

Signatures of quantum chaos in phonon-polariton billiards

Dmitri Basov

db3056@columbia.edu

Columbia University <https://orcid.org/0000-0001-9785-5387>

Yinan Dong

Columbia University <https://orcid.org/0000-0002-5996-642X>

Felix Liu

Columbia University

Ekrem Demirboga

Department of Mathematics and Joint Quantum Institute, University of Maryland

<https://orcid.org/0000-0002-1459-6963>

Andrey Grankin

University of Maryland

Dihao Sun

Columbia University

Yuchen Lin

Columbia University

Lukas Wehmeier

Brookhaven National Laboratory <https://orcid.org/0000-0001-8416-953X>

Song Liu

Columbia University

James Edgar

Kansas State University <https://orcid.org/0000-0003-0918-5964>

Cory Dean

Columbia University <https://orcid.org/0000-0003-2967-5960>

Michael Fogler

University of California, San Diego <https://orcid.org/0000-0001-8239-7221>

Victor Galitski

University of Maryland

Keywords: polariton, quantum chaos, nanoimaging

Posted Date: September 15th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10721342/v1>

License: © ⓘ This work is licensed under a Creative Commons Attribution 4.0 International License.

[Read Full License](#)

Additional Declarations: There is **NO** Competing Interest.

Signatures of quantum chaos in phonon-polariton billiards

Yinan Dong^{1,2}, Felix Liu¹, Ekrem Demirboga^{3,4}, Andrey Grankin³, Dihao Sun¹, Yuchen Lin¹, Lukas Wehmeier⁵, Song Liu⁶, James H. Edgar⁶, Cory R. Dean¹, Michael M. Fogler⁷, Victor M. Galitski³, Dmitri N. Basov^{1*}

*Corresponding author. Email db3056@columbia.edu

1. Department of Physics, Columbia University, New York, NY 10027, USA
2. Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY, 10027, USA
3. Joint Quantum Institute, Department of Physics, University of Maryland, College Park, MD 20742, USA
4. Department of Mathematics, University of Maryland, College Park, MD 20742, USA
5. National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, NY 11973, USA
6. Tim Taylor Department of Chemical Engineering, Kansas State University, Manhattan, KS 66506, USA
7. Department of Physics, University of California San Diego, La Jolla, CA 92093, USA

Abstract

We use scanning near-field optical microscopy to study hyperbolic phonon polaritons in hexagonal boron nitride (hBN) cavities with regular and chaotic geometries. In Sinai billiards, we observe quasi-periodic edge modes, quantum scarring, and irregular patterns that evolve as the probe-sample coupling is varied. The random-wave features coexist with non-chaotic one-dimensional boundary modes arising from non-trivial polariton reflection at the billiard edge. In successive iterations of Koch snowflake billiards, we find polaritonic interference patterns of progressively finer structure. The Fourier transforms of polaritonic images reveal densely populated circles in the reciprocal space, in agreement with Berry's random-wave conjecture. We develop a numerical framework based on the two-dimensional Helmholtz equation with boundary conditions that encode angle-dependent reflection phase shifts. Theoretical analysis of dissipative Green's function qualitatively reproduces the observations. The calculated level statistics exhibit a crossover from Poisson-like behavior in integrable billiards to Wigner-Dyson-like behavior in chaotic ones. These results establish van der Waals cavities as a rich platform for studying generalized chaotic dynamics of light-matter hybrid polaritons.

Keywords: polariton, quantum chaos, nanoimaging

Introduction

Quantum mechanics is a wave theory, but generic wave problems are nontrivial. Outside a small set of separable models, the specific spectrum and eigenfunctions of the Helmholtz or Schrödinger operator are controlled by geometry, making resonant cavities a canonical setting for studying wave dynamics¹. While the spectra and the eigenfunctions are specific to each cavity, they typically fall into one of two categories. The rare integrable cases exhibit Poisson-like spectral statistics and regular eigenfunctions², often displaying clear symmetries or patterns. More generally, cavities display irregular eigenfunctions³ and the eigenvalue statistics of the random-matrix theory^{4,5,6}. This dichotomy has its counterpart in classical dynamics of a particle confined in a cavity. In particular, the non-integrable quantum cases correspond to classically chaotic billiards^{7,8,9,10}. The core open problem in experimental studies of quantum chaos is to

definitively measure its universal signatures in real physical systems that are subject to external noise, environmental decoherence, losses, finite-size constraints, *etc.* not included in idealized theoretical models. Recent experiments have studied chaos in systems of photons^{11,12,13,14}, electrons^{15,16}, and excitons¹⁷. Phonon-polaritons provide another, yet unexplored arena for quantum chaos. In this work, we investigate experimentally and theoretically chaotic signatures of phonon polaritons in hexagonal boron nitride (hBN)^{18,19}. These collective modes are coherent hybrid excitations of mid-infrared photons and optical phonons distinguished by low losses and strong light-matter interaction²⁰. Optical anisotropy of hBN gives rise to hyperbolicity and multi-modal structure manifested in, e.g., super resonances²¹ and superluminal singularities²². Here, we exploit these hyperbolic phonon polaritons to reveal signatures of chaotic wave dynamics including complex spatial patterns, sensitivity to perturbations, scarring²³, and Berry's circles²⁴. Our results institute phonon polaritons in van der Waals materials as a new platform for quantum chaology.

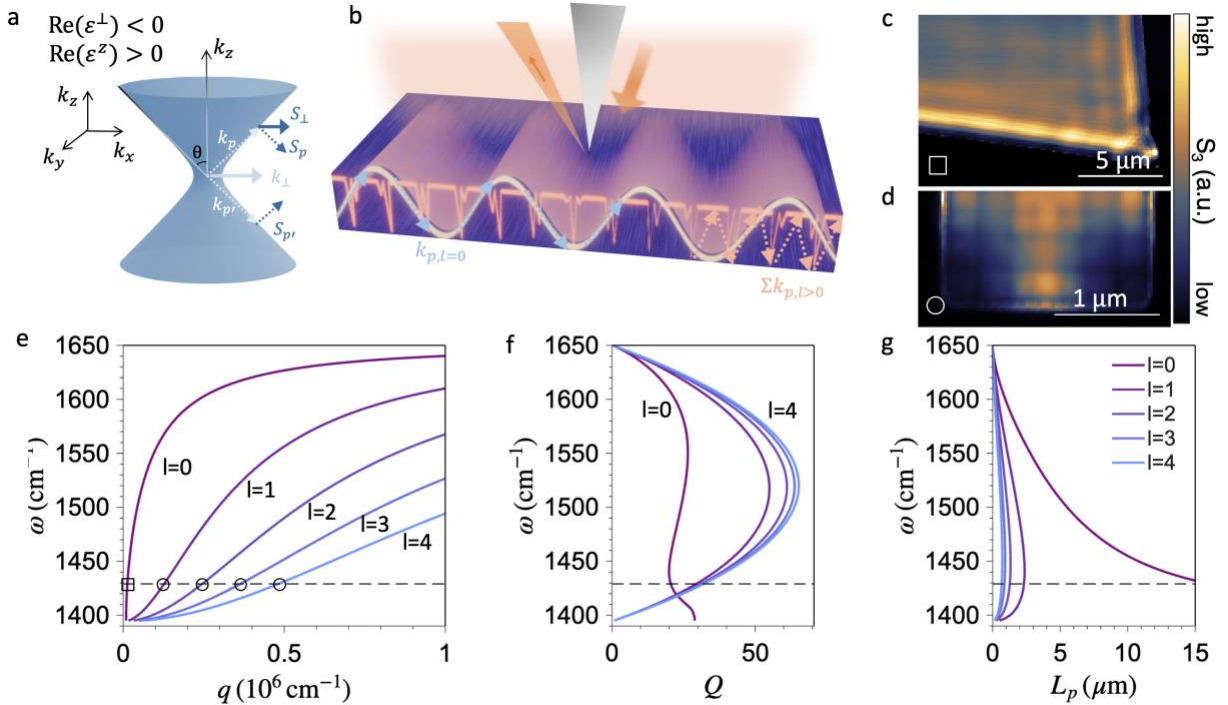


Figure 1: Nano-imaging of hyperbolic phonon polaritons in hBN. a. The isofrequency surface of the polaritons. The in-plane and the out-of-plane components of 3D vectors are designated by $\perp$ and z , respectively. The dashed light blue arrows show polariton momenta $\mathbf{k}_p$ and $\mathbf{k}_{p'}$ that have the same in-plane component $\mathbf{k}_\perp$ (the solid light blue arrow). The dashed dark-blue arrows indicate the Poynting vectors $\mathbf{S}_p$, $\mathbf{S}_{p'}$, and their 2D projection $\mathbf{S}_\perp$. b. Schematic of the experiment. The sharp tip of a near-field probe (the gray needle) scatters the infrared light (the orange arrows) and launches polaritons in hBN. The sine wave represents the $l = 0$ polariton mode with dashed light blue and orange arrows indicating $\mathbf{k}_{p,l=0}$ and $\mathbf{k}_{p,l>0}$ modes, respectively. The flattened periodic spikes depict “hot spots” due to the rays that are superpositions of multiple $l > 0$ modes. c. An s-SNOM image of a corner of a larger hBN crystal

where the $l = 0$ polariton mode dominates. d. An s-SNOM image of a smaller square-shaped sample where the $l > 0$ rays are prominent. $\omega = 1429 \text{ cm}^{-1}$ in both c and d. e. Polariton dispersion calculated for h^{10}BN of thickness $d = 68 \text{ nm}$. f, g. Calculated Q -factor and propagation length of the modes. The dashed line is $\omega = 1429 \text{ cm}^{-1}$.

Seeing hBN phonon polaritons

The phonon polaritons we study appear inside the mid-infrared Reststrahlen band (RB) $\omega_{\text{TO}} < \omega < \omega_{\text{LO}}$ of hBN (TO, transverse optical, LO, longitudinal optical) where its principal permittivities $\varepsilon^z(\omega)$, $\varepsilon^\perp(\omega)$ are of opposite signs^{17,18}. The permittivity $\varepsilon^\perp(\omega) < 0$ diverges to very large negative values near the in-plane optical phonon frequency $\omega_{\text{TO}} = 1395 \text{ cm}^{-1}$ and approaches zero at $\omega_{\text{LO}} = 1650 \text{ cm}^{-1}$, with values quoted for isotopically enriched h^{10}BN ²⁵. The permittivity $\varepsilon^z(\omega) > 0$ remains approximately constant across the RB. As shown in Fig. 1a, the polariton isofrequency surfaces are shaped as hyperboloids $\varepsilon^\perp \mathbf{k}_\perp^2 + \varepsilon^z k_z^2 = \varepsilon^\perp \varepsilon^z k_{\text{ph}}^2$ in momentum space, where $k_{\text{ph}} = 2\pi\omega$ is the photon momentum. These hyperboloids are asymptotic to the cone with an opening angle θ such that $\tan \theta = -i\sqrt{\varepsilon^\perp}/\sqrt{\varepsilon^z}$. The dashed arrows in Fig. 1a and b indicate the momentum $\mathbf{k}_p = (\mathbf{k}_\perp, k_z)$ and the energy flux (Poynting vector) $\mathbf{S}_p$ of the polaritons.

In a thin hBN layer of a thickness d , the out-of-plane momentum k_z is restricted to discrete values $\pm k_{z,l}$ where $k_{z,l} = (\pi/d)(l + \delta_z)$, $l = 0, 1, \dots$ is an integer, and $\delta_z = \delta_z(\omega)$ is a dimensionless function^{18,26}. The polariton dispersion splits into multiple branches with two-dimensional (2D) in-plane momenta $|\mathbf{k}_\perp| = k_l(\omega) = [\varepsilon^z k_{\text{ph}}^2 - (\varepsilon^z/\varepsilon^\perp)k_{z,l}^2]^{1/2}$, as illustrated by Figs. 1b,e. As detailed in SI, the dispersion of the principal ($l = 0$) mode can be mapped to that of (non-hyperbolic) plasmon-polaritons in a film with a complex conductivity $\sigma(\omega) = -\frac{i}{2}cd\varepsilon^\perp\omega$. The hyperbolicity resurfaces when modes with different l form superpositions. These multi-mode wavepackets (Fig. 1b) propagate as collimated rays²⁷, generating surface “hot spots” with the period $\frac{2\pi}{k_{l+1}-k_l} \simeq 2d \tan \theta$.

To image the propagation of the phonon polaritons, we employed scattering-type scanning near field optical microscopy (s-SNOM). As shown in Fig. 1b, a mid-infrared laser beam is focused on a metallized tip by a parabolic mirror. Incident photons at the tip apex launch polaritons that propagate outward, undergo multiple reflections from the billiard boundaries, and form standing-wave interference patterns. The tip subsequently outcouples propagating and interfering polaritons back into the far-field for detection. By operating in tapping mode with pseudoheterodyne signal demodulation²⁸, the near-field component of the signal is isolated and recorded with $\sim 15 \text{ nm}$ resolution. Figures 1c and 1d show representative s-SNOM maps for two samples of thickness $d = 68 \text{ nm}$ at frequency $\omega = 1429 \text{ cm}^{-1}$. As illustrated in Fig. 1b and presented in data (Fig. 1c-d), the principal polariton mode exhibits a smooth sinusoidal wave profile (Fig. 1c), while the higher-order polaritons produce periodic hot spots (Fig. 1d). The latter is more prominent in smaller cavities (Fig. 1d), possibly due to a stronger polariton confinement²⁹.

When the dielectric losses are included, the permittivities $\varepsilon_{\perp}(\omega)$, $\varepsilon_z(\omega)$ and the polariton momenta $k_l = k_l(\omega)$ acquire imaginary parts that determine the quality factor, $Q = \frac{1}{2} \text{Re } k_l(\omega) / \text{Im } k_l(\omega)$. For higher-order polariton modes which are more phonon-like, $Q \approx -\frac{1}{2} \text{Re } \tan \theta / \text{Im } \tan \theta$ is limited primarily by losses in hBN that depend on frequency, isotopic composition²⁴, and temperature³⁰. The calculated Q -factor of these modes displays a broad maximum $Q \sim 50$ in the middle of the RB (Fig. 1f). For the principal mode, losses in the substrate (SiO_2) also contribute to Q , especially near the bottom of the RB where this mode is more photon-like. The quality factors determine the propagation lengths $L_l = [2 \text{Im } k_l(\omega)]^{-1} = Q / \text{Re } k_l$, which are plotted in Fig. 1g for $d = 68$ nm case. Since samples studied in our work have lateral sizes L in the micron range, both $l = 0$ and 1 polaritons experience multiple bounces in billiard cavities.

The aforementioned mapping to 2D plasmons (SI) helps to simplify modeling of the principal polaritons laterally confined in a sample of a nontrivial shape. This mapping implies that the real-space distribution of the electric field $\psi(x, y)$ of the principal polariton mode just above the surface can be approximately described by the 2D Helmholtz equation

$$(\nabla_{\perp}^2 + E)\psi(x, y) = F(x, y), \quad (1)$$

$$E = k_0^2(\omega), \quad (2)$$

supplemented with the appropriate boundary conditions (BC) at the edges. Such BC differ from those at the large-area interfaces. In the thin-slab limit $k_0 d \ll 1$, they approach instead the BC for 2D plasmons^{31,32} that involve a direction-dependent reflection phase shift (SI, Sec. 2B). Function $F(x, y)$ on the right-hand side of Eq. (1) represents an external source of the polaritons corresponding to the localized excitation by the near-field probe in this work. With a δ -function-like $F(x, y)$, the solution to Eqs. (1-2) defines the polariton Green's function. On the other hand, if $F(x, y)$ is set to zero in a finite-size billiard, the solutions of Eq. (1) are eigenfunctions $\psi_m(x, y)$ corresponding to discrete real eigenvalues E_m . The spectral decomposition of the Green's function is dominated by eigenmodes with E_m close to $k_0^2(\omega)$. Observability of individual eigenmodes is limited by the dielectric losses where at most $\sim Q$ lowest eigenmodes can be resolved because $\text{Im } k_0^2(\omega) \sim E_m / Q$ becomes larger than the level spacing $s_m = E_{m+1} - E_m$ as m increases.

For a 3D hBN nanocrystal of size $d \ll L \ll L_p$, a generic shape of the boundary is expected to construct a non-integrable billiard. On the other hand, a common example of integrable billiard is a spheroidal particle²⁶. Below we will investigate a few special cases of billiards hosting chaotic hBN phonon polaritons.

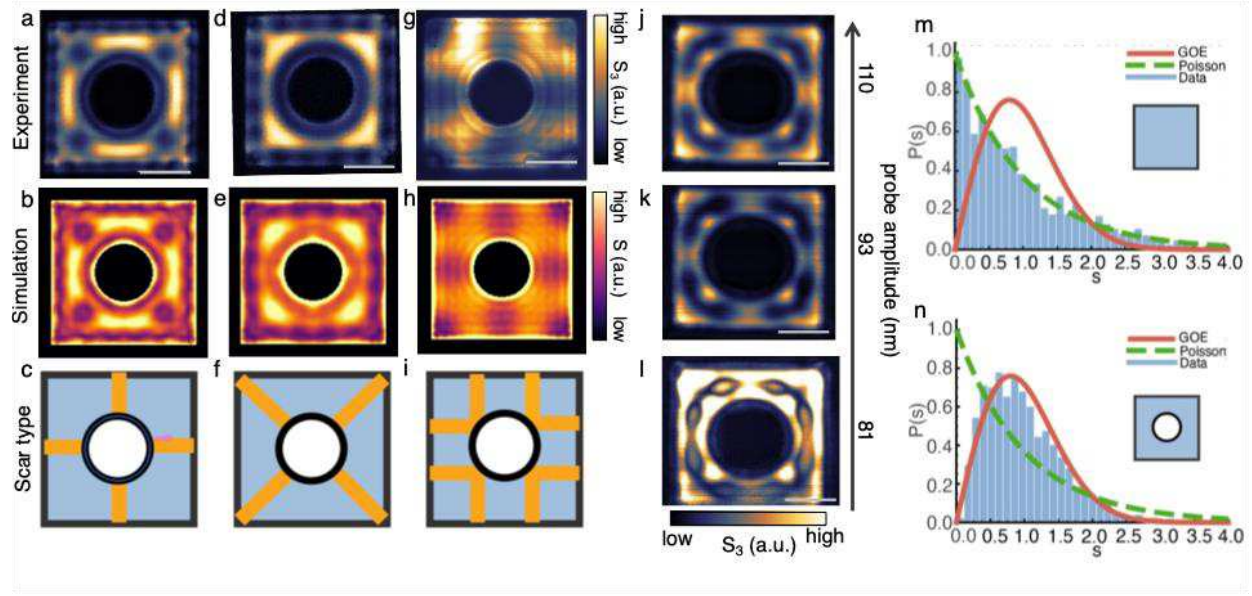


Figure 2: Polariton chaos in Sinai billiards. a.-i. Scarring effect of hBN phonon polaritons. The first row shows experimental s-SNOM data (the third-order demodulated pseudoheterodyne signal S_3). The second row shows the Green's function simulations. The third row illustrates the classical trajectories (the orange lines). The s-SNOM data in a is taken on a 76 nm thick hB¹⁰N at 1408.5 cm⁻¹. The data in d is taken on the same sample at 1413 cm⁻¹. The data in g is taken on a 60 nm thick natural hBN at 1405 cm⁻¹. All the samples have width 3 μm and hole diameter 1.2 μm. The scale bars are 1 μm. j.-l The s-SNOM data for a 72 nm-thick natural hBN sample with probe tapping amplitudes 110, 93, and 81 nm at frequency $\omega = 1493$ cm⁻¹. m, n. Calculated eigenvalue spacing distribution for a square and a Sinai billiard. The dashed green and solid red curves represent the Poisson distribution and the GOE Wigner surmise, respectively.

Signatures of chaos in a polariton Sinai billiard

The Sinai billiard³³, which can be presented as a square domain with a central hole, is an ergodic dynamical system that serves as a paradigm^{34,35} for studying of quantum chaos. The chaoticity originates from defocusing of particles or waves propagating within the domain when they reflect off the convex inner boundary. Here we demonstrate three key signatures of chaos in polariton Sinai billiards: the eigenfunction scarring, the butterfly effect, and the eigenvalue statistics.

Quantum scarring^{36,37} is a phenomenon where the eigenstates of a classically chaotic quantum system have enhanced probability density in a vicinity of unstable classical periodic orbits. In our polariton billiards, the scarring manifests itself as brightened traces in the polariton maps presented in Fig. 2a-i. We observed three types of scars in polariton Sinai billiards which are refer to as the “+” cross (Fig. 2a-c), the “x” cross (Fig. 2d-f), and the Γ -cross (Fig. 2g-i). Each leg of these four-legged crosses is an independent orbit. The top row of the 3 × 3 grid in Fig.

2a-i shows our experimental data, the middle row presents numerical simulations, and the bottom row illustrates the periodic orbits from which the naming of the scars was derived. The “+” scars are associated with the shortest orbits, which are the least unstable of the three. They are the most prevalent types of scars we observed, appearing in a large variety of eigenmodes. The $\times$ scars are also fairly common and can be observed in multiple scenarios (SI). The Γ -cross scars are relatively rare. In our experiments, they appear to be formed by the higher-order polariton modes rather than the principal ones.

We note three other features in images in Fig.2. First, there is a darkened ring along the inner perimeter in Figs. 2a and 2d. We tentatively attribute it to defocusing by the curved boundary that causes incident and reflected polariton waves to have a reduced constructive interference. Second, we observe the polariton hot spots (or rather, hot lines) due to the polariton rays: the concentric circles near the inner perimeter and the squares near the outer perimeter of the billiard (Fig. 2g). Third, we detected polariton edge modes along the outer perimeter in Figs. 2a and 2d. According to the theory³⁸, the edge modes have a slightly shorter wavelength than the surface polaritons with the result for 2D plasmons as 0.906. For the simplified BC we use for hBN phonon polaritons, the ratio of the edge and surface polariton wavelengths is $\cos(\pi/8) = 0.924$ (SI Sec. 2C). Because of the wavelength mismatch, for ideal edges, these polariton edge modes would be a separate integrable one-dimensional (1D) subsystem. In reality, edge imperfections cause mixing of the surface and edge polaritons, creating an extra dissipation channel for the former. Additional data for different frequencies and billiard geometries can be found in SI.

To model the experimental imaging data we assume that the s-SNOM signal is proportional to the Green’s function (SI), which we compute numerically. For simplicity, we account for all types of polariton losses by using a complex E with an imaginary part treated as an adjustable parameter. Overall, our simulations (Fig. 2b,e,h) show a good qualitative agreement with the experimental observations (Fig. 2a,d,g).

Next, we address the observed sensitivity to external perturbations (the butterfly effect)³⁹. Since the near-field interaction between the probe and the sample depends on their separation²⁸, changing the probe tapping amplitude modifies the initial conditions for polariton launching, and so can be considered a perturbation. As shown in Fig. 2j-l and SI, the s-SNOM maps are greatly altered by changing the tapping amplitude. Figure 2l displays a much higher contrast and additional fine features compared to Figs. 2j and k measured at larger tapping amplitudes. Our modeling indicates that these extra features can be attributed to whispering-gallery-type polariton eigenmodes. Although contrast improving with decrease in tapping amplitude is not unusual for s-SNOM, we posit that the observed sensitivity to perturbations reflects not only the local probe-sample coupling, but also the intrinsic sensitivity of chaotic polariton dynamics. We return to this point below when we discuss Fig. 4.

Another widely accepted signature of quantum chaos is the eigenvalue statistics. For integrable systems, the eigenvalue spacing distribution $P(s)$ is Poissonian. On the other hand, the spectral properties of a classically chaotic system follow the universal Wigner-Dyson statistics of Gaussian random matrices. In particular, for spinless time-reversal-symmetric systems, $P(s)$ is normally described by the Gaussian Orthogonal Ensemble (GOE) and is closely approximated by an analytical expression known as the Wigner surmise. There are two reasons to verify these conjectures for our polariton billiards. First, our problem is characterized by non-standard BC.

Second, some billiards exhibit anomalous properties even for standard Dirichlet BC because of Kramers-like degeneracies⁴⁰. For the purpose of this verification we computed several thousand eigenvalues E_m (SI) of the square and the Sinai billiards. We plotted the distribution $P(s)$ of the nearest-neighbor spacings $s_m = E_{m+1} - E_m$, measured in units of the average spacing, in Figs. 2m and n. The square billiard is integrable for the Dirichlet BC and remains such for our BC as well. The level spacing in this billiard is well described by the Poisson distribution (Fig. 2a, the dashed green curve). On the other hand, $P(s)$ for the Sinai billiard conforms to the GOE Wigner surmise (the solid red curve), corroborating its chaotic nature.

Although the finite polariton loss prevents experimental study of eigenvalue statistics, we were able to obtain spectral comparison with the intergrable 1D edge modes. To do so, in addition to the single-color imaging, we performed broadband hyperspectral near-field imaging. Figure 3a shows spectra taken along the billiard edge. They exhibit discrete increments in the total number of nodes that correspond to switching between consecutive eigenmodes. This behavior is characteristic of an integrable 1D cavity. In contrast, spectra acquired in the interior of the Sinai billiard (Fig. 3b) display complex nodal patterns which presumably arise from chaotic dynamics. The key features of Fig. 3a and b are qualitatively comparable with Green's function simulations shown in Figs. 3c and d.

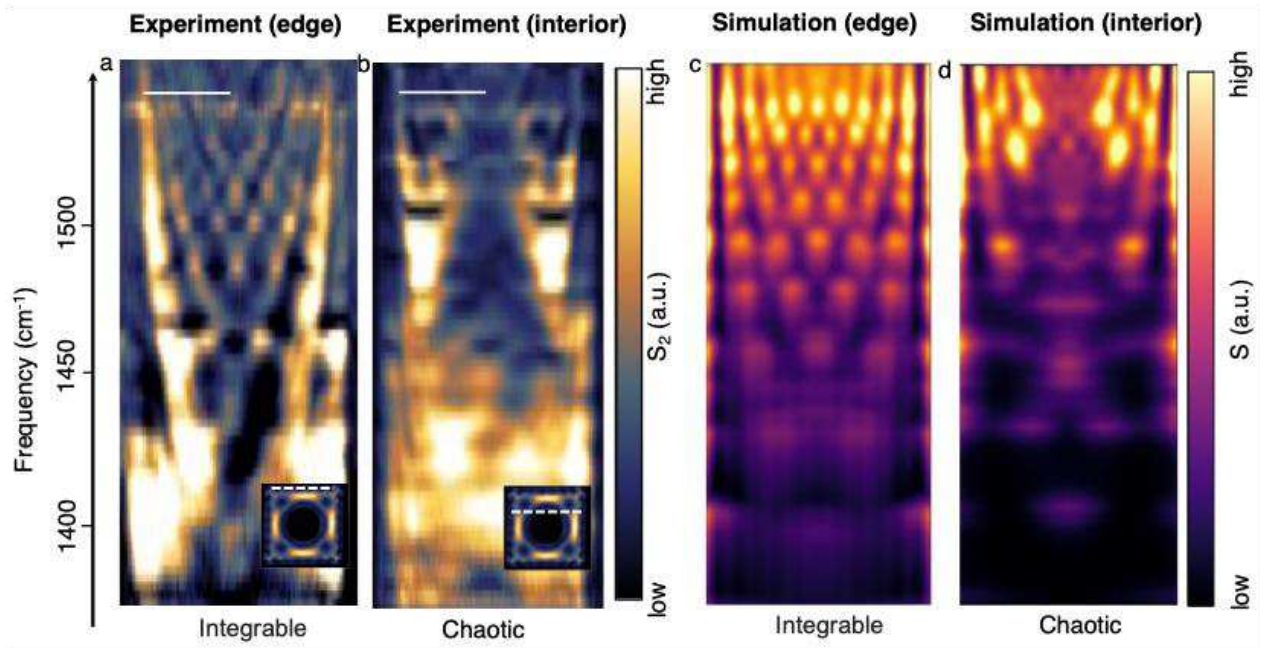


Figure 3: Hyperspectral images of phonon polaritons. a. Image taken along the edge of an hBN Sinai billiard of thickness 100 nm and width 5 μm . The location of the line scan is marked by the dashed line in the inset. b. Image taken along the line in the interior of the Sinai billiard. Scale bars in both a and b: 2 μm . c-d. Green's function simulations for a-b.

Koch snowflake billiards

Koch's fractal curve construction⁴¹ inspires another interesting example of nonintegrable

billiards featuring increasing boundary complexities. Starting from a regular hexagon (Fig. 4a-c), each subsequent iteration is generated by trisecting edges and inserting inward equilateral triangles for the boundaries. The three rows in Figs. 4 a-l present the corresponding s-SNOM data, their fast Fourier transforms (FFTs), and the FFTs of the simulated Green's functions. The dominating wave features are $l = 1$ modes measured with a fixed probe amplitude of 106 nm, (other conditions available in SI). All the FFT maps inherit C_6 symmetry from the billiard geometries, and the central bright spot is due to the finite-size effect. Interestingly, at the distance of approximately $k_{\text{ring}} = 12.56 \mu\text{m}^{-1}$ (indicated by the orange arrow and the half dashed circles), the high-intensity features corresponding to $l = 1$ phonon polariton modes behave distinctively. As the iteration order increases, this feature evolves from a set of six isolated dots and patches (Fig. 4b,e) to a nearly isotropic ring (Fig. 4h, k). This progressively increasing uniformity of the polariton momentum directions complies with Berry's random-wave conjecture³. Briefly, this conjecture states that in the semiclassical limit, eigenstates of a chaotic system can be viewed as superpositions of numerous plane waves with a statistically isotropic momentum distribution. Therefore, a chaotic cavity eigenmodes $\psi_m(\mathbf{r})$ consist of waves whose momenta densely populate the circle $|\mathbf{k}_\perp| = \text{Re } k_0$. Since the s-SNOM signal represents the Green's function, which is bilinear in $\psi_m(\mathbf{r})$, the FFT of the s-SNOM amplitude is maximized on the ring of radius $2 \text{Re } k_0$ corresponding to k_{ring} . Similar circles⁴² have been recently observed in experimental studies of plasmons in graphene on twisted hBN.

Another measure of wave randomness is the autocorrelation length⁴³ of the real-space polariton images. We have calculated this length for the data in Fig. 4a, d, g and j, obtaining 717, 484, 327, and 255 nm, respectively. As the boundary becomes increasingly more fractal, the autocorrelation length converges to $\pi/(\text{Re } k_0) \approx 250 \text{ nm}$, i.e., one half of the polariton wavelength, serving as additional evidence³⁸ for the emergence of chaos.

A few remarks on the Sinai and the Koch snowflake billiards are in order. First, while a smooth boundary predominantly supports specular reflection of polaritons, a multiscale boundary can locally scramble the reflection phase and thereby promote diffused scattering. Second, in the Sinai billiards, scarring of eigenfunctions gives rise to quasi-regular features in the momentum space, obscuring Berry's circles. In contrast, Koch snowflake billiards with fractal boundaries are a better choice to observe the random-wave effect. Third, the 0-th and 1-th iteration of Koch snowflake billiards are not classically chaotic. However, quantum wave effect allows chaotic behavior to arise⁴⁴. Therefore, if sufficiently high-order eigenmodes are accessible, quantum chaos can happen in classically non-chaotic billiards.

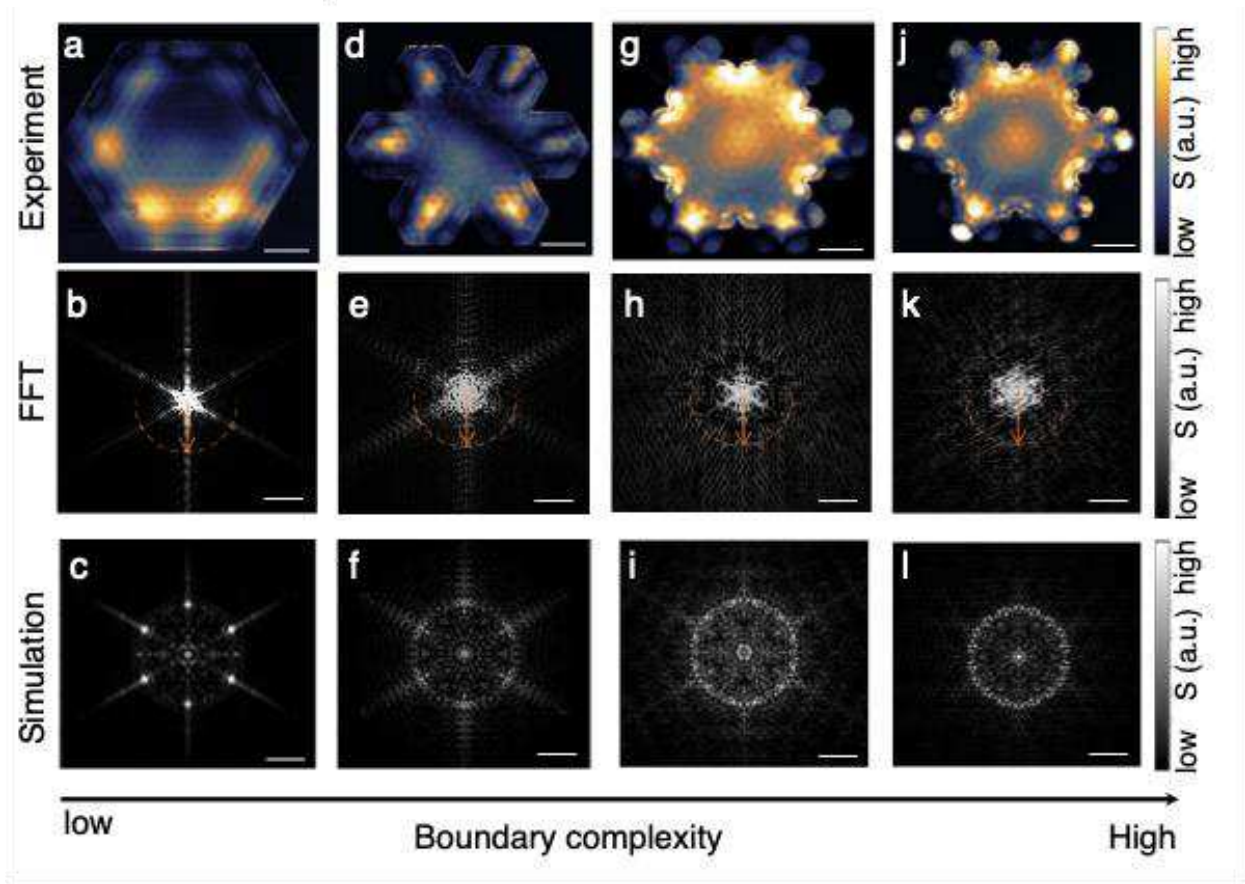


Figure 4: Berry's random-wave conjecture tested by high-order ($l = 1$) phonon polaritons in the Koch snowflake billiards. a–d. Real-space images of the experimental data taken at 1429 cm^{-1} with a probe amplitude of 106 nm . Scale bars, $1 \text{ } \mu\text{m}$. e–h. FFTs of experimental data. The orange arrow indicates the polariton momentum $k_{\text{ring}} = 12.56 \text{ } \mu\text{m}^{-1}$ sketched in dashed half circles. As the iteration order increases, discrete features along this value merge into an almost isotropic ring. i–l. Green's function simulations (FFT). Scale bars, $10 \text{ } \mu\text{m}^{-1}$.

Discussion

Our experiments establish hyperbolic phonon polaritons in hBN billiards as a versatile platform for exploring quantum chaos in hybrid light-matter systems. In Sinai billiards, we identify multiple signatures of chaotic wave dynamics, including Wigner-Dyson level statistics, quantum scars, and sensitivity to perturbations, along with the contrasting spectra of chaotic bulk modes and integrable edge modes. In Koch snowflake billiards, increasing boundary complexity drives the polariton momentum distribution towards an isotropic ring in Fourier space, consistent with Berry's random-wave conjecture. Together, these observations demonstrate that the onset and characters of quantum chaos in polariton billiards can be engineered through geometry and boundary conditions, in addition to the tunability of polariton dispersions and quality factors. These capabilities extend wave chaotic physics to deeply subwavelength collective excitations

and points towards broader realizations in plasmonic, excitonic and magnonic polariton systems.

Beyond providing a new setting for fundamental studies of quantum chaos, our findings could be harnessed for applications such as enhanced light-matter interaction²⁰, broadband coupling⁴⁵, and information scrambling⁴⁶ in quantum materials. The ability to sculpt van der Waals cavities at the nanoscale, combined with the rich dispersion of hyperbolic polaritons, may enable designer chaotic resonators for tailoring spectral and spatial properties. Future work could explore non-Hermitian effects⁴⁷, many-body interactions^{48,49}, and topological features^{50,51} in chaotic polariton systems, further bridging the gaps between quantum chaos, condensed matter physics, and nanophotonics.

Acknowledgements

Y.D., F.L., Y.L., M.F., and D.N.B are supported by NSF ECCS-2529062 for cavity research, and in part by Julian Schwinger Foundation for polariton imaging. Y.D., D.S., C.R.D., and D.N.B. are supported by Programmable Quantum Materials, an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award no. DE-SC0019443. S.L. and J.H.E. are supported by the National Science Foundation, award number CMMI 1538127 for the h^{10} BN crystal growth. D.N.B. is a Moore Investigator in Quantum Materials EPIQS no. 9455. L.W. is supported by Empire State Development Corporation of New York. V.G. was supported by the U.S. Department of Energy, Office of Science Basic Energy Sciences under Award No. DE-SC0001911. V.G. is grateful to Sir Michael Berry for helpful comments on the random wave conjecture and classical-to-quantum correspondence.

References

- ¹ Balian, R., & Bloch, C. *Annals of Physics*, 60, 2, 10 (1970-1972) Distribution of eigenfrequencies for the wave equation in a finite domain: I-III.
- ² Berry, M.V., Tabor, M. Level clustering in the regular spectrum. *Proceedings of the Royal Society A* 356, 375–394 (1977). <https://doi.org/10.1098/rspa.1977.0140>
- ³ Berry, M.V. Regular and irregular semiclassical wavefunctions. *Journal of Physics A* 10, 2083 (1977). https://doi.org/10.1142/9789813221215_0023
- ⁴ Porter, C.E. *Statistical Theories of Spectra: Fluctuations*. Academic, New York (1965).
- ⁵ Mehta, M.L. *Random Matrices and the Statistical Theory of Energy Levels*. Academic, New York (1967). <https://doi.org/10.2307/2284361>
- ⁶ Guhr, T., Müller-Groeling, A., Weidenmüller, H.A. Random-matrix theories in quantum physics: common concepts. *Physics Reports* 299, 4–6 (1998). [https://doi.org/10.1016/s0370-1573\(97\)00088-4](https://doi.org/10.1016/s0370-1573(97)00088-4)
- ⁷ Berry, M. V. (1987). The Bakerian Lecture, 1987: Quantum chaology. *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences*, 413(1844), 183–198. <https://doi.org/10.1098/rspa.1987.0109>
- ⁸ Bohigas, O., Giannoni, M.J., Schmit, C. Characterization of chaotic quantum spectra and universality of level fluctuation laws. *Physical Review Letters* 52, 1–4 (1984). <https://doi.org/10.1103/physrevlett.52.1>
- ⁹ Stöckmann, H.-J. (1999). *Quantum Chaos: An Introduction*. Cambridge University Press. <https://doi.org/10.1017/CBO9780511599989>
- ¹⁰ Haake, F. (2010). *Quantum Signatures of Chaos* (3rd ed.). Springer-Verlag. <https://doi.org/10.1007/978-3-642-05428-0>

- ¹¹ Nöckel, J.U., Stone, A.D. Ray and wave chaos in asymmetric resonant optical cavities. *Nature* 385, 45–47 (1997). <https://doi.org/10.1038/385045a0>
- ¹² Lemos, G.B., Gomes, R.M., et al. Experimental observation of quantum chaos in a beam of light. *Nature Communications* 3, 1212 (2012). <https://doi.org/10.1038/ncomms2214>
- ¹³ Cao, H., Wiersig, J. Dielectric microcavities: Model systems for wave chaos and non-Hermitian physics. *Reviews of Modern Physics* 87, 61 (2015). <https://doi.org/10.1103/revmodphys.87.61>
- ¹⁴ Wang, S., Liu, S., Liu, Y., et al. Direct observation of chaotic resonances in optical microcavities. *Light: Science and Applications* 10, 135 (2021). <https://doi.org/10.1038/s41377-021-00578-7>
- ¹⁵ Hagymási, I., Vancsó, P., et al. Interaction effects in a chaotic graphene quantum billiard. *Physical Review B* 95(7) (2017). <https://doi.org/10.1103/physrevb.95.075123>
- ¹⁶ Ge, Z., Graf, A.M., et al. Direct visualization of relativistic quantum scars in graphene quantum dots. *Nature* 635(8040) (2024). <https://doi.org/10.1038/s41586-024-08190-6>
- ¹⁷ Gao, T., Estrecho, E., et al. Observation of non-Hermitian degeneracies in a chaotic exciton-polariton billiard. *Nature* 526(7574) (2015). <https://doi.org/10.1038/nature15522>
- ¹⁸ Dai, S., et al. Tunable phonon polaritons in atomically thin van der Waals crystals of boron nitride. *Science* 343, 1125–1129 (2014). <https://doi.org/10.1126/science.1246833>
- ¹⁹ Caldwell, J.D., Kretinin, A.V., Chen, Y., et al. Sub-diffractive volume-confined polaritons in the natural hyperbolic material hexagonal boron nitride. *Nature Communications* 5(1) (2014). <https://doi.org/10.1038/ncomms6221>
- ²⁰ Basov, D.N., et al. Polaritons in van der Waals materials. *Science* 354, 1992 (2016). <https://doi.org/10.1126/science.aag1992>
- ²¹ Narimanov, E.E., Demler, E.A. Hyperbolic quantum processor (2024). <https://doi.org/10.48550/arXiv.2412.14098>
- ²² Bucher, T., et al. Superluminal Correlations in Ensembles of Optical Phase Singularities (2025). <https://doi.org/10.1038/s41586-026-10209-z>
- ²³ Kaplan, L. Scars in quantum chaotic wavefunctions. *Nonlinearity* 12, 1 (1999). <https://doi.org/10.1088/0951-7715/12/2/009>
- ²⁴ Berry, M.V. Regular and irregular semiclassical wavefunctions. *Journal of Physics A* 10, 2083 (1977). https://doi.org/10.1142/9789813221215_0023
- ²⁵ Giles, A.J., Dai, S., Vurgaftman, I., Hoffman, T., Liu, S., Lindsay, L., Ellis, C.T., Assefa, N., Chatzakis, I., Reinecke, T.L., Tischler, J.G., Fogler, M.M., Edgar, J.H., Basov, D.N., Caldwell, J.D. Ultralow-loss polaritons in isotopically pure boron nitride. *Nature Materials* 17(2), 134–139 (2017). <https://doi.org/10.1038/nmat5047>
- ²⁶ Sun, Z., Gutiérrez-Rubio, A., Basov, D.N., Fogler, M.M. Hamiltonian optics of hyperbolic polaritons in nanogranules. *Nano Letters* 15, 4455 (2015). <https://doi.org/10.1021/acs.nanolett.5b00814>
- ²⁷ Dai, S., Ma, Q., Andersen, T., et al. Subdiffractive focusing and guiding of polaritonic rays in a natural hyperbolic material. *Nature Communications* 6, 6963 (2015). <https://doi.org/10.1038/ncomms7963>
- ²⁸ Ocelic, N., Huber, A., Hillenbrand, R. Pseudoheterodyne detection for background-free near-field spectroscopy. *Applied Physics Letters* 89, 101124 (2006). <https://doi.org/10.1063/1.2348781>
- ²⁹ Herzig Sheinfux, H., Orsini, L., Jung, M. et al. High-quality nanocavities through multimodal confinement of hyperbolic polaritons in hexagonal boron nitride. *Nat. Mater.* 23, 499–505 (2024). <https://doi.org/10.1038/s41563-023-01785-w>
- ³⁰ Ni, G., et al. Long-lived phonon polaritons in hyperbolic materials. *Nano Letters* 21(13), 5767–5773 (2021). <https://doi.org/10.21203/rs.3.rs-280509/v1>
- ³¹ Jiang, B.-Y., Mele, E.J., Fogler, M.M. Theory of plasmon reflection by a 1D junction. *Optics Express* 26, 17209 (2018). <https://doi.org/10.1364/oe.26.017209>
- ³² Svintsov, D.A., Alymov, G.V. Refraction laws for two-dimensional plasmons. *Physical Review B* 108(12), 121410 (2023). <https://doi.org/10.1103/physrevb.108.121410>
- ³³ Sinai, Ya. G., Dynamical systems with elastic reflections. *Russian Mathematical Surveys*, 25, 2, 137 (1970), <https://doi.org/10.1070/RM1970v025n02ABEH003794>
- ³⁴ M.V. Berry, Quantizing a classically ergodic system: Sinai's billiard and the KKR method, *Annals of Physics*, 131, 163–216 (1981)

- ³⁵ Izrailev, F. M. Simple models of quantum chaos: Spectrum and eigenfunctions, *Physics Reports*, 196, 5-6, 11(1990)
- ³⁶ Heller, E. J. Bound-State Eigenfunctions of Classically Chaotic Hamiltonian Systems: Scars of Periodic Orbits. *Physical Review Letters*, 53, 16, 10(1984)
<https://journals.aps.org/prl/abstract/10.1103/PhysRevLett.53.1515>
- ³⁷ Kaplan, L. Scars in quantum chaotic wavefunctions. *Nonlinearity* 12, 1 (1999).
<https://doi.org/10.1088/0951-7715/12/2/009>
- ³⁸ Volkov, V.A., Mikhailov, S.A. Edge magnetoplasmons: low frequency weakly damped excitations in inhomogeneous two-dimensional electron systems. *Sov. Phys. JETP* 67(8), 1639 (1988).
- ³⁹ Lorenz, E. N. Deterministic nonperiodic flow. *Journal of the Atmospheric Sciences*, 20(2), 130–141(1963).
- ⁴⁰ Leyvraz, F., Schmit, C., Seligman, T.H. Anomalous spectral statistics in a symmetrical billiard. *Journal of Physics A: Mathematical and General* 29(22), 575–580 (1996). <https://doi.org/10.1088/0305-4470/29/22/004>
- ⁴¹ Von Koch, H. Une méthode géométrique élémentaire pour l'étude de certaines questions de la théorie des courbes planes. *Acta Mathematica* 30, 145–174 (1906). <https://doi.org/10.1007/bf02418570>
- ⁴² Zhang, S. et al. Plasmonic Polarization Sensing of Electrostatic Superlattice Potentials, *Phys.Rev.X*. 15.011019(2025) <https://link.aps.org/doi/10.1103/PhysRevX.15.011019>
- ⁴³ Lin, J., Zhuang, Y., Graf, A.M., Keski-Rahkonen, J., Heller, E.J. Shaping chaos in bilayer graphene cavities (2026). <https://doi.org/10.48550/arXiv.2512.10914>
- ⁴⁴ Rozenbaum, E.B., Bunimovich, L.A., Galitski, V. Early-time exponential instabilities in nonchaotic quantum systems. *Physical Review Letters* 125, 014101 (2020). <https://doi.org/10.1103/physrevlett.125.014101>
- ⁴⁵ Xuefeng Jiang et al. „Chaos-assisted broadband momentum transformation in optical microresonators. *Science* 358,344-347(2017).DOI:10.1126/science.aao0763
- ⁴⁶ Swingle, B. Unscrambling the physics of out-of-time-order correlators. *Nature Phys* 14, 988–990 (2018). <https://doi.org/10.1038/s41567-018-0295-5>
- ⁴⁷ El-Ganainy, R., Makris, K., Khajavikhan, M. et al. Non-Hermitian physics and PT symmetry. *Nature Phys* 14, 11–19 (2018). <https://doi.org/10.1038/nphys4323>
- ⁴⁸ Ruidas., S, Roy., S, et al. Emergence of many-body chaos in the presence of quasiparticles, *Phy. Rev.B* 114, 014306 (2026)
- ⁴⁹ Carusotto, I and Ciuti, c Quantum fluids of light, *Rev. Mod. Phys.* 85, 299 (2013)
<https://doi.org/10.1103/RevModPhys.85.299>
- ⁵⁰ Lu, L., Joannopoulos, J. & Soljačić, M. Topological photonics. *Nature Photon* 8, 821–829 (2014).
<https://doi.org/10.1038/nphoton.2014.248>
- ⁵¹ Mangold, F., Baù, E., Nan, L. et al. Phonon-polaritonic skyrmions: transition from bubble- to Néel-type. *Light Sci Appl* 15, 239 (2026).

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [20260816SI.pdf](#)