Title
Programmable Bloch polaritons in graphene.

Permalink
https://escholarship.org/uc/item/7b98h3nx

Journal
Science advances, 7(19)

ISSN
2375-2548

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Publication Date
2021-05-07

DOI
10.1126/sciadv.abe8087

Peer reviewed
Efficient control of photons is enabled by hybridizing light with matter. The resulting light-matter quasi-particles can be readily programmed by manipulating either their photonic or matter constituents. Here, we hybridized infrared photons with graphene Dirac electrons to form surface plasmon polaritons (SPPs) and uncovered a previously unexplored means to control SPPs in structures with periodically modulated carrier density. In these periodic structures, common SPPs with continuous dispersion are transformed into Bloch polaritons with attendant discrete bands separated by bandgaps. We explored directional Bloch polaritons and steered their propagation by dialing the proper gate voltage. Fourier analysis of the near-field images corroborates that this on-demand nano-optics functionality is rooted in the polaritonic band structure. Our programmable polaritonic platform paves the way for the much-sought benefits of on-the-chip photonic circuits.
toward (not shown). Next, when the lower polaritonic band is raised further into the polaritonic bandgap, polariton propagation is completely inhibited (Fig. 1D). Simulations confirm that Bloch polaritons only propagate along \( \hat{M} = \langle 10 \rangle \) directions. Inset shows an enlarged real-space lattice pattern. (E) to (G), (H) to (J), same as (B) to (D) at \( \bar{n}_s = 4.8 \times 10^{12} \text{ cm}^{-2} \) and \( \bar{n}_s = 6.0 \times 10^{12} \text{ cm}^{-2} \), respectively. At \( \bar{n}_s = 4.8 \times 10^{12} \text{ cm}^{-2} \), Bloch polaritons reside in the lower band (E), emerge in the \( K \) pockets (F), and propagate along \( \hat{K} = \langle 10 \rangle \) directions (G). At \( \bar{n}_s = 6.0 \times 10^{12} \text{ cm}^{-2} \), Bloch polaritons reside entirely in the lower band (H), exhibit circular equi-energy contours (I) and propagate isotropically in all directions (J). Inset of (I) shows the first BZ marked with symmetry points.

The metallic tip of an atomic force microscope (AFM) acts as an optical antenna that outcouples Bloch polaritons into free-space photons, enabling real-space polaritonic imaging deeply below the diffraction limit (31, 32). The tip-scattered light is registered by a detector, and the amplitude \( s(r) \) and phase \( \phi(r) \) of the corresponding near-field signal are extracted by a proper demodulation scheme (Materials and Methods). We mainly analyzed the near-field amplitude images \( s(r) \). All the near-field measurements were performed at \( T = 60 \text{ K} \) to reduce SPP losses due to phonon scattering (34) and to minimize broadening of the polaritonic band structure.

Near-field images close to a gold launcher show directional propagation of Bloch polaritons (Fig. 2). The leading edges of the launcher form a 30° angle, thus exciting Bloch polaritons propagating along \( \hat{R} \) and \( \hat{M} \) directions (arrows in Fig. 2A). In the course of our measurements, we kept the laser energy at \( \omega = 890 \text{ cm}^{-1} \) while tuning the gate voltage. It is instructive to plot the gate-voltage dependence of the band structure at \( \omega = 890 \text{ cm}^{-1} \) (Fig. 2B and fig. S1). This new representation contains the same physics as Fig. 1 (B, E, and H) but facilitates a more direct connection to our imaging data. Specifically, Fig. 2B reveals that polaritons reside near \( M \) points at lower carrier densities, whereas their \( K \) point counterparts are activated at higher carrier densities. At gate voltage \( V_g = 47.5 \text{ V} \) (Fig. 2C), Bloch polaritons only propagate along the \( \hat{M} \) direction. This phenomenon is particularly apparent when comparing the line profiles along \( \hat{M} \) (black) and \( \hat{K} \) (cyan) directions (Fig. 2F). A combination of \( V_g = 47.5 \text{ V} \) and \( \omega = 890 \text{ cm}^{-1} \) produces polaritonic fringes only in the \( \hat{M} \) direction, which also indicates that polaritons reside in the upper polaritonic band (Fig. 2B, top dashed line). At a slightly higher gate voltage \( V_g = 52.5 \text{ V} \), Bloch polariton propagation is inhibited in the \( \hat{M} \) direction, and the launcher only excites SPPs traveling diagonally along the \( \hat{R} \) direction (Fig. 2D). Line profiles in Fig. 2F confirm that Bloch polaritons propagate predominantly in the \( \hat{R} \) direction and that they can only be excited in the lower polaritonic band (Fig. 2B, middle direction).
dashed line). At even higher gate voltage $V_g = 72.5$ V, the laser energy is away from the polaritonic bandgap (Fig. 2B, bottom dashed line) and Bloch polaritons propagate isotropically in all directions (Fig. 2, E and F). Near-field images also reveal periodic dark spots, which are attributed to variation in the local polaritonic density of states (28) that is peripheral to our focus on directional Bloch polaritons. We have investigated multiple devices at a variety of gate voltages across the bandgap (fig. S2). All devices show consistent control of polariton propagation by gate voltage. The agreement between our experimental observations and band structure calculations demonstrates...
the ability to steer Bloch polaritons along the specified trajectories in our planar structure using the electric field effect.

Fourier analysis of the polaritonic patterns confirms that the directional launching is governed by the polaritonic band structure. Experimental inputs for this analysis are acquired by imaging Bloch polaritons emanating from the gold launcher in Fig. 3A, which resembles a point source for polaritonic waves (fig. S3). A gate voltage of $V_g = −45$ V implies that Bloch polaritons reside above the polaritonic bandgap and propagate along the $\tilde{M}$ direction (Fig. 3B). The symmetrized Fourier transform (Materials and Methods) of the near-field image (Fig. 3C) yields prominent features around $M$ points in the BZ, forming a hexagonal motif. A similar pattern is also observed in modeling results in Fig. 3D. The agreement between gross features in the data and the modeling corroborates that the directional Bloch polariton propagation is governed by the band structure. Propagating Bloch polaritons vanish for gate voltages within the bandgap at $V_g = −50$ V (fig. S4). At even lower gate voltages such as $V_g = −52.5$ V, polariton propagation direction is switched to $\tilde{K}$ (Fig. 3E). The Fourier transform of the near-field image reveals the formation of polaritonic pockets near $K$ points (Fig. 3, F and G), the complete field of view of the Fourier image is shown in fig. S5). Last, at $V_g = −60$ V, equi-energy contours appear as near-perfect circles within the first BZ (Fig. 3, I and J). The three selected panels of Fig. 3 (C, F, and I) are displayed again in Fig. 3K and are seen to be in close agreement with the simulated polaritonic band structure.

**Outlook**

We demonstrated programmable Bloch polaritons in a graphene polaritonic crystal platform. Bloch polaritons propagate along designated directions set by the applied gate voltage. The same general approach is well suited to the manipulation of other polaritonic systems, such as exciton polaritons and magnon polaritons. Relatively straightforward improvements of our platform aimed at reducing plasmonic damping and working gate voltages will enable operation at ambient conditions. Furthermore, our back-gated platform is compatible with ubiquitous metal oxide semiconductor field effect transistor technology and sets the stage for integrated photonic circuits (52, 53).

**MATERIALS AND METHODS**

**Cryogenic near-field imaging techniques**

Cryogenic near-field imaging measurements were performed using a home-built scattering-type scanning near-field optical microscope (s-SNOM) (54). The s-SNOM apparatus is based on a tapping mode AFM, coupled to a continuous-wave CO₂ laser (Access Laser), operating in ultrahigh vacuum (UHV) and cryogenic temperatures. The incoming laser beam is focused on the sample using a high-NA off-axis parabolic mirror inside the UHV chamber. The metallic AFM tip is tapped above the sample surface at a frequency of ~250 kHz. The tip-scattered light is demodulated using a pseudo-heterodyne detection scheme to extract both the near-field amplitude $s(\mathbf{r}, \omega)$ and the phase $\phi(\mathbf{r}, \omega)$. The images shown in the main text are the near-field amplitude $s(\mathbf{r}, \omega)$ normalized to the gold launcher. Gold deposited using thermal evaporation serves as a good reference for the demodulated signal. To properly suppress background contributions to the near-field signal, we demodulate at the third harmonic of the tip tapping frequency (28).

**Sample fabrication**

The patterned dielectric superlattices (PDSLs) (28, 39) consisting of a hexagonal array of pillars were fabricated by plasma etching the SiO₂ using a thin polymethyl methacrylate (PMMA) mask. Si substrates with thermally grown SiO₂ of thickness 285 nm were spin-coated with a layer of 495 A PMMA of thickness 50 nm. The hexagonal pattern was written by e-beam lithography in a Nanobeam nB4 system at a current of 300 to 400 pA. SiO₂ pillars were etched in an Oxford Plasmalab 80 Plus system using a mixture of CHF₃ gas (40 sccm) and Ar gas (5 sccm) to a depth of ~50 nm. The PMMA mask was removed through an O₂ plasma etching, and the PDSL was cleaned by piranha chemical etching. The resulting superlattice is an array of pillars with 46- to 48-nm diameter and 58-nm height that form a hexagonal lattice, with lattice periodicity 85 nm in the device shown in Fig. 2 and 80 nm in the device shown in Fig. 3.

A mechanically exfoliated graphene and hBN heterostructure was placed onto the PDSL by mechanical transfer (55). The heterostructure consists of a top h₁BN layer of ~4 nm, a layer of graphene, and a bottom h⁻¹BN layer of ~4 nm and was assembled using a polypropylene carbonate (PPC) transfer slide. Given the challenges involved in picking up an hBN layer as thin as 4 nm, such as the topmost layer, using PPC, we dug a circular hole (diameter 7 to 11 μm) out of a thick (30 to 60 nm) hBN flake using e-beam lithography. This thick hBN flake could be easily picked up by the PPC, and the thin h₁BN and graphene flakes were positioned so that they covered the circular hole from below and could be picked up by the van der Waals interaction between them and the thick top hBN flake. Therefore, inside the circular hole (not shown in experimental images) we had a thin h⁻¹BN-graphene-h₁BN heterostructure. Metal contacts and launchers were deposited using standard e-beam lithography processes. The graphene device is a single crystal within our field of view, as confirmed by near-field imaging results. All the reported back gate voltages are measured from the charge neutrality point of the devices. The residual charge density in the absence of gate voltage is estimated to be $n_s = 1.75 \times 10^{11}$ cm⁻².

The monoisotopic h⁻¹BN single crystals were synthesized by the metal flux method (56). High-purity elemental h⁻¹B (99.41%) was first mixed with Ni and Cr powders in a weight ratio of 1:12:12. These materials were then loaded into an alumina crucible and heated to a molten state with a flowing mixture of nitrogen (95%) and hydrogen (5%) at 1550°C. The h₁⁻¹BN crystals were precipitated on the flux surface by slowly cooling (1°C/hour) to 1500°C. After the growth process, the mixture temperature was quickly quenched to room temperature. The crystal flakes were obtained from the solidified flux with tape.

**Symmetrization of Fourier space images**

Near-field images acquired near the apex of the gold launcher were cropped to remove the gold launcher. The cropped near-field images were subsequently Fourier-transformed, using a Hann window, to get the raw image in reciprocal space. To enhance the signal-to-noise ratio and better reveal the excited polaritonic modes, we applied all the symmetry operations of the hexagonal lattice to the raw Fourier image and averaged the resulting images. More specifically, we consecutively rotated the raw Fourier image in 60° intervals, leading to six transformed images. We then applied the mirror operation to these six transformed images, resulting in six additional images. The average of all 12 transformed images results in the Fourier images shown in Fig. 3. The Fourier transform method is commonly used to analyze polaritonic equi-energy contours (57–60).
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Acknowledgments

**Funding:** Experimental research on Bloch polaritons is solely supported by the Energy Frontier Research Center on Programmable Quantum Materials funded by the US Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under award DE-SC0019443. The development of infrared near-field imaging capabilities at Columbia was supported through the Vannevar Bush Faculty Fellow ONR-VB: N00014-19-1-2630 (D.N.B). D.N.B is a Moore Investigator in Quantum Materials EPIQS #9455. The monoisotopic hBN crystal growth (J.H.E) was supported by the National Science Foundation (NSF) award CMMI #1538127. Theory and modeling results obtained by G.S. and M.J. were supported by the Office of Naval Research (ONR) under grant N00014-21-1-2056, and by NSF under grants No. DMR-1741788 and DMR-1719875. M.J. also acknowledges support by the fellowship from Kwanjeong Educational Foundation. Theory and modeling results obtained by M.F. were supported by the ONR grant N00014-18-1-2722. **Author contributions:** L.X., A.S.M., and Y.D. performed the nanoscale infrared measurements and characterizations. Y.L., C.F., S.Z., S.L., K.W., T.T., J.H.E., and C.R.D. designed and created the device structures. L.X., M.J., F.L.R., M.M.F., and G.S. provided theoretical calculations. D.N.B. supervised the project. L.X. and C.L. analyzed the data. L.X., Y.L., M.J., and D.N.B. cowrote the manuscript with input from all coauthors. **Competing interests:** The authors declare that they have no competing interests. **Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Raw experimental data can be found in the Dryad repository (61).

Submitted 17 September 2020
Accepted 19 March 2021
Published 7 May 2021

10.1126/sciadv.abe8087

**Citation:** L. Xiong, Y. Li, M. Jung, C. Forsythe, S. Zhang, A. S. McLeod, Y. Dong, S. Liu, F. L. Ruta, C. Li, K. Watanabe, T. Taniguchi, M. M. Fogler, J. H. Edgar, G. Shvets, C. R. Dean, D. N. Basov, Programmable Bloch polaritons in graphene. *Sci. Adv.* **7**, eabe8087 (2021).