Unusual Superconducting Proximity Effect in Magnetically Doped Topological Josephson Junctions

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The transport properties of a topological Josephson junction fabricated from a magnetically doped topological insulator (TI) were investigated. The conductance spectra of the Nb/Fe- Bi₂Te₂Se/Nb junction below 1 K showed an unusual trident-shaped zero bias conductance peak with a tiny peak width of \( \sim 6 \mu V \). The central peak of the trident peak represents the dc-Josephson current, and the side peaks may reflect an induced unconventional Cooper pairing. Additionally, unsaturated temperature dependence of critical current under 1 K may reflect the presence of an unconventional proximity effect. Furthermore, microwave irradiation derived a drastic change in the conductance spectra from the peak structure into oscillatory signals, a hallmark of the ac-Josephson supercurrent. The phase relation of the ac-Josephson effect under high power radiofrequency-irradiation was found to be \( 4\pi \)-periodic. The results suggest that the junction based on magnetically doped 3D TIs may realize an unconventional Cooper pairing, thus enabling access to the basic physics of Andreev bound states and unconventional superconductivity.

INTRODUCTION

Probing S/I/N/I/S junctions (S: superconductor, I: Insulator, N: normal metal) is one of the powerful tools to investigate the existence of Andreev bound states, and phase relations in unconventional superconductor candidates [1, 2]. When the barrier height of the insulator is high, namely an S/I/N junction condition, the conductance spectra of the junction reflect density of states (DOSs) of quasiparticles [3], while, Josephson supercurrent emerges in a low barrier condition (an S/N/S junction) [4]. In a conductance measurement, an unconventional superconductor is expected to show an unique peak structure called a zero bias conductance peak (ZBCP), reflecting symmetry of superconducting (SC) gap functions, and surface Andreev bound states (ABSs) [5-8]. On the other hand, the appearance of \( 4\pi \)-periodic Josephson supercurrent is expected in the topological Josephson junction [9]. Recently, such signature behaviors are observed in nanowires with strong spin orbit interaction (SOI) [10] and spin Hall insulator systems [11,12]. The results thus found to emulate \( p \)-wave superconductors, and thus Majorana fermions, whose antiparticles are identical as the fermions themselves [13], have been expected to emerge at the edges of the \( p \)-wave superconductors [14].

In the context of searching for unconventional SC states, TIs are promising material systems for the following three reasons. First, theories predict emergence of the proximity-induced chiral \( p \)-wave like SC states possessing the time-reversal symmetry (TRS), as described in ref. 15 and Fig.1(e), and the Majorana fermions on the 2D interface of TI/S. Indeed, there have been several studies that report observing ZBCP similar to that observed in unconventional superconductors [16-20]. More recently, a sign-changing order parameter implying the chiral \( p \)-wave component has also been reported [21].

Second, the 2D surface of 3D-TI is favorable for tuning experimental set up and configurations, for future braiding operations accomplished by complicated electrodes on chips [22-26]. Finally, though unconfirmed, odd-frequency Cooper pairing may exist at the boundary of the TI/S. In symmetry classification, Cooper pairs can be categorized into four types in accordance with their frequencies, spin and orbital of pairing. In fact, there exist two classes of odd-frequency pairing: Odd-Singlet-Odd (OSO) and Odd-Triplet-Even (OTE) [27]. These odd-frequency Cooper pairs are related to the physics of ABSs and Majorana fermions [27, 28]. Owing to strong SOI of TIs, spin-momentum locking on the Dirac cone are supposed to suppress the conventional s-wave (ESE) pairing and as a result, other unconventional components are expected to emerge at the interface of TI/S.

Accessing magnetic interaction is another pathway to realize unconventional superconductors. Strong magnetic fields or direct exchange interactions usually induce Zeeman splitting, and thus stabilize the triplet components instead of the conventional s-wave pairing. In an ultimate case, a dominant OTE pairing is theoretically deduced for an S/half-metal/S junction [29], and has been confirmed experimentally [30]. However, an eccentric phenomenon derived from the dominant OTE component is yet to be reported.

Combination of magnetic moments and spin-momentum locking in TIs may lead to rather robust unconventional Cooper pairs. In fact, as per a theoretical
study, majority of components depend on the alignment of magnetic moments; in-plane magnetic moments lead to OTE pairing [31]. However, a junction consisting of magnetic TI has not been explored so far.

In this paper, we report the transport properties of an S/Ferromagnetic-TI(FTI)/S junction fabricated from a magnetically-doped Bi$_2$Te$_2$Se having high bulk insulating properties. Owing to an accidentally realized moderate gap length and barrier height, both conductance spectra and Josephson current were recorded. The spectra showed an unusual trident-shaped ZBCP, and 4π-periodic Josephson supercurrent which suggests the existence of unconventional Cooper pairing caused by proximity effect in the magnetic TI junction.

**EXPERIMENT**

S/FTI/S junctions, as described in Fig. 1(a), were fabricated using exfoliated single crystals of Bi$_{1.5}$Fe$_{0.1}$Te$_2$Se (Fe-BTS), followed by Nb-sputtering. The Fe-BTS crystals were grown by a modified Bridgman method [32]. Ferromagnetism of the crystals thus obtained was checked using a commercial SQUID magnetometer (Quantum Design: MPMS-XL). The crystals were exfoliated onto a Si substrate. Larger SC-electrodes were made by sputtering with Nb (50 nm)/Ti (10 nm). The narrower SC-parts of Ti (5 nm)/Nb (90 nm)/Ti (5 nm) were deposited onto the crystal via electron beam lithography and lift-off technique. As a result, S/FTI/S Josephson junctions with approximately 100 nm length gap and 300 nm width, were obtained as shown in Fig. 1(b). The junction between terminals 1 and 3 was mainly measured in this paper. The transport properties of the junctions were measured by a standard lock-in technique with modulation frequency of 761.1 Hz. Microwave (radio frequency: rf) was irradiated via a loop antenna with diameter of $\sim$ 1 cm.

**RESULTS AND DISCUSSIONS**

The crystals of Fe-doped Bi$_2$Te$_2$Se thus grown show n-type conduction with high resistivity ($\sim$ 20 m$\Omega$cm at 300 K), a low carrier density of $5.6 \times 10^{18}$ cm$^{-3}$, and a bulk carrier mobility of $\mu_b \sim 56$ cm$^2$/Vs. These values ensure high bulk insulating nature of Fe-BTS, compatible with a typical large bulk insulating TI of Bi$_2$Te$_2$Se [33, 34]. Fig. 1(c) shows substantial ferromagnetic magnetization of the crystals similar to that of Fe-doped Bi$_2$Se$_3$ [32], indicating that magnetically doped TI with high resistivity was established. The magnetic moments of bulk crystals are expected to align along the c-axis similar to the Fe-doped Bi$_2$Se$_3$ [32]. BTS has the Dirac cone with band gap ($E_g = 215$ meV) [35] and broken TRS by Fe-doping leads to a gap opening at the Dirac point ($E_{mag} \sim 40 - 50$ meV) [32]. The electronic band structures of Fe-BTS displaying similar transport properties to those of the non-doped BTS, are estimated to have massive Dirac fermion states with high bulk insulating gap, and the Fermi level, $E_F$, may also be located above the gaped Dirac point ($E_g > E_F > E_{mag}$).

The fabricated S/FTI/S junctions exhibit metallic temperature dependence as shown in Fig. 1(d), and the measured resistances are reasonably consistent with the expected values for bulk crystals within an order of magnitude. A clear two-step transition ($T_{c1} = 7.4$ K, and $T_{c2} = 6.0$ K; both are lower than critical temperature of bulk Nb, $T_c$(Nb) = 9.2 K [36]), is widely observed in the Nb-based junctions, and is a hallmark of existence of the proximity effect. Note, the resistance below $T_{c2}$ was not zero but $\sim$ 1.0 $\Omega$, even at the lowest temperature of $T = 0.55$ K. We confirmed limited contribution of measurement condition such as noise to the finite resistance, and concluded that the resistance represents unique nature of the junction.

The small finite resistance suggests that coherence length $\xi$ of the proximity effect in Fe-BTS, is comparable with the half junction length ($\xi \sim L/2 = 50$ nm). Figure 1(e) shows a schematic model of possible current situation in the proximity effect. Similar to the ref. 15, induced triplet pairing is located at the surface and far from the electrodes, reflecting slightly longer coherence length than that of s-wave. In this picture, resistance should be originating from the normal state between the areas of triplet pairing. The small finite resistance indicates that the existing normal area should also be narrow, and thereby the dominant pairing may be p-wave. On the other hand, non-doped TIs based on Bi$_2$X$_3$ ($X = $ Se, Te) reportedly have long-range coherence length $\sim$
Anomalous features, on the other hand, were discovered small energy scale. The inset of Fig. 2(a) shows the hysteresis is derived from the conventional Ambegaokar-Baratoff theory [39]. Furthermore, other junctions did not show this hysteretic jump (not shown here). Thus, we concluded that the hysteresis is derived from the conventional s-wave and comes from electrodes between the larger SC-electrodes and narrower ones (e.g., a yellow colored electrode and an orange part in Fig. 1(a)), and thereby the jump does not reflect the junction nature of the S/TI/S junction.

Anomalous features, on the other hand, were discovered in small energy scale. The inset of Fig. 2(a) shows a bend structure in a small bias region of the $I-V$ characteristics. Conductance spectra of the discovered small bias structure are highlighted in Fig. 2(c). Owing to the $\xi < L$ condition (including longer electric trajectories between electrodes) and the tiny finite resistance (having finite barrier), our junction may behave as an intermediate state between S/I/N and S/N/S, and may show broadened dip or peak structures at the same bias voltage [3]. While the narrow structure seemed like an extraordinary sharp ZBCP in the wide range scale of Fig. 2(b), the peak had a trident-shape with a dip just outside the highlighted scale (Fig. 2(c)). The peak width was estimated as $\sim 6 \mu V$, which is quite small compared to the width of the ZBCPs of Majorana zero-mode found in other systems (e.g., nanowire: $\sim 50 \mu V$ [5, 6], and the Bi-based TI/S junctions: several hundreds of $\mu V$ [17,19,20]). The large discrepancy implies other origins. In the present situation, the Majorana fermions may not have emerged without any operations due to absence of both a magnetic edge, and a phase-slip of superconductors. The center peak, however, should come from dc-Josephson current. Fig. 2(a) shows temperature dependence of $dI/dV - I$ of the S/FTI/S junction. Some of the curves exhibiting discernable changes are picked up in Fig. 2(b) - (d) ($\Delta T = 0.25 K$ and $0.10 K$ for (b) and (d), respectively). Unique temperature evolution of the induced peak was observed as follows: at first, a typical single peak began to develop at 2 K. At 1.20 K, the peak approached a local maximum value, and the top soon started to split into M-shape. The center of the peak developed as temperature was cooled below 1.00 K. Reflecting this unique temperature evolution, temperature dependence of zero-bias resistance ($R_{0}$) had two discontinuous points at 1.K and 1.20 K as shown in the inset of Fig. 2(e). The $R_{0}$ for most part showed a monotonic decrease and was expected to become zero around 0.2 K.
The two discontinuities in the \( R_0 \) indicate existence of at least two origins of the spectra: the center ZBCP from the dc-Josephson current, and the M-shaped structure coming from other mechanism e.g., DOS of quasi-particles. In fact, this M-shaped conductance and the adjacent dip structures are almost identical to the calculated spectra for an N/I/S junction of chiral \( p \)-wave with anisotropic pair amplitude (the parameter \( C = 0.3 \)) [40]. This anisotropy is possibly induced by an in-plane magnetization component. In a chiral \( p \)-wave state, it is expected to have a ZBCP stem from the ABS. Though it exists, the weak ZBCP signal may be concealed by the strong peak signal of the dc-Josephson effect.

One inimical fact in this DOS scenario is the difficulty to explain the tiny peak-width less than the thermal energy. Such tiny signals are usually concealed by thermal fluctuations unless arising from quantum phenomena. As another hypothesis, the M-structure may reflect condensed components of the proximity effect; the two peaks represent \( \uparrow \uparrow \) and \( \downarrow \downarrow \) states, and less \( \uparrow \downarrow \) component, namely dominant triplet Cooper pairing. In this scenario, the peak width should correspond to the energy difference of the two levels. In the Fe-BST crystal, the estimated magnetic gap is 20 - 50 meV, which is three orders magnitude greater than the peak width. On the other hand, the magnetic coercive field is about 0.15 T (8.7 \( \mu \)eV) as shown in Fig. 1 (c), which is close to the peak width. This fact implies that the magnetic moments may induce unconventional SC states. Despite insufficient theoretical support, experimentally observed unique conductance-spectra unlike the conventional gap structure, clearly points toward emergence of an unconventional SC state.

The temperature dependence of \( I_c \) and \( I_{top} \), the critical currents at the local minimum and maximum of conductance spectra, respectively, are summarized in Fig. 3 (c) in order to characterize the unique temperature evolution. The product \( I_c R_{Np} \) was estimated (the normal state resistance of \( R_{Np} = 3.19 \)) to be \( \sim 2 \) \( \mu \)eV at 0.5 K, which is quite small compared with the thermal energy scale (i.e., 47.4 \( \mu \)eV at 0.55 K), implying existence of quantum phenomena such as the Josephson effect. Indeed, the calculated Josephson coupling energy \( E_{JC} = I_c h/2e \) was 0.6 meV at 0.55 K, which is significantly larger than the thermal energy. A dashed line in the Fig. 3 (e) indicates the competition between the thermal energy and Josephson coupling energy \( I_c h/2e = k_B T \). The line meets the experimental results around 1.25 K, supporting the existence of the dc-Josephson current.

One of the prominent features in the \( I_c \) is the low-temperature anomaly, which rapidly increases with cooling process. Usually, similar temperature dependence is observed in the clean-limit systems. However, the present junction is estimated to be the dirty-limit due to its finite resistance; the curves should reach a certain saturated value according to the Kulik-Omel’yanchuk theory [41].

Below 1.0 K, despite few measurement points, our results can be fitted by a function of \( I_c = a/T + b \) (where \( a = 0.49 \), \( b = -0.33 \)), as shown by a solid line in Fig. 3 (e). The low-temperature anomaly obeying \( I_c \propto T^{-1} \) is theoretically predicted in \( px \)-wave pairs [42]. Either component of a chiral \( p \)-wave pairing: \( px \) or \( py \), can be reportedly enhanced under a certain situation [21]. In the DOS scenario, one possibility is that the \( px \) component stemming from the chiral \( p \)-wave mainly contributes to the temperature dependence. On the other hand, \( T \)-dependence of \( I_c \) in triplet pairing is also known to show unsaturated \( I_c \) - \( T \) curve [43]. Since accessing further low temperature data is indispensable to determine a function of \( I_c \) - \( T \), we are planning to conduct additional experiment approaching several tens of mK using a dilution \( ^3 \)He refrigerator, as a future work.

If the junction has a certain ABS, by rf-irradiation, 4\( \pi \)-periodic Josephson current is expected to appear in \( I - V \) characteristics, as a step-like signal (upper panel of Fig. 3 (b)) with doubled step-width compared to the conventional Shapiro-step voltage \( V_f = hf/2e \), where \( h \), \( f \), \( e \) denote the Planck constant, micro wave frequency, and elementary charge, respectively [9]. However, finite resistance screened the steps, and as a result, we observed an almost linear response with a smeared kink structure in the \( I - V \) curve (shown in inset of Fig. 3 (a)).

We also performed the differential conductance \( dI/dV \) - \( V \) measurements. According to the ref. 44 , the long-range phase-coherence effect enables us to access the ac-Josephson current even in the condition of \( 2 \xi < L \). Indeed, a drastic evolution was observed as shown in Fig. 4 (a). These changes may represent transition from the TI/S contribution to the ac-Josephson cur-

FIG. 4. (Color online) (a) rf-response of induced structure at 0.60 K with frequency of 750 MHz. (b) Expected relation between \( I - V \) curve and \( dI/dV \) - \( V \) in Shapiro-step response. (c) Temperature evolutions of \( dI/dV \) with rf (750 MHz, -15 dBm) at 0.60 K. (d) Frequency variation of \( dI/dV \) at 0.60 K. The rf-powers were -15, -10, -4, -4 dBm for 750 to 1500 MHz, respectively. The horizontal axis is normalized by the conventional Shapiro-step voltage \( V_f = hf/2e \).
rent. For enhancing rf-power, the M-spectra and the adjacent dips were suppressed. Around -30 dBm, the contribution from the ac-Josephson current became gradually prominent, while the ZBCP rapidly fade away, reflecting the $n = 0$ Bessel function-like step of (dc) Josephson current. In other words, this actually ensures existence of the dc-Josephson current without the rf-irradiation, as moderate rf-power provides $n \neq 0$ Bessel signals.

Fig. 4(d) shows frequency variation of $dI/dV$ spectra under the moderate high-power rf. Since we used loop antenna of diameter $\sim 1$ cm (the antenna characteristics focused on $\sim 1$ GHz), the rf power was modulated for each frequency. The horizontal axis is normalized by the conventional Shapiro-step voltage $V_f = hf/2e$. In the $dI/dV - V$ picture, the Shapiro-step looks like an oscillating signal as shown in the lower panel of Fig. 4(b). The step-width usually corresponds to the local maximum of the oscillation.

The step widths for all measured frequencies in Fig. 4(d) were found to have almost doubled compared to voltages of standard Shapiro steps at least up to $n = 4$ step, which is suggestive of the $4\pi$-periodicity as expected for the Majorana bound states. Interestingly, the high bias signals showed further high step-widths (approximately $6\pi$-period) above $\sim 7 \mu V$. This trend is contrary to the data for other TI/S junctions and nanowire systems [10,11,12]. For example, one of the former studies has reported either first-step missing or missing few-steps in the Shapiro-step, while higher steps have transformed into the normal standard Shapiro-step pattern [11]. On the other hand, our junction showed further high step-width.

Empirically, one possible origin of doubled bias voltage is influence of the finite resistance to the bias voltage. To check the contribution of the resistance, temperature dependence of the oscillatory component in the $dI/dV$ spectra is displayed in Fig. 4(c). The oscillation periodicity remain unchanged up to $1.4$ K including the higher bias region, while the finite resistance increased by about a factor of four (see inset of Fig. 4(c)). Therefore, both the doubled step voltage and the higher region are irrelevant with the residual resistance, and are originated from intrinsic quantum phenomena driven by ac-voltage of the rf-irradiation. While mechanisms leading to $2\pi/n$-periodicity are widely understood e.g., higher harmonic contribution, the opposite situation is quite unusual. As far as we know, the only other mechanism leading to $4\pi$-periodicity except from topological origins is the Landau-Zener transition. These transitions may occur under high rf-irradiation due to phase-slip of SC states via enhanced highly transparent $2\pi$-components in clean limit junctions [45,46]. Although, the $4\pi$-periodicity by the Landau-Zener process is considered experimentally exceptional, further investigation of the rf-power dependence is needed to determine the origin of the $4\pi$-periodicity. Even though the origin is still debatable, absence of odd steps and further large periodicity of Josephson current strongly suggest realization of unconventional SC states in our junctions. Furthermore, this study may also reflect thermodynamic stability of the oscillation-periodicity, reflecting quantum or topological nature of avoiding phase-fluctuation by thermal fluctuation [47]. The previously reported $4\pi$-periodic signals of ABSs in other systems were recorded at several tens mK [10,11,12], while the present case maintained at most 1.4 K.

In order to determine the direction of the magnetic moment in the junction, field dependence of $dI/dV - I$ was evaluated. As per existing reports, the majority SC components in the proximity effect depend on the moment direction [31]. Fig. 5(a) shows the differential conductance spectra under magnetic field perpendicular to the surface. The conductance values at the zero-bias $G_{cero}$, local maximum $G_{top}$, and local minimum $G_{c}$ of the trident peak, as well as the critical currents (voltages) determined at the local maximum/minimum are shown in Fig. 5(b)-(d). Apparently, the obtained pattern of the $I_c - H$ curve is different from the typical Fraunhofer pattern. This is attributed to small field range compared with the estimated field periodicity $\Delta B = \Phi_0/S$, where $\Phi_0$ represents the flux quantum and $S$ area, is c.a. 700 G using the area of $100 \mu m \times 300 \mu m$). The important fact in the $I_c - H$ curve is the symmetry of the pattern. If the moments align along out-of-plane, the pattern of $H//c$-axis should be asymmetric due to the hysteresis nature of the ferromagnetism. However, the present symmetric pattern points to in-plane spin alignment. Another fact also supports the in-plane magnetic structure. While both critical voltages $V_{top}$ and $V_{c}$ remain unchanged, a reduction of about 15% in the conductance and the critical currents were observed. Due to $H//c$-axis, those reductions should point to diminution of states derived from the in-plane alignment, suggesting in-plane alignment of the ground spin structure in con-

FIG. 5. (Color online) (a) Field dependence of differential conductance spectra at 0.55 K. Inset is enlarged $dI/dV - I$ curves at the several magnetic fields. (b)-(d) obtained conductance values, critical currents, and critical voltages defined from Fig. 5(a).
trast to the c-axis in the bulk crystals. It is indeed very common for thin films to favor in-plane spin alignment. It may be noted that, the magnetic gap in the Dirac cone emerges for the out-of-plane situation, and the gap need not exist for the in-plane case. Furthermore, the in-plane spins support both the aforementioned scenarios, the chiral p-wave with in-plane anisotropy in the DOS scenario and the triplet Cooper pairing (OTE) driven by in-plane spins [31].

Non-doped 3D TI based Josephson junctions reported so far exhibited only the conventional Shapiro-step [38,48]. The observed discrepancy in the present case should be attributed to the doped magnetic moments, which allow unconventional SC states in the proximity effect. The pairing can be intuitively understood by considering a non-doped TI/S case as a first step; the spin-momentum locking on the Dirac cone suppresses a conventional s-wave pairing (↑↓−↓↑: ESE), and as a result, the p-wave SC state (↑↑−↓↓: ETO) emerges on the interface of TI/S to compensate the missing ↓↑ pairing. Note, that the emergent p-wave SC states hold the time-reversal symmetry. On the other hand, in the doped junctions, the pairing depends on the direction of the moments. When the moments are aligned perpendicular to the circular Fermi surface of the Dirac cone, it leads to the triplet (↑↑) p-wave pairing (ETO) in the out-of-plane component, and simultaneously, the triplet (↑↓ + ↓↑) p-pairing (ETO) with the TRS on the in-plane component is converted to an odd-frequency triplet (↑↓ + ↓↑) s-pairing (OTE), ensuring that total symmetry of the time-spins-orbital must be minus. On the other hand, for the in-plane aligned moments, the spins parallel to the moments can make triplet (↑↑) pairing between in-plane surfaces and may lead to anisotropic Fermi surface. Theoretical studies conducted on both cases in similar situations have supported the emergence of odd-pairing on magnetic TI/S junctions [31,49,50]. Interestingly, the importance of odd-pairing is not limited to dominant p-wave but has been pointed out to observe Majorana fermions as well [27,28,51]. Our results support in-plane spin alignment and thus suggest that both possibilities should still be considered, i.e., the anisotropic chiral p-wave and the dominant OTE pairing. Either case has potential to host Majorana quasi-particle on the edge by phase-slip of the superconductors via rf-irradiation. Therefore, the S/Fe-BTS/S junction, is a suitable platform to further investigate Majorana physics and unconventional SC states.

CONCLUSION

In summary, we examined the transport properties of the S/FTI/S junction consisting of magnetically doped TI, and experimentally probed the existence of unconventional states owing to the in-plane magnetic mo-

ments. The unique phenomena in the junction were the emergence of the exceptional trident-shaped conductance peak, the $I_c$-anomaly in temperature dependence, and the 4$\pi$-periodic Josephson response holding up to 1.4 K, which implies the possibility of existing Majorana bound state induced by high-power rf-irradiation. These results suggest that the magnetic Josephson junctions are promising platform for studies of unconventional superconductivity. Spatial advantage of electrodes on 2D surface of 3D TI is expected to accelerate establishment of fundamental technology utilizing vertices, for future quantum computing and observation of interplay between unconventional superconductivity and exotic yet undiscovered quasiparticles.

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