Controllable 0–π Josephson junctions containing a ferromagnetic spin valve

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Superconductivity and ferromagnetism are antagonistic forms of order, and rarely coexist. Many interesting new phenomena occur, however, in hybrid superconducting/ferromagnetic systems. For example, a Josephson junction containing a ferromagnetic material can exhibit an intrinsic phase shift of π in its ground state for certain thicknesses of the material. Such ‘π-junctions’ were first realized experimentally in 2001 (refs 2,3), and have been proposed as circuit elements for both high-speed classical superconducting computing and for quantum computing4–10. Here we demonstrate experimentally that the phase state of a Josephson junction containing two ferromagnetic layers can be toggled between 0 and π by changing the relative orientation of the two magnetizations. These controllable 0–π junctions have immediate applications in cryogenic memory, where they serve as a necessary component to an ultralow power superconducting computer11. Such a fully superconducting computer is estimated to be orders of magnitude more energy-efficient than current semiconductor-based supercomputers12.

Phase-controllable junctions also open up new possibilities for superconducting circuit elements such as superconducting ‘programmable logic’, where they could function in superconducting analogues to field-programmable gate arrays.

When a superconducting (S) material and a ferromagnetic (F) material are placed in contact with each other, the properties of both materials are modified near the S/F interface. The intriguing nature of this ‘superconducting proximity effect’ in S/F systems arises due to the exchange field in F which imposes a phase shift on the two electrons of a Cooper pair as they propagate across F. Cooper pairs in conventional superconductors consist of two electrons with equal and opposite momenta and opposite spin. When such a pair crosses the S/F boundary, one electron goes into the majority, or up-spin, band in F and the other goes into the minority, or down-spin, band, causing the two electrons to acquire a net centre-of-mass momentum ±q\(\mathbf{Q}\) = ±(\(\hbar k^\downarrow - \hbar k^\uparrow\))\(^\dagger\), where \(\hbar k^\downarrow\) and \(\hbar k^\uparrow\) are the Fermi momenta of the majority and minority bands, respectively13. Alternatively, one can say that the electron pair correlation function oscillates in F with wavevector \(\mathbf{Q}\) perpendicular to the S/F interface. In S/F/S Josephson junctions, those oscillations translate into oscillations between 0-junctions and π-junctions as the F-layer thickness is increased14–17.

Imagine now a Josephson junction with the structure S/F\(_1\)/N/F\(_2\)/S, where F\(_1\) and F\(_2\) may be different ferromagnetic materials14–17. The pair correlation function describing Cooper pairs from the left-hand S accumulates a phase \(\phi_i = Q_i d_i\) while traversing F\(_i\), where \(d_i\) is the thickness of F\(_i\). If the magnetization of F\(_2\) is parallel to that of F\(_1\), then the pair correlation function will accumulate an additional phase \(\phi_2 = Q_2 d_2\) traversing F\(_2\). If, however, the magnetization of F\(_2\) is antiparallel to that of F\(_1\), then the role of majority and minority bands is reversed, and the pair correlation function will acquire the opposite phase, −\(\phi_2\). As shown schematically in Fig. 2a, if we choose \(\phi_1\) to be close to \(\pi/2\) and \(\phi_2 < \pi/2\), then when the layers are parallel, \(\phi = \phi_2 = \phi_1 + \phi_2\) putting the junction into the π-state, and when the layers are antiparallel, \(\phi = \phi_2 = \phi_1 - \phi_2\), putting the junction into the 0-state.

Experimental verification of the prediction outlined above requires performing a phase-sensitive measurement, which we accomplish by fabricating a superconducting quantum interference device, or SQUID, containing two Josephson junctions of the structure described above. The junctions are elliptically shaped with different aspect ratios of 2.2 and 2.8 so that the magnetic layers in the two junctions will have different switching fields. We choose different ferromagnetic materials—one hard for the ‘fixed layer’ and the other soft for the ‘free layer’—so that only the free layer switches its magnetization direction in small applied magnetic fields. The free layer was chosen as Ni\(_{20}\)Fe\(_{80}\) (‘Permalloy’) of thickness 1.5 nm to put the junction close to the 0–π transition (J.A.G., M.A. Khasawneh, B.M.N., E.C.G., R.L., W.P.P.Jr, and N.O.B., in preparation). The fixed layer in the junctions is Ni of thickness 1.2 nm, which should add or subtract a small phase increment15–20. Further information about the materials can be found in Methods. Figure 1b shows a cartoon with the design of our SQUIDs and junctions, as well as the four accessible magnetic states of the junctions. We will use the figure’s labelling convention for the four states as ‘π–π’, ‘0–0’, and so on, corresponding to the states of the two junctions, JJ-1 and JJ-2 respectively. We will show that these labels accurately describe the phase states of the junctions.

We initialize the junctions into the π–π state by applying a large in-plane field of \(H_{\text{ext}} = -2.600\) Oe, which sets all four magnetic layers in the negative direction. We then measure, at zero field, a set of \(I–V\) curves with different values of the current \(I_\text{p}\) through the flux line to observe oscillations in the SQUID critical current as a function of applied flux \(\Phi\). Critical currents are obtained by fitting \(I–V\) curves to the standard form for an overdamped Josephson junction21. Note that the critical currents for the two polarities of applied current, \(I_+\) and \(I_-\), need not be the same. Next we apply a small ‘set’ field \(H_{\text{set}} = 5\) Oe, return the field to zero, and repeat the scan of \(I–V\) curves versus flux. We continue taking small steps in \(H_{\text{ext}}\), each time setting the field back to zero and repeating a full flux scan. Figure 2a shows a three-dimensional plot of \(I_+\) versus

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The SQUID is now in the ‘0–0’ state. Figure 2b shows similar data acquired for $H_m < 0$. Again there are two jumps in the plot, the first occurring at $H_m = 100$ Oe, returning the SQUID to the ‘0–0’ state, and the second at $H_m = 100$ Oe, returning the SQUID to the ‘0–0’ state as at initialization. Taken together, Fig. 2a,b corresponds to a major loop through all four accessible magnetic states of the system. The fact that the magnitudes of the switching fields for $H_m < 0$ are generally larger than for $H_m > 0$ is due to dipolar coupling between the fixed Ni layer and the free NiFe layer in each junction.

Figure 3a shows more detailed data of $I_{cH}$ and $I_c$ versus $H_m$ for four selected values of $H_m$ taken just after each jump. Several features are immediately apparent in the data. First, $I_{cH}$ and $I_c$ never approach zero; rather they oscillate with an amplitude of approximately 85 $\mu$A in all four magnetic states. Second, the oscillations of $I_{cH}$ and $I_c$ are not sinusoidal, but rather have an asymmetric saw tooth or ratchet shape. Third, the maxima in the $I_{cH}$ and $I_c$ data do not line up with each other, so in general $I_{cH}(\Phi) \neq I_c(\Phi)$. All three of these features are well understood\(^{11,22}\), the first is due to the finite geometrical inductance of the SQUID loop, whereas the second and third are due to...
asymmetries in the inductances of the two arms of the loop and in the critical currents of the two junctions.

A simple model of an asymmetric SQUID is shown in Fig. 1c, where \( I_1 \) and \( I_2 \) are the effective inductances of the two arms of the SQUID loop and \( I_1 \) and \( I_2 \) are the currents through each arm\(^{22,23} \). Our SQUIDs have an inductance asymmetry, that is, \( I_1 \neq I_2 \), because the current paths through the two sides of the SQUID have different lengths (see Fig. 1b). Our SQUIDs also have an asymmetry in the junction critical currents because the critical current is different when a junction is in the 0 versus the \( \pi \) state. Asymmetries in the SQUID loop inductances and in the critical currents of the two junctions cause horizontal shifts of the \( I_c \) versus \( \Phi \) data in opposite directions, which change when the critical current in one of the junctions changes. One can remove those shifts from the data by plotting the average magnitude of the critical current, \( I_{cav} = (I_c - I_{cav})/2 \) versus \( \Phi \), as shown in Fig. 3b for the four magnetic states represented in Fig. 3a. The \( I_{cav} \) curves have a variety of shapes depending on how much the \( I_c \) curves in Fig. 3a are shifted with respect to each other. Regardless of the shapes, Fig. 3b shows that the locations of the minima and maxima in \( I_{cav} \) line up with each other, with phase shifts of \( \pi \) between successive curves. Figure 3a also shows independent fits to the \( I_c \) and \( I_{cav} \) data, described under Methods and in the Supplementary Methods, which confirm a \( \pi \) phase shift between each magnetic state. This demonstrates that we have been able to successfully control the phase of our junctions as proposed above.

The results presented in Fig. 3 are reproducible on repeating the whole major loop. In addition, one can obtain a ‘minor loop’ data after initialization by keeping \( H_0 \) between \( +30 \) Oe and \( -35 \) Oe, so that only the free layer of JJ-1 switches its state. We have obtained similar minor loop data from several different devices; the best major loop data were obtained in the device shown here.

In conclusion, we have demonstrated unequivocally a Josephson junction whose ground state can be switched between the 0-state and \( \pi \)-state by reversing the magnetization direction of one magnetic layer contained within the junction. Transitions between these states were verified by detecting the additional phase of the \( \pi \)-state junction within a d.c. SQUID. Phase-controllable Josephson junctions have applications in superconducting electronics based on single-flux-quantum logic. The most obvious application is in superconducting memory\(^{14–17} \). A single S/F/S Josephson junction with controllable critical current amplitude could function as a superconducting memory cell, but one must find a way to address such a memory cell when it is embedded in a large memory array, and the speed at which the junction switches into the voltage state after the ‘read’ current is applied is limited by the small \( I_{Rmin} \) product of the junction. (S/F/S junctions with larger \( I_{Rmin} \) product have been demonstrated\(^24 \), but not with two ferromagnetic layers in a spin-valve configuration.) A solution to both problems is a memory cell based on a SQUID with a phase-controllable Josephson junction\(^25 \), such as the one shown schematically in Fig. 4 (ref. 10). In that cell, the S/F/S junction serves as a passive phase shifter; it has larger critical current than the two S/I/S junctions, so it stays in the supercurrent state during the read operation. The two S/I/S junctions have smaller \( I_c \) but large \( I_{Rmin} \) product, hence they respond quickly to the read current. The state of the memory cell is determined by the critical current of the whole SQUID, which is large when the phase shifter is in the 0-state and small when it is in the \( \pi \)-state. A scheme for addressing individual SQUID-based memory cells embedded in a large memory array has been proposed\(^25 \). Aside from memory applications, there are other single-flux-quantum circuits that already benefit from the use of fixed-phase \( \pi \)-junctions\(^7,8,20 \). One can now start to envisage new types of superconducting circuits using switchable 0–\( \pi \) junctions such as...

**Figure 3** | \( I_c^+, I_c^− \) and \( I_{cav} \) data with fits for all four magnetic states. **a**, Detailed plots of positive and negative SQUID critical currents, \( I_c^+, I_c^− \), versus flux-line current \( I_\Phi \), for the four magnetic states implicated in Fig. 2. The states are labelled (\( \pi-\pi \)) (0–0), and so on, according to the phase states of JJ-1 and JJ-2, respectively. \( I_c^+ \) and \( I_c^- \) both oscillate as a function of \( I_\Phi \), but with a ratchet shape due to the finite and unequal geometrical inductances of the two arms of the SQUID loop. For each magnetic state, the two curves are shifted with respect to each other in opposite directions by amounts that depend on the individual critical currents, \( I_c^+ \) and \( I_c^- \), of the two Josephson junctions. These critical currents will change depending on whether the junction is in the 0 or \( \pi \) state. The solid lines are the result of least-squares fits to the data using the asymmetric SQUID model shown in Fig. 1c, as described in Methods and Supplementary Methods. **b**, Plot of average critical current, \( I_{cav} = (I_c^- - I_c^+) \) versus \( I_\Phi \), for the same four magnetic states represented in **a**. The solid lines are derived from the fits in **a**. Whereas the shapes of the \( I_{cav} \) curves depend on the alignment between the \( I_c^+ \) and \( I_c^- \) curves, the positions of the maximum and minima in \( I_{cav} \) are immune to the shifts in \( I_c^+ \) and \( I_c^- \). This figure shows schematically the \( \pi \) phase shifts in the (0–0) and (0–0) states relative to the (\( \pi-\pi \)) and (0–0) states. The analysis presented in Methods provides unambiguous proof of the \( \pi \) phase shifts.
Figure 4 | Use of a controllable 0–π junction in a memory cell. This SQUID-based memory cell has high critical current when the controllable S/F/S junction is in the 0-state and low critical current in the π-state. If the critical current of the S/F/S junction is larger than those of the two S/I/S junctions, then only the latter switch into the voltage state when the read current is applied, whereas the S/F/S junction acts as a passive phase shifter. The read time of the memory cell, \( t_{\text{read}} = \frac{\hbar}{eI_{\text{c}}} R_N \), is then determined by the faster S/I/S junctions with higher \( I_{\text{c}}R_N \) product. Methods to address a single cell in a memory array are discussed in refs 11, 25.

the ones described here. This should open up new horizons in the nascent field of ‘superconducting spintronics’.

Methods

Methods and any associated references are available in the online version of the paper.

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Author contributions

E.C.G. and B.M.N. contributed equally to this work. The samples were designed by N.O.B., Y.W. and E.C.G. and fabricated by E.C.G. with significant help from B.M.N. and J. Willard for performing the FastHenry simulations, and B. Bi for help with fabrication using the Keck Microfabrication Facility. Preliminary work by Y.W. was supported by DOE-BES under grant #DE-FG02-06ER46341. This research is supported by the Office of the Director of National Intelligence (ODNI), Intelligence Advanced Research Projects Activity (IARPA), via US Army Research Office contract W911NF-14-C-0115. The views and conclusions contained herein are those of the authors.

Additional information

Supplementary information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to N.O.B.

Competing financial interests

The authors declare no competing financial interests.
Methods

Materials. Ni was chosen as the fixed layer material on the basis of earlier work showing that Ni layers of thickness 1.0 or 1.5 nm magnetize in applied fields of 1.0 kOe. Magnetometry measurements of Co/Ni multilayers show that such thin Ni films surrounded by Co on both sides have a remanent magnetic moment of \( M \approx 420 \text{emu.cm}^{-2} \), magnetically 'dead layers' of total thickness \( \approx 0.4 \text{nm} \), and a Curie temperature slightly above room temperature for \( d_{\text{Co}} \approx 1.0 \text{nm} \), and well above room temperature for \( d_{\text{Co}} = 1.5 \text{nm} \) (data taken by C. Klose, unpublished Bachelor's thesis). Permалloy was chosen as the free layer material based on ongoing work in our lab showing good single-domain switching of Ni/Fe nanomagnets embedded inside a Ni/Fe superlattice (a Josephson junction) [R.M.M., E.C.G., R.L., W.P.P.Jr, and N.O.B., in preparation). The nominal sputtering target our lab showing good single-domain switching of NiFe nanomagnets embedded

Sample fabrication. The Josephson junctions and SQUIDs used in this work are fabricated using ultrahigh-vacuum sputtering deposition and standard microfabrication techniques, including photolithography, e-beam lithography, and lift-off. The fabrication process provided by the manufacturer includes high-quality layers of NiFe thin films. After milling, a SiO\(_2\) layer is deposited by sputtering, again using photolithography and lift-off to define the pitch-fork-like pattern seen in Fig. 1b. Initialization of the Ni magnetizations requires applying a large in-plane field \( \mu_0 H = 560 \text{Oe} \), which is small enough not to induce any trapped flux in the Nb lines.

Data analysis. The standard model for an asymmetric dc SQUID is shown in Fig. 1c. The SQUID is characterized by the four parameters: \( I_0 \), \( I_1 \), \( I_2 \), and \( I_3 \), which are the effective inductances of the two arms and the critical currents of the two Josephson junctions. \( I_0 \) and \( I_1 \) are simply related to the geometric inductances of the two arms if the mutual inductance between them is properly taken into account. On the basis of the geometry shown in Fig. 1b, we expect that \( I_2 \gg I_1 \). In our samples \( I_1 \) and \( I_2 \) are fixed, whereas \( I_0 \) and \( I_3 \) change depending on whether the corresponding junction is in the 0 or \( \pi \) state. The externally applied flux \( \Phi \), defined as positive when the magnetic field \( B \) points out of the page. In addition, the SQUID acquires an extra phase shift of \( \pi \), or equivalently an extra flux of \( \Phi /2 \), when one of the two Josephson junctions is in the \( \pi \) state. When both junctions are in the \( \pi \) state, the two additional phase shifts cancel.
currents change. In addition to determining the shapes of the $I_c(\Phi)$ curves, non-zero values of $\beta_L$, $\alpha_L$ and $\alpha_I$ also shift the positions of the peaks in $I_c(\Phi)$, as discussed above: $\Phi_{\text{peak}} = (L_2 I_2 - L_1 I_1) / (\beta_L (\alpha_L + \alpha_I) \Phi_0 / 2).

To reduce the number of free parameters in the fits, we first determined the conversion from $I_c(\Phi)$ to $\Phi$ by fitting the $I_c(\Phi)$ curves shown in Fig. 3b with a simple Fourier series. For the $(0-\pi)$ and $(\pi-0)$ states, a single cosine wave fits the data well, whereas for the $(0-0)$ and $(\pi-\pi)$ states we used a cosine wave plus its second harmonic. From the Fourier series fits to the four data sets we determined that $I_c(\Phi) = (1.115 \pm 0.002) \mu A \times \Phi / \Phi_0$. That conversion factor was then kept fixed in all the ensuing fits. The fitting procedure was as follows. For each magnetic state, the data for $I_c^+ (\Phi)$ and $I_c^- (\Phi)$ were fitted simultaneously. The free parameters in each fit were $L_1$, $L_2$, $\beta_L$, $\alpha_L$, $\alpha_I$ and $\phi_{\text{shift}}$. Although we expected $L_1$ and $|L_2|$ to be equal to each other, the data exhibited small differences between $L_1$ and $|L_2|$, of order a few $\mu A$. The last parameter is the shift of the centre of the pattern relative to zero flux, $\phi_{\text{shift}} = \Phi_{\text{shift}} / \Phi_0$. The values of the dimensionless fitting parameters and the corresponding physical SQUID parameters extracted from the fits to all four magnetic states are shown in the Supplementary Methods. The independent fits to the four magnetic states produce remarkably consistent results for the values of the two SQUID inductances and the critical currents of the two junctions, each in two magnetic states. The physical values averaged over the fits are: $L_1 = 5.68 \pm 0.05 \text{ pH}$, $L_2 = 11.46 \pm 0.12 \text{ pH}$, $I_{c1}^{0} = 565.9 \pm 1.4 \mu A$, $I_{c1}^{\pi} = 292.8 \pm 1.2 \mu A$, $I_{c2}^{0} = 419.5 \pm 0.2 \mu A$ and $I_{c2}^{\pi} = 210 \pm 7 \mu A$. Those values are very close to the preliminary values extracted from the analysis of the peak positions described above. Most importantly, the values of $\phi_{\text{shift}}$ confirm the additional phase shifts of $\pi$ that occur at each magnetic transition. More detail is provided in the Supplementary Methods.

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