PHYSICAL SCIENCE

Quantum imaging of current flow in graphene

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Since its first discovery in 2004, graphene has been found to host a plethora of unusual electronic transport phenomena, making it a fascinating system for fundamental studies in condensed matter physics as well as offering tremendous opportunities for future electronic and sensing devices. Typically, electronic transport in graphene has been investigated via resistivity measurements; however, these measurements are generally blind to spatial information critical to observing and studying landmark transport phenomena in real space and in realistic imperfect devices. We apply quantum imaging to the problem and demonstrate noninvasive, high-resolution imaging of current flow in monolayer graphene structures. Our method uses an engineered array of near-surface, atomic-sized quantum sensors in diamond to map the vector magnetic field and reconstruct the vector current density over graphene geometries of varying complexity, from monoribbons to junctions, with spatial resolution at the diffraction limit and a projected sensitivity to currents as small as 1 μ A. The measured current maps reveal strong spatial variations corresponding to physical defects at the submicrometer scale. The demonstrated method opens up an important new avenue to investigate fundamental electronic and spin transport in graphene structures and devices and, more generally, in emerging two-dimensional materials and thin-film systems.

INTRODUCTION

Resistivity transport measurements have been a powerful tool for discovering electronic phenomena in condensed matter and, in particular, in graphene. However, this approach averages out spatial variations and therefore cannot distinguish the contributions of bulk processes and edge effects or defect-induced contributions, which are of crucial importance in the physics of microscopic devices (1, 2). Furthermore, real-space observation of many nonclassical forms of electronic transport, such as gate-controlled electron guiding and lensing (3, 4), transverse magnetic focusing (5-7), topological currents (8), or viscous electron backflow (9), to name just a few, has remained elusive. The ability to image charge currents in graphene would thus open a new era in the study of two-dimensional (2D) electronic transport. In recent years, progress toward this goal has been made following various approaches. Edge currents in graphene have been mapped in one dimension using superconducting interferometry, but extending the concept to 2D imaging is very challenging (10). Scanning gate microscopy has been used to map conductance fluctuations and even cyclotron orbits (11-14), but the invasive gating tip makes quantitative analysis difficult and limits the technique's applicability (15, 16). Scanning magnetometry based on a superconducting quantum interference device is typically limited to moderate spatial resolutions of several micrometers (17), despite holding great promise for improvement (18), and is restricted to low temperatures. Thus, a general, noninvasive method offering submicrometer resolution and capable of operating over a wide range of temperatures, including room temperature, is still lacking. Here, we fabricate an integrated quantum imaging platform where graphene devices are defined directly onto a diamond chip containing an array of nearsurface, atomic-sized magnetic sensors in the form of nitrogen-vacancy (NV) centers (19-24). Using this platform, we image the magnetic field generated by charge currents injected into graphene ribbons and junc2017 © The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science. Distributed under a Creative Commons Attribution NonCommercial License 4.0 (CC BY-NC).

tions and reconstruct the current density distribution (25–27), revealing current flow features associated with submicrometer defects in the graphene structures. Our approach can be applied to other 2D materials and thin-film systems and can be used to not only map charge currents but also spin currents and magnetic moments.

RESULTS

The principle of the experiment is illustrated in Fig. 1A. Graphene ribbons and metallic contacts are fabricated directly onto a diamond chip that hosts a layer of NV centers embedded ≈20 nm below the surface. The fabrication process involves wet transfer of monolayer graphene grown by chemical vapor deposition (CVD) on a copper foil, onto the diamond substrate, and subsequent patterning via electronbeam lithography (EBL) and plasma etching (see Materials and Methods). The graphene-diamond platform is then mounted onto a glass coverslip equipped with a microwave (MW) resonator, and placed in a wide-field microscope operating at room temperature (23). Figure 1B shows an optical micrograph of the final device, where only the metallic contacts are visible on the diamond because of the very weak intrinsic absorption of graphene. A close look at an area containing a graphene ribbon (Fig. 1C) confirms that monolayer graphene on diamond provides no measurable contrast. To visualize the graphene sheet in situ, we make use of the NV photoluminescence (PL) quenching from the graphene (28). The NV layer is illuminated with a green laser beam, and the resulting red PL is imaged with a camera (Fig. 1D). The graphene ribbon appears as a dark area because of resonance energy transfer (29). A line cut across the ribbon (Fig. 1E) indicates a PL reduction of \approx 30%. This corresponds to a mean distance between the graphene sheet and the NV layer of ≈ 20 nm (29), in agreement with the expected NV implantation depth.

We next injected a current through one of the graphene ribbons (Fig. 2A) and used the array of NV centers to map the induced magnetic field. The measurement consists of recording the PL intensity as a function of applied MW frequency to form an optically detected magnetic resonance (ODMR) spectrum (Fig. 2B). An external magnetic field of amplitude $B_{\text{ext}} = 10 \text{ mT}$ was applied to lift the degeneracy

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Fig. 1. Graphene ribbons on a diamond imaging platform. (A) Schematic of the experiment. The diamond platform consists of a diamond chip hosting a layer of nearsurface NV centers. The graphene devices are fabricated directly on the diamond chip, which is mounted on a coverslip equipped with an MW resonator. The NV centers' PL under green laser and MW excitations is imaged on a camera to form the magnetic field image. (B) Optical micrograph of the final device. Apparent on the diamond are metallic contacts, and wire bonds, used for current injection in the graphene ribbons. (C) Bright-field image recorded with the camera, focused on a graphene ribbon (not visible). (D) PL image of the same area under laser excitation. The graphene ribbon is now visible because of PL quenching. (E) Line cut across the ribbon extracted from (D) (white dashed line). a.u., arbitrary units.



Fig. 2. Magnetic field imaging and reconstruction of the current density. (A) PL image of the graphene ribbon under study, defining the xyz reference frame. (B) ODMR spectrum of the NV centers in a single pixel near the graphene under a positive (red dots) or negative (blue dots) applied current. Solid lines are data fit to a sum of eight Lorentzian functions. Inset: Energy levels of the electron spin of a single NV center, showing the Zeeman splitting $2\gamma_e B_{\parallel}$ between $m_s = \pm 1$, where m_s is the spin projection along the NV axis, B_{\parallel} is the magnetic field projection, and γ_e is the electron gyromagnetic ratio. The two electron spin resonances are indicated by green arrows. The experimental spectrum comprises eight resonances in total due to the four possible crystallographic orientations, allowing vector magnetometry. (C) Maps of the B_x (top), B_y (middle), and B_z (bottom) components of the magnetic field produced by a total current I = 0.8 mA. (**D**) Maps of the J_x (top) and J_{v} (middle) components of the current density reconstructed from (C). The bottom panel shows the norm of the current density, |J|. The black arrows represent the vector \vec{J} (length proportional to $|\vec{J}|$; threshold $|\vec{J}| > 30$ A/m). Scale bars, 10 μ m (C and D).

between the four symmetry axes of NV centers, resulting in four pairs of spin resonance transitions (two resonances per NV axis; see inset in Fig. 2B). This allows the vector components of the magnetic field, $(B_{xx}B_{yx})$ B_{z}), to be extracted in the laboratory frame (21, 24). We note that vector magnetometry could also be achieved in zero external magnetic field with little modification of the experimental procedure (30). Figure 2C shows the vector magnetic field maps for a graphene ribbon under an injected current I = +0.8 mA and normalized using a current of opposite sign, I = -0.8 mA. The current density distribution is then obtained by inverting the Biot-Savart law in the Fourier space (25, 26). This provides the vector components of the lineal current density, J_x and J_y (Fig. 2D), with a precision estimated to $\approx 5\%$ (see the Supplementary Materials). They can be combined in a single plot (bottom panel in Fig. 2D), where the color codes the norm of the current density vector, |J|, and the arrows indicate the direction (and relative norm) of J. As a consistency check, integrating the current density over the width of the ribbon gives a total current of $\approx 0.81(4)$ mA, in agreement with the applied current. On the basis of the signal-to-noise ratio observed in Fig. 2D, we project a sensitivity to current densities as low as \approx 1 A/m under similar conditions (total acquisition time of 2 hours), which corresponds to a total current of 1 µA in a 1-µm-wide ribbon.

Figure 2D reveals strong irregularities in the current density along the ribbon, mainly in the form of constrictions and holes, with feature sizes imaged down to $\approx 1 \,\mu$ m. To investigate the origins of these effects, we mapped the current density in two graphene ribbons (Fig. 3, A and B) and compared the maps with the corresponding PL images (Fig. 3C), as well as with scanning electron microscopy (SEM) images of the same areas (Fig. 3D), where the relatively high electron emission of the diamond makes a clear contrast with the graphene. A clear correlation is seen between the irregular features in the current density and bright lines or dots visible in the SEM images. These bright areas, which exhibit a range of shapes with feature sizes down to \approx 200 nm, are attributed to cracks and localized tears in the graphene sheet, through which the current cannot flow. This tearing is commonly found in graphene after it was transferred onto a substrate using a wet process (31). The tears are also visible in the PL images. They appear as bright contrast areas, with a PL intensity similar to that measured on the bare diamond surface. This indicates that no significant PL quenching occurs



Fig. 3. Current flow near defects in graphene. (**A**) Maps of the norm of the current density, $|\vec{J}|$ in two different graphene ribbons driven by a total current I = 0.8 mA. (**B**) Zooms of selected areas from (A). The black arrows represent the vector \vec{J} (length proportional to $|\vec{J}|$; threshold $|\vec{J}| > 30$ A/m). (**C**) PL images corresponding to the same areas as in (B). (**D**) Corresponding SEM images. Scale bars, 5 μ m.



Fig. 4. Current flow near metallic contacts. (**A** and **B**) SEM images of two junctions between Ti/Au electrodes and a graphene ribbon. The insets show the corresponding PL images. (**C** and **D**) Maps of the norm of the current density, $|\vec{J}|$, under a total current I = 0.8 mA, corresponding to the two junctions shown in (A) and (B). The black dashed lines indicate the edges of the metallic electrodes, as extracted from the PL images. The black arrows represent the vector \vec{J} (length proportional to $|\vec{J}|$; threshold $|\vec{J}| > 30$ A/m). Scale bars, 10 µm.

in these areas, consistent with an absence of graphene. Figure 3A also reveals a nonuniform current density in tear-free areas, where the current density is maximum at the center of the ribbon and decreases toward the edges. This indicates a nonuniform conductivity across the ribbon cross section, possibly associated with modulations in the carrier density. We note that the size of the smallest observable features in the current maps is limited by diffraction (\approx 500 nm); however, superresolution techniques could be applied, with the promise of a spatial resolution down to \approx 20 nm (*32*).

Finally, as a prelude to using the system to investigate more complex geometries, we examined the current flow at the junction between a graphene ribbon and evaporated metallic contacts (Fig. 4, A and B). The contacts are made of Ti/Au (20 nm/50 nm), with the graphene ribbon on top of the metallic leads. The current density maps are shown in Fig. 4 (C and D) for two different junctions. Along with the change of direction as the current flows from the leads (along y) into the graphene ribbon (along x), we also observe constrictions in the current flow around the contact interfaces. These features are consistent with the presence of tears along the metallic edges, which likely occurred during the transfer of graphene because it had to conform to the 70-nm step of the leads. The edge-induced tears are not visible in the SEM images (Fig. 4, A and B), illustrating the method's ability to detect and investigate defects and transport in complex geometries difficult to access with conventional techniques.

DISCUSSION

The ability to map the current flow in operating graphene devices provides an avenue for hitherto inaccessible real-space investigations of conduction in the presence of impurities, grain boundaries, or ripples or under a varying gating voltage (1, 2, 33) and of a range of nonclassical transport phenomena in the ballistic regime (3-10, 34, 35). Another intriguing application is the study of orbital magnetism (36), which has generated significant interest and requires spatially resolved probing methods. The NV centers used here can sense not only quasi-static magnetic fields but also ac fields and randomly fluctuating fields (noise) by exploiting quantum decoherence (37, 20). This has recently enabled probing of Johnson noise in metals (38) and could be used to investigate spontaneous or driven charge fluctuations in graphene. Besides electronics, diamond magnetic imaging opens exciting opportunities in graphene spintronics (39). In particular, it has the sensitivity required to

image spin injection in graphene (40) or detect the spin Hall effect (41). Diamond-based imaging could also be used to detect and investigate localized magnetic moments associated with defects, impurities, or edges, which is currently a subject of active research (42, 43). Finally, we note that our approach is not limited to monolayer graphene but is also applicable to multilayer graphene, other 2D materials, and thin-film solid-state systems including topological insulators and 2D electronic systems in silicon (44), provided that they can be transferred or fabricated onto diamond or onto a thin buffer layer of a different material, if required. Hence, the quantum imaging technique reported here could become a ubiquitous investigation tool for 2D technologies in the coming years.

MATERIALS AND METHODS

Sample fabrication

The imaging sensor used in this work was engineered from electronic grade type IIa diamond (E6). The diamonds were thinned and repolished to a $4 \times 4 \times 0.1$ -mm crystal (DDK) and then laser-cut into 2×2 -mm imaging chips. The single-crystal diamond was then implanted with ¹⁵N atoms at 6 keV to a dose of 1×10^{13} ions/cm². The implanted sample was annealed at 1000°C for 3 hours and acid-treated [sulfuric acid (1 ml) and sodium nitrate (1 g) at 300°C for 10 min] to remove any unwanted surface contamination. The density of NV centers after annealing was 1×10^{11} NV/cm², estimated by comparing the intensity from a single NV center in diamond with that obtained from the NV ensemble. The depth distribution of the NV centers with a 6-keV implantation energy is predicted to be $\approx 10 \pm 4$ nm by transport and range of ions in matter (TRIM) simulations. However, it has been shown by molecular dynamics simulations (45) and by proton nuclear magnetic resonance measurements (46) that the NV depth can be twice as large as the TRIM predictions, leading to an estimated average depth of ≈ 20 nm in our sample. The graphene devices were fabricated on the diamond substrate with a two-step EBL process. In the first step, Ti/Au (20 nm/50 nm) contacts were evaporated onto the diamond surface through a soft mask patterned in a bilayer MMA(E11)/PMMA(A4 950) resist stack. After liftoff of the resist stack in acetone, PMMA-coated CVD graphene, purchased as grown on copper foil (Graphene Supermarket), was transferred using the wet chemical etch method of Liang et al. (31) without the SC1 step. The transfer method, rather than the mechanical exfoliation method, was used because it allows relatively simple parallel fabrication of multiple devices on a single diamond chip. After drying over for 48 hours at room temperature, a layer of negative tone resist, SU8 (2002), was spun on top of the transfer PMMA(A4 950) layer. The SU8 was patterned by EBL to protect the areas of graphene to remain in the final device, whereas the exposed PMMA/graphene stack was etched away by oxygen plasma asher. Finally, the cured SU8 protective mask was removed by acetone dissolution of the underlying PMMA layer exposing the patterned graphene. The graphene-diamond chip was then glued onto a glass coverslip equipped with a gold MW resonator fabricated by photolithography. The quality of the graphene in the final device was estimated by Raman spectroscopy (fig. S1), showing signs of tearing.

Scanning electron microscopy

The SEM images in Figs. 3 and 4 were taken using an FEI Nova NanoLab system, using the secondary electrons in-lens detector at an acceleration voltage of 5 kV. Significant charging of the insulating diamond was observed, limiting the spatial resolution of the images. However, we note that charging was essential to see the graphene. Images taken with the graphene and gold electrodes connected to the ground provided

higher-resolution images of the diamond surface (because of reduced charging) but exhibited no measurable contrast between graphene and bare diamond (see comparison in fig. S2). Therefore, the images shown in Figs. 3 and 4 were taken with the sample completely isolated from the ground.

Measurement setup

The wide-field magnetic imaging was performed using the setup described by Simpson et al. (23), which is based on a modified Nikon inverted microscope (Ti-U). Optical excitation from a 532-nm Verdi laser was focused (f = 300 mm) onto an acousto-optic modulator (Crystal Technologies model 3520-220) and then expanded and collimated (Thorlabs beam expander GBE05-A) to a beam diameter of 10 mm. The collimated beam was focused using a wide-field lens (f =300 mm) to the back aperture of the Nikon $40 \times (1.3 \text{ numerical aperture})$ oil immersion objective via a Semrock dichroic mirror (Di02-R561-25x36). The NV fluorescence was filtered using two bandpass filters before it was imaged using a tube lens (f = 300 mm) onto a sCMOS camera (Neo, Andor). MW excitation was provided by an Agilent MW generator (N5182A) and switched using a Mini-Circuits RF switch (ZASWA-2-50DR+). The MWs were amplified (Amplifier Research 20S1G4) before they were sent to the MW resonator. The SpinCore PulseBlasterESR-PRO (500 MHz) was used to control the timing sequences of the excitation laser, MWs, and sCMOS camera, and the images were obtained and analyzed using a combination of custom LabVIEW/Matlab codes. The laser power density used for imaging was 30 W/mm², and all images were taken in an ambient environment at room temperature. The graphene devices were connected to the Picoammeter/Voltage Source (Keithley 6487) to operate the dc current through the devices. The magnetic images were obtained by recording ODMR spectra of the NV layer in the pulsed regime (47), with a π time of 200 ns. The acquisition time was 24 s per MW frequency (hence, 2 hours in total for the whole spectrum). A background magnetic field from a permanent magnet (strength of 10 mT at the sample) was applied in such a way that all four pairs of spin resonances, corresponding to the four possible NV center's symmetry axes, could be resolved (see an example spectrum in Fig. 2B).

Numerical methods

The ODMR spectrum recorded at each pixel was fit to a sum of eight Lorentzian functions to obtain the resonance frequencies, $\{\omega_{i\pm}\}_{i=1 \text{ to } 4}$. The Zeeman splitting for each NV axis, $\Delta \omega_i = \omega_{i+} - \omega_{i-}$, was then converted into the magnetic field projection onto the corresponding axis, $B_{\parallel,i}$ via the approximate relation $B_{\parallel,i} \approx \Delta \omega_i / 2\gamma_e$, where γ_e is the electron gyromagnetic ratio (24). To suppress the background magnetic field and retain only the Oersted contribution due to the current I, we recorded images with two opposite signs of I and used the normalized difference $\Delta B_{\parallel,i}(+I) = [B_{\parallel,i}(+I) - B_{\parallel,i}(-I)]/2$. Next, the magnetic field projections along the four NV axes, $\{\Delta B_{\parallel,i}\}_{i=1 \text{ to } 4}$, were used to infer the Oersted magnetic field in the laboratory frame, $\{B_x,$ $B_{y_2} B_z$, where z is the normal to the diamond surface and x is parallel to the edges of the graphene ribbons (see Fig. 2A). To reconstruct the 2D current density in the xy plane, we inverted the Biot-Savart law in the Fourier space based on the method described by Roth et al. (25). This yields the distribution J(x, y), which is a lineal current density expressed in units of amperes per meter, the current being confined in the 2D sheet of graphene. More detailed information about the numerical methods, including tests of robustness against parameters and a discussion of uncertainties, are available in the Supplementary Materials.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/ content/full/3/4/e1602429/DC1

- Supplementary Materials and Methods
- fig. S1. Raman spectroscopy.
- fig. S2. SEM imaging of graphene on diamond.
- fig. S3. Zeeman splitting as a function of magnetic field.
- fig. S4. Subtraction of the background magnetic field.
- fig. S5. Oersted magnetic field in the laboratory frame.
- fig. S6. Procedure to reconstruct the current density.
- fig. S7. Robustness of the reconstruction procedure.
- fig. S8. Oersted field as a function of probe distance.

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Acknowledgments: We would like to thank S. Rubanov from the Advanced Microscopy Facility of the Bio21 Institute, University of Melbourne, for assistance with SEM imaging. We acknowledge helpful discussions with C. Ritchie and L. T. Hall. **Funding:** This research was supported, in part, by the Australian Research Council Centre of Excellence for Quantum Computation and Communication Technology (project number CE110001027). L.C.L.H. acknowledges the support of the Australian Research Council Laureate Fellowship Scheme (FL130100119). D.A.S. acknowledges support from the Melbourne Neuroscience Institute Fellowship Scheme. **Author contributions:** The experiment was designed by all authors. J.-P.T. performed the NV measurements, the numerical reconstruction, and the SEM imaging. J.-P.T. and L.C.L.H. wrote the manuscript. N.D. and A.S. fabricated the graphene devices on diamond and contributed to data interpretation. N.D., D.A.B., A.S., and D.A.S. prepared and characterized the diamond sample for NV imaging. D.A.S. realized the imaging setup and contributed to the NV measurements. All authors discussed the data and commented on the manuscript. **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. Additional data related to this paper may be requested from the authors.

Submitted 4 October 2016 Accepted 1 March 2017 Published 26 April 2017 10.1126/sciadv.1602429

Citation: J.-P. Tetienne, N. Dontschuk, D. A. Broadway, A. Stacey, D. A. Simpson, L. C. L. Hollenberg, Quantum imaging of current flow in graphene. *Sci. Adv.* **3**, e1602429 (2017).

ScienceAdvances

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Sci Adv **3** (4), e1602429. DOI: 10.1126/sciadv.1602429

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