Triplet Josephson Effect with Magnetic Feedback in a Superconductor-Ferromagnet Heterostructure

V. Braude and Ya. M. Blanter

Kavli Institute of Nanoscience, Delft University of Technology, 2628 CJ Delft, The Netherlands

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We study the ac Josephson effect in a superconductor-ferromagnet heterostructure with a variable magnetic configuration. The system supports triplet proximity correlations whose dynamics is coupled to the magnetic dynamics. This feedback dramatically modifies the behavior of the junction. The current-phase relation becomes double periodic at both very low and high Josephson frequencies ω_J. At intermediate frequencies, the periodicity in $\omega_J t$ may be lost.

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Spin-dependent transport through hybrid structures combining ferromagnets (F) and normal metals has recently attracted a lot of interest, motivated by the prospect of potential technological applications in the field of spintronics [1]. Particular attention is given to two complementary effects involving mutual influence between electric current and magnetic configuration. The first is giant magnetoresistance [2] in which the conductance is much larger when different magnetic regions have their magnetic moments aligned than when they are antialigned. The opposite effect is the appearance of torques acting on magnetic moments when an electric current flows through the system [3]. These *nonequilibrium* current-induced torques appear due to nonconservation of spin currents accompanying a flow of charge through ferromagnetic regions. They allow manipulation of the magnetic configuration, including switching between the opposite directions or steady-state precession, without application of magnetic fields [4]. The two effects combined promise important practical applications in nonvolatile memory, programmable logic, and microwave oscillators.

A conceptually different situation occurs when a ferromagnet is coupled to a superconductor (S), since the spin current through the superconducting part vanishes [5]. This additional constraint modifies the nonequilibrium torques, opening the possibility of perpendicular alignment of magnetic moments. Furthermore, if a magnetic structure is contacted by *two* superconductors, the proximity effect may be present, leading to a finite Josephson current through the structure at equilibrium. The torques generated by this current correspond to an *equilibrium* effective exchange interaction between the magnetic moments which can be controlled by the phase difference between the superconductors [6]. The same mechanism enables the reciprocal effect in which the supercurrent depends on the magnetic configuration.

For a uniform ferromagnet, the observation of these effects requires a very thin magnetic layer, since the proximity effect is suppressed at short distances. However, in nonuniform ferromagnets, a long-range proximity effect can exist due to triplet superconducting correlations [7]. This triplet proximity effect (TPE) strongly enhances the associated Josephson current. It is important that TPE essentially depends on the magnetic configuration of the system [7]. Hence S/F multilayers exhibiting TPE are especially suitable for studying the Josephson-induced magnetic exchange interaction. By varying the relative magnetization directions of different magnetic regions, one can control the supercurrent flowing through the structure. Then, if the magnetic configuration is allowed to respond to the Josephson-current induced torques, it creates feedback for the supercurrent and considerably modifies it.

In this Letter, we consider the magnetic exchange interaction induced by Josephson currents in a dirty S/F heterostructure exhibiting TPE. We show that this interaction favors noncollinear magnetic configurations, and the preferred direction depends continuously on the superconducting phase difference ϕ. Thus, the static magnetic configuration can be controlled by the applied phase difference. We then consider the influence of feedback from the magnetic moments on the ac Josephson effect. We demonstrate that the magnetic system exhibits a range of different behaviors, from simple harmonic oscillations to fractional-frequency periodic behavior and chaotic motion. The magnetic feedback complicates the behavior of the current in the time domain, making it generally impossible to express it in terms of a current-phase relation. This is in contrast with the ac Josephson effect without the magnetic feedback, where the time dependence of the supercurrent J is determined by the current-phase relation ($J = J_c \sin\phi$ for TPE in diffusive systems, with the exception of the magnetic configuration with mutually perpendicular directions where a transition between "0" and "π" states occurs [8]).

On the other hand, we find that both in the low- and high-frequency limit the current-phase relation becomes meaningful, with the current exhibiting a double-phase dependence, $J \propto \sin 2\phi(t)$ [9] or $J \propto \cos 2\phi(t)$. The critical current in the low-frequency regime is of the order of the value E_{Th}/eR_n, characteristic for diffusive systems, E_{Th} and R_n being Thouless energy and normal-state resistance of the junction, respectively. The unusual cosine dependence of the Josephson current appears when Gilbert damping is important in the magnetic dynamics, breaking the time-reversal symmetry. At high frequencies ω_J, the magnetization cannot effectively follow the phase variation, leading to a $1/\omega_J^2$ suppression of the effective Josephson coupling. At even higher frequencies, the damping is dominant, and the frequency dependence becomes $1/\omega_J$. The presence of damping is expressed in the appearance of a dc component of the current leading to a finite resistance.

The system.—We consider the minimum discrete setup exhibiting TPE (Fig. 1). Two magnetic regions, 1 and 3, are adjacent to the superconducting reservoirs that induce proximity mini-gaps $\Delta_{1,3}$ in them. Between these regions there is an additional single-domain magnetic region, 2, whose length is much larger than the coherence length ξ_h (and shorter than the coherence length in the absence of exchange field ξ_N), where triplet superconducting correlations are induced. This region is assumed to be weakly polarized (metallic), so that both spin directions are present at the Fermi surface. The magnetic regions are characterized by the exchange energies h_i, while the magnetization directions $\boldsymbol{n}_i$ are specified by the angles θ_1, θ_3, and χ (Fig. 1). Assuming that the conductances of these regions are much higher than those $g^n_{1,3}$ of the connectors between them, our system can be described by a circuit-theory model for TPE [8].

Let us now discuss the minimum requirements for the magnetic feedback. If all three directions of the magnetization $\boldsymbol{n}_{1-3}$ can freely rotate, they always prefer to be aligned, thus suppressing the TPE. The same situation occurs if any two of these vectors are free. In order to induce TPE, we thus need to fix the directions of two of the vectors. If only the middle vector $\boldsymbol{n}_2$ is allowed to rotate, it will assume its equilibrium position along one of the two bisectors between the fixed directions $\boldsymbol{n}_1$ and $\boldsymbol{n}_3$. On the other hand, if the magnetic vector of one of the outer regions is allowed to rotate, an interesting situation arises when its equilibrium direction continuously depends on the superconducting phase difference across the junction. In order to explore this situation, we assume that $\boldsymbol{n}_1$ and $\boldsymbol{n}_2$ are fixed, e.g., by pinning to an antiferromagnetic substrate, or by geometrical shaping, with the angle between them being θ_1. Magnetization $\boldsymbol{n}_3$ is free to rotate, with region 3 separated by a normal spacer from region 2 in order to avoid exchange coupling between them.

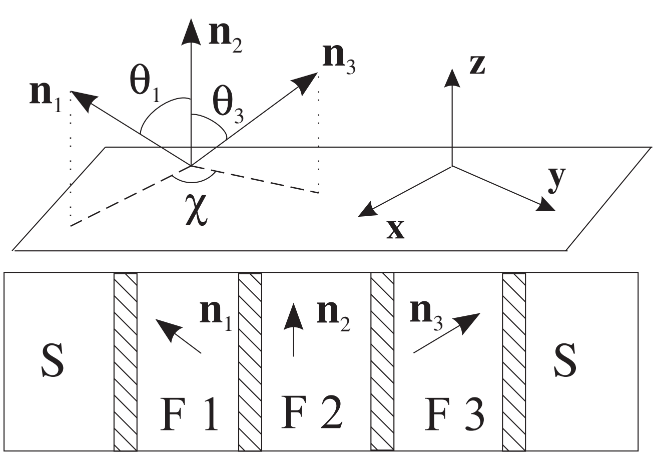

FIG. 1. The experimental setup.

In accordance with the model assumptions, regions 1 and 3 act as effective S/F reservoirs; hence, their energies are independent of the magnetic configuration. On the other hand, triplet superconducting correlations extending through region 2 are very sensitive to the magnetization directions. The configuration-dependent part of the energy can be found by integrating over the density of states (DOS) in region 2. The DOS for each spin direction is given by [8]

$$\nu^{\uparrow,\downarrow}(\varepsilon) = \frac{\nu_0}{2}\,\mathrm{Re}\left(1 - \frac{a_1^2 + a_3^2 + 2a_1 a_3\cos(\phi \pm \chi)}{(b_1 + b_3 - i\epsilon/E_{\mathrm{Th}})^2}\right)^{-(1/2)}, \tag{1}$$

where ν_0 is the normal-state DOS, $a_k = g_k^n|\Delta_k|\sin\theta_k/(g_1^n + g_3^n)\sqrt{h_k^2 - |\Delta_k|^2}$, and $b_k = g_k^n h_k/(g_1^n + g_3^n)\sqrt{h_k^2 - |\Delta_k|^2}$. The energy is given by a logarithmic integral, and the main contribution comes from $\epsilon \gg E_{\mathrm{Th}}$. In the leading order one obtains

$$E = \frac{\nu_0 v_2}{2}\log\frac{\Delta_{\mathrm{cut}}}{E_{\mathrm{Th}}} E_{\mathrm{Th}}^2(a_1^2 + a_3^2 + 2a_1 a_3\cos\phi\cos\chi), \tag{2}$$

where v_2 is the volume of the magnetic region 2 and $\Delta_{\mathrm{cut}} \simeq \min(\Delta_i, h_i - \Delta_i)$ is a cutoff energy. This expression can be written in a form presenting explicitly the dependence on the orientation angles θ_3 and χ,

$$E = p_3^2\sin^2\theta_3 + 2p_1p_3\sin\theta_3\cos\phi\cos\chi, \tag{3}$$

with $p_{1,3}$ being effective exchange couplings for the magnetic vector $\boldsymbol{n}_3$. The stable configuration is achieved when all magnetization directions are in the same $(\boldsymbol{xz})$ plane, and $\boldsymbol{n}_3$ is tilted with respect to $\boldsymbol{n}_2$,

$$\sin\theta_3 = (p_1/p_3)|\cos\phi|. \tag{4}$$

This angle depends continuously on the applied superconducting phase difference ϕ, while the angle χ assumes the values 0 or π so that the product $\cos\phi\cos\chi$ is negative. In fact, there are two stable directions, given by the angles θ_3 and $\pi - \theta_3$. In what follows we treat them as equivalent, since they correspond to the same current. The energy of the stable configuration is given by $E_{\min} = -p_1^2\cos^2\phi$. Hence, allowing the magnetization direction $\boldsymbol{n}_3$ to orient itself along the stable direction leads to the current-phase relation $J = J_c\sin 2\phi$.

Low frequencies.—When a small voltage V is applied to the structure, such that the corresponding Josephson frequency $\omega_J = 2eV/\hbar$ is much smaller than the characteristic frequency of the magnetic system ω_m (see below), the vector $\boldsymbol{n}_3$ follows the stable direction given by Eq. (4), performing slow oscillations in the $\boldsymbol{x}$-$\boldsymbol{z}$ plane. The alternating Josephson current oscillates with the double frequency

$$J = \frac{2e}{\hbar} p_1^2 \sin \frac{4eV}{\hbar} t, \tag{5}$$

while the critical current remains of the same order of magnitude as in the case with a fixed magnetic configuration.

For higher Josephson frequencies, the variation of $\boldsymbol{n}_3$ is no more limited to the $\boldsymbol{x}$-$\boldsymbol{z}$ plane. Instead, the magnetization performs a variety of nonharmonic motions whose frequency may be a multiple or a fraction of the driving frequency ω_J [Figs. 2(a), 2(c), and 2(d)]. For certain trajectories the time average of θ_3 is finite [Fig. 2(c)], corresponding to a tilt of $\boldsymbol{n}_3$ away from the equilibrium in response to an applied voltage. Within some frequency intervals, the motion is chaotic, as shown in Fig. 2(b). In these intermediate regimes, the Josephson current shows a complicated time dependence which is generally not periodic in $2\pi/\omega_J$. Hence this dependence cannot be parametrized in terms of the phase. Instead, one can speak of a Josephson current with a time-dependent coupling.

High frequencies.—If the Josephson frequency is much higher than the magnetic frequencies, the vector $\boldsymbol{n}_3$ cannot effectively follow the fast oscillations of the potential, and the time-averaged potential seen by $\boldsymbol{n}_3$ has a minimum for $\boldsymbol{n}_3 \parallel \boldsymbol{z}$. The motion of $\boldsymbol{n}_3$ can be determined by expanding $\boldsymbol{n}_3 = \boldsymbol{z} + \delta\boldsymbol{n}$ and using a linearized Landau-Lifshits-Gilbert (LLG) equation,

$$\delta\dot{\boldsymbol{n}} = \boldsymbol{z}(-\gamma \times \boldsymbol{H}_{\text{eff}} + \alpha \delta \boldsymbol{n}), \tag{6}$$

where γ is the gyromagnetic ratio, α is the damping coefficient, $\boldsymbol{H}_{\text{eff}} = -\partial E/\partial \boldsymbol{m}_3$ is the effective field, and $\boldsymbol{m}_3$ is the magnetization density of region 3.

When Gilbert damping is negligible, the trajectory of $\boldsymbol{n}_3$ has a very low aspect ratio, so that the motion is almost completely confined to the $\boldsymbol{y}$ axis. It is given by

$$\delta n_x = \frac{\gamma^2 p_1 p_3^3 \hbar^2}{e^2 V^2 m_3^2} \cos \frac{2eVt}{\hbar}, \qquad \delta n_y = \frac{\gamma p_1 p_3 \hbar}{eVm_3} \sin \frac{2eVt}{\hbar}.$$

Thus at high frequencies, $\boldsymbol{n}_3$ precesses in phase with the voltage pumping. This leads to an increase in the Josephson energy, and, correspondingly, a negative Josephson current,

$$J = -\frac{2\hbar}{e} \left(\frac{\gamma p_1 p_3^2}{V m_3} \right)^2 \sin \frac{4eVt}{\hbar}. \tag{7}$$

Hence in the high-frequency regime the system shows not only frequency doubling, but also an effective π junction behavior. The magnitude of the current is suppressed as $\sim V^{-2}$ as shown in Fig. 3.

The neglect of damping is justified as long as $\alpha\omega_J \ll \omega_m = \gamma p_3^2/m_3$. When the voltage is high enough, this condition is not satisfied anymore, and the dissipation starts to be important. In the opposite case $\alpha\omega_J \gg \omega_m$, the motion of $\boldsymbol{n}_3$ is determined by the driving against the damping force,

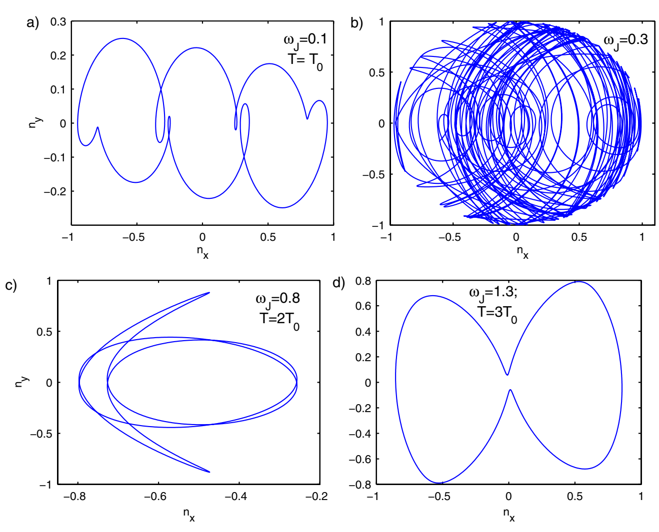

FIG. 2 (color online). Trajectories of the magnetization vector in the x-y plane for different Josephson frequencies (given in units of ω_m). Trajectory (b) is chaotic, while trajectory (c) has a finite zero-frequency component for n_x. Here T is the period of the trajectory, and $T_0 = 2\pi/\omega_J$. For comparison, the low-frequency trajectory lies entirely on the $\boldsymbol{x}$ axis.

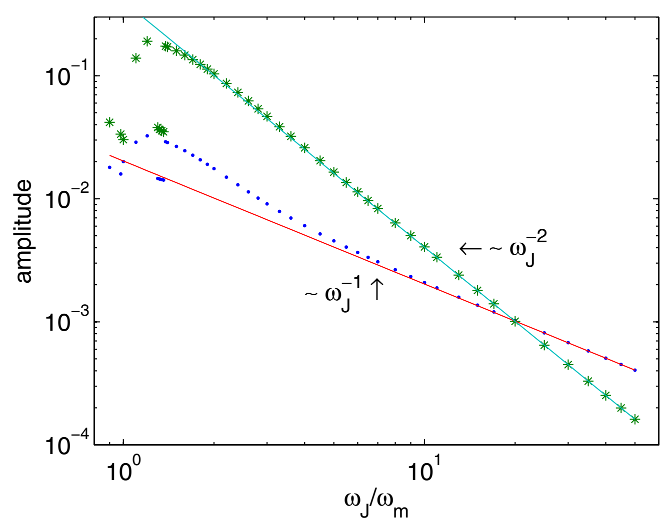

FIG. 3 (color online). The absolute value of the Josephson-current harmonics proportional to $\sin 2\omega_J t$ (asterisks) and to $1 - \cos 2\omega_J t$ (dots). Solid lines are fits $1/\omega_J$ and ω_J^{-2}. The data points are obtained from numerical integration of the full (nonlinear) LLG equation.

$$\delta \boldsymbol{n} = \frac{\gamma p_1 p_3 \hbar}{eVm_3}(-\alpha \hat{\boldsymbol{x}} + \hat{\boldsymbol{y}}) \sin\frac{2eVt}{\hbar}. \quad (8)$$

Then the Josephson current is given by

$$J = \frac{2\alpha\gamma p_1^2 p_3^2}{m_3 V}\left(1 - \cos\frac{4eVt}{\hbar}\right). \quad (9)$$

Note the unusual cosine dependence on the phase. It occurs since the time-reversal symmetry is broken by the dissipation in this regime. Due to the same reason, a zero-frequency component of the current appears, signifying the onset of a finite nonlinear resistance across the structure. Since this regime is governed by the damping, the current amplitude is proportional to α, while the suppression $\sim 1/V$ is weaker in this regime (Fig. 3).

To estimate the magnetic dynamics frequency ω_m, we use typical values $E_{\mathrm{Th}} \sim 1$ meV, $\nu_0 \sim 1/(\mathrm{eV/atom})$, and $m_3 \sim 1\mu_B/\mathrm{atom}$, where μ_B is the Bohr magneton. Then $\omega_m \sim v_2/v_3$ GHz, where $v_{2,3}$ are the volumes of the corresponding magnetic regions. As this frequency is quite low, observation of the high-frequency regimes should present no difficulty. On the other hand, the low-frequency ac regime would require extremely low voltages, below 1 μV. A reasonable alternative would be incorporating the structure in a superconducting loop and measuring the Josephson current as a function of the applied flux.

Applicability of our model requires that any magnetic anisotropy of part 3 should be smaller than the proximity-induced energy, Eq. (2). With the above values of the parameters it is of the order of $10^4 \times v_2$ J/m^3; thus, one should choose materials with a low value of the crystalline anisotropy, such as permalloy. Generally, the observation of the effect would be easier in materials with the low exchange field. Finally, we emphasize that the properties discussed above are specific for metallic systems. In half-metals, the behavior will be very different. Thus, in the low-frequency regime $\boldsymbol{n}_3$ precesses around $\boldsymbol{n}_2$ at a constant angle θ_3, while the Josephson current vanishes.

Conclusions.—We have considered the ac Josephson effect in a SFS structure with magnetic dynamics coupled to the dynamics of superconducting correlations. The magnetic configuration in the structure was assumed to be nonuniform so that the structure exhibits TPE. Variation of the magnetic configuration is shown to essentially modify the current behavior that can be observed in the appearance of fractional Shapiro steps. Thus measurement of the Josephson current would provide information about the coupling and self-consistent feedback dynamics between the superconducting and magnetic degrees of freedom. The coupling also allows control of the magnetization direction by means of applied voltage or superconducting phase. In the low-frequency limit, the magnetization follows the immediate potential minimum, leading to a $\sin 2\phi$ current-phase relation. The critical current has the same order of magnitude E_{Th}/eR_n as the one due to the usual singlet proximity effect in dirty structures. In the high-frequency regime, as long as the damping is not important, the Josephson current is negative, corresponding to a π-junction behavior. It is suppressed by a factor $(\omega_m/\omega_J)^2$ relative to the low-frequency regime. At even higher frequencies, Gilbert damping starts playing the major role in the dynamics. Then the time-reversal symmetry is broken and the current-phase relation takes an unusual cosine form. In addition, a dc component of the current appears, manifesting itself in a finite resistance. The current suppression becomes weaker in this regime. The even current-phase relation has been recently predicted in Ref. [13] for a system with spin-active interfaces. Note, however, that in our case for high frequencies the current-phase relation is not meaningful, and the even time dependence appears due to the damping.

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[9] In general, Josephson junctions with double-periodic behavior may be useful in flux-qubit design schemes [10]. Josephson frequency doubling was predicted in junctions involving unconventional superconductors [10,11] and observed in experiments involving *d*-wave grain-boundary junctions [12]. However, in contrast to the situation studied in this Letter, in all these cases the frequency doubling occurs at isolated points in the parameter space where the first harmonic vanishes, and the magnitude of the current is suppressed in comparison with the usual value $\sim\Delta/eR_n$, where R_n is the normal-state resistance.
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