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To cite this version:
Alexandre Assouline, Chéryl Feuillet-Palma, Nicolas Bergeal, Tianzhen Zhang, Alireza Mottaghizadeh, et al.. Spin-Orbit induced phase-shift in Bi2Se3 Josephson junctions. Nature Communications, Nature Publishing Group, 2019, 10, pp.126. 10.1038/s41467-018-08022-y. hal-01983043

HAL Id: hal-01983043
https://hal.archives-ouvertes.fr/hal-01983043
Submitted on 16 Jan 2019

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Spin-Orbit induced phase-shift in Bi$_2$Se$_3$ Josephson junctions

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The transmission of Cooper pairs between two weakly coupled superconductors produces a superfluid current and a phase difference; the celebrated Josephson effect. Because of time-reversal and parity symmetries, there is no Josephson current without a phase difference between two superconductors. Reciprocally, when those two symmetries are broken, an anomalous supercurrent can exist in the absence of phase bias or, equivalently, an anomalous phase shift $\phi_0$ can exist in the absence of a superfluid current. We report on the observation of an anomalous phase shift $\phi_0$ in hybrid Josephson junctions fabricated with the topological insulator Bi$_2$Se$_3$ submitted to an in-plane magnetic field. This anomalous phase shift $\phi_0$ is observed directly through measurements of the current-phase relationship in a Josephson interferometer. This result provides a direct measurement of the spin-orbit coupling strength and open new possibilities for phase-controlled Josephson devices made from materials with strong spin-orbit coupling.
I

In Josephson junctions\(^1\), the Current-Phase relationship (CPR) is given by the first Josephson equation\(^2\), \(I(\phi) = I_0 \sin (\phi + \phi_0)\). Time-reversal and spatial parity symmetries\(^3\), \(P_x P_y P_z\) impose the equality \(I(\phi \rightarrow 0) = 0\) and so only two states for the phase shift \(\phi_0\) are possible. \(\phi_0 = 0\) in standard junctions and \(\phi_0 = \pi\) in presence of a large Zeeman field, as obtained in hybrid superconducting-ferromagnetic junctions\(^4\) or in large g-factor materials under magnetic field\(^7,8\).

To observe an anomalous phase shift \(\phi_0\) intermediate between 0 and \(\pi\), both time-reversal and spatial parity symmetries must be broken\(^9\)\(\sim\)\(\sim\)\(\sim\)\(\sim\). This can be obtained in systems with both a Zeeman field and a Rashba spin-orbit coupling term \(H_{R} = \frac{\hbar}{2} (\mathbf{p} \times \mathbf{e}_z) \cdot \sigma\), where \(\sigma\) is the Rashba coefficient, \(\mathbf{e}_z\) the direction of the Rashba electric field and \(\sigma\) a vector of Pauli matrices describing the spin. Physically, these terms lead to a spin-induced dephasing of the superconducting wave-function.

This anomalous phase shift is related to the inverse Edelstein effect observed in metals or semiconductors with strong spin-orbit coupling. While the Edelstein effect consists in the generation of a spin polarization in response to an electric field\(^36,37\), the inverse Edelstein effect\(^24\), also called spin-galvanic effect, consists in the generation of a charge current by an out-of-equilibrium spin polarization. These two magneto-electric effects are predicted also to occur in superconductors as a consequence of a Lifshitz type term in the free energy\(^25,26\). Thus, in a superconductor with a strong Rashba coupling, a Zeeman field induces an additional term in the supercurrent. In Josephson junctions this term leads to the anomalous phase shift\(^20\).

Several designs of Josephson junctions leading to an anomalous phase shift have been proposed theoretically where the Zeeman field can be obtained from an applied magnetic field\(^14,16,20\) or by using a magnetic element\(^11\). These designs include the use of atomic contacts\(^12\), quantum dots\(^13,27\), nanowires\(^17,18\), topological insulator\(^19,28-30\), diffusive junction\(^20\), magnetic impurity\(^21\), ferromagnetic barrier\(^11\), and diffusive superconducting-ferromagnetic junctions with non-coplanar magnetic texture\(^31\).

Experimentally, the anomalous phase shift \(\phi_0\) can be detected in a Josephson Interferometer (JI) through measurements of the CPR. Anomalous phase shifts have been identified recently in JIs fabricated from the parallel combination of a normal ‘0’ and ‘\(\pi\)’ junction\(^22\) that breaks the parity symmetries. Because of its large g-factor \(g = 19.5\)\(^32\) and large Rashba coefficient, Bi\(_2\)Se\(_3\) is a promising candidate for observing the anomalous Josephson effect due to the interplay of the Zeeman field and spin-orbit interaction. In this topological insulator\(^33,34\), the effective Rashba coefficient of the topological Dirac states is about \(\alpha \approx 3\ eV\)\(^35\), while the Rashba coefficient of the bulk states, induced by the broken inversion symmetry at the surface, has a value in the range 0.3–1.3 eV as measured by photoemission\(^36,37\).

As detailed in refs.\(^20,26\) the amplitude of the anomalous phase depends on the amplitude of the Rashba coefficient \(\alpha\), the transparency of the interfaces, the spin relaxation terms such as the Dyakonov-Perel coefficient and whether the junction is in the ballistic or diffusive regime. At small \(\alpha\), the anomalous phase is predicted proportional to \(\alpha^2\), at large \(\alpha\) it should be proportional to \(\alpha\).

In the ballistic regime\(^11\) and for large \(\alpha\), the anomalous phase shift is given by \(\phi_0 = \frac{4\hbar \alpha E_z}{(\hbar v_F)^2}\) for a magnetic field of magnitude \(B\) and perpendicular to the Rashba electric field, where \(E_z = \frac{\hbar}{2} \mu_B B\) is the Zeeman energy, \(L\) is the distance between the superconductors and \(v_F\) is the Fermi velocity of the barrier material. For the Rashba spin-split conduction band with \(\alpha = 0.4\ eV\), \(v_F = 3.2 \times 10^5\) m/s\(^-1\) and junction length \(L = 150\) nm, a magnetic field \(B = 100\) mT generates an anomalous phase \(\phi_0 \approx 0.01\pi\), while for Dirac states\(^33\) with \(v_F = 4.5 \times 10^6\) m/s\(^-1\), \(\phi_0 \approx 0.005\pi\).

In the diffusive regime, the expected anomalous phase shift has been calculated in ref.\(^20\). For weak \(\alpha\), highly transparent interfaces and neglecting spin-relaxation, the anomalous phase shift is given by the relation:

\[
\phi_0 = \frac{\tau m^2 E_z (AL)^3}{3h^2 D}
\]

where \(\tau = 0.13\) ps is the elastic scattering time, \(D = \frac{1}{2} L^2 r\) \(= 40\) cm\(^2\) s\(^-1\) is the diffusion constant and \(m^* = 0.25\) \(m_e\) is the effective electron mass\(^38\).

To test these theoretical predictions, we fabricated single Josephson junctions and JIs from Bi\(_2\)Se\(_3\) thin films of 20 quintuple layers thick, \(\approx 20\) nm, grown by Molecular Beam Epitaxy and protected by a Se layer, see Supplementary Note 1 and Supplementary Figure 1. As described in Supplementary Figure 2, these junctions are in the diffusive regime. From the measurement of the relative phase shift between two JIs with different orientations of the Josephson junctions with respect to the in-plane magnetic field, we observed unambiguously the anomalous phase-shift predicted by Eq. (1).

**Results**

**Current-phase relationship and Shapiro steps.** The JI shown in Fig. 1a consists of two junctions in parallel of widths \(W_1 = 600\) nm and \(W_2 = 60\) nm, respectively. The phase differences \(\phi_1\) and \(\phi_2\) for the two junctions are linked by the relation \(\phi_1 - \phi_2 = 2\pi \frac{e}{\hbar} S\), where \(e = BS\) is the magnetic flux enclosed in the JI of surface \(S\), \(B_z\) is a small magnetic field perpendicular to the sample, i.e. along \(e_\phi\), and \(\phi_0\) is the flux quantum. In this situation, the Zeeman energy is negligible and oriented along the Rashba electric field, which implies \(\phi_0 = 0\). As the critical current \(I_{c1}\) is much higher than \(I_{c2}\), then \(\phi_1 = \pi/2\) and \(I_{c2} = I_{c1} + L_z e_\phi \cos(c B_{z0}) / \omega = 2n S / \phi_0\). Thus, a measurement of the critical current \(I_{c1}\) as function of \(B_z\) provides a measure of the current \(I_{c2}\) as function of \(\phi_0\), i.e. the CPR. From the voltage map as a function of current \(I\) and \(B_z\), shown Fig. 1b, the critical current \(I_{c1}\) is extracted when the voltage across the device exceeds the value \(V_{switch} = 4\) mV, as shown in Fig. 1c. We find that the CPR displays a conventional sinusoidal form \(I_{c1} = I_{c0} \sin (\phi)\), as shown by the fit in Fig. 1d. Furthermore, under microwave irradiation, the JI displays a conventional, \(2\pi\) periodic, Shapiro pattern, as shown Fig. 2, and detailed discussion in Supplementary Note 2.

**Asymmetric Fraunhofer pattern with in-plane magnetic field.** Figure 3b–d show resistance maps dV/dI of a single junction, Fig. 3a, as function of current \(I\) and \(B_{y}\) for different values of an in-plane magnetic field, \(B_{y}\). Figure 3e shows the corresponding critical current curves. A Fraunhofer pattern is observed with the first node located at \(B_{y0} = 1.2\) mT. This value is consistent with the theoretical value \(B_{y0} = \frac{4L_z}{(cW_1)^2}\), using the effective magnetic penetration depth \(\lambda_z = 0.27\) nm and taking into account flux-focusing effects, see Supplementary Note 3. While for \(B_{y0} = 0\), the Fraunhofer pattern is symmetric with respect to \(B_{y0}\), this pattern becomes asymmetric upon increasing the amplitude of \(B_{y0}\). This evolution is shown in the critical current map as a function of \(B_z\) and \(B_{y}\) in Fig. 3f. We observed a much less pronounced asymmetry when we apply the magnetic field in the \(x\) direction as shown in Supplementary Figure 5. Similar behavior has been observed recently in InAs\(^40\) interpreted as the consequence of a
combination of spin-orbit, Zeeman and disorder effects. As described in ref. 3, the generation of an anomalous phase shift requires breaking all symmetry operations \( U \) leaving \( UH(\varphi)U^\dagger = H(-\varphi) \), where \( H \) is the full Hamiltonian of the system including spin-orbit interactions. These symmetry operations are shown in Table 1 together with the parameters breaking those symmetries. This table shows that for a system with a finite spin-orbit coefficient \( \alpha \), finite \( B_y \) is sufficient to generate an anomalous phase shift. However, additional symmetry operations \( U \) leaving \( UH(B_y, \varphi)U^\dagger = H(-B_y, \varphi) \) must be broken to generate an asymmetric Fraunhofer pattern, as shown in Supplementary Table 1. In addition to non-zero values for \( \alpha \) and \( B_y \), disorder along \( y \) direction, i.e. non-zero \( V_y \), is required to generate an asymmetric Fraunhofer pattern. AFM images, as shown in Supplementary Figure 6c, show that the MBE films present atomic steps. Due to the dependence of the Rashba coefficient on film thickness35, phase jumps along the \( y \) direction of the junction can be produced by jumps in the Rashba coefficient and explains the polarity asymmetry of the Fraunhofer pattern. As detailed in Supplementary Note 4, using a simple model, the asymmetric Fraunhofer pattern measured experimentally can be simulated, as shown in Fig. 3g.

**Current-phase relationship with in-plane magnetic field.** To unambiguously demonstrate that an anomalous phase shift \( \varphi_0 \) can be generated by finite spin-orbit coefficient \( \alpha \) and finite magnetic field \( B_y \) alone, a direct measurement of the CPR with in-plane magnetic field is required. To that end, we measured simultaneously two JIs, oriented as sketched in Fig. 4a, differing only by the orientation of the small junctions with respect to the in-plane magnetic field.

The CPRs for the two JIs are measured as function of a magnetic field making a small tilt \( \theta \) with the sample plane, which produces an in-plane \( B_y = B \cos(\theta) \) and a perpendicular \( B_z = B \sin(\theta) \) magnetic field, as sketched in Fig. 4c. In this situation, the critical current for the reference JI changes as \( I_c \propto \cos(\omega_{\text{ref}} B) \) with \( \omega_{\text{ref}} = \frac{2\pi}{\varphi_0^\dagger} \sin(\theta) \) where \( S_{\text{ref}} \) is the surface of the JI. For the anomalous JI, the critical current changes as \( I_c \propto \cos(\omega B) \) with:

\[
\omega = \frac{2mS}{\varphi_0^\dagger} \sin(\theta) + C_{\varphi_0} \cos(\theta)
\]

where \( C_{\varphi_0} = \frac{m^2 g_{\varphi_0}(\omega)}{2e} \) in the diffusive regime.

In Eq. (2), the first term arises from the flux within the JI of area \( S \), the second term arises from the anomalous phase shift \( \varphi_0 = C_{\varphi_0} B \cos(\theta) \).

Figure 4b shows voltage maps for two different orientations \( \theta \), Fig. 4c. At low \( B \), the two JIs are in-phase and become out-of-phase at higher magnetic field, indicating that the frequency \( \omega \) of the anomalous JI is slightly larger than the reference JI. This is also visible on the critical current plot, Fig. 5a, extracted from these voltage maps. To see this more clearly, the average critical current, shown as a continuous line in Fig. 5a, is removed from...
the critical current curve and the result shown in Fig. 5b for the two JJs. On these curves, the nodes at \(\pi(2n + 1/2), n = 0, 1, \ldots\) are indicated by large red (blue) dots for the reference (anomalous) curve. At low magnetic field, the two JJs are in-phase as indicated by the blue and red dots being located at the same field position. Upon increasing the in-plane magnetic field, the two JJs become out-of-phase with the anomalous JJ oscillating at a higher frequency than the reference JJ, as indicated by the blue dot shifting to lower magnetic field position with respect to the red dot. This increased frequency for the anomalous device is expected from Eq. (2) as a consequence of the anomalous phase shift. Supplementary Figure 7a,b shows additional data taken from negative to positive magnetic field, across zero magnetic field. A plot of the phase difference between the two JJs as function of in-plane magnetic field, shown in Supplementary Figure 7c, demonstrates that the two JJs are in-phase at zero magnetic field and reach a dephasing approaching about \(\pi/2\) for an in-plane magnetic field of \(\approx 80\) mT.

One also sees that the oscillation period of both JJs increase with increasing \(B\). As detailed in Supplementary Note 3, this is due to flux focusing that makes the effective area of the JJs larger at low magnetic field. As the effect of flux focusing decreases with the increasing penetration depth at higher magnetic field, the effective areas of the JJs decreases upon increasing the magnitude of the magnetic field and so the period of oscillations increases.

While the two JJs have been fabricated with nominally identical areas, to exclude that the observed difference in frequencies between the two JJs is due to a difference of areas, we plot in Fig. 5c, the frequency ratio \(\frac{\omega}{\omega_{\text{ref}}}(\theta)\) measured at different angles \(\theta\). Because each curve contains several periods \(T_{\text{i}}\), the frequency ratio is obtained from the average between the \(N\) periods ratio as \(\omega/\omega_{\text{ref}} = \frac{1}{N} \sum_{n=1}^{N} \frac{T_{\text{ref}}}{T_{\text{i}}(\theta)}\), where \(T_{\text{ref}}\) and \(T_{\text{i}}(\theta)\) are the \(n\)th oscillation period for the reference and for the anomalous device respectively. This method enables ignoring the flux focusing effect because the ratio is only taken between two periods measured at about the same magnetic field. We find that the experimental data follows the relation:

\[
\frac{\omega}{\omega_{\text{ref}}}(\theta) = \frac{S}{S_{\text{ref}}} + \frac{C_{\psi} \phi_0}{2\pi S_{\text{ref}} \tan(\theta)}
\]

(3)

At large \(\theta\), this ratio is equal to the ratio of areas \(S/S_{\text{ref}} \approx 1\), however, for small \(\theta\), this ratio increases as \(1/\tan(\theta)\), indicating the presence of an anomalous phase shift \(\phi_0\).
Another way of extracting the frequency is described in the Supplementary Note 5 and leads to the same result, as shown in Supplementary Figure 8.

**Discussion**

A fit of the experimental data with Eq. (3), Fig. 5c, enables extracting the coefficient \( \frac{C_\phi \phi_0}{2 \pi \Delta} = 41 \pm 510^{-5} \). Using the expression of \( C_\phi \phi_0 \) in the diffusive regime given above, we calculate a spin-orbit coefficient \( \alpha = 0.38 \pm 0.015 \text{ eVÅ} \). This value of the Rashba coefficient is consistent with the value extracted from Rashba-split conduction band observed by photoemission measurements\(^{36,37}\). Table 2 gives the anomalous phase shift extracted from the critical current oscillations at the largest magnetic field about 80–100 mT. The phase shift is extracted from the magnetic

![Image](https://example.com/image.png)
field difference between the last nodes of the oscillations, indicated by blue and red dots on Fig. 5. At this largest magnetic field, we find an anomalous phase shift \( \phi_{\alpha} \approx 0.9 \pi \) for all three tilt angles \( \theta \). This shows that the anomalous phase shift depends only on the parallel component of the magnetic field as expected. This experimental value is compared with the theoretical values calculated in the ballistic regime, for the Rashba-split conduction states and Dirac states, and in the diffusive regime, for the Rashba-split conduction states. These theoretical values are calculated using \( \Delta = 0.4 \text{eVÅ} \) for the Rashba-split conduction states and \( \Delta = 3 \text{eVÅ} \) for the Dirac states. See main text for the formula and other parameters.

![Table 2 Anomalous phase shifts obtained at \( B \approx 100 \text{mT} \), compared to theory](image)

| \( \theta = 0.1^\circ \) | \( \theta = 0.22^\circ \) | \( \theta = 0.46^\circ \) | Ballistic | Dirac | Diffusif |
|---|---|---|---|---|---|
| \( \phi_{\alpha} \) | 0.88\( \pi \) | 1.01\( \pi \) | 0.85\( \pi \) | 0.01\( \pi \) | 0.005\( \pi \) | 0.94\( \pi \) |

The three first columns show the anomalous phase shifts extracted from the last nodes of the critical current oscillations shown in Fig. 5 for the three curves taken at different angles \( \theta \). The last three columns show the calculated anomalous phase shifts in the ballistic regime, for the Rashba-split conduction states and Dirac states, and in the diffusive regime, for the Rashba-split conduction states. These theoretical values are calculated using \( \Delta = 0.4 \text{eVÅ} \) for the Rashba-split conduction states and \( \Delta = 3 \text{eVÅ} \) for the Dirac states. See main text for the formula and other parameters.

A detailed look at Table 1 shows that the anomalous shift observed here must be the consequence of finite Rashba coefficient and in-plane magnetic field. While Table 1 shows that disorder alone \( V_y \) is sufficient to generate an anomalous phase shift, this disorder-induced anomalous phase shift should exist even at zero magnetic field and should not change with magnetic field. In contrast, as discussed above, we have seen that the two JIs are in-phase near zero magnetic field and become out of phase only for finite magnetic field. Thus, this observation implies that disorder \( V_y \) is absent, which is plausible as the small Josephson junction is only 150 nm \( \times \) 150 nm. In the absence of disorder \( V_y \), Table 1 shows that the only way for an anomalous phase shift to be present is that the coefficient \( \alpha \) be non-zero. Indeed, if \( \alpha \) were zero, the first and third symmetry operations of Table 1 would not be broken even with finite \( B_y \).

To summarize the result of this work, the simultaneous measurements of the CPR in two JIs making an angle of 90° with respect to the in-plane magnetic field enabled the identification of the anomalous phase shift \( \phi_{\alpha} \) induced by the combination of the strong spin-orbit coupling in Bi\(_2\)Se\(_3\) and Zeeman field. This anomalous phase shift can be employed to fabricate a phase battery, a quantum device of intense interest for the design and fabrication of superconducting quantum circuits.

**Methods**

**Sample preparation.** The Bi\(_2\)Se\(_3\) samples were grown by Molecular Beam Epitaxy. The crystalline quality of the films was monitored in-situ by reflection high energy electron diffraction and ex situ by x-ray diffraction, and by post growth verification of the electronic structure though the observation of the Dirac cone fingerprint in angle-resolved photoemission spectra as described in ref. 43. Following growth, the samples were capped with a Se protective layer. The Josephson junctions are...
Fabricated on these thin films with standard e-beam lithography, e-beam deposition of Ti(5 nm)/Al(20–50 nm) electrodes and lift-off. The Se capping layer is removed just before metal evaporation by dipping the samples in a NMF solution of Na2S. In the evaporator chamber, the surface is cleaned by moderate in-situ ion beam cleaning of the film surface before metal deposition. While for standard junctions, an aluminum layer 50 nm thick is usually deposited, we also fabricated junctions with 20-nm-thick electrodes to increase their upper critical field as required by the experiments with in-plane magnetic field. See Supplementary Figure 3 for a lateral sketch of the devices. After microfabrication, the carrier concentration is about 10^19 cm^-3 and the resistivity about 0.61 mΩcm, as shown in Supplementary Figure 2. A comparison between the normal state junction resistance of the order of 20–50 Ω and the resistivity of the films indicates negligible contact resistance, i.e. the junction resistance is due to the Bi2Se3 film between the electrodes.

Measurements details. The values for the normal resistance and critical current values measured on 20 devices are found to be highly reproducible, demonstrating the reliability of our procedure for surface protection and preparation before evaporation of the electrodes. The devices are measured in a dilution fridge with a base temperature of 25 mK. The IV curves are measured with standard current source and low noise instrumentation amplifiers for detecting the voltage across the junctions. The measurement lines are heavily filtered with π filters at room temperature at the input connections of the cryostat. They are also filtered on the sample stage at low temperature with 1 nF capacitances connecting the measurements lines to the ground.

Data availability
The data that support the main findings of this study are available from the corresponding author upon request. The source data underlying Fig. 5c and Supplementary Fig. 8d are provided as Source Data files.

Received: 9 July 2018 Accepted: 12 December 2018
Published online: 10 January 2019

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Acknowledgements

The devices have been made within the consortium Salle Blanche Paris Centre. We acknowledge fruitful discussions with S. Bergeret and J. Danon. This work was supported by French state funds managed by the ANR within the Investissements d’Avenir programme under reference ANR-11-IDEX-0004-02, and more specifically within the framework of the Cluster of Excellence MATISSE. We also thank L. Largeau (C22-Centre de Nanosciences et de Nanotechnologies-Universit Paris-Sud) and D. Demaille (INSP-Institut des Nanosciences de Paris-Sorbonne Universit)) for the atomic-resolved HAADF-STEM images.

Author contributions

H.A. proposed and supervised the project. M.E. and P.A. have grown the Bi2Se3 thin material in this article are included in the article appropriate credit to the original author(s) and the source, provide a link to the Creative Open Access

Additional Information

Supplementary Information accompanies this paper at https://doi.org/10.1038/s41467-018-08022-y.

Competing interests: The authors declare no competing interests.

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Journal peer review information: Nature Communications thanks the anonymous reviewers for their contribution to the peer review of this work. Peer reviewer reports are available.

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