Letter

Tunable Graphene dc Superconducting Quantum Interference Device

C#ag#lar Girit, V. Bouchiat, O. Naaman, Y. Zhang, M. F. Crommie, A. Zettl, and I. Siddiqi

More About This Article

Additional resources and features associated with this article are available within the HTML version:

- Supporting Information
- Access to high resolution figures
- Links to articles and content related to this article
- Copyright permission to reproduce figures and/or text from this article

View the Full Text HTML

Tunable Graphene dc Superconducting Quantum Interference Device

Çağlar Girit,[†,‡] V. Bouchiat,[||,¶] O. Naaman,[†,§,¶] Y. Zhang,[†] M. F. Crommie,[†,‡] A. Zettl,*[,†,‡] and I. Siddiqi*[,†,§]

Department of Physics, University of California, Materials Sciences Division, Lawrence Berkeley National Laboratory, Quantum Nanoelectronics Laboratory, University of California, Berkeley, California 94720, and Institut Néel, CNRS/UJF, BP 166, 38042 Grenoble Cedex 9, France

ABSTRACT

Graphene exhibits unique electrical properties on account of its reduced dimensionality and "relativistic" band structure. When contacted with two superconducting electrodes, graphene can support Cooper pair transport, resulting in the well-known Josephson effect. We report here the fabrication and operation of a two junction dc superconducting quantum interference device (SQUID) formed by a single graphene sheet contacted with aluminum/palladium electrodes in the geometry of a loop. The supercurrent in this device can be modulated not only via an electrostatic gate but also by an applied magnetic field—a potentially powerful probe of electronic transport in graphene and an ultrasensitive platform for nanomagnetometry.

Graphene, a single atomic layer of graphite, exhibits unique electrical and mechanical properties on account of its reduced dimensionality and "relativistic" band structure.[1] When contacted with two superconducting electrodes, graphene can support Cooper pair transport,[2,3] resulting in the well-known Josephson effect. The supercurrent of such a junction can be modulated with an electrostatic gate which tunes the carrier type and density of the graphene sheet. We report here the fabrication and operation of a two junction superconducting quantum interference device (SQUID) formed by a single graphene sheet contacted with aluminum/palladium electrodes in the geometry of a loop. The supercurrent in this device can additionally be modulated via an applied magnetic field—a potentially powerful new probe of electronic transport in graphene. Graphene SQUIDs also suggest a new modality for ultrasensitive magnetometry of nanomagnets chemically or physically attached to the carbon surface.

We obtain graphene samples by mechanical exfoliation of graphite flakes on oxidized silicon ($SiO_2(285 \text{ nm})/Si^{2+}$) wafers[4] and fabricate devices using conventional electron-beam lithography. The two graphene Josephson junctions that form the SQUID are patterned side by side on the same graphene sheet as shown in Figure 1a. The gaps between the metallic electrodes that define the junctions are patterned to be very short ($L \sim 50$ nm) and wide ($W \sim 4 \ \mu$m) to obtain large critical currents. Once the devices are patterned, a 3 nm interfacial layer of palladium, followed by a 50 nm thick aluminum layer is deposited by electron-beam evaporation. Though palladium is a normal metal, superconducting charge carriers from the aluminum top layer can transit through this thin layer that is used to obtain a reliable, low resistance contact to the graphene sheet. The Si^{2+} layer serves as an electrostatic back gate common to both junctions.

Low frequency transport measurements are performed on devices anchored to the mixing chamber of a dilution refrigerator with base temperature $T = 20$ mK and equipped with shielded, differential wiring filtered with multiple stages of copper powder and discrete element low pass filters. A small superconducting solenoid mounted above the sample is used to apply the magnetic field. Measurements of the mobility taken in the normal state indicate a mean free path ranging from $10-50$ nm which is comparable to the junction length. Below 1 K, a supercurrent is observed, and Figure 1b shows the differential resistance dV/dI of a typical device as a function of bias current and gate voltage taken at base temperature. The central dark region corresponds to the zero-voltage state. The SQUID critical current is demarcated by the yellow dashed line, and varies in magnitude from one to seven microamperes as a function of gate voltage. Transport processes which involve multiple reflections of charge carriers at the superconductor/graphene interface that result

* To whom correspondence should be addressed. E-mail: (I.S.) irfan_siddiqi@berkeley.edu; (A. Z.) azettl@berkeley.edu.
† Department of Physics, University of California.
‡ Materials Sciences Division, Lawrence Berkeley National Laboratory.
§ Quantum Nanoelectronics Laboratory, University of California.
|| Institut Néel, CNRS/UJF.
¶ These authors contributed equally to the work.
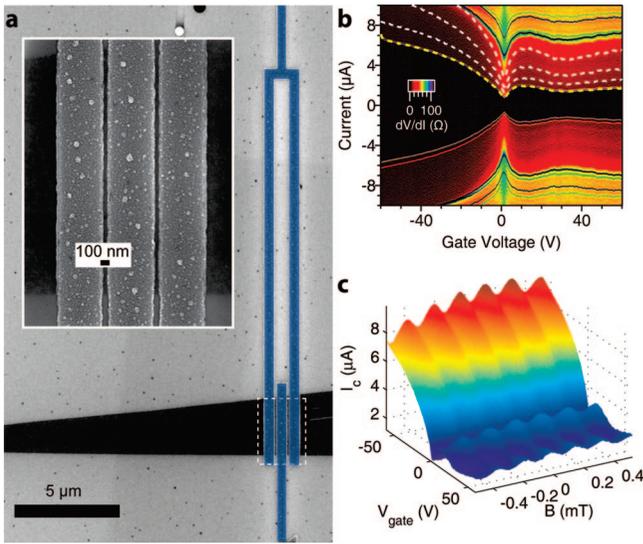
Figure 1. (a) Scanning electron microscope image of a superconducting quantum interference device made with graphene barriers (black, graphene; gray, silicon oxide substrate; blue false-color, Pd/Al electrodes). Inset: close-up of 50 nm wide graphene Josephson junctions. (b) Differential resistance of a device showing dependence of critical current on back gate voltage (yellow dashed line) at 20 mK. Traces of multiple Andreev reflections at constant voltages $2\Delta_g/ne$ for $n = 1, 2, 3$ are indicated by the white dashed lines ($n = 1$ at top, $\Delta_g \sim 75$ μeV). (c) Mean switching current as a function of magnetic field B and gate voltage V_{gate}. The sinusoidal oscillations indicate quantum interference between the two graphene Josephson junctions. For a fixed flux bias, the magnitude of the modulation can be controlled by the back gate.

in the conversion of electrons and holes to Cooper pairs (multiple Andreev reflection)[5] give rise to a subharmonic gap structure. These features are observed at submultiples of the effective energy gap $2\Delta_g/ne$ and are indicated by the white dashed lines, giving $\Delta_g \sim 75$ μeV.

Figure 1c demonstrates the effect of a magnetic field applied perpendicular to the device plane. The magnetic flux through the SQUID loop induces a phase difference between the supercurrents passing through the two Josephson junctions and interference should manifest itself as a variation of the total critical current with applied field. The standard relationship for an ideal, low loop inductance tunnel junction SQUID is

$$I_e = \sqrt{(I_{c1} - I_{c2})^2 + 4I_{c1}I_{c2} \cos^2 \frac{\pi\Phi}{\Phi_0}} \qquad (1)$$

with I_{c1} and I_{c2} the critical currents through each junction and Φ the applied flux. When a magnetic field is applied, we observe oscillations in the SQUID critical current with a periodicity of 190 μT, which in the context of eq 1 implies an effective loop area $\Delta B/\Phi_0 \sim 11$ μm^2. This value is in good agreement with the geometric loop area of 10 μm^2. The critical current at $\Phi = \Phi_0/2$ (destructive interference) is not fully suppressed to zero and the modulation depth

varies from device to device. There are many factors that may contribute to this effect including asymmetries in the metal/graphene interface as well as subtleties associated with the current-phase relation in graphene and are the subject of further study. For a ballistic superconductor—graphene—superconductor junction,[6] the current-phase relation is nonsinusoidal and depends on the peculiar Dirac nature of electrons in graphene. Our devices are in the diffusive regime where signatures of "Dirac" transport are not expected in the SQUID modulation curves. The modulation depth can be tuned with the electrostatic gate, allowing one to vary the SQUID sensitivity $dI_c/d\Phi$ at fixed flux bias, potentially useful in increasing dynamic range and varying the coupling strength in quantum measurements.

We have presented the first graphene dc superconducting quantum interference device, a powerful future platform for nanoscale magnetometry and fundamental transport studies. SQUID devices can be used to probe the current-phase relation of graphene Josephson junctions and provide unique signatures of transport mechanisms and characteristic scattering lengths in single layer graphene. Compared to conventional SQUIDs based on embedded semiconductor 2D electron gases, graphene is simple to fabricate, electrically contact with high-transparency electrodes,[7] and has the conduction layer directly accessible for functionalization. Finally, since graphene can be easily patterned by oxygen plasma or AFM oxidation and equipped with local gates, it is straightforward to fabricate multijunction circuits with tunable critical currents. For magnetometry applications, this patternability combined with the large critical currents observed indicate that higher signal-to-noise ratios can be obtained for spin detection compared to other nanoscale SQUIDs.[8]

References
(1) Geim, A. K.; Novoselov, K. S. *Nat. Mater.* **2007**, *6*, 183.
(2) Heersche, H. B.; Jarillo-Herrero, P.; Oostinga, J. B.; Vandersypen, L. M. K.; Morpurgo, A. F. *Nature* **2007**, *486*, 56.
(3) Du, X.; Skachko, I.; Andrei, E. Y. *Phys. Rev. B* **2008**, *77*, 184507.
(4) Novoselov, K. S.; Jiang, D.; Schedin, F.; Booth, T. J.; Khotkevich, V. V.; Morozov, S. V.; Geim, A. K. *Proc. Natl. Acad. Sci. U.S.A.* **2005**, *102*, 10451.
(5) Klapwijk, T. M.; Blonder, G. E.; Tinkham, M. *Physica B+C* **1982**, *109–110*, 1657.
(6) Titov, M.; Beenakker, C. W. J. *Phys. Rev. B.* **2006**, *74*, 041401.
(7) Schäpers, T. *Springer Tracts Mod. Phys.* **2001**, *174*, 21.
(8) Cleuziou, J. P.; Wernsdorfer, W.; Bouchiat, V.; Ondarçuhu, T.; Monthioux, M. *Nat. Nanotechnol.* **2006**, *1*, 53.

NL802765X