Macroscopic quantum tunnelling in spin filter ferromagnetic Josephson junctions

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The interfacial coupling of two materials with different ordered phases, such as a superconductor (S) and a ferromagnet (F), is driving new fundamental physics and innovative applications. For example, the creation of spin-filter Josephson junctions and the demonstration of triplet supercurrents have suggested the potential of a dissipationless version of spintronics based on unconventional superconductivity. Here we demonstrate evidence for active quantum applications of S-F-S junctions, through the observation of macroscopic quantum tunnelling in Josephson junctions with GdN ferromagnetic insulator barriers. We show a clear transition from thermal to quantum regime at a crossover temperature of about 100 mK at zero magnetic field in junctions, which present clear signatures of unconventional superconductivity. Following previous demonstration of passive S-F-S phase shifters in a phase qubit, our result paves the way to the active use of spin filter Josephson systems in quantum hybrid circuits.
Superconductor (S)/Ferromagnet (F) interfaces are a special class of hybrid systems where different ordered phases meet and generate new nanoscale phenomena and new forms of global order\(^1\). A ferromagnetic barrier in a Josephson junction (JJ) generates novel physics\(^3\),\(^4\) and represents a technological key for advances in weak superconductivity, spintronics and quantum computation\(^5\)–\(^7\). Recent interest in ultra-low-power, high-density cryogenic memories has spurred new efforts to simultaneously exploit superconducting and magnetic properties so as to create novel switching elements having these two competing orders\(^8\). S-F-S junctions are expected to shed light on several aspects of unconventional superconductivity, including transport through spin-aligned triplet Cooper pairs\(^4\)–\(^9\),\(^10\)–\(^14\).

The NbN-GdN-NbN junctions investigated in these experiments are spin filter devices\(^1\) with an unconventional predominant second harmonic current-phase relation (CPR)\(^1\)\(^5\). By changing the thickness of the GdN ferromagnetic insulator (FI) barrier it is possible to change its magnetic properties and hence the spin filter efficiency (SFE)\(^1\)\(^6\),\(^1\)\(^7\). In this paper we report measurements on NbN-GdN-NbN junctions for very low and very high SFE values, from almost a few percent up to 90%, with an intermediate value of 60%. By increasing the thickness of GdN we also obtain junctions with lower values of the \(I_R S_e\) product, where \(I_e\) is the critical current and \(R_s\) the normal state resistance. Following the previous demonstration by Feofanov et al.\(^1\)\(^8\) of passive S-F-S phase shifters in a phase qubit, the experiment reported here demonstrates the active quantum potential of S-F-S JJs via the occurrence of macroscopic quantum tunnelling (MQT) in spin filter devices.

**Results**

**Transport properties of spin filter JJs.** The hallmark of the spin filter effect is the decrease in resistance in the \(R\) vs \(T\) curves below the ferromagnetic transition, as shown in Fig. 1a \((T_{Curie} \approx 33\, \text{K} \text{ for GdN})\). SFE is defined as the percentage difference in the tunnelling probability for up/down spin electrons owing to the difference in barrier heights of the up/down spin channel in the FI caused by exchange splitting, so that 100% SFE corresponds to pure tunnelling of one spin sign. See Methods for details concerning the calculation of SFE. Table 1 collects the parameters of the measured junctions. All the NbN-GdN-NbN JJs present hysteresis > 90% in the current–voltage (IV) characteristics, in a wide range of Josephson coupling energies \(E_J = I_c0\phi_0/2\pi\) (where \(I_c0\) is the critical current in the absence of thermal fluctuations and \(\phi_0 = h/2e\) is the quantum flux). For all the measured devices the junction area is about 7 \(\mu\text{m} \times 7 \mu\text{m} \approx 50 \mu\text{m}^2\).

In Fig. 1d, IV curves, measured as a function of temperature \(T\) for the junction with the highest SFE, are reported. The dependence of \(I_c\) as a function of \(H\) at \(T = 4\, \text{K}\) is reported in Fig. 1c. The blue curve is the first measurement of the magnetic pattern, after nominal zero-field cooling. \(I_c(H)\) has then been measured both in the downward direction of the magnetic field sweep and in the upward direction (black and red curves, respectively). The black and red curves show a distinctive shift of the absolute maximum of \(I_c\) from \(-1\) mT to \(+1\) mT, respectively, arising from the hysteretic reversal of the FI barrier\(^1\)\(^9\). The period of \(I_c(H)\) in non-spin filter junctions for the same geometry is twice as large (3 mT), pointing to a largely predominant second harmonic in the CPR in spin filter JJs, as discussed in ref. 16.

**Measurements of switching current distributions.** We have studied the escape rate of the superconducting phase \(\varphi\) as a function of temperature and for different values of the magnetic field, through standard switching current distribution (SCD) measurements\(^2\)\(^0\)–\(^2\)\(^3\). SCDs have been performed for all the
the plasma frequency and the height of the potential barrier, respectively. The normalized bias current in the washboard potential may result in the observation of two critical currents in the CPR16 we have not found evidence of two critical currents, and hence the case of a pure second harmonic (CPR).

Table 1 | Device parameters.

| Barrier thickness (nm) | Spin filter efficiency | $I_{c0}$ ($\mu$A) | $E_2$ (meV) | $I_R^m$ (mV) |
|------------------------|------------------------|-------------------|-----------|-----------|
| 1.5                    | < 30%                  | 820               | 1,700     | 1.0       |
| 1.7                    | < 30%                  | 280               | 570       | 0.9       |
| 1.8                    | 60%                    | 120               | 250       | 0.7       |
| 3.0                    | 90%                    | 60                | 60        | 0.1       |

Spin filter efficiency has been determined at 4.2 K. $I_{c0}$ has been estimated from the fits of the switching current distributions and $I_R^m$ from the IV curves.

Figure 2 | Phase dynamics with strong second harmonic component.

Washboard potential in the case of a strong second harmonic component in the current-phase relation, for different values of the bias current $\gamma$; Both a and b refer to the case $g = 2$, for which two potential wells are present. A and B indicate the lower and the higher potential well, respectively. The blue rectangle in a is magnified in panel b, in which thermal activation (TA; black arrow) and macroscopic quantum tunnelling processes (MQT; red arrow) from the well B at $\gamma = 0.9$ are shown. $o_a$, $o_B$, and $\Delta U_0$ are the plasma frequency and the height of the potential barrier, respectively.

Figure 3 | Switching current distributions. (a) Measurements of switching current distributions (SCDs) from 4 K down to 0.3 K. When increasing the temperature, the histograms move to lower values of switching currents $I_{sw}$ and the s.d. $\sigma$ increases. Each histogram collects $10^5$ switching events. (b) Thermal behaviour of the SCDs from 0.9 K down to 20 mK. The red lines are fits of the probability density of switching, according to equation (2) in the case of a dominant second harmonic component in the current-phase relation (CPR). Below 100 mK the histograms overlap, indicating the transition to the quantum regime, definitely confirmed by their dependence in magnetic field. In panel c the function $o(\gamma)$ is plotted (black curve) and the red line is the value of the function in the limiting case of a pure second harmonic (CPR).

samples reported in Table 1. According to the Resistively and Capacitively Shunted Junction (RCSJ) model24,25, for a JJ with a conventional CPR the dynamics is equivalent to that of a particle of mass $m_\phi$ moving in a washboard potential $U(\phi) = -E_2(\cos \phi + g \sin \phi)$. The particle mass is given by $m_\phi = C(\phi_0/2\pi)^2$ with $C$ the capacitance of the junction. The normalized bias current $\gamma = I/I_{c0}$ determines the tilt of the potential.

The second harmonic component in the CPR, $I = I_1 \sin \phi + I_2 \sin 2\phi$, leads to a modified washboard potential $U(\phi) = -E_1(\cos \phi + g_2 \cos 2\phi + g_1 \gamma \sin \phi)$, $E_1 = \hbar I_1 / 2e$, which may assume the form of a double well for values of $g = I_2 / I_1$ larger than 0.5 (refs 26–29 (see Fig. 2a)). The presence of two wells in the washboard potential may result in the observation of two critical currents in the IV characteristics, since when tilting back the washboard potential the phase particle may be retrapped in one of the potential wells with finite probability27; the case of ‘$\phi$ JJ’s’ with $g < -1/2$ has been recently studied28,29. Measurements of two well-distinguished critical currents constitute a very direct criterion to estimate the $g$ factor27,28. Although spin filter JJs have a strong second harmonic component in the CPR16 we have not found evidence of two critical currents, and hence the case of $\phi$ junction with negative values of $g$ is not considered in this work.

The $\sin 2\phi$ term in the CPR on average lowers the barrier height of the washboard potential without significantly altering the asymptotic expression of the potential barrier for $\gamma$ close to $0$. The second harmonic component in the CPR, $I = I_1 \sin \phi + I_2 \sin 2\phi$, leads to a modified washboard potential $U(\phi) = -E_1(\sin \phi + g_2 \sin 2\phi + g_1 \gamma \cos \phi)$, $E_1 = \hbar I_1 / 2e$, which may assume the form of a double well for values of $g = I_2 / I_1$ larger than 0.5 (refs 26–29 (see Fig. 2a)). The presence of two wells in the washboard potential may result in the observation of two critical currents in the IV characteristics, since when tilting back the washboard potential the phase particle may be retrapped in one of the potential wells with finite probability27; the case of ‘$\phi$ JJ’s’...
For $Q>1$ and $\gamma$ close to 1 the escape rate in the quantum regime is: $\Gamma_q = a_q \exp(-\frac{\Delta U}{k_B T})$, where $a_q = \left(864\pi^2 U/h \omega_p\right)^{1/2}$. The crossover temperature between the thermal and quantum regimes is given by $33$

$$T_{\text{cross}} = \frac{\hbar \omega_p}{2\pi k_B} \left\{ \left[ 1 + \left( \frac{1}{2Q} \right)^2 \right]^{1/2} - \frac{1}{2Q} \right\}. \quad (1)$$

The experimental probability density of switching is related to the escape rate through the following equation $34$.

$$P(I) = \frac{\Gamma(I)}{\Delta I/\Delta t} \exp\left[-\int_0^t \frac{\Gamma(I')}{\Delta I/\Delta t} dt'\right] \quad (2)$$

where $\Delta I/\Delta t$ is the current ramp rate.

The measurements have been performed in a dilution refrigerator, which reaches a base temperature of 20 mK. A full description of the apparatus is given in detail in the Methods section. The bias current of the junction is ramped at a constant sweep rate $\Delta I/\Delta t = 2 \text{ mAs}^{-1}$ and at least $10^4$ switching events have been recorded using a standard technique $35$.

Figure 3a shows a set of SCDs as a function of temperature for the high-SFE JJ reported in Fig. 1. The thermal behaviour of the SCDs is typical of underdamped JJs and the s.d. $\sigma$, which is proportional to the width of the switching histograms, increasing with temperature as expected. Figure 3b shows the SCDs measured below 1 K (black circles). The dependence of the s.d. $\sigma$ on temperature is reported in Fig. 4a (right axis), along with the thermal behaviour of the mean value of the SCDs, $I_{\text{mean}}$, below 0.5 K (left axis). When decreasing the temperature, $I_{\text{mean}}$ increases while $\sigma$ decreases and both saturate at a crossover temperature of about 100 mK. Below this crossover the histograms overlap and the escape process is no longer regulated by thermal fluctuations, indicating the transition to the MQT regime $20,21$.

A further confirmation of the observation of MQT comes from measurements of SCDs in magnetic field. The behaviour of $\sigma(T)$ at $H = 1.1 \text{ mT}$ is shown in Fig. 4b. At this value of the magnetic field, which reduces $I_c$ to half of the value measured at zero field (see the blue squares in Fig. 1c), lower values of $\sigma$ have been measured and $T_{\text{cross}}$ is reduced by a factor $\sqrt{2}$, down to about 70 mK, in agreement with MQT theory $33$. In both cases of 0 and 1 mT, $T_{\text{cross}}$ has been determined by the intersection of the $T^2/3$ curve in the thermal activation regime (dashed green lines in Fig. 4b) and the mean value of $\sigma$ in the MQT regime (black full lines). The measurements in presence of magnetic field prove that the flattening of $\sigma$ at $H = 0 \text{ mT}$ is a quantum effect and is not due to noise or heating in the measurement set-up $20-23$.

**Discussion**

In the literature there are no measurements of SCDs on junctions with a dominant second harmonic component in the CPR. Numerical simulations of the phase dynamics as a function of the damping parameter $Q$, the $g$ factor and the temperature $T$ give the conditions for which a double-well potential effectively behaves in the escape process as a single well (for details see Supplementary Figs 1 and 2 and Supplementary Note 1). Namely, for values of $Q \approx 10$ SCDs with a single peak have been obtained for $g \leq 2$ or in the limiting case of pure second harmonic CPR ($I_c = 0$). In this case the washboard potential changes its periodicity but assumes the form of a single-well potential. Instead, for $g \geq 2$, two critical currents should be observed. In fact, the heights of the two barriers approach each other when increasing the $g$ factor, and the phase may be retrapped in both the potential wells with a finite probability, resulting in a bimodal switching distribution when counting many escape events $27-29$ (see Supplementary Fig. 3 and Supplementary Discussion). As this is not observed,
a pure second harmonic is the only possible explanation consistent with both measurements of magnetic field pattern (see Fig. 1c) and SCDSs. $I_{\alpha}$ can be obtained by fitting the probability density of switching $P(D)$ (red lines in Fig. 3b) in the thermal and quantum regime, $I_{\alpha} = 30.41 \pm 0.05 \, \mu A$. A quite accurate value of the capacitance $C$ can be obtained from the crossover temperature $T_{\text{cross}}$, which depends on $Q$, $C$ and $I_{\alpha}$, see equation (1). By inserting the values of $I_{\alpha}$ and $Q$ in the expression for $T_{\text{cross}}$ we get $C = 4.5 \pm 0.9 \, \text{pF}$. These values lead to $\alpha \approx 14 \, \text{GHz}$. Nevertheless, as shown in Fig. 3c, the function $a(g)$ in the equations for $\Delta U$ and $\alpha p$ is a slowly varying function for $g > 1$; thus, the junction parameters weakly depend on the $g$ factor for high values of $g$.

We expect in future additional insights coming from a comparative analysis with samples with lower SFE. For junction cross-sections of about 50 mm$^2$ the $I_c$ values for such junctions are too high to be in the conditions to observe unambiguously the transition from the thermal to the quantum regime as commonly occurring also in standard S-Insulator-S junctions [20,21,36-39]. Only further advances in fabrication able to insert S-FI-S junctions in cavities and quench architectures will give more refined feedback on the modes of the dissipative domains of the junction and on the triplet component. However, we can infer that the $Q$ values of spin filter junctions are relatively higher than one would naively expect on the basis of the properties of the low-SFE samples, which are characterized by higher values of $I_c$. Concerning the $Q$ values of spin filter JJ's, the reduction in $I_c$ is compensated by the increase of $R_{\alpha}$, as reported in Table 1.

In conclusion, we have demonstrated the occurrence of MQT in NbN-GdN-NbN spin filter JJ's. Spin filtering drives the S-F-S junction in the underdamped regime and in the appropriate window of junction parameters to observe MQT. The SCDSs, together with the period of $I_c(H)$ modulation, provide direct evidence for a pure second harmonic CPR in the junction where MQT was observed. This is clear evidence of unconventional superconductivity, and it is possible that transport occurs by means of a pair of spin-aligned triplet Cooper pairs, which may suppress magnetic sources of decoherence[40,41]. Demonstration of macroscopic quantum phenomena in spin filter devices gives promise for their application in quantum hybrid circuits[18] and also possibly as quiet memories.

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Methods

Determination of SFE. SFE at a particular temperature is calculated from the $R$ vs $T$ curve. SFE at any temperature below the Curie temperature ($T_{\text{Curie}}$) of GdN and NbN is obtained by fitting an exponential to the temperature dependence of SFE ($R_m$) (see Fig. 1c) and SCDs. The SFE $\sigma$ at a given temperature $T$ is obtained by measuring the critical current $I_c$ at temperature $T$.

Set-up for SCDSs. The SCDSs have been measured by thermally anchoring the samples to the mixing chamber of He$^4$/He$^3$ Oxford dilution refrigerator. The bias current is ramped at a constant sweep rate $\Delta I/\Delta t$ of about 2 mA s$^{-1}$. The voltage is measured using a low-noise differential amplifier and is fed into a threshold detector, which is set to generate a pulse signal when the junction switches from the superconducting state to the finite voltage state. This signal is used to trigger a fast voltmeter to record the value of the switching current. This procedure is repeated at least 10$^5$ times at each temperature, which allows to compile a histogram of the switching currents. Filtering is guaranteed by a room temperature electromagnetic interference filter stage followed by low-pass RC filters with a cutoff frequency of 1.6 MHz anchored at 1.5 K, and by a combination of copper powder and twisted pair filters thermally anchored at the mixing chamber of the dilution refrigerator.
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**Author contributions**

D.M., A.P., L.L., M.G.B. and F.T. conceived the experiments, A.P. and M.G.B. designed and realized the junctions; D.M. carried out the measurements; D.M., G.R. and F.T. worked on the theoretical modelling and data analysis; D.M., M.G.B. and F.T. co-wrote the paper. All authors discussed the results and commented on the manuscript.

**Additional information**

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