

Sensing quantum fluctuations via forbidden harmonics in quantum materials

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High-harmonic generation in solids provides a powerful window into light-driven electron dynamics on attosecond timescales, yet the role of quantum fluctuations of the driving field remains largely unexplored. Here, we show that the quantum state of light offers a nonclassical degree of freedom for controlling strong-field dynamics in quantum materials. Using graphene driven by bicircular quantum light composed of counterrotating ω and 2ω components, we theoretically predict that squeezing-induced fluctuations break the dynamical symmetry of the classical bicircular field and activate otherwise forbidden harmonics. By analyzing the helicity-resolved harmonic spectra, we uncover robust polarization signatures uniquely dictated by the squeezed mode's profile, including a relative-phase-controlled helicity switching in the lowest orders. Crucially, second-order intensity correlations reveal that these forbidden harmonics exhibit strongly enhanced, super-Poissonian photon-number fluctuations, confirming that they inherit and directly transduce the nonclassical statistics of the squeezed driving vacuum. Our findings establish forbidden harmonics as high-fidelity optical probes of the quantum state of light and provide additional opportunities at the intersection of quantum optics, strong-field physics, and lightwave engineering in quantum materials.

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Over the past decades, strong-field-driven high-harmonic generation (HHG) has emerged as a powerful platform for generating attosecond pulses and probing electronic motion on its natural timescale [1–4]. The extension of HHG from gases to solids has transformed the field by enabling the investigation of both equilibrium electronic structure and ultrafast nonequilibrium dynamics in condensed phase [5–22]. Beyond its fundamental significance, solid-state HHG offers a promising route toward compact sources of coherent extreme-ultraviolet radiation and attosecond pulses, while simultaneously providing a unique spectroscopic window into light-driven quantum phenomena in solids [23–30]. Among quantum materials, graphene has emerged as an interesting platform for exploring nonperturbative nonlinear optical interactions. Its gapless Dirac spectrum, exceptional carrier mobility, and the intricate interplay between interband excitation and intraband acceleration give rise to a remarkably rich nonlinear optical response [31–48]. Previous studies have shown that higher-order harmonic emission in graphene can be engineered by tailoring the waveform, polarization, and other properties of the driving field [49–53]. This exquisite sensitivity to the driving radiation makes graphene a suitable platform for investigating symmetry-controlled subcycle

electron dynamics and developing new paradigms for ultrafast lightwave engineering.

Until recently, HHG has been understood almost exclusively within a classical framework of the driving field, which successfully captures its defining features, including harmonic plateaus, cutoff energies, and polarization selection rules. Consequently, the quantum nature of the driving field has remained largely unexplored. Recent advances in high-photon-number nonclassical light sources, however, have opened an entirely new frontier in strong-field physics, enabling access to regimes where quantum fluctuations of the driving field can actively shape intense laser-driven HHG [54–68]. Driving solid HHG with nonclassical light reveals distinct quantum-optical phenomena that bridge strong-field physics and quantum optics. Utilizing bright squeezed vacuum and squeezed coherent states in materials such as ZnO [69,70], GaAs [71], Mg:LN, and Si substantially boosts harmonic efficiency, extends spectral cutoffs, and enables precise yield control via photon statistics [72]. In turn, strong-field nonlinearities in these solids transduce the quantum properties of the driving field into the emitted radiation, producing squeezed light near exciton resonances [73] and harmonic sidebands characterized by super-Poissonian photon bunching [74]. These developments raise a fundamental question: Can quantum fluctuations themselves serve as a new control knob for strong-field electron dynamics in solids? This question is particularly compelling in graphene, whose nonperturbative optical response is governed by Dirac-fermion symmetry and exhibits extreme sensitivity to driving-field polarization. Addressing these questions would not only uncover a relatively unexplored regime of quantum strong-field physics but also

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enable the direct extraction of various properties of the quantum state of light from the harmonic spectra.

This Letter discusses HHG in graphene driven by bicircular quantum light. In the classical limit, bicircular driving fields composed of counterrotating circularly polarized components at frequencies ω and 2ω possess threefold rotational symmetry. This dynamical symmetry imposes selection rules, allowing only the $(3n+1)$ and $(3n+2)$ harmonics while strictly forbidding the $3n$ harmonics. Additionally, the helicities of the emitted $(3n+1)$ and $(3n+2)$ harmonics are inherited from the ω and 2ω driving fields, respectively [75,76]. In the following, we demonstrate how squeezing-driven quantum fluctuations fundamentally alter this picture. Although the mean driving field retains the classical symmetry, the fluctuations break the underlying dynamical symmetry, and activating classically forbidden $3n$ harmonics. This fluctuation-induced symmetry breaking establishes a direct link between the quantum state of the driving field and the nonlinear optical response of graphene. By analyzing helicity-resolved harmonic spectra together with the second-order intensity correlation function, we show that the forbidden harmonics carry distinct polarization signatures and nonclassical photon statistics that are absent in the classical regime. These observables provide experimentally accessible fingerprints of quantum-fluctuation-driven HHG and establish properties of the forbidden-harmonic emission as a direct optical probe of the quantum state of the driving field. The generation of forbidden harmonics has also been reported in quantum-light-driven HHG from gases [77,78].

We consider the quantum state of the driving field is a displaced squeezed state, a coherent bicircular counterrotating $\omega - 2\omega$ field with one component prepared in a squeezed state along a chosen polarization direction $\mu \in \{\parallel, \perp\}$ as [64]

$$|\Phi(t_0)\rangle = \mathcal{D}_{\omega,R}(\alpha_\omega\sqrt{2}) \otimes [\mathcal{D}_{2\omega,L}(\alpha_{2\omega}\sqrt{2})\mathcal{S}_{2\omega,\mu}(\xi)]|\bar{0}\rangle, \quad (1)$$

where $\mathcal{D}$ and $\mathcal{S}$ are the displacement and squeezing operators, $\xi = re^{i\theta}$ is the squeezing parameter, and $|\alpha_\omega| = |\alpha_{2\omega}|$ ensures the retrieval of coherent limit at $r = 0$. The vector potential of the coherent bicircular laser configuration, consisting of two circularly polarized components with identical amplitudes, is

$$\mathbf{A}_{cl}(t) = g(t)A_0[\{\cos(\omega t) + \cos(2\omega t)\}\hat{\mathbf{e}}_x + \{\sin(\omega t) - \sin(2\omega t)\}\hat{\mathbf{e}}_y], \quad (2)$$

where $g(t)$ represents a $\sin^2$ pulse envelope and A_0 is the peak amplitude of the vector potential. Here, the fundamental ω component carries right-circular polarization (RCP) ($\sigma = +1$), while the second-harmonic 2ω component possesses left-circular polarization (LCP) ($\sigma = -1$), establishing the requisite transient threefold rotational symmetry. To obtain the high-harmonic spectra for a coherent driving field, we numerically solve the density-matrix equations of motion within the Houston-basis framework using a fourth-order Runge-Kutta algorithm with a time step of 20 attoseconds (as), following the approach of Refs. [79,80]. The fundamental driving field has a wavelength of 3200 nm of an eight-cycle pulse duration with a peak electric field strength of 8.7 MV/cm. The electronic structure of monolayer graphene is described by a nearest-neighbor tight-binding Hamiltonian, and the Brillouin zone is sampled on a uniform 200×200 $\mathbf{k}$ -point grid.

The total harmonic spectrum is expressed in terms of the Husimi-weighted average over coherent field configurations as [56]

$$\mathcal{I}_q(\omega) = \int d^2\alpha Q(\alpha)|\chi_{q,\mu}(\alpha)|^2. \quad (3)$$

Here, $\chi_{q,\mu}(\alpha)$ is the spectral amplitude of the q th harmonic along μ polarization of a coherent field with amplitude α , and $Q(\alpha)$ is the Husimi Q -function of the squeezed field component, which is a two-dimensional Gaussian with quadrature widths

$$\sigma_a = \sigma_0\sqrt{\frac{1+e^{-2r}}{2}}, \quad \text{and} \quad \sigma_p = \sigma_0\sqrt{\frac{1+e^{2r}}{2}}, \quad (4)$$

where $\sigma_a < \sigma_0 < \sigma_p$ reflects the asymmetric squeezing of the amplitude and phase quadratures, and σ_0 sets the overall fluctuation scale. The applicability of ensemble-based approaches to quantum light-driven HHG remains an active topic of discussion, driving the development of alternative theoretical frameworks [81–84]. Throughout this Letter, we use $\sigma_0 = 0.05$ and $r = 3.0$ unless stated otherwise.

The total vector potential for squeezing applied to the 2ω component along the parallel direction reads as

$$\mathbf{A}(t) = g(t)A_0[\{\cos(\omega t) + (1+\mathbf{u})\cos(2\omega t) + \mathbf{v}\sin(2\omega t)\}\hat{\mathbf{e}}_x + \{\sin(\omega t) - \sin(2\omega t)\}\hat{\mathbf{e}}_y]. \quad (5)$$

The mean field reproduces the classical bicircular driving field, with $\langle\mathbf{u}\rangle = \langle\mathbf{v}\rangle = \mathbf{0}$. In contrast, the field variance, $(\Delta E)^2(t) \propto [\cosh(2r) - \sinh(2r)\cos(2\omega t + \theta)]$, does not preserve the transient threefold symmetry of the coherent field. To quantify the quantum properties of the generated harmonics, we evaluate the equal-time second-order intensity correlation function [62],

$$g_{q,\mu}^{(2)}(0) = \frac{\langle|\chi_{q,\mu}|^4\rangle_Q}{\langle|\chi_{q,\mu}|^2\rangle_Q^2}. \quad (6)$$

Here, $\langle\cdots\rangle_Q$ stands for the ensemble average over quantum fluctuations. This correlation function distinguishes coherent ($g^{(2)} = 1$), and squeezed-vacuum ($g^{(2)} = 3$) photon statistics, thereby providing a direct measure of the nonclassical character of the emitted harmonics.

Let us explore the impact of squeezing-induced quantum fluctuations on HHG driven by bicircular quantum light. As illustrated in Fig. 1(a), the introduction of squeezing leads to the striking emission of the classically forbidden $3n$ harmonics, an explicit fingerprint of quantum fluctuations, which dynamically break the threefold symmetry of the classical field to unlock harmonic pathways completely inaccessible to classical light. Remarkably, this quantum-mediated symmetry breaking occurs even while the average field macroscopically retains the pristine symmetry of the classical bicircular field. To investigate whether the emergence of these $3n$ harmonics depends on the specific geometry of the quantum state, we evaluate both amplitude and phase squeezing applied along the parallel ($\parallel$) and perpendicular ($\perp$) directions of the 2ω field component [Fig. 1(b)]. Despite the distinct phase-space fluctuation profiles characteristic of these four configurations (see the top panel of Fig. 1), the resulting harmonic spectra remain qualitatively

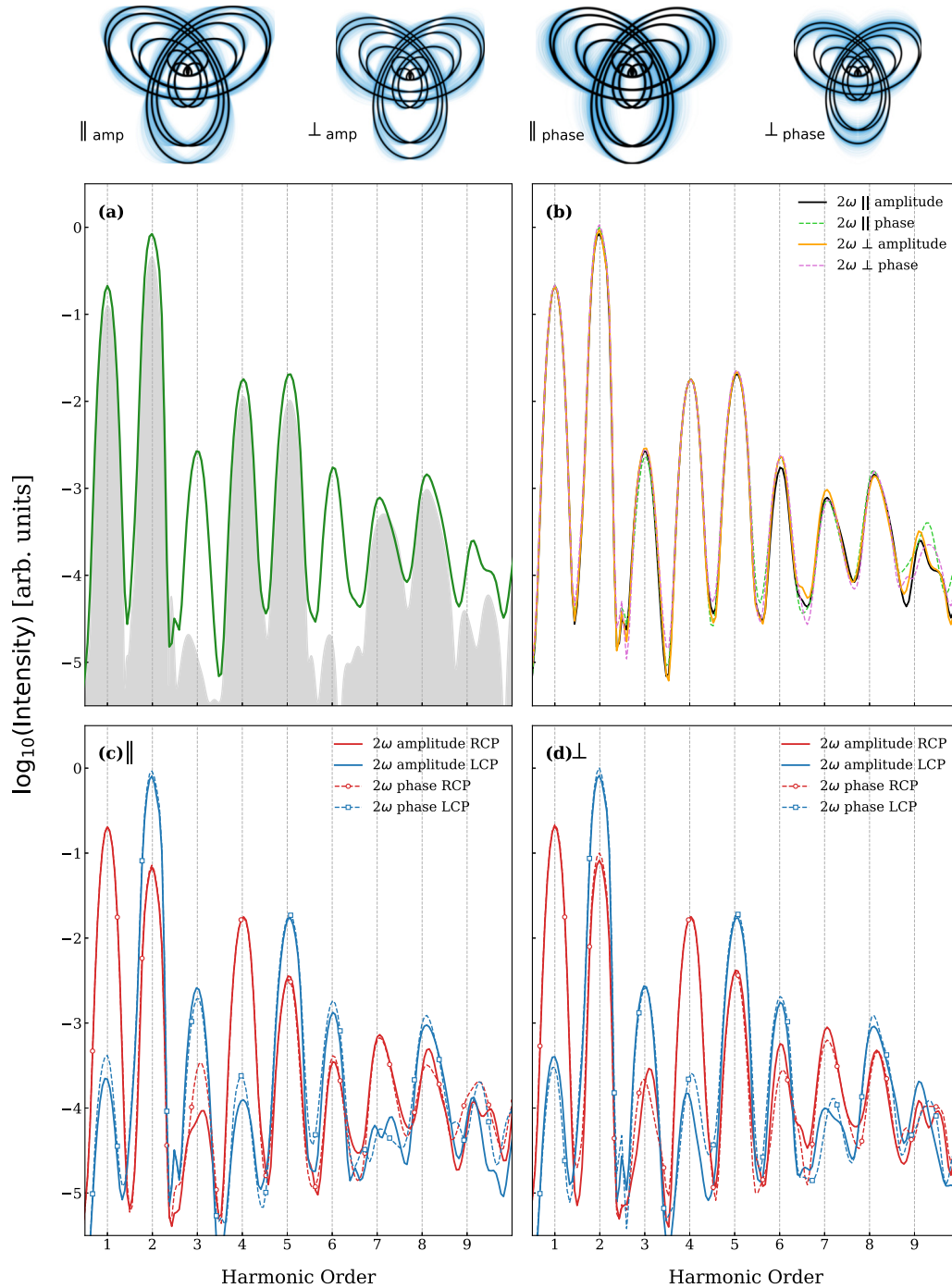


FIG. 1. Quantum-fluctuation-induced high-harmonic generation in graphene. (a) HHG spectrum of graphene driven by $\omega - 2\omega$ bicircular quantum field with amplitude squeezing applied parallel to the 2ω component. The corresponding classical spectrum is shown in gray for comparison. (b) HHG spectra for amplitude squeezing and phase squeezing applied parallel (||) and perpendicular (⊥) to the 2ω component. Helicity-resolved HHG spectra mapping the distinct signatures of (c) || and (d) ⊥ squeezing configurations. Top panels: Blue-shaded Lissajous profiles represent the quantum light for amplitude squeezing and phase squeezing applied || or ⊥ to the 2ω component; the corresponding classical light is shown by solid lines.

similar. In all instances, the $3n$ harmonics are generated with comparable yields. This uniformity demonstrates that the appearance of the $3n$ harmonics is a highly robust consequence of squeezing-driven symmetry breaking. Consequently, the macroscopically observed response is governed primarily by

the presence of quantum fluctuations rather than the specific quadrature or polarization axis along which the squeezing is initialized.

Beyond the appearance of the forbidden $3n$ harmonics, deeper insight into the quantum-light modulation of HHG is

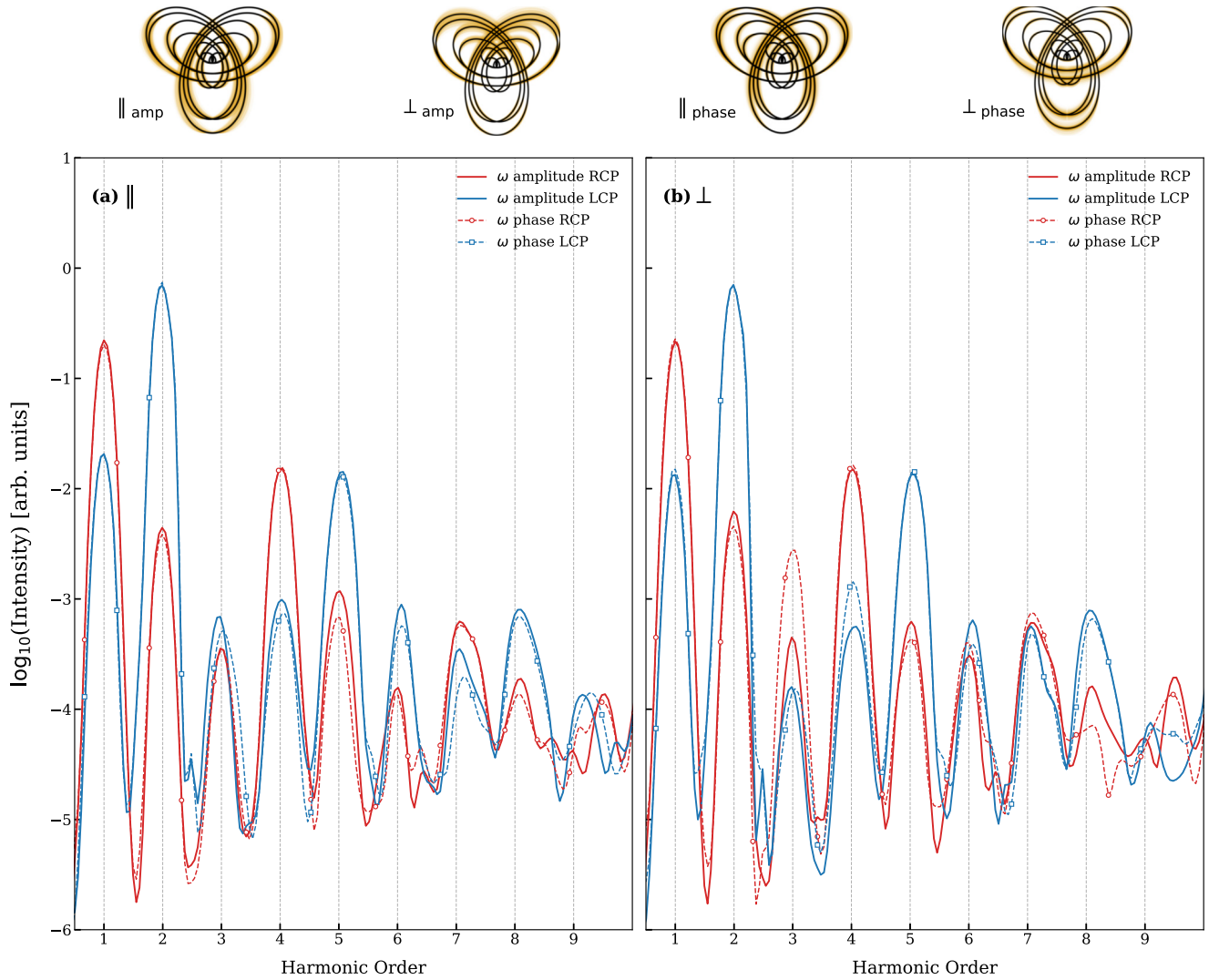


FIG. 2. (a), (b) Helicity-resolved HHG spectra analogous to Figs. 1(c) and 1(d), but with squeezing applied to the ω component of the bicircular quantum light. Top panels: Same as in Fig. 1, but with squeezing applied to the ω component of the bicircular field.

revealed by analyzing the helicity of the emitted harmonics [Figs. 1(c) and 1(d)]. A classical bicircular field enforces angular momentum conservation, dictating a highly structured, alternating helicity profile: The $(3n + 1)$ harmonics inherit the helicity of the fundamental ω field, while the $(3n + 2)$ harmonics mirror the counterrotating 2ω field [85]. While this alternating pattern remains largely preserved under quantum-light driving, a distinct departure emerges: Each allowed $(3n + 1)$ and $(3n + 2)$ harmonic acquires a finite contribution from the opposite helicity component. This polarization mixing indicates that quantum fluctuations reshape the polarization landscape of the emitted radiation.

The most compelling quantum signature manifests within the classically forbidden $3n$ channels, which display a highly asymmetric polarization characterized by a dominant LCP component alongside a weaker RCP contribution. This behavior stems from the fact that the forbidden harmonics directly inherit the polarization attributes of the squeezed field as fluctuations are introduced into the 2ω mode. The persistence of

this asymmetry across all squeezing configurations proves that the polarization properties of these forbidden $3n$ harmonics are determined exclusively by the fluctuating mode's helicity rather than the orientation of the squeezing direction. Consequently, these forbidden harmonics do not merely reveal fluctuation-induced symmetry alteration; they constitute a direct diagnostic of the quantum state encoded in the driving field.

So far, we have examined the influence of squeezing to the 2ω mode on HHG. We now consider the case in which squeezing is applied to the ω field, which uncovers a distinct regime of control over HHG (Fig. 2). Although the alternating helicity structure of the allowed $(3n + 1)$ and $(3n + 2)$ harmonics remains robust, the forbidden $3n$ orders display a markedly different polarization response. For the low-order forbidden harmonics, parallel squeezing favors the LCP component, whereas perpendicular squeezing reverses the dominant helicity. This helicity switching disappears at higher orders, where the forbidden harmonics remain predominantly LCP

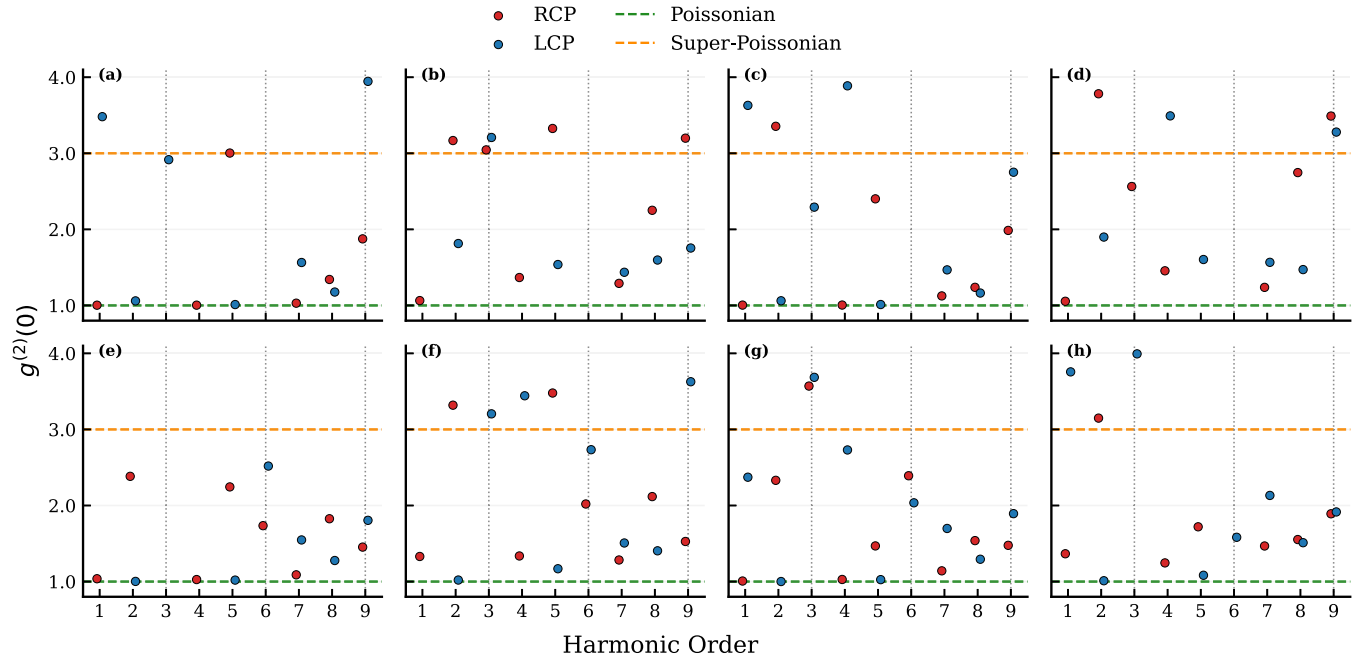


FIG. 3. Helicity-resolved second-order correlation function for the emitted harmonics. (a)–(d) and (e)–(h) correspond to squeezing applied to the 2ω and ω fields, respectively. The first and third columns show the effects of $\parallel$ and $\perp$ amplitude squeezing applied to the field component, whereas the second and fourth columns correspond to $\parallel$ and $\perp$ phase squeezing. Vertical dashed lines indicate the forbidden $3n$ harmonics, while horizontal dashed lines mark $g^{(2)} = 1$ and 3 .

regardless of the squeezing orientation. The helicity switching observed in the lowest-order forbidden harmonics reveals that the emitted polarization is controlled not only by the squeezed mode but also by the relative phase of the quantum fluctuation channels. Rotating the squeezing direction changes the phase relation between the RCP and LCP fluctuations, thereby altering the dominant photon-absorption pathways. At higher-harmonic orders, helicity-zero fluctuation channels dominate the emission, making the forbidden harmonics largely insensitive to the squeezing orientation.

Although the harmonic spectra establish the emergence and polarization properties of the forbidden harmonics, they do not reveal whether the emitted radiation retains fingerprints of the quantum state of the driving field. To address this, we analyze the second-order intensity correlation function, $g^{(2)}(0)$, across all squeezing configurations (Fig. 3). The dominant helicity channels of the classically allowed harmonics exhibit $g^{(2)} \approx 1$, indicating nearly coherent emission, whereas their weaker opposite-helicity components show $g^{(2)} > 1$, revealing their fluctuation-induced origin. More strikingly, the forbidden $3n$ harmonics exhibit strongly enhanced correlations, with $g^{(2)} \geq 3$, indicating pronounced super-Poissonian photon-number fluctuations.

While the exact value of $g^{(2)}(0)$ varies with the specific squeezing configuration and harmonic order, reflecting the distinct quantum fluctuation pathways that contribute to the emission process, Nevertheless, the overall trend remains universal across all configurations. The classically forbidden harmonics systematically exhibit substantially stronger photon-number correlations than their allowed counterparts. This behavior confirms that the forbidden harmonics originate entirely from fluctuation-induced symmetry alteration and

directly inherit the nonclassical statistics of the squeezed light. These enhanced correlations provide unambiguous evidence that the forbidden harmonics act as a high-fidelity quantum transducer, mapping the nonclassical properties of the driving light onto the emitted radiation at higher energy.

Having established the quantum origin of the forbidden $3n$ harmonics, we next examine how their emission can be controlled by the strength of the quantum fluctuations in the driving field. Figure 4(a) shows that, for small fluctuation amplitudes σ_0 , the harmonic spectrum closely resembles its classical counterpart, with the forbidden harmonics remaining strongly suppressed. The intensity of the $3n$ harmonics grows systematically as σ_0 increases, while the allowed harmonics remain unaffected. This behavior demonstrates that σ_0 directly controls the magnitude of the fluctuation-induced symmetry breaking, and, consequently, the efficiency of the forbidden emission channels.

Complementarily, as the squeezing parameter approaches zero ($r \rightarrow 0$), the driving field reduces to a coherent state, completely extinguishing the $3n$ harmonics and recovering the classical selection rules [Fig. 4(b)]. Increasing r rapidly amplifies the $3n$ harmonic yield, providing further evidence that these channels are fed explicitly by nonclassical fluctuations of the driving field as the larger values of r correspond to stronger quantum fluctuations. Taken together, Figs. 4(a) and 4(b) establish σ_0 and r constitute experimentally tunable control knobs for systematically tailoring fluctuation-induced high-harmonic emission.

In conclusion, our results establish quantum fluctuations as an alternative control resource for dynamical symmetry and strong-field electron dynamics in solids. Using

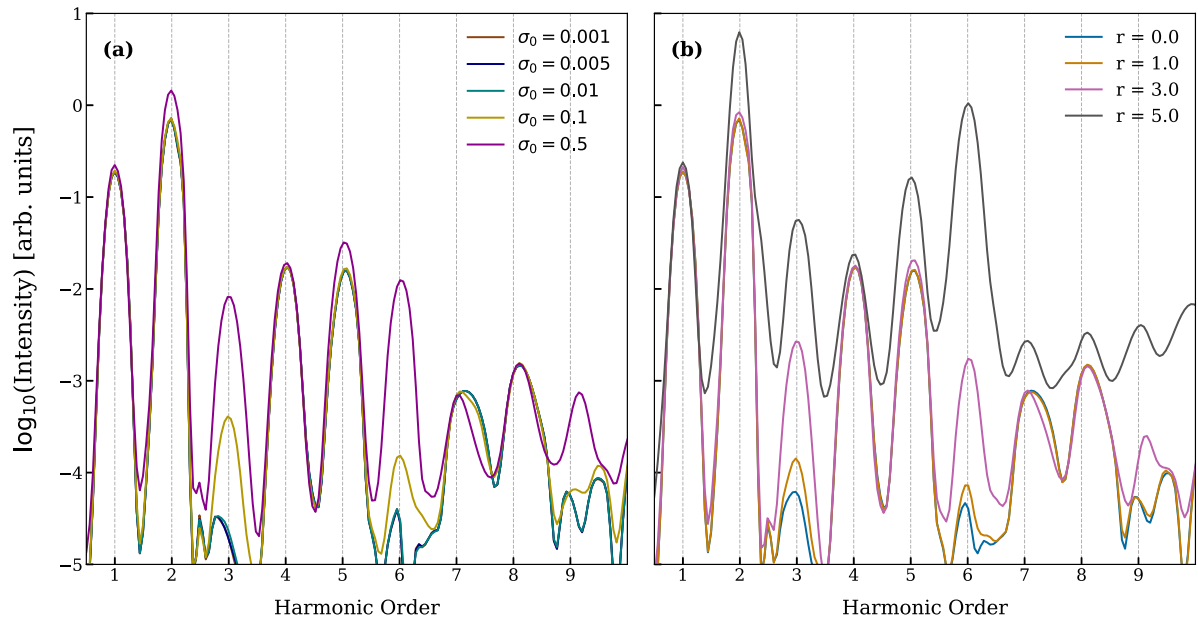


FIG. 4. Harmonic spectra sensitivity to quantum squeezing. HHG spectra as a function of (a) fluctuation strength σ_0 (at fixed $r = 1.0$), and (b) squeezing parameter r (at fixed $\sigma_0 = 0.05$). In both cases, amplitude-quadrature squeezing applied parallel to the 2ω component of the bicircular field.

monolayer graphene driven by bicircular quantum light, we demonstrate that squeezing-induced fluctuations break the dynamical symmetry of the classical field—even when the mean field retains this symmetry—thereby activating otherwise forbidden harmonics. These induced channels act as a high-fidelity quantum transducer; they exhibit distinct, phase-controlled polarization profiles and strongly enhanced, super-Poissonian statistics that inherit the nonclassical signatures of the driving field, revealing direct and measurable fingerprints of the nonclassical driving field. The robustness of these signatures across different squeezing configurations and driving modes identifies fluctuation-induced symmetry breaking as a general mechanism for engineering nonlinear optical responses with quantum light. More broadly, our theoretical results establish forbidden-harmonic emission in solids as an optical quantum sensor capable of *in situ* quantum-state

tomography and subcycle electron wave-front engineering, opening additional opportunities for quantum-light spectroscopy, and attosecond sensing of statistical fluctuations of squeezed light, while positioning solid-state HHG as a powerful interface between quantum optics and attosecond science.

Note added. Recently, we became aware of a work [86], demonstrating that bicircular quantum $\omega - 2\omega$ light can enable the generation of forbidden harmonics in 2D materials.

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Data availability. The data that support the findings of this article are available upon reasonable request.

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