanglement and squeezing of HHG light would open fascinating perspectives in quantum technology, with the emergence of a field of basic and applied research: attosecond quantum optics. Here, we measure entanglement in a tripartite solid-state HHG system through the violation of a Cauchy-Schwarz inequality. Displaced squeezing is experimentally observed and supported by numerical simulations with excellent agreement to the data. The Schmidt number is estimated, which indicates a low-dimensional multimode structure for each harmonic, an important property for most quantum technologies. At the applied level, the semiconductor HHG source is scalable, integrable, operates at room temperature with compact lasers, and thus represents a useful and exploitable resource in quantum photonics.

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I. INTRODUCTION

Nonclassical sources of light are a versatile resource in quantum information science and technology [1], thanks to their unique properties such as entanglement and squeezing to boost metrology [2–4], imaging [5,6], or to establish secure communication [7–9]. Another promising prospect is photonic quantum simulation [10–12] that uses displaced squeezed states of light, superposed to simulate arbitrary quantum systems. The main advantage of photonic quantum states is their robustness against decoherence due to the low interaction of propagating photons with the environment. Furthermore, the agility of light manipulation offers the possibility of encoding high-dimensional entanglement in time bins, frequency bins, or topological states [13].

One critical showstopper for realizing these fantastic prospects promised by quantum technology is the difficulty in increasing the amplitude of entangled quantum states of light. The scaling problem arises from the intrinsic probabilistic nature of quantum mechanics and the usual strategy to manipulate arduously isolated quantum systems with weakly interacting optical and electric fields. After decades of tremendous research effort, the generation of entangled states of light with high photon number or a high number of entangled modes still requires complex experimental setups.

High-harmonic generation (HHG) has the potential to overcome this bottleneck by generating many modes, high-dimensional quantum states of light.

Theoretical works on HHG have shown that the process could potentially be a highly versatile source of nonclassical light. HHG is a nonperturbative frequency upconversion process that can generate broadband frequency combs covering a spectral range spanning from the near-infrared down to the extreme ultraviolet [14–16]. The ultrashort time structure of the HHG radiation is inherited from the highly nonlinear interaction of electrons in atoms, molecules, or condensed matter with the periodically oscillating driving electric field potential of an ultrashort pulse of light [17,18]. This petahertz-frequency electron wave-packet pendulum then generates attosecond bursts of light as a response to the strong field interaction. Nonclassical effects in HHG open avenues to be explored, exploiting the broadband generation of nonclassical photon correlation [19,20], multipartite entanglement [19,21], and attosecond control of squeezed states of light [22], as well as conditioned generation of cat states [23,24]. The first experimental work on semiconductor HHG (SHHG) reported the generation of squeezed states of light [25]. It has also been shown that HHG can be driven by nonclassical light [26]. However, a premise in using HHG as a quantum resource is a comprehensive analysis of entanglement and mode structure. The concept of modes is well known in classical electromagnetism and refers to multiple solutions of the Maxwell equations. The electromagnetic field can propagate in, e.g., different spatial, polarization, temporal, or spectral modes. Here, we are investigating the distribution of quantum mechanical properties, which are extended to multiple optical modes [27]. The mode analysis of quantum states is of great importance, as it leverages engineering to monitor and select

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favorable modes for specific photonic quantum-state applications [28–31].

To this extent, higher-order intensity correlation functions can be analyzed to infer the underlying modal distribution of the generated state. Being closely connected to the moment-generating functions, higher-order photon number correlation functions probe properties such as the width and skewness of the underlying photon number distribution. In the case of multimode (MM) squeezed states of light, they can also, under some assumptions, be used to estimate the modal squeezer distribution [31]. Based on this, entanglement can be quantified by an analysis of the multimodal structure through an effective Schmidt mode decomposition of bipartite subsystems.

In this work, we analyze the modal distribution of nonclassical states of HHG photons generated, by measuring higher-order correlation functions. The nonclassicality of a tripartite harmonic state is demonstrated through the violation of a set of Cauchy-Schwarz inequalities (CSIs). Here, the quantized nature of the HHG light and the strong nonclassical correlations between photons of different harmonic orders are responsible for the violation of the CSI. Further, we extend existing techniques analyzing a squeezed vacuum state [31–33] to a more general one, including displacement of the state.

We explore the mode decomposition of SHHG from a cadmium telluride (CdTe) crystal with a [110] cut and a bandgap of 1.44 eV (Fig. 1). This crystal has recently reported a relatively high conversion efficiency ($\approx 10^{-10}$ in our experiment) due to its large density of states near the Fermi level and a high carrier mobility [34]. Such features make this material favorable for signal-demanding quantum information applications. By simultaneously measuring the second-order correlation function (SCF) and third-order correlation function (TCF), we extract information about the modal structure and key parameters of the generated state. Further, we confirm previous results showing multimode squeezed light [25]. The violation of a CSI is shown to be even more significant in CdTe compared to previous studies [25].

A displaced squeezed state of light is verified and supported by numerical simulation. Our analysis shows that, in addition to squeezing, displacement is necessary to explain the observed dependencies of correlation functions on the driving laser intensity. Based on the Schmidt decomposition, the dimensionality of the state is estimated.

II. MEASUREMENT OF A MULTIMODE CAUCHY-SCHWARZ INEQUALITY

The Hanbury-Brown-Twiss-like configuration allows us to retrieve the SCF expressed as

$$g_{ij}^{(2)}(\tau) = \frac{\langle \hat{a}_i^\dagger(\tau) \hat{a}_j^\dagger(0) \hat{a}_i(0) \hat{a}_j(\tau) \rangle}{\langle \hat{a}_i^\dagger(0) \hat{a}_i(0) \rangle \langle \hat{a}_j^\dagger(\tau) \hat{a}_j(\tau) \rangle}, \quad (1)$$

where $\hat{a}^\dagger$ and $\hat{a}$ denote the creation and annihilation operators of photons in the mode i or j representing two of the three central spectral modes of three harmonic states H3, H4, and H5. As the HHG pulses have a duration much shorter than the timing resolution of the detection (200 ps), we effectively

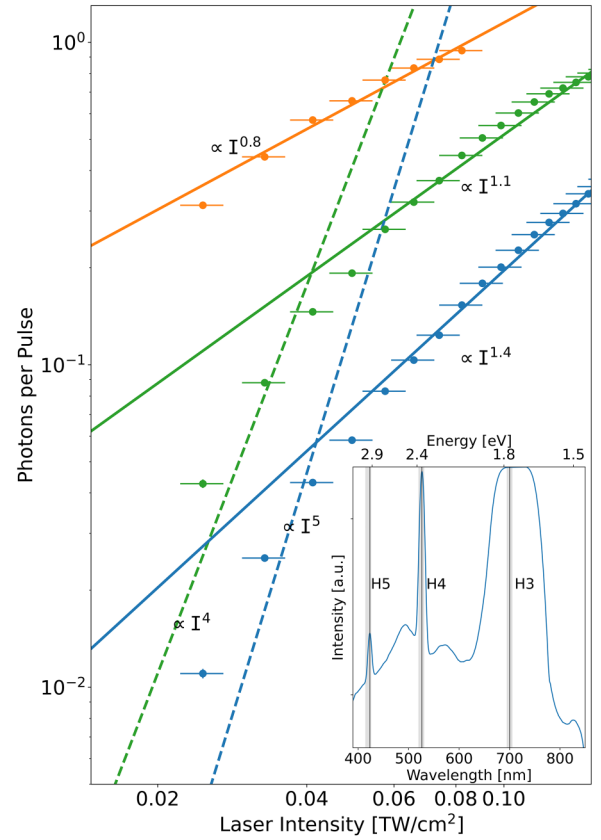


FIG. 1. Confirmation of nonperturbative HHG. The graph shows the loss-corrected number of photons per pulse of three harmonic orders (third order in yellow, fourth order in green, and fifth order in blue) over the driving laser intensity. For all recorded high-harmonic orders, an intensity crossover is apparent, at which the dependency changes from a perturbative to a nonperturbative scaling law. The fitted curves (solid lines) are the best fit of a power law, and the perturbative scaling law is depicted to ease the comparison (dashed lines). The inset shows the spectrum with the three investigated high harmonics. The gray area represents the width of the spectral filters employed in the experimental setup. Spectral filtering is already a mode selection, as we thereby reduce the width of the squeezer distribution.

perform a broadband, multimode measurement

$$g_{ij}^{(2)}(0) \equiv g_{ij}^{(2)} = \frac{\int d\omega_1 \int d\omega_2 \langle \hat{a}_i^\dagger(\omega_1) \hat{a}_j^\dagger(\omega_2) \hat{a}_i(\omega_1) \hat{a}_j(\omega_2) \rangle}{\int d\omega_1 \langle \hat{a}_i^\dagger(\omega_1) \hat{a}_i(\omega_1) \rangle \int d\omega_2 \langle \hat{a}_j^\dagger(\omega_2) \hat{a}_j(\omega_2) \rangle}, \quad (2)$$

where the convolution of the photon statistics of the spectral modes is measured [25,31,32]. In turn, this acquisition is sensitive to squeezing effects in the radiation.

We observe a characteristic transition of the SCF when adjusting the driving laser intensity from $g_{ij}^{(2)} \gg 2$ to $g_{ij}^{(2)} = 1$ [Fig. 2(c)]. Connecting the SCF to the photon statistics of the radiation, these values indicate a pronounced super-Poissonian photon number distribution in each of the measured harmonic orders. This distinctive dependence on the driving laser intensity and strong interbeam correlations is indicative of the presence of two-mode squeezing in HHG

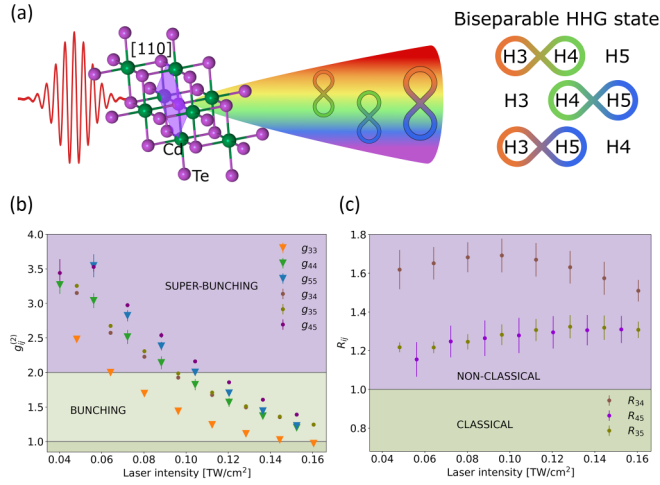


FIG. 2. Quantum Optical Characterization of HHG. (a) Schematic illustration of the HHG state. An ultrashort laser pulse is focused on a CdTe crystal. In the interaction, distinct spectral frequencies are generated. Our data suggest the presence of two-mode squeezing in the measured harmonic orders resulting in a biseparable state. (b) Intensity-dependent photon statistics. The graphic depicts the auto- and cross-correlations $g_{ij}^{(2)}$ for the three different harmonics acquired with the setup shown in Fig. 5. A clear transition from superbunching with $g^{(2)} > 2$ toward Poissonian photon statistics $g^{(2)} = 1$ is visible. This behavior is characteristic for a photonic state exhibiting multimode squeezing. As the driving laser intensity is increased, the squeeze distribution is broadened and a convolution of the photon statistics of the individual spectral modes is measured. As these are uncorrelated with each other, it results in a transition toward $g^{(2)} = 1$. For low driving laser intensities, only few squeezing modes are excited such that the super-Poissonian photon number distribution is resolved. (c) Certification of the nonclassicality of HHG. From the experimental data in panel (c), we evaluate the R parameter, as defined in Eq. (4). For all bipartite sets and across all driving laser intensities, we observe $R > 1$ with high significance, confirming that the generated state from SHHG is nonclassical. The error bars are calculated from the multiple repetitions as indicated in the experimental data. The pairwise CSI violation and the observation of two-mode squeezing signatures imply the generation of three entangled biseparable states.

as reported in previous works [32,35]. Intuitively, the scaling of the SCF with an increase in the driving laser intensity can be explained by an increase in the number of modes exhibiting squeezing effects in one harmonic mode. With an increase in the driving laser intensities, progressively squeezers are excited in additional spectral modes. As these are uncorrelated with each other, the photon statistics measured with broadband detection become steadily more random. For squeezed vacuum states, an approximated expression for the measured SCF for multimode state can be derived. In this formula, the SCF is inversely proportional to the mean number of photons in the state. Although the scaling of our data resembles this formula, we find that a correction is needed when fitting it to our experimental data. Theoretical models of SHHG include the possibility of the presence of displacement in addition to squeezing. For displaced squeezed states, it is likely that this approximate expression is not directly applicable anymore. However, in such cases, the displacement in the

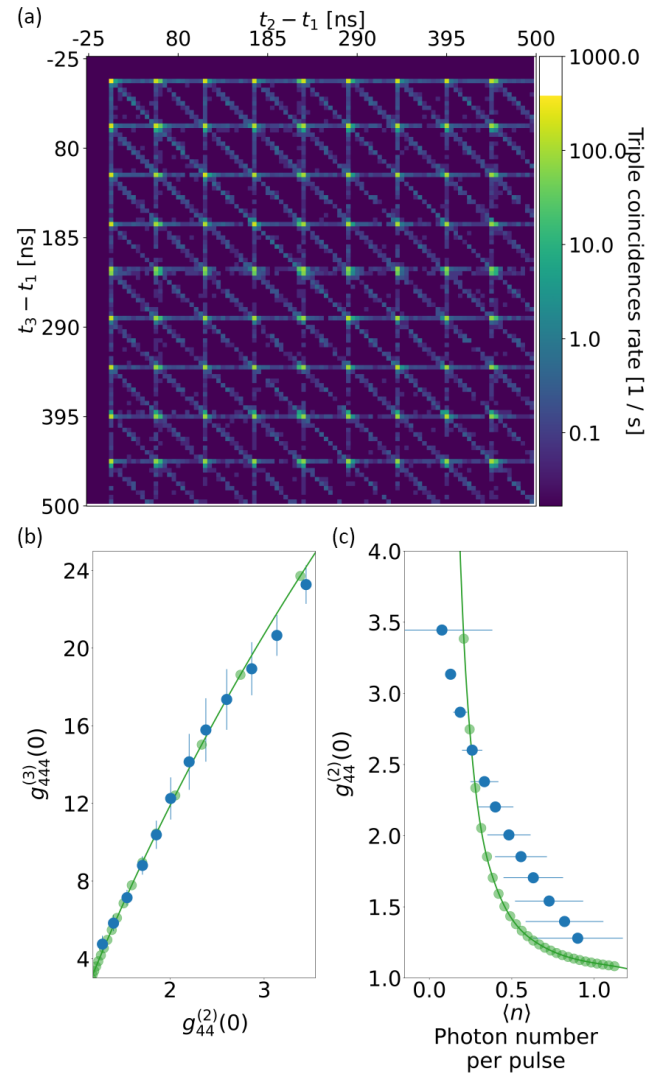


FIG. 3. Measurement of higher-order intensity correlations. Panel (a) shows the raw dataset of the triple coincidence counts for the fourth harmonic using the setup in Fig. 6. The colormap encodes the relative number of triple coincidences per second. The periodic grid is due to the pulsed excitation. The spacing between the peaks coincides with the time difference between the ultrashort laser pulses used for the excitation of the SHHG process. We calculate the TCF by normalizing the raw threefold coincidences with respect to the mean height of the peaks. Panels (b) and (c) show the results from the joint measurement of the SCF and TCF in a single-beam setup for the fourth harmonic depicted in blue. An almost linear relationship between $g_{44}^{(2)}$ and $g_{444}^{(3)}$ is apparent in panel (a). Panel (b) shows the SCF $g_{44}^{(2)}$ over the loss-corrected mean photon number per pulse $\langle n \rangle$. The green curve is the result from an optimization of a parameterized multimode, displaced squeezed thermal state [Eq. (6)] from which the respective observables are calculated. In panels (b) and (c), the theoretical model (green) fits the experimental data points (blue) within the standard deviation. The simulation reproduces well the trend of the measured values. The modeled state used to approximate the experimental results contains squeezing as well as nonzero displacement. The exact parameters are listed in the main text, and the squeezing parameters over the modes are visualized in Fig. 4. This suggests that the SHHG state is a displaced squeezed state. A similar analysis is shown for the dataset of the fifth harmonic in Fig. 7.

harmonics may grow more rapidly than any of the squeezing contributions as the intensity increases, potentially explaining the predominantly coherent behavior observed at higher intensities.

We evaluate the following Cauchy-Schwarz inequality as

$$g_{ii}^{(2)} g_{jj}^{(2)} \geq [g_{ij}^{(2)}]^2 \quad (3)$$

to confirm the presence of nonclassical correlations between the harmonic states [36]. We calculate the R parameter defined as

$$R_{ij} := \frac{[g_{ij}^{(2)}]^2}{g_{ii}^{(2)} g_{jj}^{(2)}} \quad (4)$$

from the experimental data such that a violation of the CSI corresponds to $R > 1$. The results are shown in Fig. 2(d), where we obtain a significant violation for all harmonic bipartite sets from the experimental data [complete dataset displayed in Fig. 2(c)]. The CSI is known to be violated from two-mode squeezed states, as shown by theory [36,37] and experiment [38,39], and proves the presence of nonclassical correlations between the analyzed modes [40]. Indeed, the bipartite reductions correspond most likely to a two-mode squeezed state, exhibiting entanglement between modes. The

tripartite state, as illustrated in Fig. 2(a), can therefore be interpreted as three entangled biseparable states. Genuine tripartite entanglement is out of reach here as it requires entanglement measures beyond the current study [41].

III. ESTIMATION OF THE MODE DISTRIBUTION

Further, insights into the modal structure of the generated squeezed state of light can be obtained by simultaneous measurement of the SCF and TCF [31,32]. SCF and TCF correspond to the second- and third-order correlation functions of the time-averaged intensities of a state. Being closely connected to the photon number distribution of the state, they contain information on its nature and properties. The first-order correlation corresponds to the mean value. The SCF is derived from the second moment, the variance, of the states' photon number distribution and thus measures its spread. Higher-order correlations are sensitive to more refined geometric properties of the distribution like the skewness characterized by the third moment and measurable via the TCF.

The broadband TCF is given by

$$g_{ijk}^{(3)} = \frac{\int d\omega_1 \int d\omega_2 \int d\omega_3 \langle \hat{a}_i^\dagger(\omega_1) \hat{a}_j^\dagger(\omega_2) \hat{a}_k^\dagger(\omega_3) \hat{a}_i(\omega_1) \hat{a}_j(\omega_2) \hat{a}_k(\omega_3) \rangle}{\int d\omega_1 \langle \hat{a}_i^\dagger(\omega_1) \hat{a}_i(\omega_1) \rangle \int d\omega_2 \langle \hat{a}_j^\dagger(\omega_2) \hat{a}_j(\omega_2) \rangle \int d\omega_3 \langle \hat{a}_k^\dagger(\omega_3) \hat{a}_k(\omega_3) \rangle}. \quad (5)$$

As described above, measuring higher-order correlations can give further insights into the underlying photon number probability distribution function. Indeed, for squeezed vacuum states, the analysis of the simultaneous measurement of $g^{(2)}$ and $g^{(3)}$ can be used to estimate the broadband squeezing distribution, the squeezing strength, and the effective Schmidt number or cooperativity parameter K [32,33].

The adapted experimental setup is shown in Fig. 6. As described above, we select a single harmonic order to a single-beam configuration to acquire the two- and threefold coincidence counts and simultaneously acquire the SCF and TCF.

By changing the driving laser intensity, we obtain a set of data with distinctive values for intrabeam correlations $g_{ii}^{(2)}$ and $g_{iii}^{(3)}$. Similar to the analysis carried out in Refs. [32,33], we establish a linear relationship between these two parameters (Figs. 3 and 7), from which we estimate the squeezing strength and distribution. As a result, the generated state can be interpreted as a displaced squeezed state in contrast to the squeezed vacuum state analyzed in Refs. [32,33]. Evaluating higher-order intensity correlation functions for thermal states results in the scaling $g^{(n)} = n!$ [42]. Our experiments show $g_{iii}^{(3)} > 6$, confirming that the observed superbunching does not originate from a thermal state.

Motivated by the interaction Hamiltonian modeling of the SHHG process [25], we base our analysis on a simulated multimode system of a multimodal Gaussian state. In the most general case, this includes multimode displacement,

squeezing, and rotation of the vacuum in case of a pure state. However, for the purpose of modeling of the state, we neglect rotations in a first analysis since they can be switched past squeezing and displacement operators to act on the vacuum first [43]. The actual state generated might be more refined with rotations present among the modes. However, this aspect is beyond the scope of our current explorative analysis. Consequently, we use the fact that the vacuum is an eigenstate of a rotation with eigenvalue 1, which leaves us with only a squeezing and displacement operation. For simplicity, we also omit the inclusion of two-beam squeezing between spectral modes for now such that the squeezing matrix is diagonal and the operator can be decomposed into k single-beam squeezers. In order to take losses into account, we do not restrict the subsequent analysis to pure states (squeezing and displacement) but allow for a general mixed Gaussian state, i.e., a displaced, squeezed thermal state [44].

The density matrix of such a state goes along the following standard formulation:

$$\rho = \bigotimes_{k=1}^d \hat{D}_k(\alpha) \hat{S}_k(\zeta) \rho_{\text{th},k} \hat{S}_k^\dagger(\zeta) \hat{D}_k^\dagger(\alpha), \quad (6)$$

where $\hat{D}_k(\alpha) = \exp(\alpha_k \hat{a}_k^\dagger - \alpha_k^* \hat{a}_k)$ is the displacement operator, $\hat{S}_k(\zeta) = \exp\{(\zeta_k^* (\hat{a}_k^\dagger)^2 - \zeta_k \hat{a}_k^2)/2\}$ the squeezing operator with $\zeta_k = r_k \exp(i\theta_k)$, $\rho_{\text{th},k} = \sum_n P(n_k) |n\rangle \langle n|$ the density matrix of a thermal state with occupation number n_k , and

d is the number of spectral modes constituting the corresponding SHHG mode. With this approach, we explicitly include multimode thermal states, MM squeezed vacuum states, MM displaced squeezed states, and displaced squeezed thermal states. Our parameterization of the latter includes the three former ones (and more), if the parameters are chosen adequately; e.g., $n_{\text{th}} = 0$, $\alpha = 0$, and $r \neq 0$ result in the squeezed vacuum state.

The state in Eq. (6) can be visually depicted, in the single-mode case, in the phase space spanned by the quadratures (amplitude and phase) of the electromagnetic field. The initial thermal state is located at the origin with a variance higher than the vacuum state, resembling its increased noise. The squeezing operator transforms this state into an elongated ellipse, of which major and minor axes as well as the respective orientation depend on the complex squeezing parameter ζ_k . Finally, the displacement operator $\hat{D}_k(\alpha)$ shifts the state away from the origin, increasing the state's mean photon number [45]. We discretize the integrals in Eq. (1):

$$g^{(2)} \approx \frac{\sum_l^d \sum_m^d \langle \hat{a}_l^\dagger \hat{a}_m^\dagger \hat{a}_l \hat{a}_m \rangle}{\sum_l^d \langle \hat{a}_l^\dagger \hat{a}_l \rangle \sum_m^d \langle \hat{a}_m^\dagger \hat{a}_m \rangle} \quad (7)$$

and assume uncorrelated squeezer modes. This simplifies the numerator in the case $l \neq m$ to products of expectation values of photon number operators. The integral expression in Eq. (5) is discretized analogously. Further, we parameterize the squeezing parameter as $r_k = B\lambda_k$ with $\lambda_k = \sqrt{1 - \mu^2} \mu^k$, assuming a two-dimensional Gaussian joint-spectral density distribution (JSD) [32] with an underlying, normalized thermal squeezer distribution characterized by the thermal occupation parameter μ . The assumption of a Gaussian JSD is justified through the use of narrowband spectral filters [46]. The strength of the process B , as well as the displacements, thermal excitations, and squeezing phases, is assumed to be isotropic across all modes k , i.e., $\alpha_k = \alpha$, $n_k = n_{\text{th}}$, and $\theta_k = \theta$. We note that these assumptions are primarily made to simplify the simulations in accordance with Refs. [32,33]. However, they may still serve as a rough estimation of the underlying parameters, as the state generated in SHHG is likely Gaussian in a first approximation. In total, the parameters B , μ , θ , α , and n_{th} are employed to create k states specified by Eq. (6).

Different values for the SCF, TCF, and mean photon number $\langle n \rangle$ are obtained by subsequently including up to red $d = 90$ modes to the simulation [47]. The convergence of the simulation is verified by decreasing the number of modes by 15 and confirming that the same parameters and estimated Schmidt number are obtained. Additionally, the simulated data points, shown as green points in Figs 3(b) and 3(c), exhibit good convergence to the experimental data with an increase in d . The parameters for both datasets on the fourth and fifth harmonics are found by minimizing the root-mean-square deviation (RMSD) between the data and the obtained values from the simulation on the $g^{(2)}$ vs $g^{(3)}$ and the $\langle n \rangle$ vs $g^{(2)}$ dataset. The results for the fourth and fifth harmonics are shown in Figs. 3 and 7. The uncertainties of the parameters are estimated via a perturbation-based sensitivity analysis, where the experimental data are systematically perturbed by $\pm 1\sigma$ experimental uncertainties and the optimization repeated. The

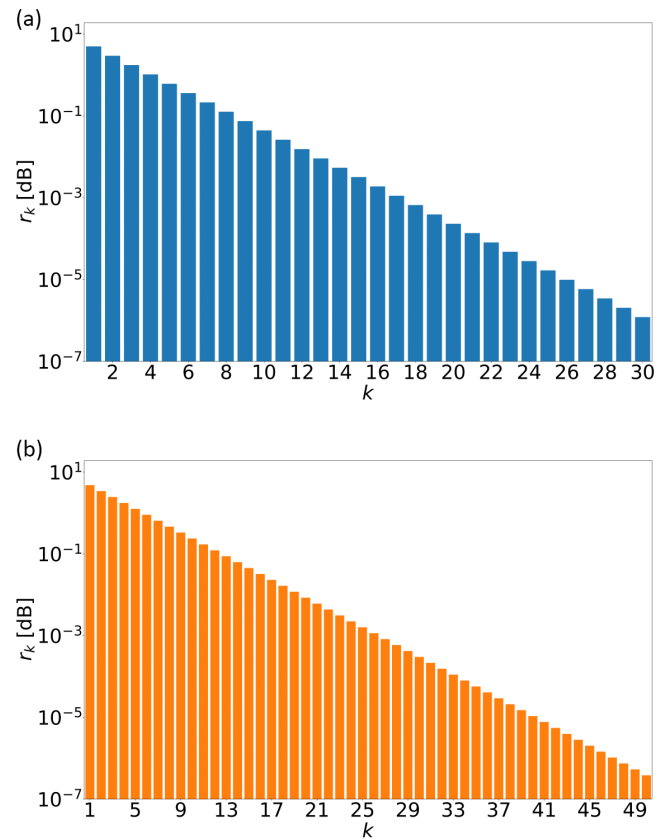


FIG. 4. Reconstructed Mode Distribution. First modes of the estimated squeezer distribution for the fourth (blue) and fifth harmonics (inset in orange), calculated for the simulated displaced squeezed states in Figs. 3 and 7. Multiple modes exhibit non-negligible squeezing with a maximum squeezing level of 5.00 ± 0.22 dB for the fourth harmonic and 4.73 ± 0.23 dB for the fifth harmonic. Additionally, we calculate the effective Schmidt numbers to be $K_{\text{H4, eff}} = 2.08 \pm 0.13$ ($K_{\text{H5, eff}} = 3.18 \pm 0.50$).

reported parameter values represent the mean across all optimizations, with uncertainties given by the standard deviation.

The simulation reproduces well the experimentally observed dependence. For both measured harmonic orders, we find that a state with non-negligible displacement and squeezing is needed to interpret the experimental data, visualized in the retrieved mode distribution (Fig. 4). From the numerical analysis, we estimate the effective Schmidt number $K_{\text{eff}} = 1/\sum_k \lambda_k^4$ [48] as $K_{\text{H4, eff}} = 2.08 \pm 0.13$ and $K_{\text{H5, eff}} = 3.18 \pm 0.50$. This suggests that the fourth and fifth harmonics exhibit moderate multimode character, with the fourth harmonic closer to the single-mode limit while the fifth harmonic shows more significant mode mixing. The amount of squeezing is estimated to be significant, with approximately 5.00 ± 0.22 dB for the fourth harmonic and 4.73 ± 0.23 dB for the fifth harmonic. For both datasets, a small amount of displacement with magnitudes of $|\alpha_{\text{opt},4}| = 0.0609 \pm 0.0026$ and $|\alpha_{\text{opt},5}| = 0.0642 \pm 0.0131$ is needed. These estimations support the interpretation of the SHHG state exhibiting squeezing and displacement effects.

Despite restrictive parameter assumptions, we find the simulations of the model to be in good agreement with

the experimental data and fitting well the interpretation of the observed scaling of the correlation functions with the driving laser intensity due to the broad squeezer distribution, as previously suggested in Ref. [25]. The deviations of the experimental data could originate from the finite data size introducing a statistical uncertainty, which becomes more pronounced for a low number of mean photons [33]. Additionally, the assumption of constant state parameters over the modes and intensity regime is restrictive. For low intensities, the generation of the SHHG orders seems to be dominated by a perturbative generation mechanism, which makes it likely that the generated state differs from the one created in the nonperturbative regime at higher driving laser intensities (Fig. 1). For example, while squeezing effects could be present at all driving laser intensities, additional displacement could be induced only at higher intensities. However, including mode- and intensity-specific parameters would drastically increase the dimensionality of the optimization problem and likely lead to overfitting. We note that it could well be that the measured state is more complex than we assume. Here, we make an educated guess based on theoretical treatment of SHHG and conclude that the creation of a squeezed displaced thermal state as described above is compatible with our dataset.

Still, our interpretation and numerical analysis provides a good explanation. As the SHHG process is highly complex and the theoretical work on understanding the emergence of quantum features has just recently started, we expect that other intrinsic effects such as light-matter entanglement in atoms [49] might contribute to the observed features of the data. Further, as electronic carrier relaxation times in semiconductors are typically longer than the duration between pump pulses, excited carriers may persist and contribute to interband dynamics. Theoretical work has shown that such higher-band excitations and resulting electron correlations can lead to squeezing effects in solid-state HHG [50].

IV. METHODS

Our experimental setups are based on a 200 mW all-fiber pulsed laser system with a central wavelength of 2100 nm and a repetition rate of 18.66 MHz. The ultrashort laser pulses with a duration of 80 fs are linearly polarized and focused on the CdTe[110] crystal with a focal length of 2.5 cm. Before, a half-wave plate and polarizer combination allowed us to adjust the driving laser intensity. After the sample, a slightly closed aperture and an infrared filter separate the driving laser light from the generated high-harmonic orders. An additional broadband polarizer removes the luminescence background of the SHHG signal. The SHHG radiation is collimated, and the harmonic modes 3–5 spatially are separated by means of dichroic mirrors and spectral filters.

For the measurement of the SCF, a beam splitter is placed behind each spectral filter, and the two outputs are focused on single-photon avalanche diodes (SPADs), as shown in Fig. 5. They are connected to a time-to-digital converter (TDC), recording the arrival time of the photons registered on the SPADs. From the time stamps, the mean photon number as well as intra- and interbeam correlation is calculated. To record the threefold coincidence counts, a single harmonic order is selected, and two beam splitters are placed behind the

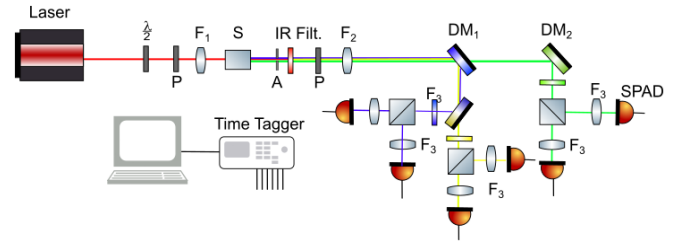


FIG. 5. Experimental setup for measuring the SCF. The ultra-short pulses are directed through a half-wave plate followed by a polarizer to control the driving laser intensity. The near-infrared pulses are focused on the CdTe[110] sample in which the SHHG process is induced. The nonperturbative regime of SHHG is carefully monitored by observation of the intensity crossovers, which are clearly visible for all of the harmonic orders [Fig. 2(b)]. An aperture and an infrared filter are placed behind the sample to filter the pump light. A second polarizer is placed to filter the luminescent background created in the sample. The three harmonic orders are collimated with an achromatic doublet and directed toward the dichroic mirrors to spatially separate different frequencies. Behind the spectral filters, a beam splitter is placed and the photons are measured with single-photon avalanche diodes. The detectors are connected to a time-to-digital converter to record the arrival times of the photons, from which the SCF is calculated.

spectral filter (Fig. 6). Behind the three beam splitter outputs, the SPADs are placed and threefold coincidence is recorded via the TDC. At the same time, the two outputs of the second beam splitter are used to record coincidence counts. In both setups, care is taken to not saturate the binary detectors by having an event rate of less than one photon per pulse. The raw coincidence counts are recorded as histograms over the time delay τ between the detectors. With an electronically created delay, the number of coincidence, or simultaneous events, between the detectors is recorded over the experiment duration. To retrieve the intensity correlation function, the events from two detectors are correlated with each other. For the third-order correlation function, triple coincidences between three detectors need to be recorded. The raw data have a periodic structure due to the pulsed excitation [Fig. 3(a)]. At a time delay $\tau = 0$, coincidence events from SHHG photons created in the same driving laser pulse are recorded. At time

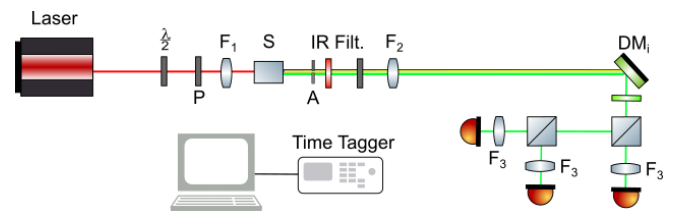


FIG. 6. Experimental setup to measure the SCF and TCF. The setup is similar to Fig. 5 up to the collimation lens. Here, only one high-harmonic order is selected with a dichroic mirror and a spectral filter. Behind the spectral filter, two beam splitters are placed and the radiation at the three outputs of the beam splitter focused onto the single-photon avalanche diodes. These are connected to a time-to-digital converter to record the arrival times of the photon and calculate the SCF as well as TCF.

delays $\tau_n = n \times (1/f_{\text{rep}})$, where f_{rep} is the repetition rate of the laser and $n \in \mathbb{Z} \setminus \{0\}$, true coincidence events arise from photons created from subsequent laser pulses. This leads to a periodic structure, as for time delays $\tau \neq \tau_n$, only accidental counts are recorded resulting in a low signal. The peaks at time delays τ_n are sometimes called satellite peaks. Assuming that coincidence events from photons created in subsequent pump pulses are uncorrelated, yielding $g^{(2)} = 1$, we normalize the raw coincidence counts with respect to the satellite peaks. A peak-finding algorithm is employed to identify the satellite peaks and the height retrieved with a Gaussian fit. For the calculation of the SCF, Gaussian peaks with a coefficient of determination of at least 90% are taken into account. In total, the normalization is then performed over 40 satellite peaks with a percentage coefficient of variation of the RMSD of approximately 15%. The maximum value of a fitted Gaussian to the central peak at $\tau_0 = 0$ is used as the final value of $g^{(2)}(0)$. The error bars are calculated from around ten repetitions of the experiment. By normalizing the raw triple coincidences [Fig. 3(a)] to the height of the off-diagonal peaks, as $g^{(3)} = 1$ for uncorrelated events, we calculate the third-order correlation function $g_{iii}^{(3)}$. Only off-diagonal satellite peaks are accounted for, and the first row and first column are neglected as well. For these, photon events from the same laser pulse could create a threefold coincidence event. A peak-finding algorithm is used to identify the maxima and, again, about 50 satellite peaks with a total coefficient of variation of less than 15%. The value $g^{(3)}(0)$ is read as the maximum value of the central peak in the normalized dataset. The error bars for $g^{(3)} = 1$ are likewise calculated as the standard deviation over the same number of repetitions. We estimate the mean photon number $\langle n \rangle$ by correcting for the detector efficiency and transmission of the optics.

V. NUMERICAL SIMULATION AND ANALYSIS

We simulate the multimode displaced squeezed thermal state from Eq. (6) using the QuTiP Python framework [47]. A set of $k_{\text{max}} = 90$ displaced squeezed thermal states is initialized with an initial guess for the fifth parameter characterizing the state mentioned in the main text [Eq. (6)]. It is ensured that the distribution λ_k , specifying the mode distribution, is normalized. The Fock space dimension is set to $N = 10$, much larger than the photon numbers simulated. For a gradually increasing number of modes k up to k_{max} , the discretized second- and third-order correlation functions are calculated over all modes. Additionally, the mean number of photons of the state is retrieved. We fit these values with a linear function in the $g^{(2)}$ - $g^{(3)}$ space and a fourth-order polynomial in the $\langle n \rangle$ - $g^{(2)}$ space. The experimental data points are initially fitted with the same functions. We calculate the root-mean-square error (RMSE) to the experimental data. As the experimental data for the $\langle n \rangle$ - $g^{(2)}$ datasets follow an inverse power law, the fits diverge for low number of photons. To correctly estimate the error and be less sensitive to enormous differences at low photon numbers, we weight the RMSE in this case by $\langle n \rangle^2$. The total error is calculated by the sum of the RMSE from both fits to the respective experimental dataset. An optimization algorithm is used to find the parameters that approach our dataset the best. The Nelder-Mead algorithm is used to

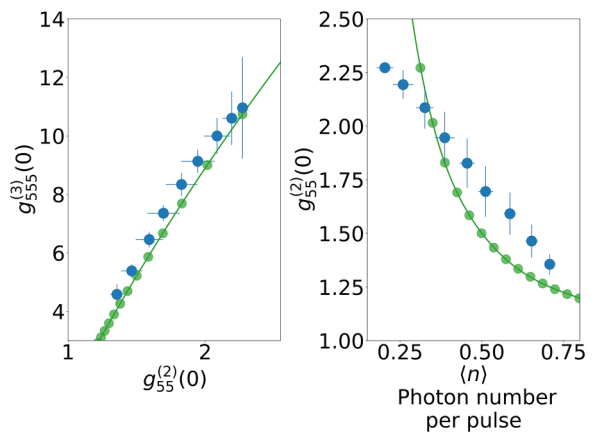


FIG. 7. SCF and TCF for the fifth harmonic. Similar to Fig. 3, the measured SCF over TCF at time delay zero is shown as the blue points in the left graph. The right graph depicts the SCF over the mean photon number per pulse. Again, an optimization algorithm is employed to retrieve the parameters, for which a displaced squeezed thermal state reproduces the observed behavior. The result of the simulation is shown as the green curves. The trend is reproduced well by the numerical comparison.

minimize the loss function described above. We optimize the parameters for both datasets simultaneously. A range of initial guesses is tested, which always converge to the optimal values stated below. Convergence is ensured by running the simulation for a reduced number of modes.

To estimate the uncertainties in our fitted parameters, we employ a perturbation-based sensitivity analysis approach. We choose this approach, as the derivative-free Nelder-Mead optimization and the nonlinearity of our loss function do not allow for a Hessian matrix estimation. Further, a Monte Carlo simulation is too computationally expensive, as it would require thousands of optimization repetitions. Our method directly assesses how sensitive the parameters are to the uncertainties in the input data ($g^{(2)}$, $g^{(3)}$, and photon number measurements). In a baseline optimization, we use the original data as input to establish a nominal, best-fit parameter set as described above. Subsequently, additional optimizations are conducted using input datasets where each experimental measurement is systematically perturbed by its estimated $\pm 1\sigma$ experimental uncertainty. This includes scenarios where all measurements are perturbed concurrently by $\pm 1\sigma$ and where individual input datasets are perturbed independently, while others remain at their baseline values. This generates a total of nine distinct datasets. For each perturbation, the nominal parameter set is used as the initial guess. The uncertainty for each parameter is quantified by the standard deviation over the optimal parameters across all perturbed optimizations, and the final parameter estimates are taken as the mean values across all optimization runs (baseline plus perturbed cases). In the case of the fourth harmonic, the optimal parameters are found to be $B_{\text{opt},4} = 0.3094 \pm 0.0270$, $\mu_{\text{opt},4} = 0.5172 \pm 0.0340$, $\alpha_{\text{opt},4} = 0.0735 \pm 0.0040$, $n_{\text{th,opt},4} = 0.001 \pm 0.0001$, and $\theta_{\text{opt},4} = 0$. For the fifth harmonic, $B_{\text{opt},5} = 0.3425 \pm 0.0444$, $\mu_{\text{opt},5} = 0.7164 \pm 0.0490$, $\alpha_{\text{opt},5} = 0.0642 \pm 0.0131$, $n_{\text{th,opt},5} = 0.002 \pm 0.0002$, and $\theta_{\text{opt},4} = 0$. For both measured harmonic orders, we find that a state with non-negligible

displacement and squeezing is needed to interpret the experimental data, visualized in the retrieved mode distribution (Fig. 4). From the numerical analysis, we estimate the effective Schmidt number $K_{\text{eff}} = 1/\sum_k \lambda_k^4$ [48] as $K_{\text{H4, eff}} = 2.08 \pm 0.13$ and $K_{\text{H5, eff}} = 3.18 \pm 0.50$. This suggests that the fourth and fifth harmonics exhibit moderate multimode character, with the fourth harmonic closer to the single-mode limit while the fifth harmonic shows more significant mode mixing. The amount of squeezing is estimated to be significant, with approximately 5.00 ± 0.22 dB for the fourth harmonic and 4.73 ± 0.23 dB for the fifth harmonic.

VI. DISCUSSION

In summary, our work explores and develops a strategy based on laser-driven electron recollision in the process of high-harmonic generation in semiconductors to produce multipartite entangled states of light. Here, we evidence nonclassicality of three harmonic biseparable states by measuring the violation of a combination of Cauchy-Schwarz-type inequalities, a required step toward certifying genuine tripartite entanglement. As a more general prospect, the quantum nature of high-harmonic generation can potentially generate an N -partite entangled frequency comb, a central resource in quantum technologies. Here, the harmonic mode structure is recovered through a Schmidt decomposition to estimate the effective Schmidt number and squeezing strength of the spectral modes of individual harmonics. Thorough analysis of second- and third-order correlation functions shows that harmonics are emitted in displaced squeezed thermal states, with non-negligible contributions from additional modes. We observe superbunching ($g^{(2)} > 2$) of the high harmonics at low laser intensity that tends to reach the coherent states ($g^{(2)} = 1$) for higher intensities. In contrast to other approaches investigating quantum HHG, which rely on inducing nonclassical features by conditional measurements or mixing the driving laser with a nonclassical field, we show that the SHHG can be nonclassical without these additional nonclassical resources. Compared to other nonclassical sources of light, SHHG is unique as it potentially creates nonclassical light in a frequency comb spanning multiple THz of spectral bandwidth. Although similar techniques exist based on parametric processes, the properties and potential tunability of SHHG states are unique. The high-harmonic generation process can be finely controlled to manipulate multiple complex dimensionalities. The spatial profile can be shaped to carry nontrivial topology via the orbital angular momentum. The spectral

phase can be addressed by its dependence on the carrier-envelope phase of the driving laser pulses. These techniques support SHHG as a highly promising candidate for applications in the broad field of quantum-state engineering, gaining possible quantum advantage. However, complete quantum-state tomography of SHHG is essential to fully determine the quantum-state properties. Still, the challenge is to perform it simultaneously over the SHHG frequency comb for each harmonic order. Novel quantum-state tomography techniques might be suitable to solve this experimental problem. Also, the exploration of higher nonperturbative generation regimes for, e.g., higher-harmonic orders or higher driving laser intensities seems like a sensible step. The measurement of higher-order moments using multiplexing by, e.g., beam splitter arrays and photon number resolving detection are suitable techniques regarding this and can be applied to analyze complementary nonclassical criteria.

In conclusion, high-harmonic generation represents an innovative resource of nonclassical light, interesting for fundamental research and potentially for quantum information science and emerging technologies, such as quantum metrology, computing, and cryptography.

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D.T. conceived the study and performed the experiments. D.T., V.C., and P.H. analyzed the data. D.T., P.H., and R.S. conducted the theoretical analysis. D.T. wrote the simulation and provided the plots. D.T., H.M., P.H., and R.S. wrote the manuscript with input from all authors. D.T. drafted the original answer to the referees. H.M. initiated the quantum HHG research program and acquired the funding for the experiments. All authors discussed the results.

There are no competing interests to declare.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available because of legal restrictions preventing unrestricted public distribution. The data are available from the authors upon reasonable request.

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