Zeeman effect induced 0-π transitions in ballistic Dirac semimetal Josephson junctions

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Abstract

One of the consequences of Cooper pairs having a finite momentum in the interlayer of a Josephson junction, is π-junction behavior. The finite momentum can either be due to an exchange field in ferromagnetic Josephson junctions, or due to the Zeeman effect. Here, we report the observation of Zeeman effect induced 0-π transitions in Bi1−xSbx, 3D Dirac semimetal-based Josephson junctions. The large g-factor of the Zeeman effect from a magnetic field applied in the plane of the junction allows tuning of the Josephson junctions from 0- to π- regimes. This is revealed by sign changes in the modulation of the critical current by applied magnetic field of an asymmetric superconducting quantum interference device (SQUID). Additionally, we directly measure a non-sinusoidal current-phase relation in the asymmetric SQUID, consistent with models for ballistic Josephson transport.
Introduction

Electrons in a Dirac semimetal possess a linear dispersion in all three spatial dimensions, and form part of a developing platform of novel quantum materials. Bi$_{1-x}$Sb$_x$ is particularly interesting as it possesses a linear dispersion relation either for three-dimensional Dirac bulk states [1] or two-dimensional topological surface states [2] depending on the Sb doping concentration ($x$). Bi$_{1-x}$Sb$_x$ supports a three-dimensional Dirac cone at the Sb-induced band inversion point around $x = 0.03$.

It was recently shown for a Dirac semimetal-based Josephson junction, that the Josephson supercurrent is partly carried by $4\pi$-periodic Andreev bound states [3]. Furthermore, it was shown that the giant g-factor of the electron pocket (the Dirac cone) leads to a large Zeeman effect for an in-plane magnetic field along one of the elongated axes. The considerable shift of the Fermi surfaces in opposite direction for opposite spins provides a large enough finite total momentum of the Cooper pairs to enter the $\pi$-junction regime at low magnetic fields. The Zeeman effect corresponds to a shift in energy of this Dirac cone by $g\mu_B B$. Because of the linear dispersion of a 3D Dirac semimetal, i.e. $E = \hbar v_F k_F$, the cone shifts in $k$-space and a Cooper pair acquires a finite momentum of $\Delta k = \frac{g\mu_B B}{\hbar v_F}$. Such a finite momentum leads to sign changes of the superconducting pair potential [4–6] when $\Delta k L$ increases (where $L$ is the length of the piece of superconductor), giving rise to sign changes of the supercurrent when embedded in a Josephson junction.

A $\pi$-junction can be the energetic ground state for a large variety of physical reasons, such as the presence of magnetic field [4, 5], magnetic impurities [7], magnetic exchange fields in superconductor-ferromagnet-superconductor Josephson junctions [8], unconventional order parameter symmetries, such as in corner junctions employing the d-wave order parameter symmetry in high-$T_c$ cuprates [9], and non-equilibrium population of quasiparticles [10]. Only recently the scenario of the Zeeman effect induced $\pi$-junction was verified experimentally for junctions with the topological insulator, HgTe [11], or the Dirac semimetal, Bi$_{1-x}$Sb$_x$ [3], as interlayer. Whereas Li et al. showed oscillations in the supercurrent as $\Delta k$ increases with the magnetic field, the associated sign changes between the 0 and $\pi$ states have not yet been revealed.

Here, we report the observation of 0–$\pi$ transitions by incorporating a Zeeman $\pi$-junction in an asymmetric superconducting quantum interference device (SQUID), by which we can
measure the current-phase relation (CPR) of the junction directly. The in-plane magnetic field can shift the phase of the SQUID oscillations of the out-of-plane magnetic field by $\pi$. Moreover, the CPR was measured to be non-sinusoidal, which shows to be quantitatively consistent with models for ballistic Josephson junctions.

**Ballistic Josephson junction in a SQUID configuration**

We fabricated a direct current (dc) superconducting quantum interference device (SQUID) to measure the CPR in a Dirac semimetal-based Josephson junction. For this purpose Bi$_{0.97}$Sb$_{0.03}$ single crystals were grown using a modified Bridgeman method (see Ref. [3] for details). Thin flakes were exfoliated and put on a Si wafer with a SiO$_2$ oxide layer on top. Nb leads were fabricated by e-beam lithography and lift-off. The Dirac semimetal Josephson junction is incorporated in a square Nb loop with an area of about 1.2 $\mu m^2$. As the second junction in the SQUID we made a Nb constriction that acts as a reference junction with a critical current of about $I_r = 7 \mu A$, much larger than the Dirac semimetal critical current of $I_s = 1.2 \mu A$. When the SQUID is strongly asymmetric ($I_s \ll I_r$), the modulation in the critical current is representing the CPR of the sample junction [12].

The shape of the $I_c(\phi)$ curves can be affected by the inductance of the superconducting loop. The inductance parameter is defined as $\beta_L = \frac{2\pi LI_c}{\phi_0}$. At low temperature, the inductance $L$ is dominated by the kinetic inductance ($L_k$) in superconducting devices. We estimate the $L_k$ in our experiment as $L_k = \left(\frac{m}{2n_s c^2}\right)(\frac{l}{A}) \lesssim 1pH$ at 10mK, where $m$ is the Cooper pair mass, $n_s$ is the density of Cooper pair, $l$ is the length of the Nb wire and $A$ is the cross-section of the wire. In general the superconducting loop in our experiment is very small. We then find $\beta_L \ll 1$, for which the deformation of the CPR by the inductance effect is negligible.

In Fig[1], we show the measured CPR when the parallel field $B_{//} = 0$. In an asymmetric SQUID, the total critical current does not drop to zero but oscillates around the $I_r$. To better capture the CPR of the sample junction, we subtract the background current, obtained by a strong smoothing. A clear saw-tooth shaped CPR is observed in our experiment.

The CPR in an SIS or SNS junction in different limits has been intensively studied. In the tunneling region, the CPR is represented by $I_s = I_c \sin(\phi)$, where $\phi$ is the phase difference between the two superconducting electrodes and the non-sinusoidal CPR was found in both
FIG. 1: Ballistic junction in SQUID configuration. (a) The extracted critical current $I_c$ of the Bi$_{0.97}$Sb$_{0.03}$ junction as a function of the phase across the junction at 20 mK. The sawtooth shape indicates the higher harmonics components in the supercurrent. The dashed blue lines indicate the maximum and the minimum of the critical current, which we used to estimate the skewness of the CPR shape in the main text. The black dashed line is the fitting curve with different weight of $2\pi$-periodic and higher modes supercurrent. (b) Scheme of the sample configuration. The crystalline orientations are indicated in the orange box. The contacts are always aligned in such way that the current flow is along the bisectrix axis of the Bi$_{0.97}$Sb$_{0.03}$ crystals. (c) Fourier transform of the periodic $I_c(B)$, higher order harmonics are visible.

the dirty [14] and the clean limit [15] of the Kulik-Omelyanchuk model (see also the review of [16]). With a perfect transparency ($D = 1$), the CPR of the point contact junction in the clean limit is just a segment of a sine function. The non-sinusoidal effect is strongest at high transparency and decreases as the interface becomes more opaque, until the CPR eventually becomes sinusoidal at very low transparency. Later, the theory was extended to the SNS junction, with a normal metal material as the interlayer. It was shown that, for a long SNS junction (i.e. $\xi_0 \ll d \ll \xi_T$, where $\xi_0 = \hbar v_F/\Delta$ is the superconducting coherence
length in the normal metal and $\xi_T = \xi_0 T_c/T$ is the thermal coherence length), the CPR becomes linear within one period: $I_s(\phi) = eN_2v_2\Delta_1\{\frac{\phi}{2} - \pi \text{Int}\left[\frac{\phi}{2\pi} + \frac{1}{2}\right]\}$. Thus, in clean SNS junctions, the CPR has a saw-tooth shape at low temperature. If disorder is included (diffusive/dirty limit), then the CPR will be smoother and closer to a sinusoidal shape. The current amplitude also decreases rapidly with the increase of temperature and the saw-tooth shape changes gradually to a sinusoidal shape [13].

To fit the data, we used a model [23] that is based on the Eilenberger equations for ballistic transport at arbitrary junction length and arbitrary interface transparencies. We first fit the temperature dependence of the extracted critical current amplitude $I_c(T)$ (Fig.2a). A very high transparency $D \simeq 0.98$ and induced superconducting gap $\Delta \simeq 4.5 K$ are obtained. The coherence length $\xi_s \sim 100 nm \sim L$. Then, using the same parameters, we plot the simulated CPR data with the experimental data at different temperatures (Fig.2b-f). We find a good and self-consistent agreement between the simulated and experimental results, confirming the ballistic transport in the junction.
FIG. 2: Current-phase relation at different temperature. (a) Temperature dependence of the extracted critical current amplitude $I_c(T)$. Black line, fitting curve. (b)-(f) Current-phase relation at different temperatures. Black lines, fitting curves (using same fitting parameters from (a))

**Critical current oscillations and Zeeman effect-induced $\pi$-junction**

In Fig 3a we show the CPR for parallel magnetic fields (along the current flow direction in the junction). In the 2D color plot, horizontal and vertical lines are drawn to indicate the nodes of the oscillations in parallel and perpendicular field respectively. In Fig 3b, the CPRs for different parallel field values (indicated by arrows) are plotted. From these CPRs, it is apparent that at specific $B_\perp$ values, the CPR changes from a maximum to a minimum, suggesting a $\pi$ phase-shift each time.

In a previous work [3], it was observed that, in a single Bi$_{0.97}$Sb$_{0.03}$ Josephson junction, the critical current oscillates with the applied parallel magnetic field (along the z-axis). Such oscillations can be attributed to a finite-momentum pairing mechanism. Similar phenomena were extensively studied in superconductor-ferromagnet-superconductor (SFS) junctions:
the critical current oscillates with increasing ferromagnetic interlayer thickness due to sequential 0-\(\pi\) transitions. In SFS junctions, the exchange energy \(E_{ex}\) is characteristic for the ferromagnetic layer. We can describe our S-Bi\(_{0.97}\)Sb\(_{0.03}\)-S junction with the same formalism by replacing the exchange energy \(E_{ex}\) by the Zeeman energy \(E_z = g\mu_B B\). When a magnetic field is applied on Dirac semimetals, the Zeeman term shifts the two Weyl cones in opposite directions in momentum space, due spin-momentum locking. Such a change in band structure introduces a phase shift in Cooper pair potential\([3]\). This Zeeman shift is illustrated in Fig.3d.

The g-factor of the electron pocket at the L-point in Bi is found to be highly anisotropic and extremely large along the bisectrix \([21, 22]\). Since the Zeeman effect in Bi\(_{0.97}\)Sb\(_{0.03}\) is very similar to that in pure Bi, we use a value typically found in literature \((g \sim 800 - 1000\) along the highly elongated direction). Because of this large g-factor, the Zeeman energy is already relevant at very low magnetic fields.

The critical current changes with \(B\) as \(I_s(B) \propto e^{-B/B_{eff}},\) where \(B^{-1}_{eff} = B_1^{-1} + iB_2^{-1}\).

Here, the two characteristic field scales are \(B_1\), which corresponds the amplitude decay of \(I_c\), and \(B_2\), which describes the period at which the junction alternates between the 0- and the \(\pi\)-states. In the clean limit (\(B_1 \gg B_2\)), we expect no decay in the critical current amplitude, while in the diffusive regime (\(B_1 \sim B_2\)), the oscillations are periodic in \(\sqrt{B}\). In the intermediate regime, the critical current can be described as a cosine oscillation with an exponential decay: \(I_s(B_{||}) \propto e^{-B/B_1} \cdot \cos (B/B_2).\) Fitting this expression for \(I_s(B_{||})\) to the data, as in Fig.3c, we obtain that \(B_1 = 41mT\) and \(B_2 = 3.2mT\). This indicates that the junction is almost in the clean limit.
FIG. 3: **Zeeman effect induced 0-π transition.** (a) 2D color-plot of the Current-phase relation of Bi$_{0.97}$Sb$_{0.03}$ junction in a parallel magnetic field. The π-shift in phase occurs periodically as the parallel field ($B_{//}$) increases. The horizontal dashed lines separate the map into different 0 or π regions. (b) CPR curves at different parallel field values (indicated by the arrows). The junction alternates between a 0 or π state. (c) Extracted $I_s$ as a function of the parallel field $B_{//}$. Blue circles: $I_s(B_{//})$ at $B_{\perp} = 0$; gray dashed line: fitting curve, exponential decay; red solid line: fitting curve, decay+oscillation. (d) Schematic illustration of the Zeeman effect induced finite-momentum pairing. At zero field (upper panel), the Dirac Fermi surface (electron pocket at L-point) is spin degenerate. As an in-plane field is applied along the large g-factor direction (lower panel), the Dirac Fermi surface splits with polarized spin. Consequently, the formed Cooper pairs acquire a finite total momentum, driving the 0 − π transition.

**Discussion**

Interestingly, we found that the critical current amplitude of the first π-state is smaller than the 0-state. And the shape of the CPR is gradually changing from the regular CPR as a function of magnetic field. This deformation of the CPR during the 0 − π transition has been observed in superconducting quantum dots and is understood as a combination of the 0- and π-states as the intermediate transition state [18] [19]. This coincides with the fact that part of the contribution to the critical current is from the hole pocket at the T-point in Bi$_{0.97}$Sb$_{0.03}$. The in-plane g-factor of the hole pocket is very small (< 2) [20]. Therefore
the Zeeman effect in this channel should be negligible and no $0 - \pi$ transition should occur in our measured field range. This may explain why the critical current amplitude at the $\pi$-state (which originates from the electron pocket) dominates the total critical current and is slightly reduced at low field in $\pi$-state.

Conclusion

In summary, by incorporating a S-Bi$_{0.97}$Sb$_{0.03}$-S Dirac semimetal Josephson junction into an asymmetric SQUID, we have been able to measure the CPR as well as the $0 - \pi$ transitions associated with the finite momentum of the Cooper pairs due to the Zeeman effect in parallel magnetic field. The junction critical current as a function of temperature as well as the CPR at all temperatures were self-consistently fitted by models for ballistic transport.

[1] H.J. Kim et al., Phys. Rev. Lett. 111, 246603 (2013).
[2] D. Hsieh et al., Nature 452, 970 (2008).
[3] Chuan Li et al., arXiv:1707.03154 (2017).
[4] P. Fulde, R.A. Ferrell, Phys. Rev. 135, A550 (1964).
[5] A.I. Larkin, Y.N. Ovchinnikov, Zh. Eksp. Teor. Fiz. 47, 1136 (1964).
[6] E.A. Demler, G.B. Arnold, M.R. Beasley, Phys. Rev. B 55, 15174 (1997).
[7] L.N. Bulaevskii, V.V. Kuzi, A.A. Sobyanin, JETP Letters 25 290 (1977).
[8] V.V. Ryazanov, V.A. Oboznov, A.Yu. Rusanov, A.V. Veretennikov, A.A. Golubov, J. Aarts, Phys. Rev. Lett. 86, 2427 (2001).
[9] D.J. Van Harlingen, Rev. Mod. Phys. 67, 515 (1995).
[10] J.J.A. Baselmans, A.F. Morpurgo, B.J. van Wees, T.M. Klapwijk, Nature 397, 43 (1999).
[11] S. Hart et al., Nature Phys. 13, 87 (2017).
[12] Della Rocca, M.L. et al. Measurement of the current-phase relation of superconducting atomic contacts. Phys. Rev. Lett. 99, 127005 (2007).
[13] J. Bardeen and J.L. Johnson. Josephson current flow in pure superconducting-normal-superconducting junction. Phys. Rev. B 5, 72 (1972).
[14] Kulik, I. O., and A. N. Omelyanchuk. Current flow in long superconducting junctions. Sov.
[15] Kulik, I. O., and A. N. Omelyanchuk. Properties of superconducting microbridges in the pure limit. *Sov. J. Low Temp. Phys.* **3**, 459 (1977).

[16] A.A.Golubov *et al.* The current-phase relation in Josephson junctions. *Rev. of Mod. Phys.* **76**, 411 (2004).

[17] Chikara Ishii. Josephson Currents through Junctions with Normal metal Barriers. *Prog. Theor. Phys.* **44**, 1525 (1970).

[18] R.Delagrange *et al.* $0 - \pi$ Quantum transition in a carbon nanotube Josephson junction: Universal phase dependence and orbital degeneracy *Physica B* **236**, 211 (2018).

[19] Romain Maurand *et al.* First-Order $0-\pi$ Quantum Phase Transition in the Kondo Regime of a Superconducting Carbon-Nanotube Quantum Dot *Phys. Rev. X* **2**, 011009 (2012).

[20] G.E.Smith, G.A.Baraff and J.M.Rowell, Effective $g$ factor of electrons and holes in Bismuth. *Phys. Rev.* **135**, 4A (1964).

[21] L.M.Roth, B.Lax and S. Zwerdling, Theory of Optical Magneto-Absorption Effects in Semiconductors. *Phys. Rev.* **114**, 90 (1959).

[22] Zengwei Zhu, *et al.* Angle-resolved Landau spectrum of electrons and holes in bismuth. *Phys. Rev. B* **84**, 115137 (2011).

[23] Artem V. Galaktionov and Andrei D.Zaikin, Quantum interference and supercurrent in multiple-barrier proximity structures. *Phys. Rev. B* **65**, 184507 (2002).