Steady Floquet–Andreev states in graphene Josephson junctions

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Engineering quantum states through light–matter interaction has created a paradigm in condensed-matter physics. A representative example is the Floquet–Bloch state, which is generated by time-periodically driving the Bloch wavefunctions in crystals. Previous attempts to realize such states in condensed-matter systems have been limited by the transient nature of the Floquet states produced by optical pulses1–3, which masks the universal properties of non-equilibrium physics. Here we report the generation of steady Floquet–Andreev states in graphene Josephson junctions by continuous microwave application and direct measurement of their spectra by superconducting tunnelling spectroscopy. We present quantitative analysis of the spectral characteristics of the Floquet–Andreev states while varying the phase difference of the superconductors, the temperature, the microwave frequency and the power. The oscillations of the Floquet–Andreev-state spectrum with phase difference agreed with our theoretical calculations. Moreover, we confirmed the steady nature of the Floquet–Andreev states by establishing a sum rule of tunnelling conductance4, and analysed the spectral density of Floquet states depending on Floquet interaction strength. This study provides a basis for understanding and engineering non-equilibrium quantum states in nanodevices.

Light is a powerful tool for tailoring new quantum states by driving them out of equilibrium14–15. One prominent example is Floquet engineering, which involves dynamic control of the properties of quantum matter through a time-periodic electromagnetic drive. Similar to the space-periodic potential that copies Bloch energy bands along the crystal momentum direction, the time-periodic potential copies Floquet states along the energy direction. The Floquet state promises rapid and comprehensive control of the excitation spectrum and topological nature of physical systems by simply tuning the intensity, frequency and polarization of the light. Therefore, there has been a great deal of research effort expended on the realization and manipulation of a wide variety of long-sought-after quantum states, such as chiral topological orders with no equilibrium counterpart2, including Floquet Majorana fermions4 and a new braiding protocol in the energy dimension5. For example, the realization and control of Floquet dynamics have been reported in both photonic and ultracold atomic systems6–11. Another important class of platforms are solid-state systems, in which transient Floquet states have been mainly investigated. As Floquet interaction strength is proportional to the ratio of the electric field to the square of the frequency of light12, previous condensed-matter experiments realizing Floquet states at the optical frequency of 30–50 THz have applied pulsed lasers to achieve a large electric field of $2.4 \times 10^7 - 4.0 \times 10^7 \text{ V m}^{-1}$ (refs. 13–15). However, Floquet states persist no longer than an order of picoseconds owing to the transient nature of the pulsed laser and the inherently short lifetime of the Floquet states. This short timescale makes it difficult to fully investigate and understand these novel light-driven states and exploit them for practical applications. Another critical issue affecting most experimental realizations of Floquet states is heating by the energy absorption from time-periodic driving. The driving increases the entropy density of the system and eventually brings it to a featureless state with no local correlations13. Although a number of ways to reduce or avoid heating effects have been suggested14–16, the heating problem still remains in photonic and ultracold gas systems because they are well isolated from the outside environment. By contrast, condensed-matter systems have well defined electron cooling paths, such as electron–phonon coupling and Wiedemann–Franz cooling of conducting electrons. However, a large electric field in an optical domain, which has been used in previous condensed-matter experiments, requires pulsed measurements to avoid heating problems. One strategy to avoid thermal problems is to use lower-frequency driving that requires a smaller electric field. There have been experimental studies in the microwave domain, but they have relied on indirect methods for probing Floquet states either by tracing their time evolution17,18, qubit–resonator resonance conditions19, magnetic resonance conditions20 or the a.c. Josephson effect17,18,21.

In contrast to previous studies, we report the experimental realization of truly steady Floquet–Andreev (F–A) states based on Andreev bound states (ABSs) formed in a graphene Josephson junction (GJJ) and their spectra based on direct tunnelling spectroscopy. Here the graphene offers an ideal platform for studying Floquet physics, as
such low-dimensional electronic systems can couple to an external electric field without screening. Here we generated steady F–A states by continuously applying a monochromatic microwave drive and probed them by superconducting tunneling spectroscopy with high energy resolution. We investigated the behaviour of the F–A states at various microwave frequencies and powers, and showed that their spectral features agreed well with our theoretical calculations. For example, we corroborated the steady nature of the F–A states by establishing a sum rule of the measured tunnelling conductance. This study clearly demonstrates steady Floquet states and will have a substantial impact on different areas of physics, including topological condensed matter, cold atoms in optical traps and non-equilibrium quantum statistical mechanics.

A GJJ consists of a non-superconducting graphene layer sandwiched between two superconductors, as shown in Fig. 1a. The GJJ allows the Josephson supercurrent by forming ABSs of electron-like and hole-like quasiparticles in the graphene, which are correlated by Andreev reflection at the graphene/superconductor interfaces. In the short junction limit where the superconducting coherence length $\xi$ is much shorter than the superconducting gap $\Delta$, a single pair of ABSs forms within the superconducting gap of ohmic-contacted aluminium (Al) electrodes $\Delta_{\text{AB}}$, and oscillates with the macroscopic phase difference between the superconductors $\varphi$. The GJJ has length $L = 0.51 \mu m$ and width $W = 3.0 \mu m$. A similar technique was used for making tunnel contact on multiwalled carbon nanotubes with aluminium or indium electrodes. In this structure, the reduced Planck constant $\hbar = h/2\pi$ is applied between the Al tunnel probe and graphene, such that the occupied DOS peak of the tunnel probe matches the empty DOS peak ($E^+$) of the ABS. Colour-coded plot of differential conductance $dI/dV$ as a function of $V$ and $\varphi$ for device 1. The top horizontal axis shows the superconducting phase difference $\varphi$ corresponding to $B$. The circles and the solid lines represent the $dI/dV$ peaks and corresponding theoretical fittings in a short junction limit, respectively. The line cut at $\varphi = 0$ plotted in the right panel shows $dI/dV$ peaks coming from the upper ($E^+$) and the lower ($E^-$) bands of the ABS.

We measured the voltage difference ($V$) between the Al tunnel probe and graphene while biasing the current ($I$). The output impedance of the current source (1 GΩ) was much larger than the largest tunnelling resistance (16 MΩ) measured in this experiment. We obtained the tunnelling differential conductance ($dI/dV$) as a function of $V$, which represents the convolution of the DOS of the ABS in the GJJ and the tunneling process $V < 0$ is applied between the tunnel probe and graphene, such that the occupied DOS peak of the tunnel probe matches the empty DOS peak ($E^+$) of the ABS. The top horizontal axis shows the superconducting phase difference $\varphi$ corresponding to $B$. The circles and the solid lines represent the $dI/dV$ peaks and corresponding theoretical fittings in a short junction limit, respectively. The line cut at $\varphi = 0$ plotted in the right panel shows $dI/dV$ peaks coming from the upper ($E^+$) and the lower ($E^-$) bands of the ABS.

Figure 1d shows the oscillation of ABS in a short junction limit and the corresponding density of state (DOS) at a fixed $\varphi$. In this study, we performed tunnelling spectroscopy on ABS formed in graphene using an Al superconducting tunnel contact with the graphene edge, as shown schematically in Fig. 1c (Supplementary Fig. 1d). Direct deposition of Al onto the graphene edge forms a sufficiently high potential barrier for the tunnel probe owing to the large interatomic distance between graphene and the Al atoms. A similar technique was used for making tunnel contact on multiwalled carbon nanotubes with aluminium or indium electrodes. In this structure, the reduced Planck constant $\hbar = h/2\pi$ is applied between the Al tunnel probe and graphene, such that the occupied DOS peak of the tunnel probe matches the empty DOS peak ($E^+$) of the ABS. Colour-coded plot of differential conductance $dI/dV$ as a function of $V$ and $\varphi$ for device 1. Direct deposition of Al onto the graphene edge forms a sufficiently high potential barrier for the tunnel probe owing to the large interatomic distance between graphene and the Al atoms.

For example, we corroborated the steady nature of the F–A states by establishing a sum rule of the measured tunnelling conductance. This study clearly demonstrates steady Floquet states and will have a substantial impact on different areas of physics, including topological condensed matter, cold atoms in optical traps and non-equilibrium quantum statistical mechanics. Our technique, when combined with rapidly developing microwave technologies, can be extended to Floquet-based novel quantum device applications.

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background subtracted \( dI/dV \) (Supplementary Fig. 3), which is three orders of magnitude better than that of the time-resolved photoemission method\(^{12}\). This corresponds to the electron temperature \( T_e \) of 73 mK, assuming that the energy resolution is determined by the broadening of Fermi–Dirac distribution as 3.5 \( k_B T_e \), where \( k_B \) is the Boltzmann constant\(^{30}\).

Figure 1e shows \( dI/dV \) measured at 20 mK as a function of \( V \) and magnetic field \( B \), the latter of which controls \( \phi_B = 2m(B - B_0)/\Phi_0 \) with a magnetic field offset \( B_0 \) of a superconducting solenoid magnet and an effective area of the superconducting ring \( A \). A clear oscillation of the \( dI/dV \) peak as a function of \( \phi_B \) is shown, which fits well with our theoretical calculations (Supplementary Fig. 4). In addition, the particle–hole symmetry of the superconductor manifests itself as a symmetric \( dI/dV \) with respect to zero-bias voltage \( V = 0 \). The oscillation of \( dI/dV \) is also observed in device 2 (Supplementary Fig. 5).

To generate steady F–A states of the GJJ, the devices were continuously irradiated with microwaves (Supplementary Fig. 6) and the tunnelling spectrum of graphene was measured using a superconducting tunnel probe. As shown in Fig. 2a, additional peaks in \( dI/dV \) emerged above and below the original ABS peaks as the microwave power \( P \) increased. The average voltage spacing between the two adjacent \( dI/dV \) peaks \( \Delta V = 51.7 \pm 11.0 \mu eV \) at \( P = -5.2 \) dBm corresponds to the microwave energy quanta \( hf = 49.6 \mu eV \) with microwave frequency \( f = 12 \) GHz. The error in \( \Delta V \) is estimated by the half-width at half-maximum of background subtracted \( dI/dV \). This behaviour can be understood by the emergence of Floquet replica states of the ABS, with the microwave irradiation acting as a time-periodic perturbation (Fig. 2b).



**Fig. 2 | Microwave power dependence of F–A states.** a, Colour-coded plot of differential conductance \( dI/dV \) as a function of bias voltage \( V \) and microwave power \( P \) measured in device 2 with microwave frequency \( f = 12 \) GHz at temperature lower than 50 mK. Here the superconducting phase difference is fixed at \( \phi = 0 \). Floquet replicas of an ABS indicated by arrows emerge as \( P \) increases. b, Schematics of the evolution of the spectral density of the F–A states at different \( P \). c, Line cuts of the plot in a at various \( P \), from -12 dBm to 0 dBm with 3-dBm intervals. The inset shows the integrated area of \( dI/dV \) line cuts at a given \( P \). d, Line cuts of the plot in a at dimensionless Floquet interaction strength parameter \( \beta \) = 0.64, \( \beta \) = 0.92, \( \beta \) = 1.24 and \( \beta \) = 1.64. The red circles are the experimental data and the black lines are the theoretical fittings. Note that each line has an offset of 0.75 mV. e, \( dI/dV \) values corresponding to the F–A states with the index \( n = 0, \pm 1, \pm 2 \) as a function of \( P \). The colour of each line corresponds to the arrows in a.
Fig. 3 | Phase and frequency dependence of Floquet–Bloch states.
a. Background-subtracted differential conductance (\(dI/dV\)) measured in device 1 as a function of bias voltage \(V\) and phase difference \(\varphi\), with various microwave power \(P\). The first panel shows \(dI/dV\) oscillations taken without microwave irradiation and the others are taken at microwave frequency \(f=12\) GHz with different powers. The theoretical calculation is overlaid as red solid lines. The colour scale ranges of \(-2\) dBm and \(0\) dBm are changed to \([-30\) nS, \(100\) nS] and \([-10\) nS, \(40\) nS], respectively.

\[
\begin{align*}
\frac{dI}{dV} &\equiv \frac{1}{2}\int_0^{\pi/2} |J_n(\beta |\cos \theta|)|^2 d\theta \\
\beta &= \frac{\alpha \sqrt{V}}{\hbar \omega} \\
\Delta &\equiv \frac{1}{2} \int_0^{\pi/2} \frac{d\theta}{|J_n(\beta |\cos \theta|)|^2}
\end{align*}
\]

(b) \(dI/dV\) measured in device 1 as a function of \(V\) and \(P\) at various \(f\). The original ABS peak and its Floquet replicas are denoted by red and black arrows, respectively. 

c. The average voltage spacing \(\langle dI/dV \rangle\) between adjacent peaks in (b) is plotted as a function \(hf\). The errors are estimated by the half-width at half-maximum of background subtracted \(dI/dV\). The red dotted line is a linear fit crossing the origin.

\((dI/dV)_{\text{loc}}\) from the proximity-induced superconductivity in graphene.

First, the contribution of the F–A states can be obtained by solving time-dependent Schrödinger equations of the GJJ under a periodic, unpolarized electromagnetic field. We also used the equilibrium Fermi–Dirac distribution of electrons for the F–A states as an approximation, which is expected to be valid when the electric field is small. The spectral weight of the \(n\)th F–A replica follows the squared Bessel function averaged by polarization angle \(\theta\).

\[
\left|J_n(\beta |\cos \theta|)\right|^2 \equiv \frac{2}{\pi} \cos \theta \int_0^{\pi/2} |J_n(\beta \cos \theta)|^2 d\theta
\]

The red dotted line is a linear fit crossing the origin.
[(dI/dV)] are plotted as a function of P in Fig. 2e, and feature non-monotonic dependence on the power P. This effect is well captured by our theoretical calculations up to about -7 dBm. Our theoretical modelling of the F-A states and dI/dV is largely based on the static solutions for the ABS, and assumes local equilibration of electrons (Supplementary Section 10). The experimental data gradually deviate from the theoretical fitting at P > -7 dBm (β = 1.7); this can be attributed to, for instance, the substantial heating of electrons that leads to a non-Fermi–Dirac distribution of electrons, a change in the DOS of the proximity-induced gap and the enhancement of the pairing gap by the microwaves. (See Supplementary Section 10 for more detailed discussions.)

We discuss the superconducting phase dependence of the F-A states. Figure 3a shows synchronized oscillations of the original ABS and the Floquet replica states with phase difference φ, which further confirms that the F-A states are replications of the original ABS. Similar oscillation of the F-A states is also observed in device 1 (Supplementary Fig. 11). For better visualization of the oscillations, we subtracted the background obtained by moving-averaging dI/dV over several peaks in V (Supplementary Fig. 12). With increasing P, the peak values of the background-subtracted differential conductance (dI/dV)φ of the Floquet replica (n ≠ 0) states became larger, whereas that of the original ABS (n = 0) became smaller (Fig. 2e). In addition, the oscillation amplitude, which corresponds to ∆ABS, becomes smaller with increasing P and vanished at P = 0 dBm. This can be explained by the significant electron heating owing to strong microwave irradiation, such that the electron temperature reaches the critical temperature of the ohmic contacted Al superconductor (Supplementary Fig. 2). Solutions of the time-dependent Schrödinger equation of the GJ under the unpolarized electromagnetic field allowed us to compute the quasi-energy spectrum of the F-A states. We found that our theoretical calculations well reproduced the observed oscillation of the F-A spectrum, as shown in Fig. 3a. Here, dI/dV for V < 0 represents only the replicas of Eφ (φ), as electrons in the superconducting tunnel probe can hop only to replicas of Eφ (φ) that are empty (Supplementary Fig. 13). The detailed features near φ = ±π are discussed in Supplementary Section 14 in detail.

Finally, we discuss the microwave frequency dependence of the F-A states. The power dependence of (dI/dV)ABS at various microwave frequencies is shown in Fig. 3b. For all frequencies, the decreasing intensity of the peak from the original ABS (n = 0, red arrow) with increasing P is accompanied by increasing intensity of the Floquet replica states (n ≠ 0, black arrows), as expected from the sum rule and Bessel function behaviour. The overall peak position shift to lower voltages with increasing P is attributed to the heating of electrons. In Fig. 3c, the voltage spacing eAV between adjacent (dI/dV)ABS peaks is linearly proportional to h/µ with a slope close to unity (1.05 ± 0.11), which confirms the nature of the F-A states.

In sum, we have realized steady F-A states in a Josephson junction device by continuous microwave irradiation without significant heating, and directly measured their energy spectra by superconducting tunnelling spectroscopy. Our technique can be used to engineer a steady chiral Floquet topological state and investigate its physics.24 Previously, pump–probe experiments indirectly illustrated the occurrence of a chiral Floquet topological state by measuring non-quantized, anomalous Hall conductance3 or gap openings in energy spectra2,1,3. By irradiating a circularly polarized microwave to graphene, our setup can readily generate the steady chiral Floquet topological state and also verify its topological character unambiguously. More precisely, the steadiness of the state would enable the simultaneous measurement of its energy spectrum and time-averaged Hall conductivity, the latter of which should be quantized and thus manifests the topology3. We also note that the same setup allows us to investigate the novel proximity coupling between superconductivity and the chiral Floquet topological state, which may serve as a platform for a Floquet analogue of chiral Majorana modes8. Such proximity coupling naturally requires robust superconductivity coexisting with a constant driving, which previous technologies in the optical domains could not achieve. Finally, this technique is not limited to graphene and can be applicable to other low-dimensional topological materials, such as topological insulators and topological semi-metals, for studying and engineering topological Floquet physics. Realization of even more exotic spatiotemporal-driven quantum states by circularly polarized or quasi-periodic driving, which have been pursued only in theory26–30, is now experimentally within reach. Our observations open up new avenues to design, build, measure and exploit the exotic far-from-equilibrium quantum states, and should lead to novel electronic device applications for microwave engineering of condensed matter.

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Methods

Device fabrication

Graphene and hexagonal boron nitride (hBN) flakes were exfoliated on separate silicon oxide wafers. A graphene monolayer was encapsulated by hBN flakes using a dry transfer method. The top hBN was 48 nm and the bottom hBN was 32 nm. The graphene stack was shaped by reactive ion etching with CF$_4$ and O$_3$ plasmas, and the electrodes were patterned by electron-beam lithography and deposited by electron-beam evaporation onto freshly etched edges of graphene. Ohmic-contact electrodes consisted of a 5-nm titanium adhesion layer and a 70-nm Al superconducting layer; the latter of which was deposited with BN-TiB$_2$ crucibles at a rate of 1.5 Å s$^{-1}$. A superconducting tunnel probe electrode consisting of a 70-nm Al layer without any adhesion layers was deposited with BN-TiB$_2$ crucibles at a rate of 0.3 Å s$^{-1}$. The evaporation chamber pressure during the evaporation was kept at less than 1 × 10$^{-7}$ mtorr.

Theoretical calculation of differential conductance

We theoretically calculated the time-averaged differential conductance of the ABS. We first modelled the GJJ without any external driving and obtained the ABS. We then included microwave driving within the graphene region and obtained the Floquet states for a given polarization of the light. Next, we computed the spectral function and differential conductance of the Floquet states using the Floquet–Kubo formula. Finally, we averaged the differential conductance over the polarization for comparison with our experiments where the microwaves were unpolarized. For further details, see Supplementary Fig. 10, Supplementary Section 13.

We first calculated the ABS spectra and corresponding wavefunctions using standard methods. Essentially, the model is equivalent to a ‘Dirac’ particle in a box’ under the superconducting boundary conditions. Formally, the Hamiltonian of graphene with superconducting electrodes is given as the Dirac–Bogoliubov–de Gennes equation. When the GJJ has a finite width along the $y$ direction, an ABS with non-zero $y$-directional momentum can be formed. Such a state requires a long-enough superconducting coherence length so that the proper interference can occur to form a bound state, that is, the ABS, even after multiple specular Andreev reflections while the electron travels along the $y$ direction. However, in experiment, the superconducting coherence length is roughly 500 nm, which is much shorter than the $y$-directional width of the GJJ and is comparable to the space between two superconductors. Thus, the finite coherence length will strongly suppress any modes with finite $y$-directional momentum. Next, we obtain the Floquet states of the GJJ in the presence of the electromagnetic gauge field $A(t)$ as a function of time $t$ in the graphene region. We obtain the Floquet states under the field by directly solving the time-dependent Schrödinger equation. We then calculate the time-averaged spectral function of the Floquet states. The time-averaged spectral function $B_l(\omega)$ of a Floquet state with quasienergy $\xi_l$ is given as

$$B_l(\omega) = \sum_{m=-\infty}^{m=\infty} |J_m(\beta)|^2 \delta(\omega - (\xi_l + m\Omega)),$$

where $J_m(\beta)$ is the Bessel function of the first kind, $\beta = \frac{\epsilon_B}{\Delta_k}$, and $\Omega$ is the frequency of $A(t)$.

Finally, we compute the differential conductance of the Floquet states. Here our starting point is the standard expression for electronic tunnelling current in terms of Green’s functions for electrons in the graphene and superconducting tunnel probe:

$$I = \frac{2e}{h} \int_{-\infty}^{\infty} dt \Phi(t-t') e^{i2\pi \epsilon_B t'} \sum_{l,r} \left[ G_{l,rr}(t,t') G_{r,lr}(t,t') \right].$$

Derivation of the sum rule of differential conductance

Here we describe our derivation of a sum rule for differential conductance, by following methods in the literature that discuss closely related Floquet sum rules. The detailed derivation of the sum rule is given in Supplementary Section 13.

The sum of the time-averaged differential conductance can be computed as

$$S = \int_{-\infty}^{\infty} dV \left( \frac{dI}{dV} \right) = \lim_{V \to \infty} \langle I(V) \rangle - \langle I(-V) \rangle,$$

where $\langle I(V) \rangle$ is the distribution function of the $\tau$-th F–A state at energy $\xi_j$.

Note that $\int_{-\infty}^{\infty} d\omega \sum_{r} B_r(\omega)$ is the sum of the DOSs of the Floquet states; it is known to respect a sum rule and $S$ should therefore satisfy a sum rule (see Supplementary Section 13-3 for details). Finally, invoking the particle–hole symmetry of the overall spectrum, we conclude that $\int_{0}^{\infty} dV \left( \frac{dI}{dV} \right) = \frac{1}{2} S$ also satisfies the sum rule, which was confirmed in our experimental data.

Data availability

The data supporting the findings of this study are available from the corresponding authors upon reasonable request.

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