Subharmonic Shapiro steps in the a.c. Josephson effect for a three-dimensional Weyl semimetal WTe$_2$

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Abstract – We experimentally study electron transport between two superconducting indium leads, coupled to a single WTe$_2$ crystal, which is a three-dimensional Weyl semimetal. We demonstrate Josephson current in 5 $\mu$m long In-WTe$_2$-In junctions, as confirmed by the observation of integer (1, 2, 3) and fractional (1/3, 1/2, 2/3) Shapiro steps under microwave irradiation. The demonstration of the fractional a.c. Josephson effect indicates the multivalued character of the current-phase relationship, which we connect with the Weyl topological surface states contribution to the Josephson current. In contrast to topological insulators and Dirac semimetals, we do not observe $4\pi$ periodicity in the a.c. Josephson effect for WTe$_2$ at different frequencies and power, which might reflect the chiral character of the Fermi arc surface states in a Weyl semimetal.

Introduction. – Like other topological materials [1–4], Weyl semimetals are characterized by topologically protected surface states. These states originate as Fermi arcs, which connect the projections of Weyl points on the surface Brillouin zone in the $k$-space [5]. In contrast to helical surface states in topological insulators [1], Weyl states inherit the chiral property of the Chern insulator edge states [5]. Fermi arcs have been experimentally demonstrated by angle-resolved photoemission spectroscopy, e.g., for MoTe$_2$ and WTe$_2$ three-dimensional crystals [6,7].

The concept of Fermi arcs has been used to explain the magnetotransport experiments [8,9]. Unfortunately, Weyl and Dirac semimetals are conductors with gapless bulk excitations [5], so it is a problem to reliably distinguish the bulk and surface transport properties. On the other hand, the edge current contribution can be retrieved even for systems with conducting bulk by analyzing Josephson current behavior [10–12]. Edge state transport is responsible for Josephson current in 1–2 $\mu$m long superconductor-normal-superconductor (SNS) junctions in graphene [13,14]. For the Cd$_3$As$_2$ Dirac semimetal, the observation of the $\pi$ and 4$\pi$ periodic current-phase relationship has been reported in Al-Cd$_3$As$_2$-Al and Nb-Cd$_3$As$_2$-Nb junctions [15,16].

In this case, the fractional a.c. Josephson effect ($\pi$ periodicity) is connected with interference between the bulk and surface supercurrent contributions, while the disappearance of the $N = 1$ Shapiro step (4$\pi$ periodicity) reflects the helical nature of topological surface states in Dirac semimetals [15,16]. Thus, it seems to be reasonable to study the a.c. Josephson effect in SNS junctions, fabricated on a Weyl semimetal surface.

Here, we experimentally study electron transport between two superconducting indium leads, coupled to a single WTe$_2$ crystal, which is a three-dimensional Weyl semimetal. We demonstrate the Josephson current in 5 $\mu$m long In-WTe$_2$-In junctions, as confirmed by the observation of integer (1, 2, 3) and fractional (1/3, 1/2, 2/3) Shapiro steps under microwave irradiation. The demonstration of the fractional a.c. Josephson effect indicates the multivalued character of the current-phase relationship, which we connect with the Weyl topological surface states contribution to the Josephson current. In contrast to topological insulators and Dirac semimetals, we do not observe 4$\pi$ periodicity in the a.c. Josephson effect for WTe$_2$ at different frequencies and power, which might reflect the chiral character of the Fermi arc surface states in a Weyl semimetal.

Samples and technique. – The WTe$_2$ compound was synthesized from elements by reaction of metal
with tellurium vapor in a sealed silica ampule. The WTe$_2$ crystals were grown by the two-stage iodine transport [17], that previously was successfully applied [17,18] for the growth of other metal chalcogenides like NbS$_2$ and CrNb$_3$S$_6$. The WTe$_2$ composition is verified by energy-dispersive X-ray spectroscopy. The X-ray diffraction (Oxford diffraction Gemini-A, MoK$_\alpha$) confirms $Pmm2_1$ orthorhombic single crystal WTe$_2$ with lattice parameters $a = 3.48750(10)$ Å, $b = 6.2672(2)$ Å, and $c = 14.0629(6)$ Å. We check by standard magnetoresistance measurements that our WTe$_2$ samples demonstrate large, non-saturating positive magnetoresistance $\rho(B) - \rho(B = 0)/\rho(B = 0)$ in normal magnetic field, which goes to zero in the parallel one, as has been shown for the WTe$_2$ Weyl semimetal [8], see ref. [19] for details of magnetoresistance measurements.

A sample sketch is presented in fig. 1. Superconducting leads are formed by lift-off technique after thermal evaporation of 100 nm indium on the insulating SiO$_2$ substrate, see fig. 1(a). A WTe$_2$ single crystal ($\approx 0.5 \text{mm} \times 100 \text{µm} \times 0.5 \text{µm}$) is placed on the indium leads pattern, and is weakly pressed by another Si/SiO$_2$ substrate. The substrates are kept strictly parallel by external metallic frame to avoid sliding of the WTe$_2$ crystal, which is verified in optical microscope. As a result, planar In-WTe$_2$ junctions are formed at the bottom surface of the crystal WTe$_2$, being separated by 5 µm intervals, as depicted in fig. 1.

The obtained In-WTe$_2$-In SNS structures should be regarded as long $\xi \ll L$ diffusive $L > l_c$ ones: the $L$ value exceeds the mean free path [20] in WTe$_2$ $l_c \approx 1$ µm, so it should be compared [21,22] with the coherence length of the diffusive SNS junction $\xi = (l_c \cdot h v_F^2 / \pi \Delta_m)^{1/2} \approx 200$ nm, where the Fermi velocity is $v_F \approx 1.5 \cdot 10^7$ m/s from ARPES data [23], and $\Delta_m = 0.5$ meV is the indium superconducting gap [24]. This estimation is even stronger for smaller $l_c$, which can be expected [25] from the magnetoresistance behavior of our samples [12,19].

Charge transport is investigated between two superconducting indium leads in a four-point technique. An example of electrical connections is presented in fig. 1(a): the S1 electrode is grounded; a current $I$ is fed through the S2 electrode; a voltage drop $V$ is measured between these S1 and S2 electrodes by independent wires because of low normal In-WTe$_2$-In resistance. (b) Image of the leads pattern without a WTe$_2$ crystal.

Fig. 1: (Color online) (a) Top-view image of the sample with sketch of electrical connections. Ten µm wide indium superconducting leads (S1-S4) are separated by 5 µm intervals on the insulating SiO$_2$ substrate. In-WTe$_2$-In junctions are fabricated on the bottom surface of a WTe$_2$ crystal by weakly pressing a crystal ($\approx 0.5 \text{mm} \times 100 \text{µm} \times 0.5 \text{µm}$) to the indium leads pattern. Charge transport is investigated between two superconducting electrodes in a four-point technique: the S1 electrode is grounded; a current $I$ is fed through the S2 electrode; a voltage drop $V$ is measured between these S1 and S2 electrodes by independent wires because of low normal In-WTe$_2$-In resistance. (b) Image of the leads pattern without a WTe$_2$ crystal.

Fig. 2: (Color online) Examples of $I$-$V$ characteristics for different samples, obtained for 5 µm long In-WTe$_2$-In junctions between the superconducting leads S1 and S2, as depicted in fig. 1. A clear Josephson behavior can be seen in the zero magnetic field at 1.4 K $< T_c$: there is no resistance at low currents, it appears above $\pm I_c \approx 2 - 8$ mA for different samples. The jump positions are subjected to small hysteresis with the sweep direction, so they are slightly different for two $I$-$V$ branches. Inset: evolution of $dV/dI$ for the S1-WTe$_2$-S2 junction (depicted by the blue curve in the main field): at minimal $T = 1.4$ K $< T_c$ (the blue curve) and at $T = 3.5$ K $> T_c$ (the red curve) in zero magnetic field, and at the critical field $B = 31$ mT at minimal $T = 1.4$ K (the green curve).

**Experimental results.**

$I$-$V$ curves. To obtain $I$-$V$ characteristics, we sweep the d.c. current $I$ and measure the voltage drop $V$. Figure 2 presents $I$-$V$ examples for different samples in zero magnetic field and at low temperature 1.4 K $< T_c$.

The curves in fig. 2 clearly demonstrate the Josephson effect in unprecedentedly long $L = 5$ µm long In-WTe$_2$-In
Subharmonic Shapiro steps

Fig. 3: (Color online) a.c. Josephson effect in an In-WTe$_2$-In SNS junction at minimal 1.4K temperature. Shapiro steps appear at $V = Nh_f/2e$. Integer steps at $N = 1, 2, 3, \ldots$ are typical for SNS junctions, while the fractional $N = 3/2, 2/3, 1/2, 1/3$ ones indicate a multivalued nonsinusoidal character of the current-phase relationship [15,16,27]. (a) At fixed frequency 2 GHz, the $N = 1$ step appears first. At higher power, the $N = 2, 3$ ones appear together with the fractional $N = 1/2$ step. The fractions $N = 3/2, 2/3, 1/3$ can only be seen at maximum power, and they can be suppressed by the lowest $17$ mT magnetic field, as demonstrated in the inset. (b) The $N = 1$ is the most robust also when decreasing the frequency at fixed power 13 dBm. The curves are shifted for clarity in (b).

The obtained $I_c$ values differ within 10% in different coolings for a given sample. They are much smaller than the critical current for the indium leads, which can be estimated as $\approx 30$ mA for the leads’ dimensions and the known [26] maximum value $j \approx 3 \times 10^8$ A/cm$^2$ for indium. There are also small jumps in the resistive state at intermediate currents $I_c < I < 30$ mA for some samples, see fig. 2.

$I$-$V$ curves under microwave irradiation. The main experimental finding is the observation of fractional a.c. Josephson effect, as is depicted in fig. 3. The sample is illuminated by microwave (rf) radiation through an open coaxial line. For the fixed frequency, see fig. 3(a), rising of the radiation power shifts $I_c$ to lower currents. Simultaneously, Shapiro steps appear, which are placed at $V = Nh_f/2e$, as it should be expected for typical SNS junctions with trivial $2\pi$ periodicity in current-phase relationship $I_f = I_c \sin(\phi)$.

In addition to the steps at integer $N = 1, 2, 3, \ldots$, we observe half-integer $N = 1/2, 3/2$ ones, i.e., $\pi$ periodicity in the a.c. Josephson effect. It usually appears due to interference effects [15,16], for example, for the double-slit geometries in superconducting quantum interference devices (SQUID) [27,28]. The situation is even more complicated at high power: there are clear-visible $N = 1/3, 2/3$ steps, which indicates a multivalued nonsinusoidal character of the current-phase relationship, see discussion in refs. [27–29].

Figure 3 also demonstrates the dependence of integer and fractional Shapiro steps on the microwave power (a), magnetic field (inset to (a)), and microwave frequency (b). The fractional $N = 1/3, 2/3$ steps are the weakest: they can be suppressed by the lowest magnetic field, while the $N = 1/2$ one is as robust as the integer $N = 2, 3$ Shapiro steps, see the inset to fig. 3(a). Also, while decreasing the frequency at constant power, $N = 1/3, 2/3$ steps disappear first, which is demonstrated in fig. 3(b). On the other hand, the integer $N = 1$ step is the most stable: it appears at the lowest power and frequency, see fig. 3, and it is the strongest at highest ones. This robustness of the $N = 1$ Shapiro step is just the opposite to the observed $4\pi$ periodicity ($N = 1$ disappearance) in Al-Cd$_3$As$_2$-Al or Nb-Cd$_3$As$_2$-Nb junctions [15,16].

Check of possible fabrication defects. First of all, we should experimentally exclude artificial reasons for the observed multivalued character of the current-phase relationship, particularly possible fabrication defects, like multiple indium shortings in the junction plane. The thickness of the indium film is chosen to be much smaller than the leads separation (100 nm $\ll 5$ $\mu$m) to avoid parasitic shorting of In leads. Moreover, we do not see Josephson current in 5 $\mu$m long In-Cd$_3$As$_2$-In junctions, prepared in the same technique [30], so the observed behavior is specific for WTe$_2$.

The crucial arguments can be obtained from the maximum supercurrent $I_c$ behavior with temperature or magnetic field.

To analyze $I_c(B,T)$ behavior, we use $dV/dI(I)$ characteristics like the ones presented in the right inset to fig. 2: the d.c. current is additionally modulated by a low a.c. component (100 nA, 10 kHz), an a.c. part of $V$ ($\sim dV/dI$) is detected by a lock-in amplifier. We have checked, that the lock-in signal is independent of the modulation frequency in the $6$ kHz–$30$ kHz range, which is defined by the applied a.c. filters. To obtain $I_c$ values with high accuracy for given $(B,T)$ values, we sweep the current $I$ ten times from zero (superconducting state) to above $I_c$ (resistive state), and then determine $I_c$ as the average value of $dV/dI$ jump positions in different sweeps. The results are presented in fig. 4. All the experimental points are well reproducible, the variation of $I_c$ in different sweeps is below the symbol size for data in fig. 4.
The symbol size. The inset demonstrates the maximum parallel orientation (magnetic field is parallel) for the parallel orientation (magnetic field is parallel to the In-WTe plane) for two different samples. The curves are obtained at minimal 1.4 K temperature. All the experimental points are well reproducible, the variation of $I_c$ is below the symbol size. The inset does not demonstrate strong decay in the high-temperature limit $T \sim T_c$.

i) The experimental $I_c(T)$ in the inset to fig. 4 is inconsistent with indium shortings, because $I_c(T)$ does not demonstrate strong decay in the high-temperature limit $T \sim T_c$, which is expected [21,22] for long diffusive SNS junctions. Instead, the experimental $I_c(T)$-dependence is even slower than the linear function of $T$ in fig. 4(a). Similar behavior has been also demonstrated in long (1.5–2 μm) graphene SNS junctions [13,14], where it has been attributed to topological edge state transport.

ii) To our surprise, the $I_c(B)$ pattern crucially depends on the magnetic-field orientation to the In-WTe-In junction plane, see fig. 4. If the magnetic field is perpendicular to the plane, strong suppression of $I_c(B)$ is observed, as can be expected for standard Josephson junctions due to the pair breaking effect [31]. In contrast, $I_c(B)$ is diminishing very slowly (within 10% until the critical field) for the parallel magnetic field, which indicates interference effects, like in non-symmetric double-slit SQUID geometries [10,11]. It indicates that the effective SQUID area is perpendicular to the junction plane and, thus, it cannot be formed by parasite In shortings: if several fabrication defects connected the leads on the SiO$_2$ surface, they could form SQUID-like geometry in the junction plane only.

Discussion. – Since we can exclude parasite shortings in the junction plane, we should connect the observed multivalued character of the current-phase relationship with non-trivial distribution [10–12,15] of the Josephson current within the WTe$_2$ crystal, i.e., with topological surface states [5,6,8,9].

The interference ($\pi$ periodicity) can appear if both surface and bulk carriers transfer Josephson current in parallel, see fig. 5, as has been proposed for a Cd$_3$As$_2$ Dirac semimetal [15,16]. This picture gives qualitatively reasonable results: i) Parallel magnetic field induces a phase shift between surface and bulk channels for Josephson current, which leads to slow $I_c(B)$ damping, as we observe in fig. 4. If the magnetic field is perpendicular to the WTe$_2$ crystal plane, both surface and bulk channels are in phase. ii) Half-integer $N = 1/2, 3/2$ Shapiro steps appear in fig. 3 due to interference between bulk and surface channels, similarly to the Cd$_3$As$_2$ Dirac semimetal [15,16]. However, the clear visible Shapiro steps at fractional $N = 1/3, 2/3$ indicates that the interference scheme for the Josephson current is more complicated [27,28] for WTe$_2$. This is the reason to consider also the surface states on the opposite sample surface, see fig. 5, which are not independent in a Weyl semimetal [5]. Due to the necessity of the surface state coupling, the contribution of this channel can only be seen at maximum microwave power and frequency in fig. 3.

Another evidence on the Weyl specifics of the WTe$_2$ surface states is the fact that we do not observe 4π periodicity in the a.c. Josephson effect: the integer $N = 1$ Shapiro step is the strongest one at the highest power in fig. 3, while the maximum power value covers the range of $N = 1$ disappearance in Al-Cd$_3$As$_2$-Al and Nb-Cd$_3$As$_2$-Nb junctions [15,16]. In the latter case, 4π periodicity is connected [15,16] with the helical nature of topological surface states in Dirac semimetals, while Weyl surface states inherit the chiral property of the Chern insulator edge states [5].

Because of topological protection, Weyl surface states can efficiently transfer the Josephson current, which appears in slow $I_c(T)$ decay in the inset to fig. 4. This is another argument for the surface states, since the bulk supercurrent contribution should demonstrate strong exponential decay [21,22] in the high-temperature limit $T \sim T_c$ for long diffusive SNS junctions.

Conclusion. – As a conclusion, we experimentally study electron transport between two superconducting
indium leads, coupled to a single WTe$_2$ crystal, which is a three-dimensional Weyl semimetal. We demonstrate the Josephson current in 5 μm long In-WTe$_2$-In junctions, as confirmed by the observation of integer (1, 2, 3) and fractional (1/3, 1/2, 2/3) Shapiro steps under microwave irradiation. The demonstration of the fractional a.c. Josephson effect indicates the multivalued character of the current-phase relationship, which we connect with the Weyl topological surface states contribution to the Josephson current. In contrast to topological insulators and Dirac semimetals, we do not observe 4π periodicity in the a.c. Josephson effect for WTe$_2$ at different frequencies and power, which might reflect the chiral character of the Fermi arc surface states in a Weyl semimetal.

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