

Infrared nano-imaging of Dirac magnetoexcitons in graphene

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Michael Dapolito  ^{1,2}, Makoto Tsuneto¹, Wenjun Zheng  ¹, Lukas Wehmeier  ^{1,3}, Suheng Xu², Xinzhang Chen  ^{1,2}, Jiacheng Sun¹, Zengyi Du¹, Yinming Shao  ², Ran Jing  ², Shuai Zhang  ², Adrien Bercher⁴, Yinan Dong  ², Dorri Halbertal  ², Vibhu Ravindran^{1,5}, Zijian Zhou¹, Mila Petrovic  ¹, Adrian Gozar  ^{6,7}, G. L. Carr  ³, Qiang Li^{1,8}, Alexey B. Kuzmenko  ⁴, Michael M. Fogler⁹, D. N. Basov  ²✉, Xu Du  ¹✉ & Mengkun Liu  ^{1,3}✉

Magnetic fields can have profound effects on the motion of electrons in quantum materials. Two-dimensional electron systems subject to strong magnetic fields are expected to exhibit quantized Hall conductivity, chiral edge currents and distinctive collective modes referred to as magnetoplasmons and magnetoexcitons. Generating these propagating collective modes in charge-neutral samples and imaging them at their native nanometre length scales have thus far been experimentally elusive. Here we visualize propagating magnetoexciton polaritons at their native length scales and report their magnetic-field-tunable dispersion in near-charge-neutral graphene. Imaging these collective modes and their associated nano-electro-optical responses allows us to identify polariton-modulated optical and photo-thermal electric effects at the sample edges, which are the most pronounced near charge neutrality. Our work is enabled by innovations in cryogenic near-field optical microscopy techniques that allow for the nano-imaging of the near-field responses of two-dimensional materials under magnetic fields up to 7 T. This nano-magneto-optics approach allows us to explore and manipulate magnetopolaritons in specimens with low carrier doping via harnessing high magnetic fields.

When a magnetic field is applied to a material with free charge carriers (electrons or holes), the orbital motion of the charge carriers is affected by the formation of quantized Landau levels (LLs). This Landau quantization leads to a plethora of physical phenomena, most famously the quantized Hall effect in the d.c. transport of two-dimensional systems. In optical spectroscopy, optical transitions between LLs are observed

when the field-controlled cyclotron energy $\hbar\omega_c$, where ω_c is the cyclotron frequency defined below, exceeds the LL smearing due to thermal excitation or scattering processes. The cyclotron frequency is inversely proportional to the effective cyclotron mass of the electron. As such, the largest cyclotron energy gap at a given magnetic field is apparently attained in quantum materials with a linear energy-momentum

¹Department of Physics and Astronomy, Stony Brook University, Stony Brook, NY, USA. ²Department of Physics, Columbia University, New York, NY, USA.

³National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, NY, USA. ⁴Département de Physique de la Matière Quantique,

Université de Genève, Genève 4, Switzerland. ⁵Department of Physics, University of California, Berkeley, CA, USA. ⁶Department of Physics,

Yale University, New Haven, CT, USA. ⁷Energy Sciences Institute, Yale University, West Haven, CT, USA. ⁸Condensed Matter Physics and Materials

Science Division, Brookhaven National Laboratory, Upton, NY, USA. ⁹Department of Physics, University of California at San Diego, La Jolla, CA, USA.

✉e-mail: db3056@columbia.edu; xu.du@stonybrook.edu; mengkun.liu@stonybrook.edu

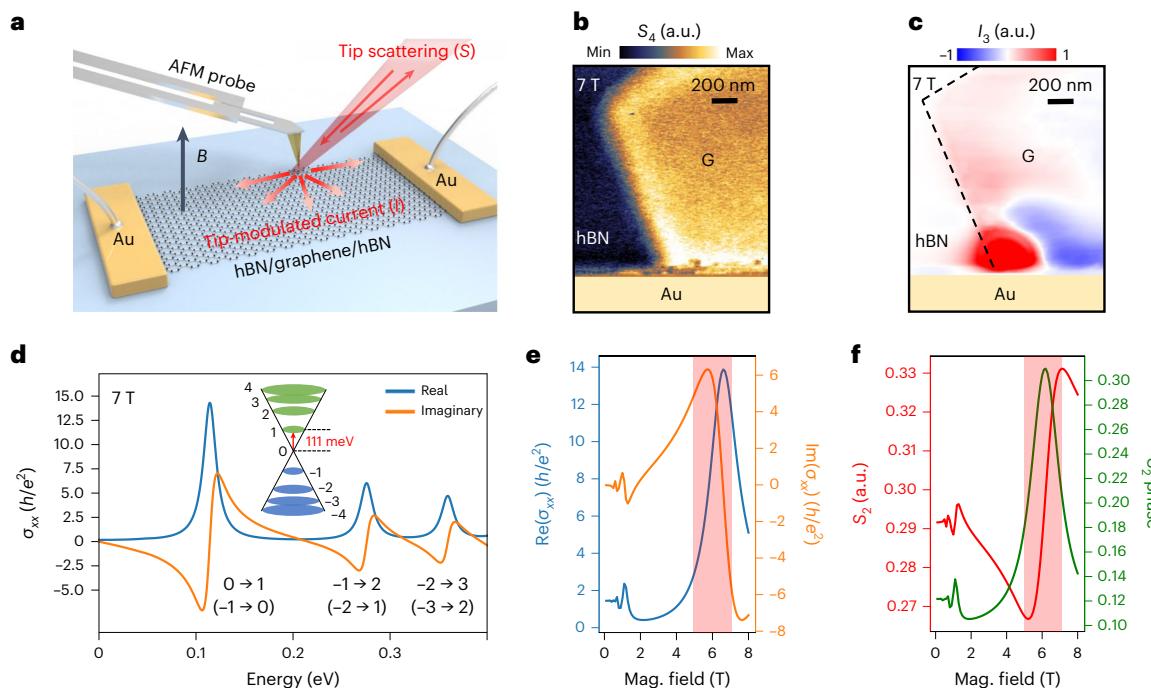


Fig. 1 | Magnetic field-dependent m-SNOM and near-field photocurrent DiME measurements of hBN-encapsulated graphene at 200 K. **a**, Schematic of the experimental setup. The near-field scattering amplitude (S) and photocurrent (I) can be measured simultaneously. **b**, m-SNOM image (S_4) of the hBN-graphene boundary. Graphene is labelled with G . **c**, m-SNOM near-field photocurrent images (I_3) of graphene, taken simultaneously with **b**. The graphene edge is marked with a dashed line. **d**, Calculated real ($\text{Re}(\sigma_{xx}) \equiv \sigma_1$, blue) and imaginary ($\text{Im}(\sigma_{xx}) \equiv \sigma_2$, orange) optical conductivities of graphene as functions of energy

at $B = 7.0$ T. Parameters: $I = 8.0$ meV, $v_F = 1.19 \times 10^6$ m s $^{-1}$ and $T = 200$ K. The inset depicts the LLs superimposed on the Dirac cone of the quasiparticle dispersion in graphene. **e,f**, Calculated optical conductivity (**e**) and S_2 amplitude (**f**) as functions of the magnetic field at photon energy 111 meV (or 900 cm $^{-1}$) with the same parameter values used in **d**. The pink-shaded region indicates where the DiMEs are visible in our near-field imaging experiments. a.u. arbitrary units; mag., magnetic; max, maximum; min, minimum.

dispersion and large Fermi velocity, including graphene, as the electrons behave as massless Dirac fermions. Thus, graphene has emerged as an exemplary material for magneto-optics studies^{1–5}. It exhibits the quantum Hall effect and sharp cyclotron resonances at room temperature in moderately high fields^{6–8}.

The applied magnetic field also alters the collective modes of electrons and holes in graphene. A unifying term for such modes is magnetoexcitons. However, their real-space features have been experimentally elusive thus far. We remark here that magnetoexcitons originating from optical transitions between adjacent LLs are also commonly referred to as magnetoplasmons^{9,10}. Previous experimental work examined how plasmon eigenmodes of graphene nanostructures (for example, discs and ribbons) evolve into magnetoplasmons as the magnetic field increases. Two types of excitations have been uncovered, namely surface and edge magnetoplasmons that appear above and below the cyclotron gap, respectively^{11–14}. These prior studies were carried out exclusively on doped graphene, which behaves like conventional doped semiconductors. In this work, however, we focus on graphene near the charge-neutrality point (CNP), where the charge response is qualitatively different. For example, at the CNP, the Hall conductivity σ_{xy} vanishes due to electron–hole symmetry^{4,15}. To describe these modes in graphene near the CNP, we introduce the terms Dirac magnetoplasmons and Dirac magnetoexcitons (DiMEs) to emphasize the important role of the linear Dirac energy–momentum electronic dispersion near the CNP. Precisely at the CNP, ‘DiME’ appears to be the more appropriate term and we use it below. The DiMEs in charge-neutral graphene have been discussed theoretically^{10,15,16} but have remained experimentally unexplored at infrared frequencies, as probing them with conventional diffraction-limited optics is challenging. We accomplished this task by harnessing near-field nano-infrared

techniques that involve the confinement of light to nanoscale dimensions matching the momentum and wavelength of the collective mode of interest^{17,18}. Here we report a visualization of the DiMEs and a previously unreported mapping of the DiME dispersion at infrared frequencies using a home-built cryogenic magneto scanning near-field optical microscope (m-SNOM)¹⁹. By coupling light resonantly to the inter-LL transitions, we observed magnetic-field-tunable DiME signatures in the scattered m-SNOM signal and an accompanying photocurrent modulation at the edges of graphene.

Our technique is based on scattering-type scanning near-field optical microscopy (s-SNOM), which has made substantial progress over the last decade and led to numerous experimental achievements including polariton sensing^{20–23}, characterization of phase transitions^{24–27} and nanoscale photocurrent detection^{28–31}. m-SNOM brings forth new opportunities to explore quantum materials with broken time-reversal symmetry using nano-optics. Like traditional s-SNOM, the spatial resolution of m-SNOM is not subject to the diffraction limit of light. This is especially important at photon energies below 1 eV, where magneto-optical phenomena can now be studied on length scales as short as a few tens of nanometres. As shown below, this enables us to directly visualize the DiMEs and the associated edge-dominated d.c. photocurrent.

As in traditional s-SNOM, the key element in m-SNOM is a sharp metalized tip that concentrates incident light onto a sample, as shown schematically in Fig. 1a. The confined light interacts with the sample and is scattered back into the far field. The oscillating tip causes time-periodic oscillations of the total scattering amplitude S . The scattered signal is demodulated into its corresponding Fourier harmonics S_j of sufficiently high orders, $j = 3, 4$, to represent the desired near-field signals. Under common simplifying assumptions (Supplementary Note 1), one can quantitatively relate these ‘demodulated’ amplitudes S_j to

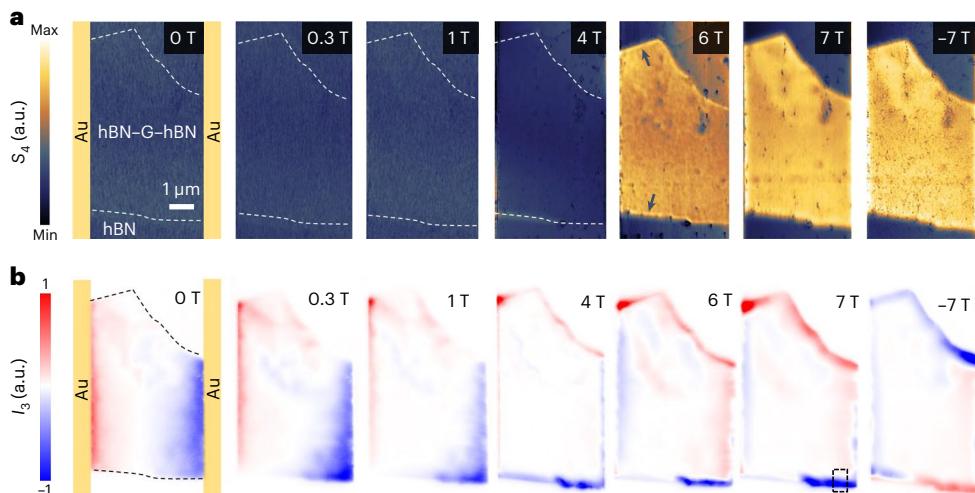


Fig. 2 | Nano-infrared images of near-charge-neutral graphene in the regime of the field-tuned $0 \rightarrow 1\text{LL}$ transition. **a**, m-SNOM images. **b**, Near-field photocurrent images. The incident light has a wavelength of $11.2\text{ }\mu\text{m}$ (111 meV or

$~900\text{ cm}^{-1}$) and the sample was kept at 200 K . The box in the bottom right corner of the sample at 7 T (**b**) shows the region we image in Figs. 3 and 4. The white dashed lines in **a** outline the boundaries of the graphene.

the optical reflection coefficient $r_p = r_p(q, \omega)$ of the sample, which is a function of the incident photon frequency ω and in-plane momentum q . In general, a larger r_p produces a larger S_4 . The s-SNOM signal is strongly enhanced when ω is resonant with collective modes of the system, which give rise to the poles of $r_p(q, \omega)$. Additionally, the collective modes produce spatially periodic interference fringes along the sample edges in the s-SNOM images, as discussed below.

Resonant excitation of Dirac magnetoexcitons

To study DiMEs in real space, we use monolayer graphene encapsulated between two thin crystals of hexagonal boron nitride (hBN) with thicknesses of 5 nm (top) and 28 nm (bottom). The data presented below were taken at temperature $T = 200\text{ K}$, although similar phenomena have been observed in a wider range of temperatures from 70 to 300 K . Using electrodes fabricated on the two side edges of an hBN-graphene-hBN heterostructure, we performed scanning near-field photocurrent microscopy simultaneously with the m-SNOM measurement²⁹. Representative images of these measurements are shown in Fig. 1b (S_4) and Fig. 1c (I_3), the photocurrent demodulated at the $j = 3$ harmonic of the tip tapping frequency).

Electrons in graphene near the CNP are treated as Dirac fermions with LL energies that scale as $E_{\text{CNP}} + \text{sgn}(n)\sqrt{|n|}\hbar\omega_c$, where n is the LL index, E_{CNP} is the Fermi energy at the CNP and \hbar is the reduced Planck constant. Here, $\hbar\omega_c = \sqrt{2\hbar v_F/l_B}$ is the gap between the $n = 0$ and $n = 1$ LLs, where $l_B = \sqrt{\hbar/e|\mathbf{B}|}$ is the magnetic length, v_F is the graphene Fermi velocity and \mathbf{B} is the magnetic field. At the CNP, all $n < 0$ LLs are filled, all $n > 0$ LLs are empty and the $n = 0$ level is half filled. The allowed inter-LL optical transitions are $-|n| \rightarrow |n| + 1$ and $-|n| - 1 \rightarrow |n|$, yielding resonances of the complex optical conductivity $\sigma_{xx}(\omega) = \sigma_1 + i\sigma_2$ at energies $\Delta E = \hbar\omega = (\sqrt{|n|} + \sqrt{|n| + 1})\hbar\omega_c$. The calculated real and imaginary conductivities as a function of the photon energy are plotted in Fig. 1d. Here we assume a phenomenological momentum dissipation rate of $\Gamma = 8.0\text{ meV}$. This dissipation rate was chosen to best match the experimental magnetic-field-dependent near-field contrast and DiME dispersion of Figs. 2 and 4a, respectively. In principle, Γ will vary for different LL transitions³². However, since here we report only the $0 \rightarrow 1$ transition, we kept the dissipation rate constant for these calculations. At 7 T , the energy of the incident light in our experiment, 111 meV , is close to the principal cyclotron resonance of a bare graphene sample at $\hbar\omega_c \approx 114\text{ meV}$. In Fig. 1e, where σ_{xx} is plotted versus B , weaker resonances due to higher- n transitions at fields $B \simeq 1\text{ T}$ may also be

discerned. Many-body effects, which were neglected in these calculations, cause B -dependent corrections to v_F (and, therefore, ω_c) that scale with the dimensionless coupling parameter $g = e^2/h\hbar v_F \approx 1$ (refs. 33–35). Here $\kappa = \kappa_1 + i\kappa_2$ is the effective dielectric permittivity of the graphene surroundings. To account for these sample-dependent corrections, we treat v_F as an adjustable parameter. Shown in Fig. 1f, the m-SNOM signal S_2 is computed using a lightning rod model (Supplementary Note 1) with σ_{xx} as an input. The results shown in Fig. 1e,f indicate that the B -field dependence of the near-field signal faithfully reproduces the field-tunable effects rooted in cyclotron resonance physics and the attendant collective modes.

The collective mode dispersions of graphene can be derived from the magnetic-field-dependent σ_{xx} , which determines the near-field reflection coefficient $r_p(q, \omega)$ of the system. As we are measuring near-charge-neutral graphene, the off-diagonal conductivity components are omitted in our calculation. The mode dispersions are defined by the poles of $r_p(q, \omega)$ at complex $q = q_1 + iq_2$, which are distinct from highly doped graphene. The mode wavelength is $\lambda = 2\pi/q_1$ and an analytical approximation for λ of the DiMEs associated with the $0 \rightarrow 1$ LL transition is¹⁵

$$\lambda(\omega) \simeq \frac{g(\omega)}{2\pi} \frac{v_F}{(\omega - \omega_c)}, \quad (1)$$

where g is the frequency-dependent coupling parameter defined above. This equation, valid for $\lambda \gg l_B$ and $\omega \rightarrow \omega_c$, implies that the group velocity of the DiMEs is $\lesssim v_F$. In contrast, the group velocity of plasmons in doped graphene at $B = 0$ is typically much higher than v_F . Equation (1) also suggests that measuring the DiME wavelength $\lambda(\omega)$ directly probes the Fermi velocity v_F . This is in stark contrast with the plasmon polaritons in highly doped graphene, whose wavelength depends on the carrier density. The mode damping is characterized by the quality factor $Q = q_1/q_2 \approx (\sigma_1/\sigma_2 + \kappa_2/\kappa_1)^{-1}$ (refs. 11,12). Assuming that σ_1/σ_2 is the dominant quantity in this expression, Fig. 1d,e indicates that $Q > 1$ starting from frequencies exceeding ω_c by an amount $-\Gamma$. This is the lower bound for the range of ω where propagating DiMEs exist. The term κ_2/κ_1 plays an important role when either σ_1 or σ_2 is close to zero. A theoretical upper bound of ω where DiMEs are observable with m-SNOM is set by the condition $\lambda \simeq a$. Here $a \simeq 15\text{ nm}$ is the radius of curvature of the tip, which defines the spatial resolution of m-SNOM. In comparison, $l_B \approx 10\text{ nm}$ at $B = 6\text{ T}$.

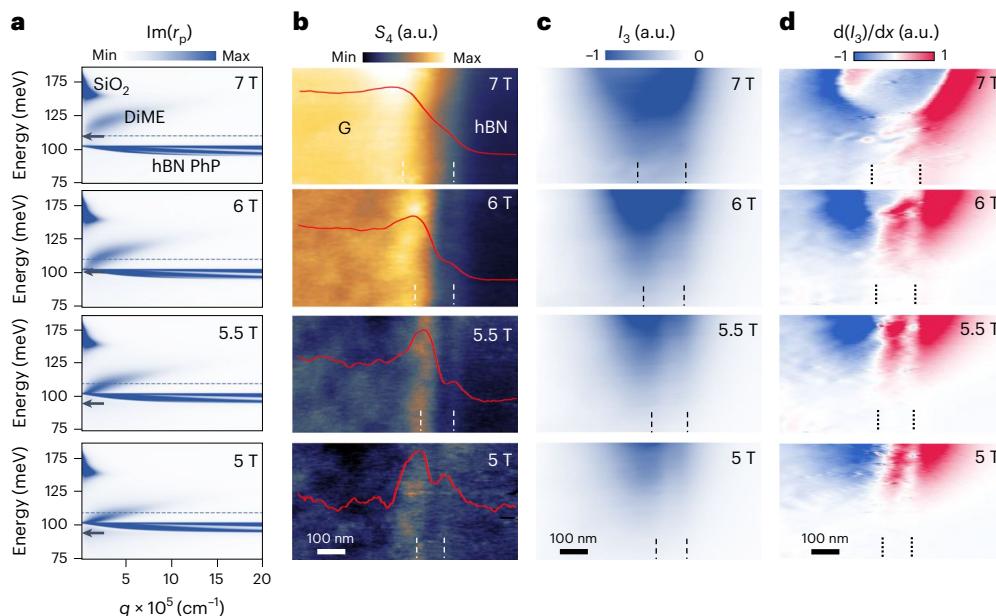


Fig. 3 | Magnetic field tuning of DiMEs. **a**, Calculated imaginary part of the reflection coefficient r_p illustrating the field-dependent dispersion of the collective modes in the system. The dashed lines denote our incident light frequency of $\sim 900 \text{ cm}^{-1}$. The broadened curve intersecting the dashed lines is the DiME dispersion. The black arrows indicate the principal cyclotron resonances ($0 \rightarrow 1\text{LL}$ transition) at each magnetic field. Parameters: $\Gamma = 8.0 \text{ meV}$, $v_F = 1.19 \times 10^6 \text{ m s}^{-1}$ and $T = 200 \text{ K}$. **b,c**, Field-dependent m-SNOM image (**b**) and

near-field photocurrent image (**c**) at the graphene–hBN boundary. The red curves in **b** are the averaged line profiles of the near-field signal (S_4). **d**, Derivative of the photocurrent signal in **c** highlighting the modulation of the edge photocurrent by the DiME. The region where the data was taken is marked by the black rectangle in Fig. **2b** (note the scan area is rotated by 90° here). The dotted and dashed lines in **b**, **c** and **d** label the observed interference fringes as guides for the eyes. PhP, phonon polariton.

These unique collective modes of graphene can be directly imaged as a function of the magnetic field B , as shown in the m-SNOM images (S_4) of Fig. **2a**. For $B \leq 4 \text{ T}$, there is little contrast between hBN and graphene, indicating that the coupling of light to the electronic system of graphene is weak. At 6 T , the contrast between hBN and graphene becomes clearly visible and the real-space signature of propagating plasmons, namely an interference fringe, along the edge of graphene (the black arrows in the 6 T panel; also see Fig. **3b**) becomes evident. At our largest field of $B = \pm 7 \text{ T}$, the S_4 signal is strongly enhanced over the entire graphene sheet. Since hBN is an insulator and inert to a magnetic field, the observed evolution of the contrast must be attributed to graphene. Indeed, the observed changes in S_4 can be explained by an increased scattering of light due to the $0 \rightarrow 1\text{LL}$ transitions in graphene. As the B field grows, first the high-frequency tail and then the central part of this resonance become accessible at the frequency of our incident light. This interpretation is corroborated by the calculations reported in Supplementary Note 1.

Another perspective of the DiME signatures can be obtained using complementary information on the magneto-optical effects provided by imaging a nano-photocurrent with the same m-SNOM instrument. The tip-induced part of the photocurrent is captured by the third-order demodulated photocurrent signal I_3 (Fig. **2b**). As asymmetry is required for local net photocurrent generation, the detected I_3 is almost negligible away from the gold electrical contacts (labelled by ‘Au’ in Fig. **2b**) and graphene edges (dashed lines in Fig. **2b**), where the graphene is largely homogeneous and, thus, possesses inversion symmetry. At the contacted and contact-free edges, the inversion symmetry is broken, leading to a finite photocurrent²⁹. The photocurrent traces I_3 display a strong dependence on the field. Without the magnetic field, I_3 is uniform along the contacts. As the magnetic field increases until 4 T , shrinking photocurrent ‘hot spots’ at the opposite corners of the sample appear (top left and bottom right in Fig. **2b**). Assuming that the photocurrent is due to the photo-thermoelectric effect^{29,36} and invoking the Shockley–Ramo formalism^{37–39}, we surmise that these hot

spots mark the locations where an enhanced current density would exist in a d.c. transport experiment. Current crowding at the corners of the sample is known to occur when the d.c. Hall conductivity σ_{xy} becomes comparable to or exceeds the diagonal d.c. conductivity σ_{xx} (Supplementary Note 3). Such a finite σ_{xy} can arise in our sample due to the residual doping of graphene.

At higher fields ($B \geq 6 \text{ T}$), we observed a strong photocurrent extending along the contact-free edges of graphene. This photocurrent reverses its sign when the magnetic field is flipped from $+B$ to $-B$. We attribute this edge photocurrent to a DiME-enhanced photo-Nernst effect^{40,41}, as detailed in Supplementary Note 3. The DiME enhancement of the photocurrent was confirmed by applying inter-LL resonant and non-resonant light and comparing the resulting photocurrent signals. For the non-resonant light (wavelength $6.59 \text{ }\mu\text{m}$ and photon energy 188 meV), the photocurrent was reduced by more than an order of magnitude compared to what is seen in Fig. **2b**.

Visualizing and mapping the DiME dispersion

The images in Fig. **2** contain rich insights into how magnetic-field-induced collective modes alter the real-space optical and optoelectronic properties of charge-neutral graphene. By zooming in on the graphene boundary, we can analyse the behaviour of the excitonic fringes and study the dispersion of the collective modes. Theoretically, one can visualize the collective mode dispersion by plotting the numerically computed $\text{Im}(r_p)$ as a function of real q and ω . From such a plot (Fig. **3a**), we find three classes of modes in our system: (1) waveguide phonon polaritons of hBN encapsulating layers at $92 \text{ meV} < \hbar\omega < 103 \text{ meV}$, (2) the surface phonons of the SiO_2 substrate at $\hbar\omega = 146 \text{ meV}$ and (3) the DiMEs of graphene residing between (1) and (2). The DiME dispersion curve exhibits flattening and/or avoided crossings with the other modes. This behaviour originates from a decrease of parameter $g(\omega) \propto \kappa^{-1}(\omega)$ near the phonon resonances where the effective permittivity $\kappa(\omega)$ diverges. The momentum q where the DiME dispersion intersects our laser frequency (blue dashed line in Fig. **3a**) decreases

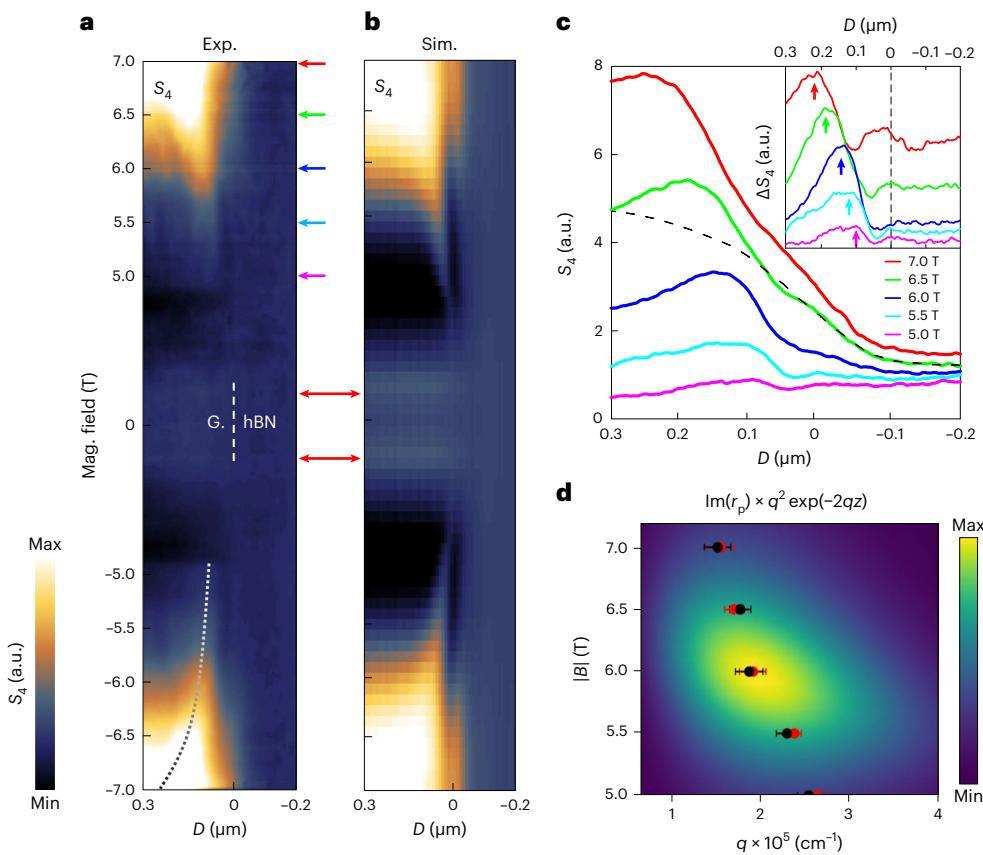


Fig. 4 | Magnetic field-dependent DiMEs dispersion. **a,b**, Measured (a) and simulated (b) m-SNOM line scans at the graphene–hBN boundary while sweeping the magnetic field from 7 T to −7 T. The line scans clearly show that the DiME appears between ± 5 and ± 7 T and disappears for magnetic fields between 0 and ± 5 T. Note that the sweep of the magnetic field is reduced in speed between ± 5 and ± 7 T. The simulation uses the same parameter values as in Fig. 3. The double-headed red arrows indicate the faint signature of the $-1 \rightarrow 2$ ($-2 \rightarrow 1$) transition. The dashed curve in a is a guide for the eye. **c**, Line cuts from a showing the change in DiME wavelength from 7 to 5 T. The black dashed line shows one example of the integrated Lorentz-shaped background subtracted from the line

profile of the s-SNOM signal measured at 6.5 T. The inset shows the extracted oscillatory part of the signal, from which we extract the spacing d between fringes. **d**, Colour map showing the magnetic-field-dependent $\text{Im}(r_p)$ multiplied by the tip model response function $q^2 \exp(-2qz)$, where $z \approx 50$ nm is the average tip-sample distance³⁶. Black and red dots are the measured DiME wavevector $q = 2\pi/\lambda = 2\pi/2d$ at positive and negative fields, respectively. Data points are presented as the optimal wavevector plus or minus the standard deviation determined from the fitting of the peak positions in the inset of c. Exp., experimental; G., graphene; Mag., magnetic; Sim., simulated.

as the magnetic field increases, yielding a correspondingly larger λ at higher fields.

As shown in Fig. 3b, the fringes, spaced by $d = \lambda/2$, are caused by interference of tip-launched DiME polariton waves with their reflections off the sample boundaries⁴². The fringes become evident at magnetic fields $B \gtrsim 5$ T with the spacing increasing from ~ 100 nm at ± 5 T to ~ 200 nm at ± 7 T, in agreement with the rough estimates of λ we extracted from the DiME dispersion curve in Fig. 3a. Fringe-like features are also present in the photocurrent images (Fig. 3c,d). This is because oscillations of the DiME electric field intensity modulate the local thermal energy dissipation, which in turn leads to oscillations in the near-field photo-thermal electric current⁴³. In general, we found that the photocurrent signal extends into the interior of the sample over a slightly longer length scale compared to the DiME fringes. This length scale, referred to as the cooling length, is determined by the interplay of the thermal conductivity of graphene and the thermal resistance of the graphene–hBN interface²². Note that in addition to the tip, the collective modes in graphene can be launched by any sharp boundaries or metallic structures, for example, electrical contacts. This gives rise to λ -periodic rather than $\lambda/2$ -periodic fringes. At the top of the images in Fig. 3b–d there is an Au contact oriented horizontally (right side of the box in Fig. 2b (7 T)). Although the photocurrent is enhanced near this contact, we do not observe contact-launched fringes there. This is

possibly due to the contact-doping effect causing graphene to become doped away from charge neutrality in the vicinity of the electrodes⁴⁴.

To quantify the field-dependent DiME dispersion, we took line scans across the graphene–hBN boundary while sweeping the magnetic field from 7 to −7 T. As shown in Fig. 4a, the onset of the fringes occurs at around ± 5 T. The curvature of the interference fringes (dashed trace in Fig. 4a) suggests that there are longer wavelengths (lower q) at higher fields. We used a commercial finite-element solver to simulate the DiME interference fringes with the same parameter values as in Fig. 3a⁴⁵. The simulation results, depicted in Fig. 4b, qualitatively agree with our experimental observations in Fig. 4a. This provides clear evidence that the observed changes in the edge fringes originate from the field-dependent DiME dispersion. Note that a slight brightening of the s-SNOM signal at around 1.1 T can also be seen in Fig. 4a,b (double-headed red arrows). We attribute this to the weaker $-1 \rightarrow 2$ ($-2 \rightarrow 1$) LL transition (Fig. 1d). With an improved signal-to-noise ratio, future experiments can explore this and other higher order LL transitions. In addition, potential modifications to the DiME dispersion due to the bending of the LLs caused by quantum Hall edge states can also be examined, preferably at lower temperatures.

Shown in Fig. 4c are line cuts of the s-SNOM signal taken along the horizontals corresponding to the coloured arrows in Fig. 4a. These are used to determine λ quantitatively. By fitting the derivative of these

line cuts to an asymmetric Lorentz function⁴⁶, the background can be subtracted to reveal readily apparent fringes, as shown in the inset of Fig. 4c. The estimates of λ obtained from this fitting were converted to the wavevector $q = 2\pi/\lambda$ and are shown as dots in Fig. 4d. The DiME wavelength increases monotonically from ~240 nm at 5 T to ~400 nm at 7 T, in good agreement with the theoretical predictions represented by the colour map in Fig. 4d.

Conclusions

The propagation of DiMEs results in the formation of standing wave fringes and modulated photocurrent pathways at the sample edges. These can be observed only by using sub-diffractive imaging systems coupled to high magnetic fields. The DiME dispersion is extremely responsive to magnetic fields as it originates from inter-LL transitions. Consequently, the collective mode energies are expected to be adjustable across a wide frequency range from microwave and terahertz (millielectronvolts) to infrared and visible (electronvolts), providing an extensive scope for exploration in future research using m-SNOM. Given that the DiME exists at charge neutrality, it can be used to study many-body effects rooted in electron-electron interactions or hydrodynamic physics under the influence of a magnetic field⁴⁷. Chiral polaritons⁴⁸, proximity effects⁴⁹ or possibly strong mode coupling⁵⁰ can be investigated with DiME-based systems. Additionally, the frequency-dependent absorption edge of the DiMEs is highly sensitive to the LL filling and scattering rate. This can be useful for studying the flat band physics in graphene and other low-dimensional systems (for example, moiré modulated ν_F) subject to magnetic fields^{51,52}. Thus, the m-SNOM technique developed for this work is the first step toward the study of the magneto-optical properties of quantum materials at the nanoscale. In combination with terahertz near-field microscopy or terahertz near-field emission spectroscopy, m-SNOM can also be performed with various types of Dirac and Weyl semimetals^{23,30}, in which a magnetic field is predicted to induce chiral anomalies, along with other intriguing phenomena such as non-reciprocal polaritons, multi-photonic bands, and frequency and polarization conversion^{11,12,53–55}.

Online content

Any methods, additional references, Nature Portfolio reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at <https://doi.org/10.1038/s41565-023-01488-y>.

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Methods

Closed-cycle m-SNOM

Infrared nano-imaging capabilities at cryogenic temperatures and under magnetic fields up to 7 T were demonstrated using a closed-cycle cryostat (Opticool, Quantum Design) in Mengkun Liu's lab at Stony Brook University. The scanning stages and positioners were from Attocube (ANSxyz100/LT/UHV and ANPxxyz101/LT/UHV, respectively). The mechanical noise in s-SNOM was less than 0.5 nm in the z direction and less than 20 nm in the xy direction (resolution limited) at 100 K. Our atomic force microscope (AFM) has a spatial resolution of about 15 nm and a scan area of $50 \times 50 \mu\text{m}^2$ at room temperature. The accessible temperature range of our home-built scanning probe system is -10–350 K. A CO₂ laser centred at -11 μm and a quantum cascade laser centred at 6.4 μm , both operating in continuous wave operation at room temperature, were used for the photocurrent and s-SNOM imaging. The laser power before entering the cryostat was around 12 mW and was reduced to below 10 mW at the tip due to reflections from the cryostat window. The laser was focused onto the tip with a spot size of approximately 20 μm . We used a self-homodyne detection scheme^{18,57}, which yielded stable detection of the scattered signal and good signal contrast. A schematic of the m-SNOM and cryostat is shown in Extended Data Fig. 1.

Device fabrication

All samples used in this work were hBN-encapsulated monolayer graphene field-effect devices, fabricated through the standard dry-transfer⁵⁸ and nanolithography techniques. The top and bottom hBN and monolayer graphene crystals were first mechanically exfoliated onto SiO₂(285 nm)/Si substrates. A polydimethylsiloxane/poly-carbonate (PC) stamp was then used to pick up the top hBN at -120 °C, followed by graphene at -100 °C and finally the bottom hBN at -120 °C. The assembled hBN-graphene-hBN stack was then released onto a clean SiO₂(285 nm)/Si substrate by melting the PC at -180 °C. After removing the bulk part of the PC in chloroform, the transferred stack was annealed in forming gas (5% H₂/95% Ar) at 350 °C for 2 h to further remove the PC residue. Standard electron beam lithography was used to define the source and drain electrodes using an A4 950PMMA resist. The exposed pattern was developed in a water/isopropyl alcohol (1:3) solution at a temperature of 5 °C for 2 min. The top hBN and graphene at the contact area were then etched in CHF₃/O₂(10:1) plasma (40 mTorr and 60 W), exposing the side of the graphene layer. Cr(5 nm)/Au(40 nm) was then thermally evaporated to form side contacts for the graphene⁵⁹.

Reporting summary

Further information on the research design is available in the Nature Portfolio Reporting Summary linked to this article.

Data availability

The data represented in Figs. 1–4 are available as Source Data 1–4. All other data that support the findings of this study are available from the corresponding authors upon reasonable request.

Code availability

All codes underlying this study are available from the corresponding authors upon reasonable request.

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Author contributions

M.D., M.T., X.C., L.W., W.Z., D.N.B. and M.K.L. designed the experiment. M.D., M.T., L.W., S.Z., X.D. and M.K.L. performed the measurements. X.D. and J.S. fabricated the samples. M.D., M.T., Z.D., X.C., L.W., W.Z., S.X., Y.D., V.R., M.P., Z.Z., A.B., G.L.C., Q.L., A.B.K., A.G., X.D., M.M.F. and M.K.L. analysed the data. M.T., X.C., W.Z., S.X., Y.S., R.J. and D.H. performed the simulations. M.D. and M.K.L. wrote the paper. All authors contributed to the scientific discussions and paper revisions.

Competing interests

M.D., X.C., M.K.L. and A.G. have a patent pending related to the design of the m-SNOM. The other authors declare no competing interests.

Additional information

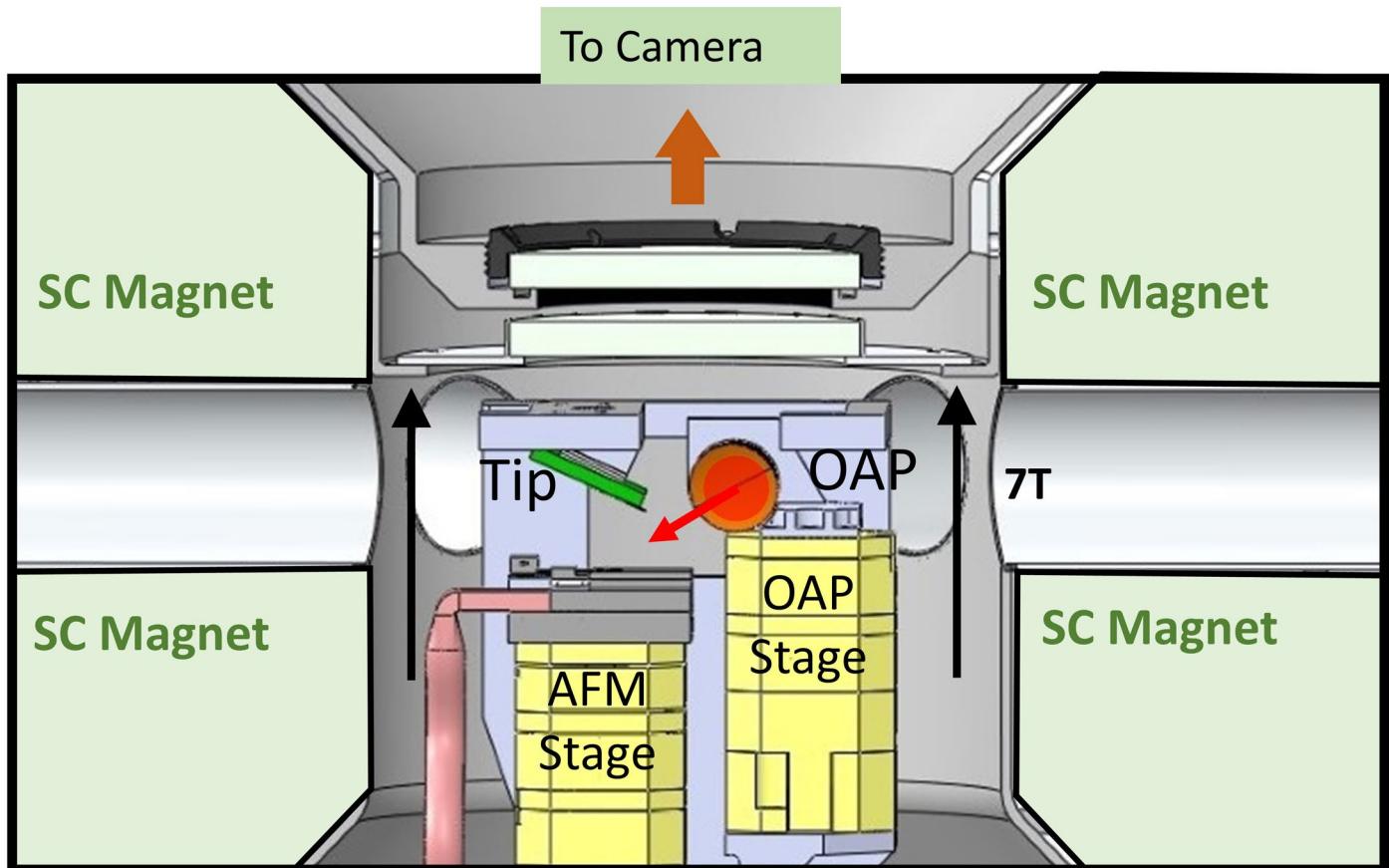
Extended data Extended data are available for this paper at <https://doi.org/10.1038/s41565-023-01488-y>.

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1038/s41565-023-01488-y>.

Correspondence and requests for materials should be addressed to D. N. Basov, Xu Du or Mengkun Liu.

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Extended Data Fig. 1 | Closed-cycle m-SNOM setup. OAP: off-axis parabolic mirror. Tip: Akiyama probe. Light enters the chamber in the horizontal direction (into the field of view) and is focused onto the sample with an OAP.

Lasing Reporting Summary

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Experimental design

Please check: are the following details reported in the manuscript?

1. Threshold

Plots of device output power versus pump power over a wide range of values indicating a clear threshold

 Yes*State where this information can be found in the text.* No*Not relevant to the experiment.*

2. Linewidth narrowing

Plots of spectral power density for the emission at pump powers below, around, and above the lasing threshold, indicating a clear linewidth narrowing at threshold

 Yes*State where this information can be found in the text.* No*Not relevant to the experiment.*

3. Coherent emission

Measurements of the coherence and/or polarization of the emission

 Yes*State where this information can be found in the text.* No*Can be obtained from Access Laser.*

4. Beam spatial profile

Image and/or measurement of the spatial shape and profile of the emission, showing a well-defined beam above threshold

 Yes*State where this information can be found in the text.* No*Not necessary to reproduce experiment.*

5. Operating conditions

Description of the laser and pumping conditions

Continuous-wave, pulsed, temperature of operation

 Yes*Methods section* No*Explain why this information is not reported/not relevant.* Yes*State where this information can be found in the text.* No*Not necessary to reproduce experiment.*

6. Alternative explanations

Reasoning as to why alternative explanations have been ruled out as responsible for the emission characteristics
e.g. amplified spontaneous, directional scattering; modification of fluorescence spectrum by the cavity

 Yes*State where this information can be found in the text.* No*Not relevant to the experiment.*

7. Theoretical analysis

Theoretical analysis that ensures that the experimental values measured are realistic and reasonable
e.g. laser threshold, linewidth, cavity gain-loss, efficiency

 Yes*State where this information can be found in the text.* No*Not relevant to the experiment.*

8. Statistics

Number of devices fabricated and tested

 Yes*State where this information can be found in the text.* No*Not relevant to the experiment.*

Statistical analysis of the device performance and lifetime (time to failure)

 Yes*State where this information can be found in the text.* No*Can be obtained from Access Laser.*

Further reading

We also suggest that authors read the following literature, which describes the important principles and signatures of laser emission and discusses some of the common mistakes that can occur during laser characterization.

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