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LETTER

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Manipulating the anomalous Josephson effect by interface valley-polarized mixing

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Abstract – We theoretically investigate the supercurrent through a Josephson junction with at least one superconducting electrode directly coupled to a valley-polarized graphene sheet. The anomalous Josephson effect is shown and remarkably manipulated by interface valley-polarized mixing together with combination of the static staggered potentials and off-resonant circularly polarized light field inducing the valley polarization. The unconventional Josephson effect results from the breaking of underlying chiral (polarized-valley rotational) and time-reversal symmetries, which in turn shows the equivalence of the valley and spin freedoms. The valley-polarized interface-tunable phase offset will be of great interest in the designing and fabrication of such novel devices based on valleytronics as superconducting flux- and phase-based quantum bits, phase batteries and rectifiers.

Recently, a continuously growing interest has been kept in unconventional Josephson effects, particularly the anomalous Josephson effect [1]. The Josephson effect describes supercurrent flowing through a junction with a thin barrier sandwiched by two superconducting electrodes, which is driven by a superconducting phase difference ϕ between the electrodes. In the context of chiral (spin-rotational) and time-reversal symmetries of the Cooper pair tunneling process [2], the current *vs.* ϕ satisfies the sinusoidal function and is strictly zero under vanishing of ϕ. However, only if these underlying symmetries are broken, can the supercurrent be finite, which corresponds to a ground state of the junction being offset by a phase ϕ_0, rather than 0 or π [3–6]. A variety of ways for creating junctions have been theoretically put forward to break the underlying symmetries, such as the ones based on non-centrosymmetric or multilayer ferromagnets [4,7], quantum point contacts [5], topological insulators [8,9], nanowires [10,11], and quantum dots [6,12,13]. Experimentally, only until very recently, was the anomalous Josephson effect demonstrated by the superconducting quantum interference device (SQUID) [3].

On other hand, the past few years have witnessed remarkable progress in the study of graphene (G)-superconductor (S) hybrids, including the Josephson junctions composed of the G nanaoribbon or valley-polarized G [14]. The ability to combine high-quality G with S's via clean interfaces has fiercely stimulated the surge in interest, leading to several experimental advances. In particular, a fully gate-tunable G SQUID was recently used to determine the current-phase relation (CPR) of ballistic G Josephson junctions [15]. However, to date, there has been no report on the anomalous Josephson effect about such G-based hybrids. And then, considering that the valley polarization can induce the 0-π transition in only valley-polarized G Josephson junction like the spin polarization in conventional ferromagnetic ones [14], could there exist the anomalous effect in the former, which has no breaking of the underlying spin-rotational symmetry? In almost all the previous works on the G Josephson junctions, superconducting metal electrodes were deposited on the G sheet, having G honeycomb lattice structure, and thus, one would consider whether the direct coupling of a bulk superconducting metal electrode to the G layer exerts a

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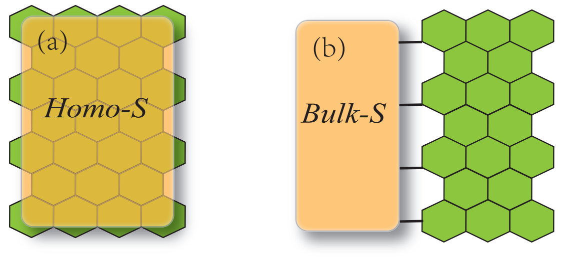

Fig. 1: The schematic diagram for the Home- and Bulk-S electrodes, where the Homo-S means the S electrode deposited on the G sheet with Cooper pairs leaking into the G lattice, whereas the Bulk-S indicates the one directly connected with the G sheet by a coupling strength t'.

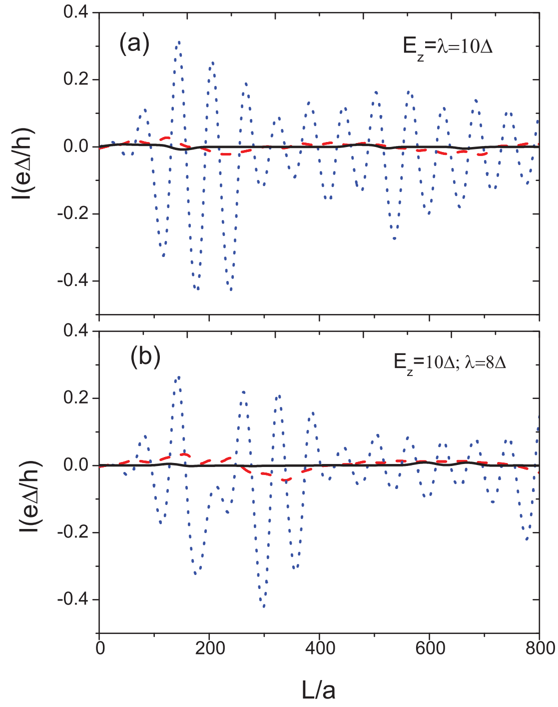

Fig. 2: The Josephson currents I at zero phase shift *vs.* L are respectively illustrated for Home-S/G/Home-S (solid line), Home-S/G/Bulk-S (dashed line), and Bulk-S/G/Bulk-S (dotted line) structures under the situations of (a) $E_z = \lambda = 10\Delta$ and (b) $E_z = 10\Delta$ as well as $\lambda = 8\Delta$.

significant influence on the Josephson supercurrent features or not, such as the anomalous Josephson effect. Although such a Josephson junction for both the two electrodes with direct coupling was only studied by Wang *et al.* [14], they focused on the critical supercurrent other than the ground-state phase difference ϕ_0 together with corresponding CPR. Especially, the one consisting of the two above-mentioned different superconducting electrodes has not hitherto been dealt with.

Therefore, in this letter, we consider two Josephson junctions with the two S electrodes sandwiched by a valley-polarized G layer of the width W of the junction, the length L, and the lattice constant a. There are two types of S electrodes in the two structures as shown in fig. 1(a) and (b), but at least one is termed as "Bulk-S" electrode, where a bulk superconducting metal electrode directly couples to the G layer, and thus does not have G honeycomb lattice structure. As a result, the intervalley scattering at the interface between the superconducting electrode and G sheet is induced due to lattice mismatch. The other is abbreviated as "Homo-S" with the S metal electrode deposited on top of the G sheet, where a finite pairing amplitude and the shift of the Fermi level in the G layer far away from the Dirac point are brought about and no intervalley scattering at the interface comes into being. The valley polarization of the G sheet could be realized in a silicene system by the interplay of the static staggered potentials and the off-resonant circularly polarized light field could give rise to the valley polarization [16]. Due to the interface valley mixing and valley polarization in the G sheet, the underlying chiral (polarized-valley rotational) and time-reversal symmetries are simultaneously broken, leading to the anomalous Josephson effect. The unconventional Josephson effect is found to be sensitively tuned by interface valley mixing as well as the valley-polarization strength.

Following Wang *et al.*, the Hamiltonian for the present systems is given by [14] $H = \sum_i (U_i - \mu) b_i^\dagger b_i - t \sum_{\langle ij \rangle} b_i^\dagger b_j + \frac{i\lambda}{3\sqrt{3}} \sum_{\langle\langle ij \rangle\rangle} \nu_{ij} b_i^\dagger b_j - \sum_i \eta_i E_z b_i^\dagger b_i + H_s$ with $b_i^\dagger(b_i)$ being the electron creation (annihilation) operator at site i and $\langle ij \rangle$ ($\langle\langle ij \rangle\rangle$) denoting the summation over the nearest (next-nearest) neighbor lattice sites. The first and second terms depict the energy band of a normal G sheet together, where t and U_i respectively stand for the hopping integral and the site energy denoting the doping level. The third and fourth terms represent the off-resonant circularly polarized light field with the strength λ and the static staggered potentials E_z in the G sheet, respectively, giving rise to a valley polarization with the two valley bands of a normal G being splitted and a Fermi momentum difference δq, where ν_{ij} (η_i) is 1 or -1. The last term H_s describing the Home or Bulk electrode can be seen in ref. [14]. We perform the numerical evaluation of the Josephson current flowing in the S/G/S junction by using a recursive method following the work [14]. For comparison, the corresponding results for the two S electrodes being both the Home-S will be also presented in the following parts.

In fig. 2, the Josephson currents at zero phase shift as a function of the valley-polarized G layer length L are respectively illustrated for the Home-S/G/Home-S, Home-S/G/Bulk-S, and Bulk-S/G/Bulk-S structures at $E_z = \lambda = 10\Delta$ and $E_z = 10\Delta$ together with $\lambda = 8\Delta$. In the case of $E_z = \lambda = 10\Delta$, it is found that the Josephson current I at zero phase for the Home-S/G/Home-S structure is zero as in the usual situation. However, very interestingly, it is no longer 0 and exhibits oscillations with the L for the Home-S/G/Bulk-S and Bulk-S/G/Bulk-S structures. Particularly, for the Bulk-S/G/Bulk-S one, the oscillation is very strong, *i.e.*, the values of not only peaks and valleys but also of the period are larger than those for the Home-S/G/Bulk-S one. At the $\lambda = 10\Delta$ and $E_z = 8\Delta$, the features, especially those of the periods, are shown to be the same as at $\lambda = 10\Delta$ and $E_z = 10\Delta$ except that magnitudes of the peaks, valleys, and the period are slightly different. These indicate that the phase ϕ_0 has been found in the junctions with the Bulk-S, exhibiting the anomalous behaviors with non-zero current at the zero phase shift, which is the main result of the work. The reason for inducing this anomalous Josephson effect is the existence of the Bulk-S induced valley-polarized mixing, and the mechanism will be presented in the following, which is thoroughly different from the above-mentioned usual mechanism of leading to the phase ϕ_0 junction, the breaking of chiral (spin-rotational) and time-reversal symmetries of the Cooper pair tunnelling process.

The physical mechanism of the anomalous Josephson current stemming from the Bulk-S induced valley-polarized mixing can be described as follows. Due to the valley-polarized mixing scattering at the interfaces, there exist valley-triplet currents in the Home-S/G/Bulk-S and Bulk-S/G/Bulk-S structures. At zero phase shift, only the $\mathbf{KK}$ and $\mathbf{K'K'}$ valley-triplets contribute to the supercurrent $I(\phi + \sum_{\sigma=\pm 1} \sigma\phi_{\sigma 0})$ with $\phi_{\sigma 0}$ indicating the anti-phase shifts caused by the $\mathbf{KK}$ and $\mathbf{K'K'}$ valleys. In generating the valley polarization, the corresponding light or electric field is applied. The light field can result in the fact that the next-nearest-neighbor hopping is clockwise or counterclockwise, and applying the electric field can lead to the fact that the A and B sublattices are not on the same surface. Therefore, the valley polarization leads to the time-reversal symmetry breaking, while the valley-polarized mixing at the interface between the Bulk S and G sheet does bring about the chiral (valley-rotational) symmetry breaking. Due to the two underlying broken symmetries, the phase shifts contributed by the $\mathbf{KK}$ and $\mathbf{K'K'}$ valleys are not identical, and resultantly cannot cancel each other out, giving rise to the anomalous Josephson current. Owing to the degree of chiral (valley-rotational) symmetry breaking caused by two Bulk-S electrodes stronger than that by one Bulk-S electrode, the corresponding anomalous effect for the former becomes more outstanding.

Since applying the light field or the static staggered potential could change the energy band structure, the Josephson currents at zero phase shift in fig. 3(a) are respectively shown as a function of the staggered potential E_z for the Home-S/G/Home-S, Home-S/G/Bulk-S, and Bulk-S/G/Bulk-S structures. In the Home-S/G/Home-S one, the Josephson current I is not varied with the E_z and equals zero. Then, for the Home-S/G/Bulk-S one, with the enhancement of E_z, we find that the I first increases from zero and exhibits oscillation behaviors alternatively along the positive and negative directions. However, for the Bulk-S/G/Bulk-S one, the properties

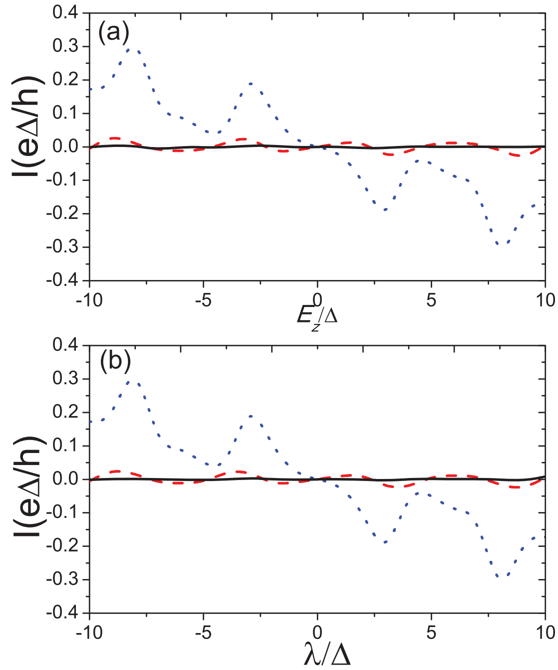

Fig. 3: The Josephson currents I at zero phase shift for the same three structures as in fig. 2 are respectively plotted as functions of (a) the staggered potential E_z and (b) the light field strength λ. Here, $L/a = 800$ and the other parameters are the same as those in fig. 2.

of the oscillation are much different from those for the Home-S/G/Bulk-S one. More specifically, the values of not only peaks but also valleys are larger than those for the Home-S/G/Bulk-S structure. Particularly, I with the increase of E_z first increases from a large non-zero value and exhibits a damped oscillation along a certain direction, and then an increasing oscillation along the opposite direction. In fig. 3(b), the Josephson currents $I(\phi)$ at $\phi = 0$ are also respectively illustrated as a function of the light field strength λ. The curves are shown to be almost the same as those in fig. 3(a), originating from the Fermi momentum difference δq-induced valley polarization, specifically, non-zero λ and E_z both open an energy gap $\sim |\kappa\lambda + E_z|$ in the same manner and shift the valley band to vary δq, giving rising to the same effects.

It is necessary for us to exhibit the currents corresponding to different phase shifts, namely, the CPR $I(\phi)$, which can present not only the effect of the ground-state phase from another aspect but also the other properties of the Josephson current such as 0-π transitions. This could be in favor of getting a considerable insight into the anomalous Josephson effect. The Josephson currents $I(\phi)$ at different junction lengths L for the Home-S/G/Home-S structure without the Bulk-S, which are shown in fig. 4(a). At $E_z = \lambda$ (left column), the Josephson currents $I(\phi)$ not only are all zero at $\phi = 0$ as in fig. 3, but also exhibit a 0-π transition with the sine function at some lengths L, which has been displayed in ref. [14]. The transition originates from the formation of the valley-singlet Cooper pairs

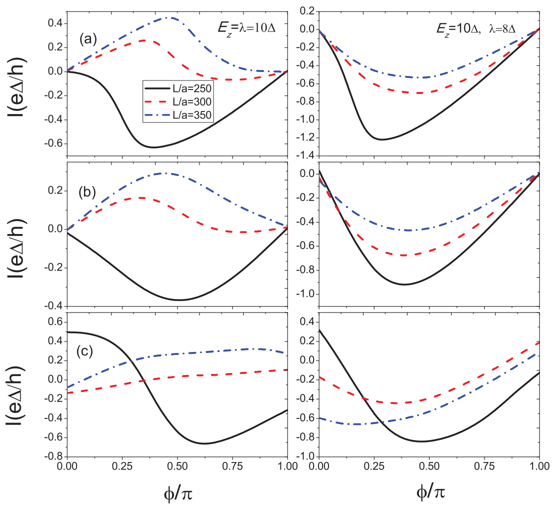

Fig. 4: Current-phase relations $I(\phi)$ for (a) Home-S/G/Home-S, (b) Home-S/G/Bulk-S, and (c) Bulk-S/G/Bulk-S structures at $E_z = \lambda = 10\Delta$ (left column) and $E_z = 10\Delta$ with $\lambda = 8\Delta$ (right column). Here, the various L are marked in the figure and the other parameters are the same as those in fig. 2.

with non-zero momentum δq induced by the valley polarization, which is the same as that by spin-polarization in the S/ferromagnet/S structure. However, at $E_z \neq \lambda$ (right column), the 0-π transition cannot be found, because the δq caused by valley polarization is too small to give rise to the transition in the usual normal G layer length range. This is very different from that in $E_z = \lambda$ case, where the normal G layer between Home-S electrodes is the valley half-metal with one valley band $\mathbf{K}$ ($\mathbf{K}'$) being insulating and the opposite valley band $\mathbf{K}'$ ($\mathbf{K}$) being metallic, *i.e.*, the valley is fully polarized [14]. For the Home-S/G/Bulk-S one, at $E_z = \lambda$ (left column in fig. 4(b)), the 0-π transition still remains with $I(\phi)$ being the sine function at some normal G layer lengths, however, although $I(\phi)$ at zero phase shift is very small, it is no longer zero. Furthermore, compared with the case of fig. 4(a), $I(\phi)$ is smaller. These imply that the singlet supercurrent still predominates but the triplet one emerges. The features at $E_z \neq \lambda$ are fully similar to those for the Home-S/G/Home-S for the same reasons. For the Bulk-S/G/Bulk-S one, at $E_z = \lambda$, there is not the 0-π transition any longer, and $I(\phi)$ has a deviation from the standard sine function, particularly, at $\phi = 0$ it is very large, which means that the triplet supercurrent predominates. The feature at $E_z \neq \lambda$ that there exists a large zero phase shift supercurrent is fully different from that in fig. 4(b). Therefore, it is concluded that when the 0-π transition occurs, one needs to break the time-reversal symmetry by valley polarization, whereas only if the underlying chiral (polarized-valley rotation) and time-reversal symmetries are simultaneously broken, can the anomalous Josephson effect arise as in conventional ferromagnetic Josephson junctions [17], which indicates the equivalence of the valley and spin freedoms.

Finally, we comment on the experimental execution of the present hybrids. The high-quality graphene was successfully combined with superconducting electrodes via clean interfaces [3,15]. The valley polarization is easily manipulated electrically by virtue of the staggered potential from E_z, which can be controlled by using a perpendicular electric field, regardless of whether it is the buckled silicene or BN/SiC substrated G [18]. Besides, the valley polarization can also be generated by other methods such as the interplay of the lattice strain, the magnetic barrier and the spin-orbital interaction plus the staggered magnetization [14]. Particularly, upon interfacing monolayer $MoSe_2$ with G, the room temperature degrees of valley polarization could be exhibited [19]. Thus, the fabrication of the present G hybrid structure is experimentally feasible and the anomalous Josephson effect could be probed.

To conclude, we have studied the valley-polarized G Josephson junctions comprising either two Bulk-S electrodes or both Bulk- and Home-S electrodes. It is demonstrated that due to the combination of the valley mixing at the interface between the S electrode and G sheet with the valley polarization, the breaking of the underlying chiral (polarized-valley rotational) and time-reversal symmetries is induced, thus leading to an anomalous Josephson effect. This in turn shows that valley freedom is equivalent to the spin one as well. The presence of at least one interface with the valley-mixing scattering is the criterion for the existence of anomalous Josephson current. However, the unconventional Josephson effect is found to be very much stronger in the presence of the valley-mixing scattering at both the two interfaces compared with that at only one interface. The valley-polarized interface-tunable phase offset could pave the new road for the realization of superconducting flux- and phase-based quantum bits [5,20], "phase batteries", and rectifiers [5,21] based on superconducting valleytronics. In particular, a low-energy dissipation is exhibited due to the definite chiralities of the quasiparticles in the G sheet making these devices immune against weak disorder. Here, it is pointed out that, although the Josephson effect in a graphene-based S/ferromagnetic insulator (FI)/S junction was theoretically studied in ref. [22], the ground-state phase difference ϕ_0 is absent due to no breaking of chiral (spin-rotational) symmetry. If the FI in such junction is replaced by several ones with spin-rotational arrangement as in ref. [7], the two underlying symmetries, the chiral (spin rotational) and time-reversal symmetries, could be concurrently broken and the resultant ϕ_0 is exhibited. And the influence on the breakings of the underlying symmetries exerted by the spin freedom is studied, while in this work, we presented that exerted by the valley freedom.

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