High-energy quasiparticle injection into mesoscopic superconductors

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At non-zero temperatures, superconductors contain excitations known as Bogoliubov quasiparticles (QPs). The mesoscopic dynamics of QPs inform the design of quantum information processors, among other devices. Knowledge of these dynamics stems from experiments in which QPs are injected in a controlled fashion, typically at energies comparable to the pairing energy. Here we perform tunnel spectroscopy of a mesoscopic superconductor under high electric fields. We observe QP injection due to field-emitted electrons with 10⁶ times the pairing energy, an unexplored regime of QP dynamics. Upon application of a gate voltage, the QP injection decreases the critical current and, at sufficiently high electric field, a field-emission current (<0.1 nA in our device) switches the mesoscopic superconductor into the normal state, consistent with earlier observations. We expect that high-energy injection will be useful for developing QP-tolerant quantum information processors, will allow rapid control of resonator quality factors and will enable the design of electric-field-controlled superconducting devices with new functionality.

Traditional superconducting information-processing devices are actuated by substantial currents (on the microamp or larger scale), including cryotrons, transmons, single-flux quantum processors and others. The challenges of scaling transmon-style qubits to useful numbers of bits have motivated recent work to actuate superconducting particle detectors, and recent progress in microfabricated particle accelerators may ultimately present opportunities for high-energy superconducting microelectronics. A conveniently fabricated, high-energy QP source is therefore of practical and fundamental interest.

We fabricated the device shown in Fig. 1a via electron-beam lithography and in situ double-angle shadow evaporation. We deposited and oxidized a 10-nm-thick aluminium film to form a tunnel probe, followed by a 30-nm-thick titanium film to form a 60-nm-wide nanowire channel and nearby gates.

Figure 1c shows the critical current and tunnel conductance of the titanium wire as a function of applied gate voltage, measured at 20 mK. As the bias current is swept past the zero resistance state at the retrapping and critical current (I), respectively. Application of gate voltage decreases the critical current before eliminating superconductivity, consistent with the results of ref. 4. In the same device and gate range, we measured the tunnel spectrum (at zero current bias along the nanowire), as shown in the right-hand side of Fig. 1c. The spectrum broadens with gate voltage, implying an increase of the QP population.

We analyse the data using a conventional model for tunnel spectroscopy. Tunneling from superconductors can reveal the electronic, magnetic and phonon structure of materials due to the highly non-linear superconducting density of states. The current through a tunnel junction is

\[ I_{TJ} = \frac{g_0}{N_1^{(0)} N_2^{(0)}} \int N_1(E) N_2(E + eV_{bias}) f(E) - f(E + eV_{bias}) dE \]

(1)

where \(g_0\) is the normal-metal conductance, \(N_1(E)\) and \(N_2(E)\) are the densities of states for each side of the junction, \(N_1^{(0)}\) and \(N_2^{(0)}\) are the normal-metal densities of states, \(V_{bias}\) is the bias voltage on the tunnel probe and \(f(E) = (1 + \exp(E/k_B T_{QP}))/2\) is the Fermi function, which defines the QP temperature, \(T_{QP}\). The magnetic field serves as a convenient experimental parameter, in the presence of which each density of states is split into spin subbands, \(N_{\uparrow\downarrow}\), treated separately in the limit of low spin-flip scattering, and given in the Abrikosov–Gorkov model as

\[ N_{\uparrow\downarrow} = \frac{N^{(0)}}{2} \sgn(E) \Re \left( \frac{u_\pm}{(u_\pm - 1)^{1/2}} \right) \]

(2a)

\[ u_\pm = \frac{\epsilon \mp \mu_B B}{\Delta} + \alpha \frac{u_\pm}{(1 - u_\pm^2)^{1/2}} \]

(2b)

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where \( u_\alpha \) are defined implicitly, \( \mu_\alpha \) is the Bohr magneton, \( B \) is the external magnetic field, \( \Delta \) is the superconducting gap and \( \alpha \) is the depairing energy. The depairing energy reflects the typical energy difference between time-reversed electron states. The functions \( N_{\alpha}(E) \) can be obtained analytically by solving equation (2b) for the \( u_\alpha \). The total current for both spin channels is then calculated numerically according to equation (1), and the derivative with respect to \( V_{\text{bias}} \) gives the conductance

\[
G(V) = \frac{dI_{\text{bias}}}{dV_{\text{bias}}} \bigg|_{\Delta \mu_1 \Delta \mu_2, a_0, a_2, B, T_{\Omega}}
\]

for given gap energies, depairing energies, magnetic field and QP temperature. Moreover, in the approximation that the two layers of the junction are in thermal equilibrium, we can calculate the QP fraction for either layer, \( x = \int_0^\infty N(E)f(E)dE/(N(0)\Delta) \). This value is defined as the ratio between the density of occupied excited states, \( \int_0^\infty N(E)f(E)dE \), and the density of Cooper pairs, \( N(0)\Delta \) (ref. 23).

The tunnel conductance thus depends on the convolution of the aluminium and titanium densities of states, causing the peak at the sum-of-gaps seen in Fig. 1b,c. The peak in Fig. 1c does not move greatly when \( I_c \) becomes zero at \( V_c = 37 \) V, indicating that the aluminium gap is much larger than the titanium gap.

We fit the model of equation (3) to the data of Fig. 1c, taking \( T_{\Omega} \) and \( \Delta_{\Omega} \) as free parameters, and fixed \( \Delta_{\Omega} = 320 \) \( \mu \)eV (ref. 23). The resulting \( \Delta_{\Omega} \) correlates with \( I_c \) as plotted in Fig. 1d. We also plot the resulting QP fraction in the aluminium and titanium layers. The accuracy of the fit (see Fig. 1b) and the adherence of \( \Delta_{\Omega}(T_{\Omega}) \) to the self-consistency relation (see Extended Data Fig. 1) indicate that QP injection, rather than a change in the underlying electronic or crystal structure, destabilizes superconductivity.

Nevertheless, it might be argued that a magnetic mechanism may lead to broadening like that observed. For instance, under high electric fields, oxygen ions might accumulate and present a large moment at the surface of the titanium25,26.

Figure 2 explores the effects of magnetic field to characterize the films and consider the possibility of a magnetic gating effect. Figure 2a shows the spectrum at several values of magnetic field. The model accurately captures the orbital depairing and Zeeman splitting. The critical current data (Fig. 2b, left) show a critical magnetic field in the titanium at \( B_s = 0.7 \) T, and inspection reveals a discontinuity at this field in the respective tunnel data (Fig. 2b, right). To fit the model to the tunnelling data, we first consider the region \( 1 \text{T} < B_s < 2.5 \text{T} \), in which the titanium is normal and the aluminium is superconducting. Here the model
has free parameters $\Delta_{\text{Al}}$ and $\alpha_{\text{Al}}$ which follow quadratic trajectories shown as dashed lines in Fig. 2c ($T_{\text{QP}} = 20 \text{ mK}$). These trajectories are used to fit the low-field region, where only $\Delta_{\text{Al}}$ and $\alpha_{\text{Al}}$ are taken to be free parameters. The resulting titanium parameters also follow quadratic dependences, and from the relation $\alpha = (eH_0^2d^2/6h)D_{\|}c/\hbar$ (where $H_0$ is the in-plane field, $H_{\perp}$ is the out-of-plane field, $d$ is the film thickness, $\hbar$ is the reduced Planck constant, $D$ is the diffusion constant, and $c$ is the speed of light in vacuum) we find the diffusion constants $D_{\|} = 0.6 \text{ cm}^2 \text{s}^{-1}$ and $D_{\perp} = 2.4 \text{ cm}^2 \text{s}^{-1}$ with a $2.5^\circ$ field misalignment. The diffusion constants are typical of thin polycrystalline metal films.

Further, Fig. 2c shows the critical current for magnetic fields along all device axes and for both zero and high gate voltages, illustrating the predominantly isotropic nature of the gate effect (complete data are provided in Extended Data Fig. 2). If we try to account for the broadening due to the gate (Fig. 1b) with the depairing parameter $\alpha$ rather than $T_{\text{QP}}$, the best fit does not match the lineshape of the data (Extended Data Fig. 3). By distinguishing between the effect of magnetic fields ($\alpha$) and the effect of the gate voltage ($T_{\text{QP}}$) we exclude any pair-breaking origin of the gate effect that is ergodic in the sense defined by de Gennes.

Next, in Fig. 3, we vary the gate voltage with a fixed field $B_x = 2T > B_{\text{c,Ti}}$. These data admit simpler analysis: with the titanium normal, equation (3) reduces to $g_{\tau_1}(V_{\text{bias}}) \sim \int N_{\text{Al}}(E - eV_{\text{bias}})f(E - eV_{\text{bias}})dE$. The spectra directly represent the thermally broadened aluminium density of states. The observed broadening with $V_{\text{G}}$ confirms the hypothesis of QP generation, and is largely independent of applied magnetic field.

We note two differences between this experiment and classical QP injection experiments. First, the injected QPs originate at energies ($-eV_{\text{c}}$) inaccessible to the spectroscopy; and, second, the injection current is so low in our case it can be difficult to measure. (The model below indicates a current of at most 0.1 nA; see also Extended Data Fig. 4.) Nonetheless, we can examine $x(V_{\text{c}})$ for clues to the underlying transport mechanism. We plot $\log (x_{\text{Al}}(V_{\text{G}}))$ in Fig. 3b, as calculated from the data of Figs. 3a and 1c. The aluminium data roughly follow $x \sim \exp(-V_{\text{c}}/V_{\text{G}})$ for $V_{\text{G}} = 2kV$, which suggests field emission. But, in our geometry, current from
the gate most directly enters the titanium. Consequently, we focus on the QP population in the titanium within the following simple kinetic model.

Given an injection current $I_G$, the number of QP in the titanium portion of the device is approximately

$$n_{QP} = \frac{I_G V_G \tau_{\text{eff}}}{\Delta}$$

where $\tau_{\text{eff}} = \tau_0/x$ is the effective QP lifetime\(^{14}\). That is, each electron from the gate exciting $V_G/\Delta$ thermal QPs which decay in proportion to their density due to recombination. In terms of $x$, the value

$$n_{QP} = x \Omega N(0) \Delta_{Ti},$$

where $\Omega$ is the volume of the injection region. The current between two metals separated by a vacuum can be calculated from the Fowler–Nordheim equation,

$$I = a V^2 \exp(-b/V).$$

Here $a = e S (16 \pi^2 \hbar^2 \phi F)$ and $b = 4 l (2 m \phi^3)^{1/2} / (3 \hbar)$ with $S$ the emission area, $\phi$ the work function and $l$ the separation\(^{27}\). Substituting into equation (4), we expect the following relation:

$$\frac{x^2 \Delta^2}{V_G^2} = A e^{-b/V_G}$$

with $A = a \tau_0/(N(0) \Omega)$. In Fig. 3f, we plot the left-hand quantity (from the data of Fig. 1c) on a log scale versus $V_G^2$ and find a nearly lin-
ear dependence. The dashed line corresponds to $l = 33$ nm and $\phi = 2.3$ eV. The value of $A = 6 \times 10^{-4} \text{V}^{-1}$ accords with the realistic estimates $\tau = 5$ ns, $S = 30 \text{nm} \times 0.6 \mu\text{m}$, $N_{D2}/N_{D1} = 7 \times 10^{18} \text{cm}^{-3} \text{eV}^{-1}$, $\Omega = 4 \mu\text{m} \times 60 \text{nm} \times 30 \text{nm}$, validating the field emission hypothesis and indicating that the current impinges on the superconductor with the energy of the electrode. A model taking into account QP diffusion along the wire is described in Extended Data Fig. 5, and captures the non-linearity in Fig. 3f.

To confirm the results, we study a second device in Fig. 3c, which is similar to the first except that the titanium wire is terminated on one side. In Fig. 3d we compare $g(V_{\text{bias}},B)$ of the two devices and attribute the comparative sharpness of the second device to microscopic differences in the films. The results of fitting the tunnel data in applied magnetic field and electric field are similar to those of the first device (Fig. 3e–f). The higher diffusion constant, $D_2 = 3.2 \text{cm}^2\text{s}^{-1}$, in the second device may contribute to its having a lower $A$ parameter ($A = 6 \times 10^{-5} \text{V}^{-1}$). In addition, several devices with simpler geometries were measured to directly reproduce the results of ref. 1 (Extended Data Fig. 6). In all devices, the effect is found to be stable over week-long time scales.

With sufficient electric field, mesoscopic superconductors can be switched into the normal state with very low quiescent currents. Introducing a tunnel junction, we have directly observed the governing process in which field emission generates QPs. The effect may be suited as a generally applicable switch of superconductivity because field emission minimizes dissipation outside of the channel, in contrast to prior three-terminal superconducting devices\textsuperscript{9}. For quantum information processors, voltage-controlled coupling resonators would have more scalable interconnects so long as the QPs may be suited as a generally applicable switch of superconductivity with simpler geometries were measured to directly reproduce the results of ref. 1 (Extended Data Fig. 6). In all devices, the effect is found to be stable over week-long time scales.

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Methods

The devices are fabricated beginning with a bilayer resist (950 kDa poly(methyl methacrylate), 4% in anisole, on (MMA (8.5) MAA) copolymer (MMA = methyl methacrylate), 11% in ethyl lactate) on dry-oxidized (90 nm SiO2), degenerately doped (001) Si, which is exposed using a 150 kV electron beam pattern generator to create a suspended mask. Then 10 nm of aluminium is thermally evaporated at 30° from normal incidence and oxidized in 100 torr medical-grade air for 30 min, after which 30 nm of titanium is deposited at normal incidence. The devices are cleaned by ultraviolet–ozone treatment (Samco UV-1, 40 °C, 3 min) after acetone liftoff. The zero-field values of the gaps, $\Delta_{\text{Ti}} = 320 \mu eV$ and $\Delta_{\text{Al}} = 50 \mu eV$, are consistent with typical enhancement of superconductivity in environmentally exposed aluminium films and degradation of titanium relative to bulk materials ($\Delta_{\text{Ti,bulk}} = 180 \mu eV$ and $\Delta_{\text{Al,bulk}} = 75 \mu eV$). The diffusion constants measured in the main text correspond to free paths $l = 3D/\nu$ of 1–5 Å, consistent with near-unity residual resistance ratios in these films (RRRAl = 1.3, RRRTi = 1.05). Four terminal measurements of the devices are performed in a dilution refrigerator at 17–22 mK equipped with low-temperature low-pass filtering, using a Keithley 2400 as gate voltage source. To tolerate a slight misalignment in the shadow evaporation process, the gate voltage is applied to the right-hand gate in fig. 1a, as detailed in Extended Data Fig. 4.

To model a given sequence of tunnelling data ($g_B$ versus either $B$ or $V_G$), 400 random initial points in the parameter space are taken, from each of which a gradient descent is performed. The objective function is calculated with 0.1 μeV resolution from equation (3). Subsequent fits start from 100 initial parameter points near to the best of the previous optimization. The results are found to be robust to constraints and details of the fitting procedure. The uncertainties in the fit parameters are typically 2 μeV and are calculated by the delete-m jackknife method. The spectroscopy below the critical field of the titanium consistently contains a fixed normal metal component which we attribute to inhomogeneities in the titanium and incorporate into the model as detailed in Supplementary Fig. 1. Even at the lowest temperatures, the linewidth of $N_L$ exceeds $k_B T$ to an extent attributable to gap anisotropy in aluminium. This is accounted for in the model by uniformly convolving the $N_L$ with a normalized Gaussian of width 18 μeV for device 1 and 9 μeV for device 2. The simple convolution introduces negligible error as compared to a more realistic model in which the gap is distributed uniformly according to the known gap anisotropy in aluminium as shown in Supplementary Fig. 2.

Data availability

The transport data and device images that support the findings of this study are publicly available in the Harvard Dataverse with the identifier doi:10.7910/DVN/LHCDHV. Source data are provided with this paper.

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Author contributions

L.D.A., A.K.S., C.G.L.B., A.T.P., S.H.L. and S.P.H. performed the low-temperature measurements. L.D.A. and A.K.S. fabricated the devices and performed the data analysis. L.D.A. and A.Y. designed the experiments. All authors discussed the results and commented on the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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Extended Data Fig. 1 | Self-consistency calculation. Comparison of the spectroscopy fitting to the self-consistency relation for weak-coupling superconductors. This relation links the zero temperature gap to the critical temperature as \( \Delta_0 = 1.764 k_B T_c \), and the temperature dependence of the gap implicitly through the relation \( \ln \left( \frac{2e^\gamma \omega_D}{k_B T_c} \right) = \frac{\ln \left( \frac{2e^\gamma \omega_D}{k_B T_c} \right)}{\pi} \). Above we plot the titanium gap energy versus \( T_{QP} \), where \( k_B T_c = 0.24 - 0.31 \) K, the grey region, obtained by solving the foregoing equation numerically for those two bounds. The conformity to the quasiparticle population to the self-consistency relation is therefore quite good, which further excludes an exotic dissipationless gate effect. The slight departure of the data from a typical BCS dependence may be due to variations in the non-equilibrium state due to the energy of the impinging electrons, which varies dramatically over this data set. The error bars are calculated as described in the methods section.

\[
\ln \left( \frac{2e^\gamma \omega_D}{k_B T_c} \right) = \frac{\ln \left( \frac{2e^\gamma \omega_D}{k_B T_c} \right)}{\pi}
\]

where \( \gamma \) is Euler’s constant, and \( \hbar \omega_D \) is the Debye energy. For weak coupling \( \hbar \omega_D \gg k_B T \), so that \( \Delta(T) \) is parametrized only by \( T_c \).
Extended Data Fig. 2 | Titanium wire magnetotransport. Resistance of the Ti wire of Fig. 1 as a function of magnetic field applied along x, y, and z directions for zero applied gate voltage (top row) and 35 V (bottom row).
Extended Data Fig. 3 | Broadening mechanism comparison. Tunnel junction conductance vs bias data (dots) for gate voltages from 20 V (most peaked) to 43 V (least peaked) as compared to best fits with $T_{QP}$ as free parameter (lines) and best fits with $\alpha_\Delta$ as free parameter (dashed lines).
Extended Data Fig. 4 | Electric field calculation and current measurements. The device of the main text (a) has a slight asymmetry in the \( y \)-direction. As a result, the left and right gates produce different electric field distributions, the magnitude of which we calculate numerically in (b) for 40 V applied to either gate. As a result of this asymmetry, we use the right gate in the data of the main text, since this most effectively applies field to the Ti. (c) To look for current flow through the gate, we measure gate current in devices identical to the devices in the text, but at 4.2 K. We observe breakdown at 45 - 60 V in such devices, above the region of stable emission in the text. Below breakdown, the small amount of current detected is ohmic and almost certainly takes place in the contacts in this measurement. In the devices of Extended Data Figure 6, measured in a separate dilution refrigerator with high line-isolation, current was measured to be less than 1 pA for \( V_G = 52 \) V, but these devices had a different gate geometry (90 nm SiO\(_2\) back gate). The Fowler-Nordheim model described in the main text predicts current of \(-0.1\) nA at the highest voltage.
Extended Data Fig. 5 | 1D Quasiparticle diffusion model. The minimal model of quasiparticle dynamics presented in the main text does not account for diffusion of QP along the Ti wire, away from the injection region. Correcting this can account for the departure of the data from equation (5) as follows: We assume that the QP distribute along the length of the Ti wire according to $\rho (y) = \rho_0 \int_{-l}^{l} \exp ((y - y_{TJ})^2 / 2D_{Ti} \tau_{eff}) ds$ where $\rho_0$ is such that $\int \rho (y) dy = 1$, and $l$ is the injector length. Moreover, we relax the assumption that $\tau_{eff} = C x^{-2}$. The new model amounts to replacing $A$ on the right hand side of equation (5) with $A_{\rho(y)} = A \rho(y_{TJ}) x_{TJ} \tau_{eff}/(l_{I} w_{TJ})$ where $y_{TJ}$ is the distance from the tunnel junction to the centre of the injection electrode, $l_{I}$ is the length of the injection electrode, $t_{TJ}$ is the thickness of the tunnel junction portion of the Ti wire, $w_{TJ}$ is the width of that portion, and now $\tau_{eff} = \nu \tau_0$. The values $l_{I} = 600 \text{ nm}$, $t_{TJ} = 5 \text{ nm}$, $w_{TJ} = 7 \text{ nm}$, and $\nu = 2.5$ correspond to the curved line in Fig. 3f of the main text, which closely follows the data. However, the junction dimensions here are lower than expected, and a finite element model would be still more realistic. (a) $\rho (y)$ plotted at the relevant gate voltages, with the sharpest distribution corresponding to the highest voltage. (b) $\rho (y)$ evaluated at the location of the detector as a function of the gate voltages of Fig. 3f. (c) The effective lifetime of quasiparticles implied by this model (filled circles) as compared to the minimal model described in the main text (open circles).
Extended Data Fig. 6 | Additional Al and Ti devices. Wide, back-gated critical current devices are measured at 50 mK in the geometry shown in (a). In a device composed of a 10 nm Al film on a 90 nm SiO₂ gate dielectric, the gate effect is observed only at elevated magnetic field. Critical current measurements are performed both at fixed gate voltage while sweeping magnetic field (b), or at fixed magnetic field while sweeping gate voltage (c). Oscillations observed at high $B_x$ are likely to be related to Weber blockade and can be adjusted by the gate voltage. Hysteresis is observed in the gate effect, as can be seen in (d) in which the gate is swept from high to low voltage (over 20 minutes). A reversed behavior occurs when the gate is swept from low to high. (e) A similar Ti device shows a gate effect at zero magnetic field and the hysteresis takes the form of an overshoot of the gate effect (peak at positive $V_G$). The thickness of the Ti here is 30 nm.