Vortex detection and quantum transport in mesoscopic graphene Josephson junction arrays

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We explore the interplay between normal-state and superconducting phase coherence in mesoscopic Josephson junction arrays created by patterning superconducting disks on monolayer graphene. We find that the mean conductance and universal conductance fluctuations are both enhanced below the critical temperature and field of the superconductor, with greater enhancement away from the graphene Dirac point. We also observe features in the magnetoconductance at rational fractions of flux quanta per array unit cell, which we attribute to the formation of vortices in the arrays. The applied fields at which the features occur are well described by Ginzburg-Landau simulations that take into account the number of unit cells in the array.

Superconducting vortices in a two-dimensional gas of chiral massless Dirac fermions are predicted to harbour excitations that resemble hypothetical elementary particles known as Majorana zero modes [1, 2]. Developing ways to create, control, and interfere vortices in candidate Dirac conductors—such as graphene and the surface states of three-dimensional topological insulators—is therefore important for detecting non-Abelian statistics and for implementing topologically protected quantum information processing [3].

In the absence of intrinsic pairing of electrons through the Cooper channel, attention has focused on using the proximity effect to induce a Dirac condensate in these materials [4]. In the proximity effect, a normal metal/superconductor interface generates phase-correlated quasiparticle pairs via Andreev reflection, which opens an effective gap in the normal metal. These pairs communicate phase information between superconductors in Josephson junctions, leading to supercurrent flow. Using graphene as the normal conductor also allows the normal-state resistance to be tuned via the carrier density [5, 6]. This leads to a superconducting-to-insulating transition in disordered arrays [7], and the addition of flux pinning in ordered arrays creates a superconducting glass state at low temperatures [8, 9]. The presence of flux vortices in such devices causes a phase frustration between array islands, which interacts with the normal-state quantum interference as well as Andreev corrections to the conductance in these mesoscopic, two-dimensional devices.

In this Letter we study how the mean value and fluctuations of the conductance of monolayer graphene are modified in the presence of ordered arrays of superconducting disks. We find that both are enhanced below the critical temperature and field of the superconductor due to the proximity effect, with greater enhancement away from the graphene Dirac point. We also detect the presence of proximity vortices in the graphene through changes in the magnetoconductance whenever the magnetic field generates an integer number of flux quanta through the arrays.

Our graphene flakes are mechanically exfoliated from natural graphite onto degenerately doped Si substrates with a 300 nm oxide layer. We identify monolayer flakes by their optical contrast and quantum Hall measurements, then pattern superconducting electrodes and square arrays of disks using electron beam lithography followed by thermal evaporation of a Ti/Al bilayer (5 nm/60 nm) [Fig. 1]. We present measurements of two array geometries: device A has disks with diameter 2a = 200 nm and center-center separation b = 700 nm, device B has disks with 2a = 500 nm and b = 750 nm.

Two-, four-, and quasi-four-terminal differential conductance was measured using standard low frequency AC lock-in techniques, in a pumped ⁴He cryostat at 340 mK.

FIG. 1. (a) Schematics of array unit cells for device A (upper) and device B (lower). (b) Schematic of device A and two-terminal measurement circuit. (c) Atomic force microscope image of a similar device (disk diameter = 200 nm).
and a dilution refrigerator between 1.05 K and 100 mK. A voltage $V_{bg}$ applied to the doped Si substrate controlled the carrier density. For devices A and B, the graphene exhibited a Dirac point at $V_{bg} \approx 2$ V and 6 V and a carrier mobility of $\sim 3500$ cm$^2$(Vs)$^{-1}$ and $\sim 5400$ cm$^2$(Vs)$^{-1}$ for $n = 5 \times 10^{11}$ cm$^{-2}$ and $T = 350$ mK. The electron mean-free path $l_e = 2D/v_F$ is approximately 130 nm for device A and 90 nm for device B, where $D$ is the diffusion constant ($\approx 0.03$ m$^2$s$^{-1}$ and 0.05 m$^2$s$^{-1}$).

Figure 2(a) shows the zero-field resistance of both devices as a function of temperature. We observe a pronounced decrease in both cases below the critical temperature of the Ti/Al bilayer, which is around 1 K. A similar decrease in resistance was recently reported [9] and attributed to the absence of dissipation from regions under and around the disks due to the proximity effect [10]. The percentage decrease we see is not proportional to the percentage of the graphene covered by the disks, limited perhaps by interface conductance or potential barriers between covered and uncovered regions of graphene. However, the temperature at which the resistance drops is higher for device B. Since the edge-to-edge separation of the disks is smaller in this device, this would indicate a proximity effect in the arrays [11]. We do not observe a fully superconducting state, even at the lowest temperatures. Although in theory the transition to the full proximity effect can be tuned by the back-gate in graphene [3], in practice the low critical temperature of aluminium restricts the $T/T_c$ range of our measurements.

Figure 2(b) plots the conductance of both devices above and below the critical field of the disks as a function of back-gate voltage. In Fig. 2(c), we find that the conductance enhancement due to the presence of superconductivity in the leads and islands depends linearly on the carrier density (in contrast to Ref. [9]). We attribute this to the greater diffusion of carriers around the contacts at higher carrier density, and a corresponding increase in the area of the effectively gapped region. As the temperature is increased [inset of Fig. 2(c)] the linear dependence on back-gate voltage persists, and by $T = 750$ mK the magnitude of $\Delta G$ has approximately halved. Above the critical temperature of the leads, $\Delta G$ is small and negative due to the suppression of weak localisation [12].

FIG. 2. (a) Temperature dependence of the zero-field differential resistance in devices A (left) and B (right). (b) Back-gate voltage ($V_{bg}$) dependence of the differential conductance at zero field (thin black) and above the critical field of each device (thick blue/red), $T = 350$ mK, for devices A and B, respectively. Grey boxes indicate data in insets. (c) Difference between the zero-field and critical-field conductance, $\Delta G$, as a function of back-gate voltage relative to the Dirac point (thick dark blue: device A, thin dark red: device B). Inset: Temperature dependence of $\Delta G$, device B. $T = 100$ mK (black), 350 mK (dark red), 750 mK (red) and 1.05 K (grey).

FIG. 3. (a) Magnetoconductance at temperatures between 250 mK and 950 mK (100 mK and 150 mK traces also shown for device B). Traces offset for clarity, grey dashed lines highlight features plotted below. (b) Plots of the critical field of the Ti/Al bilayer (stars), outermost (squares) and innermost (circles) temperature-dependent features, and temperature-independent features (diamonds). Grey horizontal lines are rational fractions of flux quanta per array unit cell; $\frac{1}{4}$ to $\frac{1}{4}$ in left panel, $\frac{1}{9}$ to $\frac{1}{9}$ and $\frac{1}{9}$ in right panel.
Having established that the modulation of the resistance is consistent with proximity effects around the islands, we now describe the magnetoconductance. Figure 3(a) shows the field-dependent differential conductance, captured at different temperatures, for devices A and B. In both devices, the application of a small magnetic field below \( T_c \) decreases the conductance of the arrays up to a critical field and for a wide range of carrier densities in both the electron and hole conduction regimes. Close inspection of these data reveals both temperature-dependent and -independent features, plotted in Fig. 3(b). Also plotted, for reference, is the critical field of branched contacts on device A. Note that sub-micron Al islands have been shown to have the same properties as continuous films \([14]\), suggesting the disks have the same critical field and temperature as the leads.

The first set of temperature-dependent features closely follows the critical field of the Ti/Al bilayer in both devices. The second set appears at lower fields and has a similar temperature dependence, but differs in magnitude between devices, occurring at smaller fields and temperatures in device A. This correlation indicates that the two sets of features are related to the extent of the proximity effect in the graphene. Below the critical field of the disks it is likely that Andreev reflection at the interfaces affects transport, explaining the first set of features. The second set corresponds to phase coherence between neighbouring pairs of disks. The dephasing length estimated from the weak localisation peak is approximately 200 nm at \( T = 950 \text{ mK} \) in both devices, and therefore spans the region between disks first in device B, then in device A as the temperature decreases. Phase coherence between neighbouring pairs of disks implies a full proximity effect and hence dissipationless path between them, leading to the observed increase in conductance below the second set of temperature-dependent features.

Below the second temperature-dependent feature, we observe temperature-independent features at fields which appear to be related to the area of the array unit cell. These have been highlighted in Fig. 3(b). Such features at fixed fields are expected in two-dimensional Josephson junction arrays \([14, 15]\), due to low-energy configurations of vortices within the array, such as integer numbers of flux quanta per unit cell, or a superlattice of empty and occupied cells. We observe temperature-independent features at \( \frac{1}{3} \) and \( \frac{5}{8} \) for device A and \( \frac{5}{8} \) for device B, whereas in a large, square array the lowest energy configurations correspond to fields of \( \frac{5}{8} \) flux quanta per unit cell \([16]\).

To examine these features in more detail, and eliminate universal conductance fluctuations and any normal state conductance dependence, we average over a 20V range of back-gate voltage, and plot the results in Fig. 4(a). We find features at fields of \( \frac{1}{9} \) flux quanta per unit cell for device A, and \( \frac{5}{9} \) for device B. Figure 4(b) and (c) shows the absolute numerical derivative of the back-gate–averaged resistance of device A as a function of flux quanta per unit cell. Grid lines at \( \frac{5}{9} \) in Fig. 4(c) align with the features, confirming our earlier observation of temperature-independent features in the raw magnetoconductance.

To understand why the features occur at fields of \( \frac{5}{9} \) flux quanta per unit cell in device A, rather than \( \frac{5}{8} \), we perform simulations based on the Ginzburg-Landau equations. The model system is a square, \( 3 \times 3 \) unit cell array of superconducting disks (\( 2a = 200 \text{ nm}, b = 700 \text{ nm} \)) embedded in a normal metal matrix. The proximity effect is described by a spatially-dependent, anisotropic expansion coefficient of the Gibbs free energy functional, \( \alpha = \alpha_0 g(r) \), which equals 1 inside the superconducting
disks and less than 1 in the metallic regions. The square simulation region has periodic boundary conditions in all directions in the two-dimensional plane, and is exposed to a homogeneous magnetic field \( B \). Figure 4(b), panels 1–4 show ground-state vortex configurations in the form of local magnetic field density, obtained in field-cooled simulations, since the initial vortex structure directly influences the response of the system to an applied current. The critical depinning current exhibits peaks at fields of \( \frac{\Theta}{2\pi} \) flux quanta per unit cell [Fig. 4(d)], suggesting that the field values of the features in device A are due to the odd number of unit cells.

Along with the phase frustration effects detailed above, the presence of reproducible, aperiodic conductance fluctuations in our mesoscale flakes is a precursor of the superconducting glass state predicted and recently observed in graphene Josephson junction arrays. In Fig. 5 we explore the effect of the superconducting disks on universal conductance fluctuations (UCFs). Figure 5(a) shows UCFs in device B as a function of back-gate voltage and applied field, at \( T = 100 \) mK and \( V_{bg} = V_{DP} = -14 \) V. The overall conductance of the array is enhanced below the critical field of the leads (middle panel), however, the UCFs themselves are also enhanced, as seen in the detrended conductance (lower panel). Figure 5(b) shows the standard deviation of the UCFs, \( \delta G = \sqrt{\langle G^2 \rangle - \langle G \rangle^2} \), as a function of magnetic field. Below the critical field of the leads, \( \delta G \) increases linearly in magnitude towards zero-field. We observe this linear trend at different carrier densities and temperatures. Comparison of \( \delta G \) with the differential conductance of the device [Fig. 5(c)] reveals that \( \delta G \) is enhanced over a larger field range than the mean conductance.

Figure 5(d)-(f) quantifies the enhancement of \( \delta G \) between \( B = 0 \) (\( \delta G_S \)) and \( B = 10 \) mT (\( \delta G_N \)), and compares data taken at different back-gate voltages and temperatures. Upper panels show the values of \( \delta G_S \) and \( \delta G_N \), and lower panels their ratio. The form of the temperature dependence is the same at both high and low carrier density [Fig. 5(d) and (e)]. Figure 5(f) shows the back-gate voltage dependence at 100 mK, and above the critical temperature of the leads at 1.05 K. The enhancement of \( \sim 1.4-1.7 \) is close to that observed in InAs nanowires and graphene contacted by Ti/Al bilayers. At 100 mK, \( \delta G \) is more enhanced away from the Dirac point. At 1.05 K, we observe the opposite trend in \( \delta G_N \), with no enhancement at zero-field.

The enhancement of UCFs in mesoscopic devices with superconducting contacts is associated with Andreev reflection at the normal/superconductor interface. The temperature-dependence of the UCF enhancement, and its onset at larger applied fields compared to the mean conductance, can be linked to the superconducting gap in the Ti/Al bilayer. A larger gap implies more Andreev reflection at the interfaces, and therefore greater enhancement of the UCFs. Our observation of a smaller enhancement close to the Dirac point is more intriguing, since suppression of superconductivity near the graphene Dirac point has been seen before in large-area devices. So far, the suppression has only been seen in the global conductance, but it also appears here in conductance fluctuation data. Loss of phase coherence caused by the formation of electron-hole puddles at low carrier densities may be responsible for the suppression in both global conductance and fluctuations. A simple normal-state resistance dependence is unlikely, since the ratio of \( \delta G_S \) and \( \delta G_N \) is the same at \( V_{bg} = V_{DP} = -7 \) V and the Dirac point, nor is our square resistance high enough for a metal-superconductor transition. Another possibility is the loss of Andreev pairs due to specular Andreev reflection at the graphene/disk interfaces, though a measurable contribution is unlikely given the diffusive transport in our substrate-supported graphene.

In summary, we have explored the interplay between
normal-state and superconducting phase effects in ordered arrays of mesoscopic superconductors on graphene. Even without a full proximity effect spanning the device, magnetococonductance features at rational fractions of flux quanta per array unit cell indicate the presence of proximity vortices. We also observe suppression of the proximity effect at the Dirac point through the mean conductance and conductance fluctuations, due to the phase-breaking effect of disorder-induced charge puddles in our large-area devices, or perhaps a contribution from specular Andreev reflection at the graphene/graphene interfaces.

These measurements of graphene-based Josephson junction arrays demonstrate the feasibility of detecting and manipulating superconducting vortices in Dirac conductors through charge transport. Extension of this approach to topological insulators could be a route to detection of non-Abelian statistics and implementation of topological quantum computing.

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