

Magnetic Josephson Junctions: New Phenomena and Physics with Diluted Alloy, Conventional Ferromagnet, and Multilayer Barriers

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SUMMARY We review a new superconducting element, called “magnetic Josephson junctions” with a magnetic barrier instead of the insulating barrier of conventional Josephson junctions. We classify the three types of magnetic barrier, i.e., diluted alloy, conventional ferromagnet, and magnetic multilayer barriers, and introduce various new physics such as the π -state arising in magnetic Josephson junctions due to the interaction between superconductivity and magnetism.

key words: Josephson effect, magnetism, superconducting spintronics

1. Introduction

The interplay between superconductivity and magnetism is an important research topic as superconducting spintronics from the perspectives of both fundamental and applied physics [1]. The coexistence of superconductivity and ferromagnetism in uranium- and iron-based superconductors is a major topic in condensed matter physics [2], [3]. Novel phenomena induced by their interaction or competition emerge also in artificial hybrid structures of superconducting and magnetic materials. Representative examples are the π -state and long-range Josephson effect in the “magnetic Josephson junctions” presented in this review. Although theoretical concepts have been presented for many years, the experimental observations have been reported in recent years following progress in nanofabrication techniques. Furthermore, in recent years, a variety of challenges to apply these phenomena to superconducting devices, such as logic circuits, cryogenic memory, and quantum computing elements (qubits), are also going on.

In this review, we describe new phenomena and physics in magnetic Josephson junctions with three different types of magnetic barrier. In the next section, we review the diluted alloy barriers with weak ferromagnetism which played an important role in realizing the π -junctions experimentally. In Sect. 3, conventional ferromagnetic barriers (e.g., Ni, Co) are introduced, as well as a half-metallic ferromagnetic barrier. In Sect. 4, we discuss the magnetic multilayer (MML) barriers providing the long-range Josephson effect via spin-triplet Cooper pairs and a new way to control the Josephson critical current by changing the magnetization configuration.

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2. Diluted Ferromagnetic Alloy Barriers

2.1 Transition between 0-State and π -State

The magnetic Josephson junction consists of a hybrid structure, i.e., a magnetic barrier sandwiched by superconducting electrodes (superconductor/magnetic barrier/superconductor junctions). By replacing the insulating barrier in conventional Josephson junctions (superconductor/insulator/superconductor [SC/I/SC] junctions) with a magnetic barrier, new physics appears due to the interplay between superconductivity and magnetism. One of the typical phenomena is the so-called π -state (π -junction) in the ferromagnetic Josephson junction (superconductor/ferromagnet/superconductor [SC/FM/SC] junction) [4]. In this junction, the order parameter of the superconductor penetrates into the ferromagnetic barrier, and the order parameter decays with an oscillation due to the effect of the exchange field in the ferromagnet as shown in Fig. 1 [5]. When the thickness of the ferromagnetic barrier is around a half-integer multiple of the oscillation period, the signs of the order parameters in the two superconductors become different. This leads to a ground state of the junction with a phase difference of π , and the current-phase relation becomes $I = I_C \sin(\varphi + \pi)$, where I_C is the Josephson critical current and φ is the phase difference between the superconductors. This unconventional state is called the π -state, contrary to the ground state with zero phase difference (the so-called 0-state or 0-junction) and the current-phase relation $I = I_C \sin \varphi$ in conventional Josephson junctions. More details of the theoretical background were provided previously [6], [7].

Although the theoretical concept of the π -state in the ferromagnetic Josephson junctions have been proposed by Buzdin in 1982 [4], a number of experimental reports have been presented since 2000. The key for successful demonstration of ferromagnetic Josephson junctions was to adopt “weak” ferromagnetic alloys for the barrier of the junction [8]–[23]. The surviving length of the order parameters penetrating into the ferromagnetic barrier is characterized by the coherence length in the ferromagnet, ξ_{FM} . ξ_{FM} is inversely proportional to the exchange energy which is the difference in energy between up- and down-spin bands in the ferromagnet. The exchange energy of typical ferromagnetic materials such as Co, Ni, and Fe is several hundred meV, and ξ_{FM} becomes in the order of a few nm [24]. The very

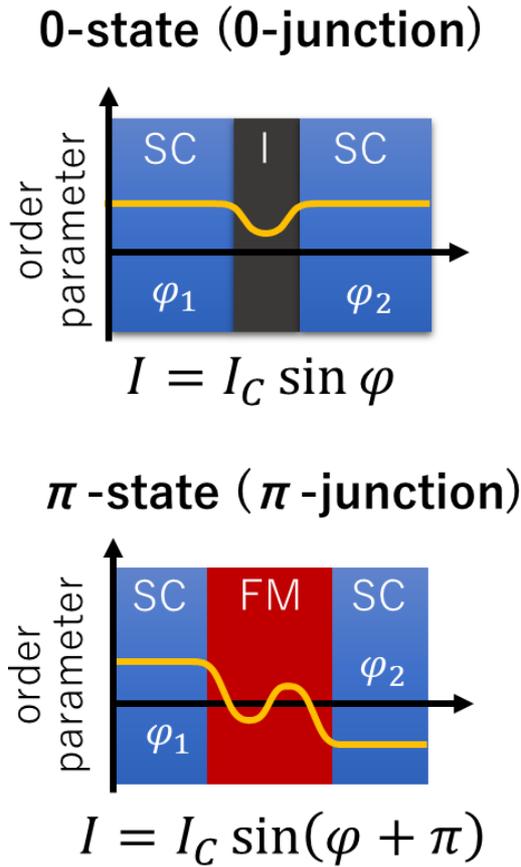


Fig. 1 Schematics of the 0-state (0-junction) in a superconductor/insulator/superconductor (SC/I/SC) junction and the π -state (π -junction) in a superconductor/ferromagnet/superconductor (SC/FM/SC) junction. Solid yellow curves indicate the order parameter. I_C and $\varphi = \varphi_1 - \varphi_2$ indicate the Josephson critical current and the phase difference between SCs, respectively.

short coherence lengths require an ultrathin ferromagnetic barrier for Josephson coupling between the superconducting electrodes. It is also very difficult to control the 0-state and π -state by tuning the ferromagnet thickness, because of the short period of oscillation of the order parameter in the ferromagnet.

Use of a diluted ferromagnetic alloy can resolve this issue in sample preparation. Using the diluted ferromagnetic alloys such as $\text{Cu}_{1-x}\text{Ni}_x$ and $\text{Pd}_{1-x}\text{Ni}_x$, the exchange energy can be controlled by the ferromagnet concentration x in the alloy. For example, in the PdNi alloy with around 10% Ni concentration, the exchange energy is a few tens of meV, which is one order of magnitude smaller than those of the conventional ferromagnets such as Co [8]. The first experimental demonstrations of the transition between the 0-state and π -state (0- π transition) in ferromagnetic Josephson junction have been reported by using $\text{Cu}_{0.48}\text{Ni}_{0.52}$ (Curie temperature of ~ 20 K) [9], [10] and $\text{Pd}_{0.88}\text{Ni}_{0.12}$ (Curie temperature of ~ 100 K) [11] as weak ferromagnetic barriers. Experimentally, the 0- π transition can be observed by measuring the Josephson critical current, I_C , with changes in the operation temperature or ferromagnetic barrier thickness.

Ryazanov *et al.* reported I_C oscillation in the temperature dependence for Nb/ $\text{Cu}_{0.48}\text{Ni}_{0.52}$ /Nb junctions [9], [10]. The coherence length, ξ_{FM} , is a complex and can be divided to the real term ξ_{FM1} and imaginary term ξ_{FM2} , corresponding to decay and oscillation of the order parameter in the ferromagnet, respectively. By increasing the temperature ξ_{FM1} (ξ_{FM2}) decreases (increases), so temperature-induced 0- π transition occurs if the transition thickness determined by ξ_{FM1} and ξ_{FM2} crosses the fixed ferromagnetic barrier thickness of the junction. As shown in Fig. 2 (a), reentrant behavior of I_C in the temperature dependence was observed, providing clear evidence of the 0- π transition [9], [10]. Around the 0- π transition, the sign of the critical current changes. In the measurement, however, only the absolute value of the critical current is observed and thus the reentrant dependence shown in Fig. 2 (a) appears. Generally, the temperature-induced 0- π transition is observed only near the transition thickness, so precise tuning of the ferromagnetic barrier thickness of the sample is required to observe this behavior between the base and superconducting critical temperatures.

Kontos *et al.* reported the 0- π transition in the ferromagnet thickness dependence for Nb/Al/ Al_2O_3 /Pd $_{0.88}\text{Ni}_{0.12}$ /Nb junctions shown in Fig. 2 (b) [11]. The authors fabricated samples with various PdNi thicknesses of 4-15 nm, and observed reentrant PdNi thickness dependence of $I_C R_N$ (R_N is the normal resistance of the junction) with a transition thickness of ~ 6.5 nm. Also for the CuNi-barrier junctions, the thickness dependent 0- π transition have been reported by the several groups later [6], [12]–[14]. In particular, Oboznov *et al.* presented the transitions of “0 to π ” and “ π to 0” by increasing the CuNi thickness from 8 to 28 nm, as shown in Fig. 2 (c) [12]. They also found that the temperature-induced transition occurred for samples with CuNi thickness of 11 and 22 nm, which were close to the first (0 to π) and second (π to 0) transition thicknesses, respectively. Subsequently, as shown in Fig. 2 (d), Khaire *et al.* reported an oscillating critical current in the wider ferromagnetic barrier thickness range of 32-100 nm for Nb/Pd $_{0.88}\text{Ni}_{0.12}$ /Nb junctions [15].

2.2 π Phase Shift Characteristics

Although observation of the 0- π transition is important to show the existence of the π -state, demonstration of the π phase shift in the superconducting loop or current-phase relation provides more direct evidence of the π -state. Guichard *et al.* performed a phase-sensitive experiment based on the quantum interference of two Josephson junctions [16]. They have fabricated a superconducting quantum interference device (SQUID) with Nb/ NbO_x /Pd $_{0.82}\text{Ni}_{0.18}$ /Nb junctions and measured the magnetic field modulation of the critical current. By changing the PdNi thickness, the junction could be in the 0-state or π -state. As shown in Fig. 2 (e), they demonstrated a half-flux-quantum ($\Phi_0/2$, Φ_0 is a flux quantum) shift of the modulation pattern for the 0- π SQUID from that for the 0-0 SQUID. The observed half-flux-quantum shift results from the interference of the 0- and

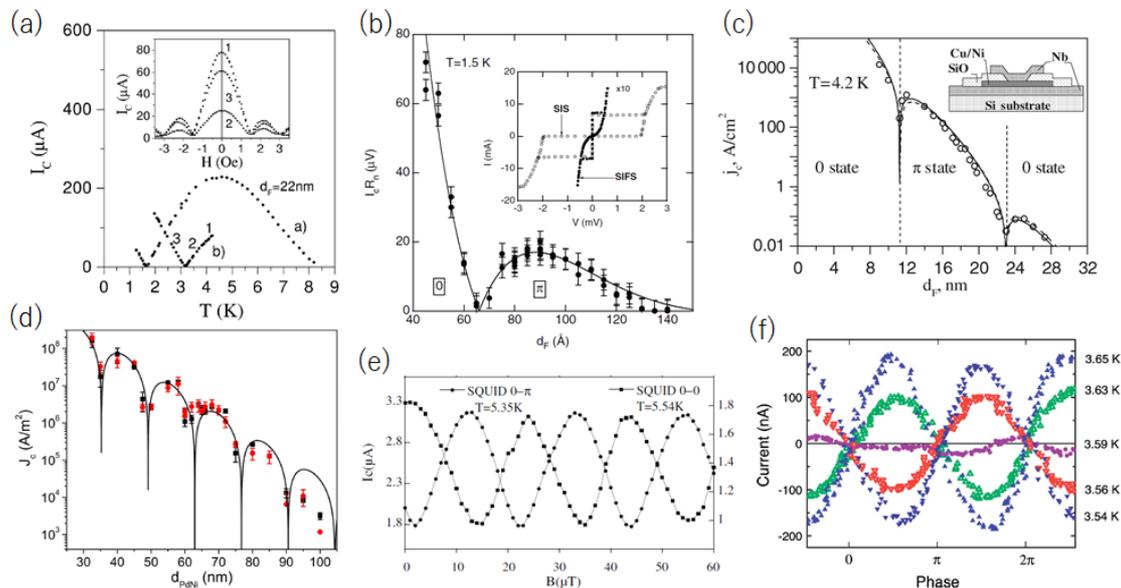


Fig. 2 Experiments on the π junctions with diluted ferromagnetic barriers. (a) Temperature dependences of the critical current for Nb/CuNi/Nb junctions. The temperature induced $0-\pi$ transition was observed for two samples with 22-nm-thick CuNi. Inset shows the magnetic field dependences of the critical current at temperatures denoted by 1-3 in the temperature-dependent curve b. From Ryazanov *et al.* [10]. Copyright (2001) by the American Physical Society. (b) Ferromagnetic barrier thickness dependence of $I_C R_N$ for the Nb/Al/Al₂O₃/PdNi/Nb junctions. The solid line is the fitted curve by the theory. Inset shows the current-voltage characteristics of SC/I/SC and SC/FM/SC junctions. From Kontos *et al.* [11]. Copyright (2002) by the American Physical Society. (c) The CuNi thickness dependence of the critical current density of Nb/CuNi/Nb junctions. Two-time ($0-\pi$ and $\pi-0$) transitions have been observed. Solid and dashed lines were fitted curves based different theoretical models. Inset shows the schematics of the cross section of the junctions. From Oboznov *et al.* [12]. Copyright (2006) by the American Physical Society. (d) The dependence of the critical current density of Nb/PdNi/Nb junction on a wide range of the PdNi thickness at 4.2 K. Squares (black) and circles (red) were the data measured before and after magnetizing the samples, respectively. The solid line is the fitted curves for the squares except the data with the PdNi thicknesses of 95 and 100 nm. From Khaire *et al.* [15]. Copyright (2009) by the American Physical Society. (e) Magnetic field modulations of the critical current of the $0-0$ and $0-\pi$ Nb/NbO_x/PdNi/Nb SQUIDS. From Guichard *et al.* [16]. Copyright (2003) by the American Physical Society. (f) Current-phase relation of Nb/CuNi/Nb junction measured at near $0-\pi$ transition temperatures. From Frolov *et al.* [20]. Copyright (2004) by the American Physical Society.

π -junctions in the loop, and represents direct evidence of the π -state. The half-flux-quantum shift has also been reported for a three-junction loop with the π -junction [17], [18]. In addition, the spontaneous magnetic flux, which was induced by the π shift, in the superconducting loop with a single π -junction was measured using a micro-Hall sensor [19].

Another experiment to show the π -state is the measurement of the current-phase relation of the ferromagnetic Josephson junction. Frolov *et al.* made a SQUID coupled circuit with an Nb/Cu_{0.47}Ni_{0.53}/Nb junction to measure its current-phase relation [20]. The CuNi thickness was set to 22 nm, at which the temperature-induced $0-\pi$ transition occurred. The results indicated a clear π -shift current-phase relation at lower than the $0-\pi$ transition temperature of around 3.6 K, as shown in Fig. 2 (f). The authors also investigate a possibility of the higher-order harmonics predicted in metallic Josephson junctions, and concluded that there was no higher-order harmonics for the junctions. Later, the existence of the second-order harmonics for the Cu_{0.52}Ni_{0.48}, Cu_{0.47}Ni_{0.53} and Cu_{0.4}Ni_{0.6} barrier junctions was reported

based on systematic measurements of the temperature dependence of the critical current, Shapiro step, and Fraunhofer pattern [21]–[23].

The π phase shift characteristics of the π -junction are also attractive from an application standpoint. In superconducting logic circuits, such as single-flux-quantum (SFQ) and adiabatic quantum-flux-parametron (AQFP) circuits, the consumption power or circuit area is expected to be reduced by the π -junction [25], [26]. For the flux type of superconducting qubits, the π -junction offers the advantage of flux-bias-free operation [27]–[29]. With regard to the superconducting material of the ferromagnetic Josephson junctions, Nb which is often used for various superconducting devices such as SFQ logic circuits, has been adopted. Recently, with a view toward the application of the above devices, NbN-based junctions, which enable higher temperature operation of the logic circuit and/or an improved coherence time in the qubits, have been reported [14], [18]. Also for the NbN-based ferromagnetic Josephson junctions, the $0-\pi$ transitions in the temperature and ferromagnetic

barrier thickness dependences and half-flux-quantum shift have been observed.

3. Conventional Ferromagnetic Barriers

Josephson coupling in ferromagnetic Josephson junctions with conventional (strong) ferromagnetic barriers such as Ni and Co also has been reported from several groups [24], [30], [31]. As described in the previous chapter, the large exchange energy of several hundred meV easily destroys the order parameter penetrating into the ferromagnetic barriers, so a thinner barrier (a few nm thick) is required to achieve Josephson coupling. Blum *et al.* presented the non-monotonic oscillatory behavior of the critical current for Ni thickness dependence of Nb/Cu/Ni/Cu/Nb junctions [30]. From the theoretical analysis, they estimated an oscillation period of 5.4 nm with an exchange energy of 107 meV for Ni. Also for the Nb-based ferromagnetic Josephson junctions with Py (NiFe), Co, and Fe barriers, the similar oscillation of the critical current has been reported. Robinson *et al.* systematically measured and analyzed the ferromagnet thickness dependence of the critical current for Co, Py, Ni, and Fe barrier junctions [24], [31]. They found that Co junctions could be described by the clean-limit theory and were adequate for device applications (Fig. 3 (a)). Although precise control of barrier thickness is required, such a single-element ferromagnet is an attractive choice for realizing spatially uniform π -junctions for practical application.

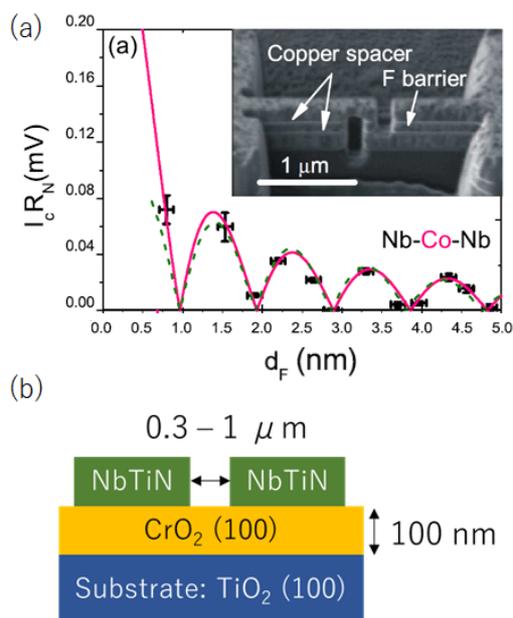


Fig. 3 Experiments on the ferromagnetic Josephson junctions with strong (not diluted) ferromagnetic barriers. (a) Co thickness dependence of $I_c R_N$ for Nb/Co/Nb junctions at 4.2 K. The solid and dashed lines are the fitted curves based on the different theoretical models. Inset shows a focused ion beam micrograph of the junction. From Robinson *et al.* [24]. Copyright (2006) by the American Physical Society. (b) Schematics of Josephson junctions with a half-metallic ferromagnet CrO_2 barrier [32]. 100-nm-thick CrO_2 (100) single crystal film was epitaxially grown on TiO_2 (100) substrate. The distance between two NbTiN electrodes was 0.3-1 μm .

In 2006, Keizer *et al.* reported that a finite Josephson current flowed through the ferromagnetic CrO_2 between NbTiN electrodes (Fig. 3 (b)) [32]. This was surprising because CrO_2 is known to be a “half-metallic” ferromagnet in which only the density of state for the up-spin electrons exists at the Fermi level, such that only the up-spin electrons can participate in transport. The conventional singlet Cooper pair with up- and down-spin electrons cannot survive in CrO_2 because Andreev reflection at the interfaces of the superconductor/half-metallic ferromagnet is prohibited [6]. Furthermore, they observed the Josephson current over a very large distance of around 1 μm and attributed this long-range Josephson current to the transport by the “triplet” Cooper pairs (also described in the next section) [33]–[35]. As the triplet Cooper pair is a pair of electrons with parallel spins, in contrast to the singlet Cooper pair, the supercurrent carried by the triplet Cooper pair can flow through strong ferromagnets such as CrO_2 .

4. Magnetic Multilayer Barriers

4.1 Long-Range Coupling Via Spin-Triplet Cooper Pairs

Over the past decade, as a new trend of the magnetic Josephson junctions, long-range Josephson coupling via spin-triplet current which was indicated in the experiment of the CrO_2 described in the previous section. Theoretically, spin-triplet correlations appear when Cooper pairs pass through the non-collinear (inhomogeneous) magnetization region [33]–[35]. To achieve this experimentally, Josephson junctions with an MML shown in Fig. 4 (a) have been studied so actively. Khaire *et al.* fabricated Nb/MML/Nb junctions with MML of a Cu/X/Cu/Co/Ru/Co/Cu/X/Cu structure, where Cu is a buffer layer to magnetically isolate the layers from the Co layers and Ru is for providing the antiparallel coupling between the Co layers to eliminate net magnetization in the junction [36]. In MML, X denotes $\text{Pd}_{0.88}\text{Ni}_{0.12}$ or $\text{Cu}_{0.48}\text{Ni}_{0.52}$ alloy. The authors systematically investigated the total Co thickness dependences of $I_c R_N$ for the junctions with and without X layers. As shown in Fig. 4 (b), $I_c R_N$ did not decay with Co thickness of 12-28 nm with a 4-nm-thick PdNi layer (red circles), whereas $I_c R_N$ decayed quickly with increasing Co thickness without PdNi (black squares). The same trend of enhanced Josephson current was observed also for CuNi layers with thicknesses of around 2-5 nm. Such a long-range Josephson current is evidence of the spin-triplet correlation. As a possible origin of the spin-triplet correlation, the authors assumed the non-collinear magnetization in the neighboring domains inside the X layer and/or between X and Co layers. Robinson *et al.* performed a similar experiment using simpler Nb/Ho/Co/Ho/Nb junction [37]. Here the key is that Ho has antiferromagnetic spiral magnetizations, which induces non-collinear magnetizations. In the fabricated junctions, $I_c R_N$ remained finite for the Co layer with more than 15 nm whereas the sample without Ho layers decayed exponentially with a characteristic length of around 1 nm. These

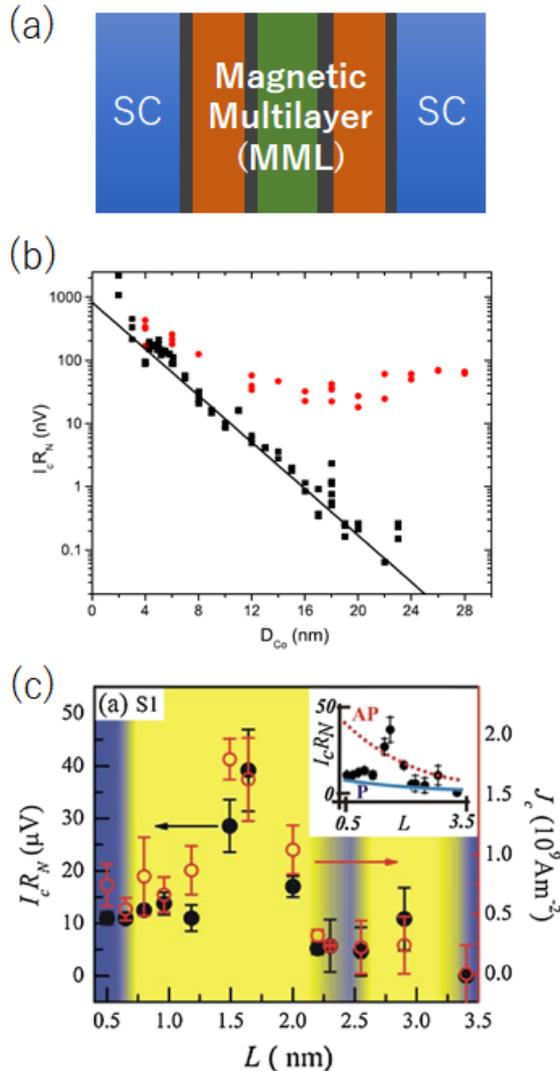


Fig. 4 (a) Schematics of Josephson junctions with magnetic multilayers (MMLs). (b) $I_c R_N$ vs. Co thickness for the junctions with MMLs of Cu/PdNi/Cu/Co/Ru/Co/Cu/PdNi/Cu (red circles) and Cu/Co/Ru/Co/Cu (black squares) at 4.2 K. The solid line shows the fitted curves for the data without PdNi layers. From Khaire *et al.* [36]. Copyright (2010) by the American Physical Society. (c) Cr thickness dependences of $I_c R_N$ (filled black circles) and critical current density (open red circles) of Nb/Fe/Cr/Fe/Nb junctions at 4.2K. Dark blue and light yellow regions indicate the parallel (P) and antiparallel (AP) couplings, respectively. Inset: $I_c R_N$ and predicted curves by the theoretical model for P (solid blue) and AP (dotted red) configurations. From Robinson *et al.* [41]. Copyright (2010) by the American Physical Society.

results indicated that the non-collinear magnetizations in Ho provided the inhomogeneity required to generate the spin-triplet correlation in the junction.

Later, Banerjee *et al.* developed “controllable” magnetic Josephson junctions using MML consisting of Cu/Py/Cu/Co/Cu/Py/Cu layers [38]. Although an inhomogeneous magnetic configuration was fixed in previous works, in this junction the relative orientation between the Py and Co layers could be controlled by applying the magnetic field. When the magnetic field is zero or low,

non-collinear alignment was achieved between the Py and Co layers, which induced the spin-triplet Cooper pairs converted in MML. On the other hand, no spin-triplet Cooper pairs were generated for high magnetic fields because the magnetizations in the Py and Co layers aligned in parallel. The authors observed reasonable magnetic field dependence of the Josephson critical current which could be explained by the theoretical models.

4.2 Josephson Critical Current Switching

In addition to spin-triplet Cooper pairs, another way to control the Josephson current was presented in a Josephson junction with a pseudo-spin-valve (PSV) MML structure [39]–[45]. PSV consists of two different ferromagnetic layers separated by a normal (non-magnetic) metal. In PSV, the relative orientation of the magnetizations in the ferromagnetic layers can be set to parallel (P) or antiparallel (AP) alignment by applying the magnetic field because of their different coercive fields. Theoretically, it has been proposed that the Josephson critical current could be enhanced with the AP alignment in this structure; in the AP configuration of two ferromagnetic layers of the same thickness, the total change in the phase of a Cooper pair becomes zero due to compensation of the phase shift in the two ferromagnetic layers of PSV [39]. On the other hand, in the P configuration, the Josephson critical current is reduced because the phase shift in PSV becomes finite, as in SC/FM/SC junctions.

For several Nb/PSV/Nb junctions, it has been reported that the Josephson critical current can be changed depending on the magnetization alignment of PSV. Bell *et al.* measured the magnetic field dependence of R_N and I_C for the junction with Co/Cu/Py layers as PSV [40]. R_N was obtained to be larger for AP alignment than that for P alignment, i.e., magnetoresistance. Simultaneously, they found that I_C was also enhanced in the AP alignment compared to that in the P alignment, and showed a hysteretic behavior in the magnetic field dependence as with R_N . In Ref. [41], the authors adopted Fe/Cr/Fe layers for PSV and controlled the P and AP alignments of the Fe layers by changing the Cr thickness due to the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction. They observed enhanced critical currents in the AP alignment in PSV, which was theoretically predicted, as shown in Fig. 4 (c). More recently, toward the realization of cryogenic memory, various devices such as $0-\pi$ junctions controlled by the magnetic field [42] or spin transfer torque [43], $0/\pi-0/\pi$ switchable dc-SQUIDs [44], and a junction with perpendicular magnetic anisotropy [45] have been presented.

5. Conclusion

In this review, we introduced the novel phenomena and physics emerging in magnetic Josephson junctions with three types of magnetic barrier, i.e., diluted alloy, conventional ferromagnetic, and MML barriers. In the last 20

years, fundamental physics pertaining to the interaction between the superconductivity and magnetism, e.g., π -state and spin-triplet correlation, has been discovered and clarified via a combination of deep theoretical insights and progress in nanofabrication techniques. Simultaneously, attempts to develop superconducting spintronic devices also started toward the realization of improved performance and/or new functions of superconducting logic, cryogenic memory, and quantum computing. In this growing research field of superconducting spintronics, further discoveries of novel physics and their device applications are highly expected.

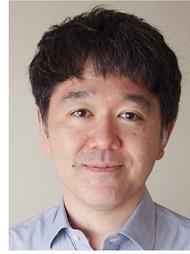
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