## Possible Evidence for Spin-Transfer Torque Induced by Spin-Triplet Supercurrents \*

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## (Received 21 June 2018)

The mutual interplay between superconductivity and magnetism in superconductor/ferromagnet heterostructures may give rise to unusual proximity effects beyond current knowledge. Especially, spin-triplet Cooper pairs could be created at carefully engineered superconductor/ferromagnet interfaces. Here we report a giant proximity effect on spin dynamics in superconductor/ferromagnet/superconductor Josephson junctions. Below the superconducting transition temperature  $T_{\rm C}$ , the ferromagnetic resonance field at X-band ( $\sim 9.0\,{\rm GHz}$ ) shifts rapidly to a lower field with decreasing temperature. In strong contrast, this phenomenon is absent in ferromagnet/superconductor bilayers and superconductor/insulator/ferromagnet/superconductor multilayers. Such an intriguing phenomenon can not be interpreted by the conventional Meissner effect. Instead, we propose that the strong influence on spin dynamics could be due to spin-transfer torque associated with spin-triplet supercurrents in ferromagnetic Josephson junctions with precessing magnetization.

PACS: 74.45.+c, 74.78.Fk, 76.50.+g DOI: 10.1088/0256-307X/35/7/077401

Spin-triplet supercurrents combining superconducting and magnetic orders provide a great opportunity to enhance the functionality and performance of spintronic devices by offering the possibility of long-range spin-polarized supercurrents. [1-4] As triplet Cooper pairs, unlike singlet pairs, can carry a net spin component, a spin-polarized current is naturally associated with the triplet supercurrents. Meanwhile, triplet Cooper pairs are immune to pair breaking by the exchange field in ferromagnets so that they sustain long-range correlations in spintronic devices. To use such triplet supercurrents in spintronics it is necessary to effectively generate and manipulate triplet pairs in devices.

In the past decade, a number of theoretical models have been proposed to explain how spin-polarized supercurrents can be created and controlled in superconductor/ferromagnet (S/F) heterostructures, [5-14] with key ingredients ranging from non-uniform superconductor, inhomogeneous and noncollinear magnetization to strong spin-orbital coupling, etc. The first experimental evidence for long-range triplet supercurrents was reported by Keizer et al.[15] from the observation of supercurrent passing through the halfmetallic ferromagnet CrO<sub>2</sub>. Then a series of experiments demonstrated systematic evidences for triplet supercurrents in S/F/S Josephson junctions. [16-20] Although these existing experiments provide compelling evidences for triplet pairing in S/F heterostructures, they are not directly probing or using the spin carried

by triplet supercurrents.

A well-known and useful phenomenon in spintronics is the spin-transfer torque induced by spinpolarized currents, which has been widely used to switch magnetization and control magnetization dynamics.<sup>[21]</sup> Similarly, the triplet supercurrents are anticipated to induce spin-transfer torques when passing through a ferromagnet. The demonstration of spin-transfer torques due to triplet supercurrents would not only confirm the net spin of triplet pairs but also pave the way for the emergence of superconducting spintronics. Recently, there are a number of works addressing on the spin-transfer torques and magnetization dynamics related to triplet supercurrents. [22-31] However, the experimental studies in this topic lie well behind theoretical progresses. So far no clear experimental evidence for spin-transfer torques produced by triplet supercurrents has been reported.

In this Letter, we investigate the ferromagnetic resonance (FMR) spectra in a series of S/F/S Josephson junctions. The results demonstrate a significant influence of superconductivity on magnetization dynamics: the resonance field  $H_r$  shifts rapidly to a lower field below superconducting transition temperature  $T_{\rm C}$ . In contrast, such an effect is absent in S/F bilayers and S/insulator/F/S multilayers. The conventional Meissner effect can not account for this phenomenon. Alternatively, we propose that the strong influence on spin dynamics by superconductivity in

<sup>\*</sup>Supported by the National Natural Science Foundation of China under Grant Nos 51725104 and 11534015, and the National Key Research and Development Program of China under Grant No 2016YFA0300700. Y.S. also acknowledges the support from Chinese Academy of Sciences under Grant No XDB07030200.

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ferromagnetic Josephson junctions could be due to spin-transfer torque induced by spin-triplet cooper pairs.  $^{[29-31]}$ 

The superconductor/ferromagnet heterostructures including Nb/Ni<sub>80</sub>Fe<sub>20</sub>/Nb Josephson junctions and Nb/Ni<sub>80</sub>Fe<sub>20</sub> bilayer are fabricated using dc magnetron sputtering on glass substrates. The base pressure of the sputtering system is about  $10^{-6}$  Pa. The films are deposited at an Ar pressure of 0.5 Pa. The MgO layer in the Nb/MgO/Ni<sub>80</sub>Fe<sub>20</sub>/Nb multilayer is deposited by radio frequency sputtering with an Ar pressure about 0.8 Pa. The FMR spectra at a fixed frequency (X-band,  $f \approx 9.0\,\mathrm{GHz}$ ) are measured in a JEOL FA-200 spectrometer with a cavity resonator. The system is equipped with a variable temperature unit down to liquid helium temperature.

The geometry of the ferromagnetic Josephson junctions and the schematic of FMR experiments are illustrated in Fig. 1(a). When a DC magnetic field  $\boldsymbol{H}_{\rm DC}$  is applied not along the direction of magnetization, the magnetization will rotate to the direction of  $\boldsymbol{H}_{\rm DC}$  along spiral path by the driven torque and damping torque. If a microwave field with magnetic component  $\boldsymbol{h}_{\rm mw}$  perpendicular to  $\boldsymbol{H}_{\rm DC}$  is applied, the magnetization can absorb microwave energy and precess continuously in balance with the damping torque. This is the basic principle of FMR.

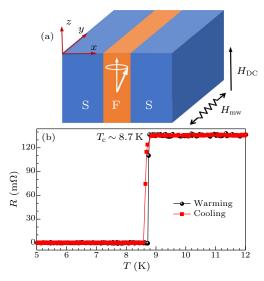


Fig. 1. Geometry of the ferromagnetic Josephson junctions and the configuration of FMR measurements. (a) The Josephson junctions in this study consist of a ferromagnetic layer (Ni $_{80}$ Fe $_{20}$ , 20 nm in thickness) and two superconductor layers (Nb, 100 nm in thickness). The FMR is measured at a fixed microwave frequency ( $\sim$ 9.0 GHz) while scanning the DC magnetic field applied in the film plane. (b) The resistance of the top Nb layer of a Nb/Ni $_{80}$ Fe $_{20}$ /Nb junction as a function of temperature. The superconducting transition temperature  $T_{\rm C}$  is  $\sim$ 8.7 K.

In our study, the ferromagnetic Josephson junctions are made of two superconducting layers of Nb (100 nm in thickness) and an FM layer of  $Ni_{80}Fe_{20}$  (20 nm in thickness). As shown in Fig. 1(b), the trans-

port measurement suggests a superconducting transition temperature  $T_{\rm C}\sim 8.7\,{\rm K}$  of the Nb layers in the Nb/Ni<sub>80</sub>Fe<sub>20</sub>/Nb junctions.

The FMR spectra of a Nb(100nm)/Ni<sub>80</sub>Fe<sub>20</sub>(20nm)/Nb(100 nm) Josephson junction measured at X-band ( $\sim 9\,\mathrm{GHz}$ ) in a cavity are shown in Fig. 2(a). All the resonance lines exhibit a single Lorenz lineshape. The resonance field  $H_r$  changes little with temperature above  $T_\mathrm{C}$ . However,  $H_r$  shifts rapidly to a lower field with decreasing temperature below  $T_\mathrm{C}$ . The temperature dependence of  $H_r$  is plotted in Fig. 2(b). As temperature decreases from  $T_\mathrm{C}$  to 4.2 K,  $H_r$  shifts by  $\sim 70\,\mathrm{mT}$ , indicating a strong influence on magnetization dynamics by superconductivity.

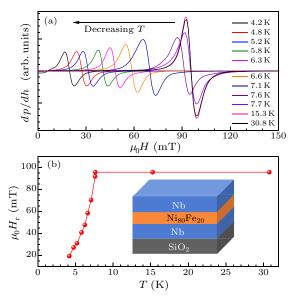


Fig. 2. FMR spectra of the Nb(100 nm)/Ni<sub>80</sub>Fe<sub>20</sub>(20 nm)/Nb(100 nm) Josephson junction. (a) The FMR spectra as a function of temperature. Below the superconducting transition temperature  $T_{\rm C}$ , the resonance field  $H_r$  shifts rapidly to a lower field with decreasing temperature. (b) The resonance field  $H_r$  as a function of temperature. The inset plots the structure of the sample. The significant shift of  $H_r$  below  $T_{\rm C}$  evidences a strong proximity effect on magnetization dynamics induced by superconductivity.

For comparison, we also measured the FMR spectra of a Nb(100 nm)/Ni<sub>80</sub>Fe<sub>20</sub>(20 nm) bilayer. As shown in Fig. 3(a), for the S/F bilayer,  $H_r$  does not shift obviously below  $T_{\rm C}$ . From 10 K to 4.2 K,  $H_r$  only shifts about 1 mT. We note that this observation is similar to a previous FMR study of S/F bilayers.<sup>[22]</sup> This control experiment clarifies that the shift of  $H_r$  is closely related to the geometry of ferromagnetic Josephson junctions rather than one S/F interface. According to previous studies, the saturation magnetization  $M_s$  of ferromagnetic layer changes little (<1%) below  $T_{\rm C}$  by the static magnetic interaction with superconductivity in the S/F/S trilayers and multilayers.<sup>[32,33]</sup> Thus, it cannot account for the significant shift of  $H_r$  (~70 mT) below  $T_{\rm C}$ .

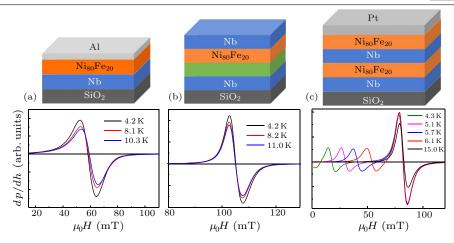


Fig. 3. Control experiments on S/F bilayer, S/I/F/S and S/F/S/F multilayers. (a) FMR spectra of a Nb(100 nm)/Nis<sub>0</sub>Fe<sub>20</sub>(20 nm) bilayer. The resonance field  $H_r$  shifts little below the superconducting transition temperature  $T_{\rm C}$ . (b) FMR spectra of a Nb(100 nm)/MgO(10 nm)/Nis<sub>0</sub>Fe<sub>20</sub>(20 nm)/Nb(100 nm) multilayer. The resonance field  $H_r$  does not shift below  $T_{\rm C}$ . (c) FMR spectra of a Nb(100 nm)/Nis<sub>0</sub>Fe<sub>20</sub>(20 nm)/Nb(100 nm)/Nis<sub>0</sub>Fe<sub>20</sub>(20 nm) multilayer. Above  $T_{\rm C}$ , there is only a single resonance line. Below  $T_{\rm C}$ , there are two separated lines from the top Nis<sub>0</sub>Fe<sub>20</sub> layer and the lower Nis<sub>0</sub>Fe<sub>20</sub> layer in the S/F/S junction, respectively.  $H_r$  of the former does not shift but of the latter shifts to lower field with decreasing temperature. These control experiments confirm that the shift of  $H_r$  is caused by supercurrents passing through the ferromagnetic layer in Josephson junctions rather than a local proximity effect at a single S/F interface or Meissner-like effect.

The shift of  $H_r$  to a lower field indicates that an effective inner magnetic field parallel to the external magnetic field is produced in the superconducting state. In other words, there should be an extra torque induced by superconductivity to assist the external field torque to keep the magnetization precession. As this extra torque below  $T_{\rm C}$  is observed in S/F/S junctions but not in the S/F bilayer, it implies that supercurrents passing through Josephson junctions, rather than a local proximity effect at one S/F interface, play a critical role. To verify this viewpoint, we have performed another control experiment in a Nb(100 nm)/MgO(10 nm)/Ni<sub>80</sub>Fe<sub>20</sub>(20 nm)/Nb(100 nm) multilayer where the supercurrents are blocked by the insulating MgO layer.

For typical S/MgO/S Josephson junctions, thickness of an MgO layer is usually below 2 nm. Above 2 nm the wave function can not overlap and tunneling supercurrents will be blocked. The FMR spectra of this insulating S/I/F/S multilayer is presented in Fig. 3(b). No obvious shift of  $H_r$  is observed below  $T_{\rm C}$ . From 11 K to 4.2 K,  $H_r$  only shifts about 1 mT. This second control experiment further suggests that the extra torque below  $T_{\rm C}$  is due to supercurrents passing through the ferromagnetic layer. Since singlet supercurrents do not carry a net spin and should not cause a spin-transfer torque, it is naturally concluded that the extra torque below  $T_{\rm C}$  is induced by triplet supercurrents.

In order to further confirm that the observed giant effect on magnetic dynamics is restricted to ferromagnetic Josephson junctions rather than a single F/S interface, we prepared an S/F/S/F multilayer which can be considered as a combination of an S/F/S junc-

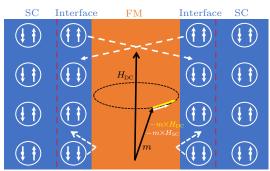
tion and an S/F bilayer. The FMR spectra of this  $Nb(100 \,\mathrm{nm})/Ni_{80}Fe_{20}(20 \,\mathrm{nm})/Nb(100 \,\mathrm{nm})/Ni_{80}Fe_{20}(20 \,\mathrm{nm})$  multilayer is shown in Fig. 3(c). Above  $T_{\rm C}$ , there is only a single resonance line because the two F layers have the same resonance field. Below  $T_{\rm C}$ , there are two separated resonance lines, one from the top  $Ni_{80}Fe_{20}$  layer and the other from the lower  $Ni_{80}Fe_{20}$  layer in the S/F/S junction, respectively.  $H_r$  of the former does not shift with temperature, but  $H_r$  of the latter shifts rapidly to lower field with decreasing temperature, similar to that observed in the S/F/S junctions. Overall, these control experiments confirm that the shift of  $H_r$  below superconducting  $T_{\rm C}$  only occurs in conducting ferromagnetic Josephson junctions.

In the following, we discuss the possible mechanism of the giant proximity effect observed in the S/F/S Josephson junctions. First, the Meissner effect can be excluded thoroughly by the control experiments. The giant shift of resonance field is only observable in the S/F/S junctions but absent in the S/F bilayer and S/I/F/S multilayer. If the change in resonance field is induced by flux focusing due to the Meissner effect of the superconducting Nb layers, one should have observed similar effects in the S/I/F/S multilayer because the thin MgO layer does not block the flux.

The FMR experiment in our study is a situation of ferromagnetic Josephson junctions with precessing magnetization. Several theoretical models<sup>[28-31]</sup> have discussed on this situation and predicted that the long range triplet supercurrents can be stimulated by varying *in time* (rather than *in space*) the orientation of the magnetization in the ferromagnet, and the triplet

supercurrent is pumped by using FMR in a ferromagnetic Josephson junction.

Figure 4 presents a schematic illustration of the dynamic process in the ferromagnetic Josephson junctions. Away from the S/F interfaces in the superconductors, only spin-singlet Cooper pairs can exist below  $T_{\rm C}$ . A conversion from spin-singlet pairs to spin-triplet pairs occurs due to the spin-mixing and spin-flip scattering at the interfaces.<sup>[2]</sup> The dynamically precessing magnetization plays an important role for the conversion process, [29] by which the coherent charge and spin transport takes place through the junction due to the conservation of total spin angular momentum carried by triplet pairs and magnons.<sup>[28–30]</sup> For the triplet pairs with up spins (parallel to the external DC magnetic field), they can pass through the F layer. However, for the triplet pairs with down spins, they will be reflected back to the interface. Then triplet pairs passing through the ferromagnetic metal between two S/F interfaces at the precessing frequency produce a high-density AC triplet supercurrent.<sup>[32]</sup> Since the triplet supercurrent is spin polarized, it exerts a strong torque on the precessing magnetization. This torque has the same direction with the torque generated by the external DC magnetic field. As a consequence, the resonance field  $H_r$  shifts to a lower field.



Triplet pairs transport between two interfaces at the resonance frequency

Fig. 4. Schematic illustration of AC triplet supercurrents induced spin-transfer torque in S/F/S Josephson junctions with precessing magnetization. Triplet cooper pairs are generated at the interfaces due to the precessing magnetization. Away from the S/F interfaces in the superconductors (SC), only singlet Cooper pairs can exist. The triplet pairs with up spins (parallel to the external DC magnetic field) can transport through the ferromagnetic (FM) layer periodically at the precessing frequency. The induced AC triplet supercurrent exerts a strong spin-transfer torque on the magnetization, causing a giant shift of the resonance field

In summary, our FMR experiments in the S/F/S Josephson junction demonstrate a significant modification on magnetization dynamics induced by superconductivity. In contrast, such a phenomenon is absent in S/F bilayers and S/I/F/S multilayers. These

results can not be understood by the simple Meissner effect of superconductivity. Instead, we propose that there could be magnetization-precessing-induced AC triplet supercurrent in the ferromagnetic Josephson junctions and the resonance field shift is a consequence of the spin-transfer torque produced by the AC triplet supercurrent.

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