Functional Superconductor Interfaces from Broken Time-Reversal Symmetry

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The breaking of time-reversal symmetry in a triplet superconductor Josephson junction is shown to cause a magnetic instability of the tunneling barrier. Using a Ginzburg-Landau analysis of the free energy, we predict that this novel functional behavior reflects the formation of an exotic Josephson state, distinguished by the existence of fractional flux quanta at the barrier. The crucial role of the orbital pairing state is demonstrated by studying complementary microscopic models of the junction. Signatures of the magnetic instability are found in the critical current of the junction.

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Introduction.—The Josephson effect between superconductors separated by a tunneling barrier remains of fundamental interest, in particular, as a phase-sensitive test of the pairing symmetry of unconventional superconductors [1] and in the study of the interplay of superconductivity with magnetism [2]. Although the qualitative features of the Josephson effect are fixed by the quantum nature of the superconductors, the modification of the superconducting state at the junction interface, the so-called proximity effect [1,2], must often be included in the description of the supercurrent transmission. In contrast, the properties of the barrier are usually regarded as fixed [1]. Recently, however, it has been shown that a thin ferromagnetic (FM) layer on a singlet superconductor (SSC) can show novel behavior [2], such as a suppression of the uniform magnetization [3] or the appearance of a domain structure [4]. These effects result from the competition between the SSC and FM phases, and indicate that the tunneling barrier in a Josephson junction is not necessarily independent of the superconductors. The Josephson coupling can also produce novel spin dynamics in a magnetic tunneling barrier [5].

In this Letter, we consider the possibility of using the presence of the two superconductors to induce a magnetic instability of a nonmagnetic tunneling barrier in a Josephson junction. In particular, by both general phenomenological arguments and solution of specific microscopic models, we show that such a novel functionality of the barrier can develop for time-reversal symmetry (TRS) breaking configurations of two spin-triplet superconductors on either side. The Josephson coupling across the tunneling barrier is essential to this effect, which manifests itself as an exotic state distinguished, for example, by the existence of fractional flux quanta at the barrier [6]. Moreover, such a junction displays an anomalous temperature dependence of the critical current.

Phenomenological theory.—The intrinsic spin structure of the Cooper pairs in a triplet superconductor (TSC) requires a quasiparticle gap with three components $\tilde{\Delta}_{S_z}$ for each z component of spin $S_z = -1, 0, +1$, each of which has odd orbital parity, i.e., $\tilde{\Delta}_{S_z}(-\mathbf{k}) = -\tilde{\Delta}_{S_z}(\mathbf{k})$. The TSC order parameter is the so-called $\mathbf{d}$ vector, defined in spin space $\mathbf{d} = \frac{1}{2}(\tilde{\Delta}_{-1} - \tilde{\Delta}_1)\hat{\mathbf{x}} - \frac{i}{2}(\tilde{\Delta}_{-1} + \tilde{\Delta}_1)\hat{\mathbf{y}} + \tilde{\Delta}_0\hat{\mathbf{z}}$. Although many different triplet pairing states are allowed by symmetry, only a few examples of TSCs have been discovered so far, e.g., Sr_2RuO_4 [7], UGe_2 [8]. In Sr_2RuO_4 the pairing state has been identified as unitary and equal spin- pairing [9]; i.e., the spins of the Cooper pairs lie in the same plane perpendicular to $\mathbf{d}$, but the condensate does not have a net spin. Restricting ourselves to such pairing states, we write $\mathbf{d} = \tilde{\mathbf{d}}e^{i\phi}$ where $\tilde{\mathbf{d}}$ is a real vector. The vector form of the TSC order parameter reveals a novel degree of freedom in Josephson junction physics: in addition to the phase difference between the condensates to the left and right of the barrier, which controls the Josephson supercurrent, there is also the mutual alignment of the left ($\tilde{\mathbf{d}}_L$) and right ($\tilde{\mathbf{d}}_R$) vectors, which controls the magnetic aspects of the transport [10]. In particular, when $\tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R \neq 0$, a Cooper pair tunneling across the barrier undergoes a reconstruction of its spin state. This causes the formation of a nonunitary state localized at the interface, which supports an effective spin through the TRS breaking combination $\langle \mathbf{S} \rangle = i\mathbf{d}_L \times \mathbf{d}_R^* + \text{H.c.}$.

The violation of TRS at the tunneling barrier by the misaligned $\tilde{\mathbf{d}}$ vectors allows the TSCs to directly couple to a magnetization $\mathbf{M}$ of the interface. As we will see, the TSCs may in fact change the electronic properties of the interface so as to stabilize a spontaneous FM order. This novel behavior can be understood on a phenomenological level by a Ginzburg-Landau analysis of the free energy. Introducing the TSC order parameters for each side of the interface as $\mathbf{d}_L = \tilde{\mathbf{d}}_L e^{i\phi_L}$ and $\mathbf{d}_R = \tilde{\mathbf{d}}_R e^{i\phi_R}$, we write the free energy of the junction F to lowest order in $\mathbf{M}$ and $\tilde{\mathbf{d}}_{L,R}$ as

$$F = \frac{|\mathbf{M}|^2}{2\chi} - t\tilde{\mathbf{d}}_L \cdot \tilde{\mathbf{d}}_R \cos(\phi) + 2\gamma \mathbf{M} \cdot (\tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R)\sin(\phi). \tag{1}$$

Here $\phi = \phi_R - \phi_L$ is the phase difference, χ denotes the intrinsic uniform spin susceptibility of the barrier, and t and γ are phenomenological parameters. The ground state of the junction is obtained by minimizing F with respect to $\mathbf{M}$ and ϕ. We find that the nonmagnetic state of the barrier is unstable for $\chi\gamma^2|\tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R|^2 > t\tilde{\mathbf{d}}_L \cdot \tilde{\mathbf{d}}_R$: the coupling to the intrinsic spin $\langle \mathbf{S} \rangle = 2\tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R \sin(\phi)$ of the junction instead realizes a TRS breaking state [11], characterized by a nonvanishing magnetization $\mathbf{M} \parallel \tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R$, and a phase $\phi = \phi_{\min} \neq 0, \pi$. The junction is then in an exotic *fractional* state [6], where the magnetic barrier is capable of carrying flux lines with noninteger multiples of the flux quantum $\Phi_0 = hc/2e$. The observation of this characteristic feature is discussed below.

Microscopic models.—Although the misalignment of the $\tilde{\mathbf{d}}$ vectors is essential to the magnetic instability, it is also controlled by the specific details of the junction through the susceptibility χ and the parameters t and γ. The latter are fixed by the orbital part of the bulk TSC state. To elucidate the crucial role this plays in the magnetic instability, we examine two complementary microscopic models of the junction. The first, shown in Fig. 1(a), has the TSCs in a p_z-wave orbital state (the p_z-p_z junction), and the left and right $\tilde{\mathbf{d}}$ vectors are aligned parallel to the barrier but at an angle 2η with respect to each other. In the second model the orbital state is p_y wave (the p_y-p_y junction), and the $\tilde{\mathbf{d}}$ vectors lie in the x-z plane but are again misoriented by the angle 2η [Fig. 1(b)]. The TSCs extend indefinitely along the z axis, and we have translational invariance in the x-y plane. The TSC gaps are spatially constant, and the maximum gap magnitude $\Delta(T)$ displays weak-coupling temperature dependence with $\Delta(T=0) = \Delta_0$. The width of the tunneling barrier is regarded to be much smaller than the coherence length of the TSCs, and so it is approximated as a δ function with normal-state height $U_P = Z\hbar v_F$. Here Z is a dimensionless quantity and $v_F = \hbar k_F/m$ is the Fermi velocity in the bulk TSCs, which are assumed to have spherical Fermi surfaces of radius k_F. If the barrier supports a magnetic moment $\mathbf{M}$, the effective barrier height for spin-σ quasiparticles with spin parallel to $\mathbf{M}$ in dimensionless units is $Z - \sigma M$, where $M = g\mu_B\mu_0|\mathbf{M}|/\hbar^2 v_F$.

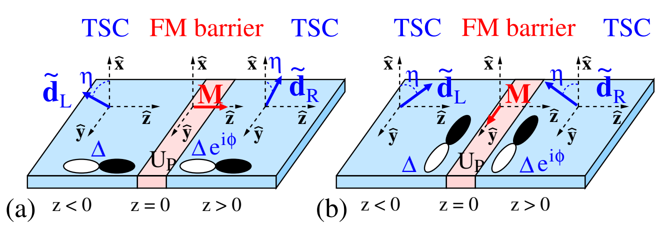

FIG. 1 (color online). (a) The p_z-p_z junction, with p_z-wave orbital pairing in the two superconductors, $\tilde{\mathbf{d}}$ vectors in the x-y plane, and an induced magnetic moment along the z axis. (b) The p_y-p_y junction, with p_y-wave orbital pairing in the two superconductors, $\tilde{\mathbf{d}}$ vectors in the x-z plane, and an induced magnetic moment along the y axis.

The Josephson coupling between the two TSCs is dominated by tunneling through Andreev bound states (ABSs) localized at the junction interface [1]. The ABSs result from the hybridization of the surface wave functions of each TSC, and hence implicitly describe the local nonunitary state and its coupling to the barrier moment. We solve the Bogoliubov–de Gennes equations [12] to obtain the ABS energies $E_{\mathbf{k},\sigma}$:

$$E_{\mathbf{k},\sigma} = \begin{cases} |\Delta(T)k_z|\sqrt{\mathcal{T}_\sigma(\mathbf{k})}\cos(\phi/2 - \sigma\eta) & p_z\text{-}p_z \\ |\Delta(T)k_y|\sqrt{1 - \mathcal{T}_\sigma(\mathbf{k})\sin^2(\phi/2 + \sigma\eta)} & p_y\text{-}p_y. \end{cases} \tag{2}$$

The ABSs are indexed by the component of spin $\sigma = \pm 1$ parallel to $\tilde{\mathbf{d}}_L \times \tilde{\mathbf{d}}_R$ and the wave vector $\mathbf{k}$ for incident trajectories from the left-hand side. Each state has two branches at $\pm E_{\mathbf{k},\sigma}$. $\mathcal{T}_\sigma(\mathbf{k}) = k_z^2/(k_z^2 + [Z - \sigma M]^2 k_F^2)$ is the transparency of the barrier to spin-σ quasiparticles [13]. We plot the ABS spectrum as a function of ϕ in Fig. 2. Note that the ABSs always intersect the line $E = 0$ in the p_z-p_z junction. These so-called zero-energy states are guaranteed by the arrangement of the p-wave orbitals, such that all specularly reflected quasiparticles experience a sign change of the gap. As the gap does not change sign upon reflection in the p_y-p_y junction, in contrast, zero-energy states only occur here at $\mathcal{T}_\sigma(\mathbf{k}) = 1$ [1].

The electronic contribution to the free energy is a weighted sum over the incident trajectories [1,14]

$$F_{\rm el} = -k_B T \sum_{\mathbf{k}} \sum_{\sigma} \frac{|k_z|}{k_F} \log\left[2\cosh\left(\frac{E_{\mathbf{k},\sigma}}{2k_B T}\right)\right]. \tag{3}$$

As in Eq. (1), the magnetic free energy of the barrier is included to lowest order $F_{\rm mag} = M^2/2\chi$, where χ is given in units of $(g\mu_B\mu_0/\hbar^2 v_F)^2/\Delta_0$. We vary ϕ and M to find the global free energy minimum of $F_{\rm el} + F_{\rm mag}$. Typical minimizing values for the p_z-p_z and p_y-p_y junctions are plotted as a function of temperature in Figs. 3(a) and 3(b), respectively. The barrier undergoes a magnetic instability at sufficiently large χ, and below a critical temperature $T_M < T_c$ such that the FM state appears only in the pres-

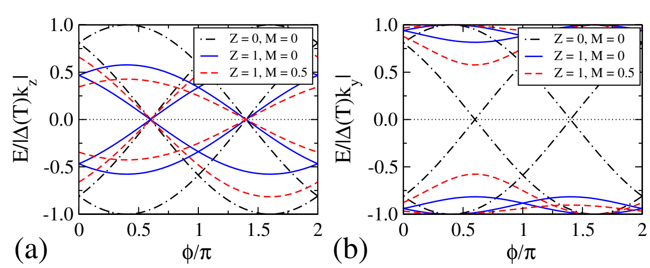

FIG. 2 (color online). The Andreev bound state spectrum in (a) the p_z-p_z and (b) the p_y-p_y junction for $k_z = k_y = k_F/\sqrt{2}$.

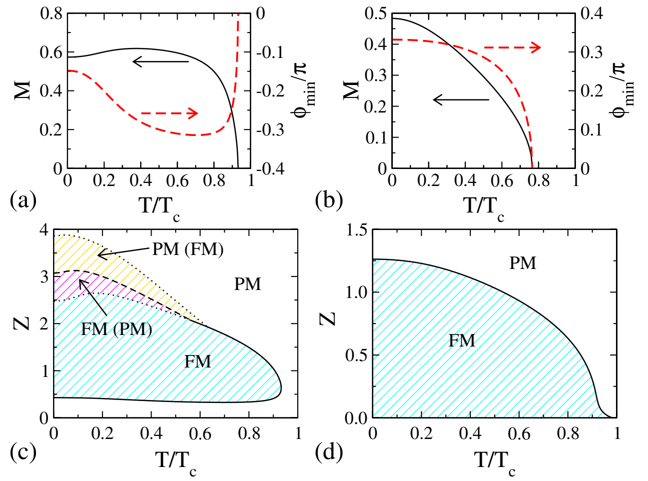

FIG. 3 (color online). Induced magnetic moment M and stable phase difference $\phi_{\min}$ as a function of reduced temperature for (a) the p_z-p_z and (b) the p_y-p_y junction. In both panels we set $\eta = 0.2\pi$, $Z = 0.7$, and $\chi = 20$. Magnetic phase diagram for the (c) p_z-p_z and (d) p_y-p_y junctions as a function of Z and T. A magnetic moment is stable in the region labeled FM, while the region of nonmagnetic behavior is denoted as PM; metastable states are shown in brackets. Second-order transitions are indicated by a solid line, first-order transitions by a dashed line, and the limits of the metastable states by a dotted line. η and χ are as in (a) and (b).

ence of superconductivity. For $Z \neq 0$ the junction is in a fractional state below T_M with two degenerate free energy minima $(M, \phi_{\min})$ and $(-M, -\phi_{\min})$ (broken TRS).

In Figs. 3(c) and 3(d) we show the phase diagram as a function of Z and T at fixed χ for the p_z-p_z and p_y-p_y junctions, respectively. The qualitatively different form of these phase diagrams follows from the dependence of the ABS spectra Eq. (3) upon the $\mathcal{T}_\sigma(\mathbf{k})$: due to the zero-energy states in the p_z-p_z junction, each bound state $E_{\mathbf{k},\sigma}$ monotonically shifts towards the middle of the gap with decreasing $\mathcal{T}_\sigma(\mathbf{k})$; for the p_y-p_y junction, in contrast, the states move towards the gap edges. From Eq. (3), the free energy contributed by $E_{\mathbf{k},\sigma}$ in a p_z-p_z junction will therefore increase (decrease) as the transparency $\mathcal{T}_\sigma(\mathbf{k})$ decreases (increases), while the opposite is true for the p_y-p_y junction.

It is immediately clear that the p_z-p_z junction is nonmagnetic at $Z = 0$, as $M \neq 0$ would reduce the transparency for both spin orientations and hence raise $F_{\rm el}$. The $M = 0$ state remains stable below some critical value of Z; increasing Z beyond this, a magnetic moment appears as the free energy decrease from increasing the transparency in the $\sigma = +1$ sector outweighs the increase in the $\sigma = -1$ sector. Although the decrease in $F_{\rm el}$ favors the indefinite growth of M with increasing Z, this is limited by the increase of $F_{\rm mag}$. For sufficiently large χ and low temperatures, the transition back into the nonmagnetic state is first order; M continuously vanishes at higher temperatures or smaller χ.

In contrast, the p_y-p_y junction displays a spontaneous magnetization at $Z = 0$ for all $T < T_c$, stabilized by the reduction in $F_{\rm el}$ from the decreased transparency in each spin sector. This effect is absent from the Ginzburg-Landau expansion of $F_{\rm el}$ in Eq. (1), as we have only kept terms to first order in $\mathbf{M}$; from Eq. (3), however, we see that the magnetization only enters $F_{\rm el}$ as $|\mathbf{M}|^2$ when $Z = 0$. Despite $|\mathbf{M}| \neq 0$, the junction is not in a fractional state and $\phi_{\min} = 0$. Turning on a finite tunneling barrier strength $(Z > 0)$ at fixed T, $|\mathbf{M}|$ decreases to compensate for the free energy increase from the enhanced transparency in the $\sigma = +1$ sector; $\phi_{\min}$ simultaneously takes on a fractional value. As Z is further increased, the barrier moment is monotonically suppressed, while the stable phase difference passes through an extremum before returning to its $Z = 0$ value.

Experimental tests.—The characteristic signature of the magnetic instability is the appearance of fractional flux quanta at the junction interface. In Fig. 4 we show a proposal for their observation in a Josephson junction with a tunneling barrier consisting of two materials, one of which undergoes the magnetic instability proposed here, while the other remains nonmagnetic at all temperatures. The stable phase difference across the magnetic region is $\tilde{\phi}$, while it is 0 across the nonmagnetic region. If a magnetic flux line is trapped at the boundary between the barrier segments, a line integral along the contour $\mathcal{C}$ in Fig. 4 shows that the enclosed flux Φ is

$$\frac{\Phi}{\Phi_0} = n + \oint_{\mathcal{C}} d\mathbf{s} \cdot \nabla\phi = n + \frac{\tilde{\phi}}{2\pi}, \qquad n \in \mathbb{Z}. \tag{4}$$

As $\tilde{\phi}$ takes a fractional value below T_M, there can exist a flux line with $\Phi \neq \Phi_0$. Experimentally, such a flux line could be observed by local magnetic probes like scanning SQUID microscopy, or inferred from the asymmetric Fraunhofer pattern of critical current versus applied field. Our proposal resembles the devices used to observe half-integer flux quanta in Ref. [15], where SSCs were used instead of TSCs, and differing widths of a FM barrier fix $\tilde{\phi} = \pi$. Although other schemes exist for the creation of fractional flux quanta [6,16], their detection in our pro-

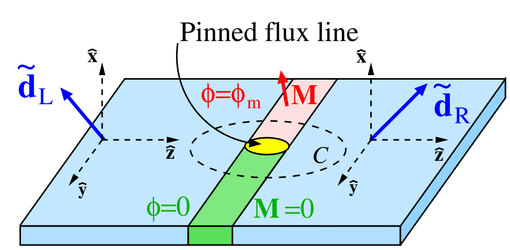

FIG. 4 (color online). Proposed experiment for the observation of fractional flux quanta at the interface between magnetic and nonmagnetic regions of a tunneling barrier. The contour $\mathcal{C}$ is used in evaluating Eq. (4).

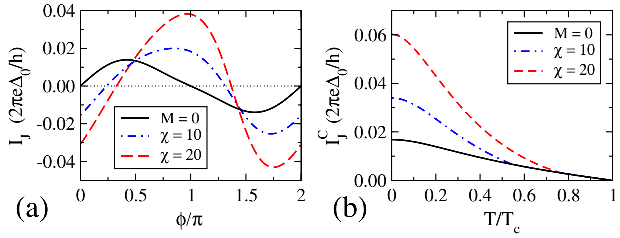

FIG. 5 (color online). (a) Current versus phase relationships in the p_y-p_y junction at $T = 0.2T_c$ both with and without ($M = 0$) the magnetic instability. (b) Critical Josephson current as a function of reduced temperature in the p_y-p_y junction. In both panels we take $\eta = 0.2\pi$ and $Z = 0.7$.

posed junction would be unambiguous confirmation of the magnetic instability.

The supercurrent transmission through the junction is radically altered by the appearance of a barrier moment. The Josephson current versus phase relationship $I_J = (2e/\hbar)\partial F/\partial\phi \propto \sin(\phi - \phi_{\min})$ is shifted from its usual form for a nonmagnetic barrier, as is clearly seen in Fig. 5(a) for the p_y-p_y junction (the p_z-p_z junction results are qualitatively identical). Note that the different magnitudes of $\max\{I_J\}$ and $\min\{I_J\}$ are due to higher-order harmonics in ϕ which are not included in the free energy expansion Eq. (1). The critical current $I_J^C = \max\{|I_J|\}$ is also strongly enhanced below the magnetic instability [Fig. 5(b)], as the increased current through the spin sector with the raised transparency overcompensates for the decreased current through the spin sector with the lowered transparency. The behavior of I_J^C below T_M is reminiscent of the "low-temperature anomaly" of d-wave Josephson junctions [1].

For simplicity, we have neglected the suppression of the TSC state near the interface due to the proximity effect. Although including this effect is not expected to alter the free energy expansion Eq. (1) or the main features of the ABS spectrum, it will increase $F_{\rm el}$ and hence shrink the parameter space where the moment is stable. This will be most pronounced in the p_z-p_z junction due to the sign change of the gap upon reflection [1]. Furthermore, the likely strong anisotropy effects in the thin FM layer do not favor the orientation of the induced moment normal to the interface. The situation in the p_y-p_y junction is more promising, however, as here the proximity effect is much less important and the induced moment lies in the easy plane parallel to the interface.

A likely candidate material for the TSCs in the p_y-p_y junction is Sr_2RuO_4, where the proposed pairing state is $\mathbf{d} \propto (k_x + ik_y)\hat{\mathbf{z}}$ [9]. At interfaces $\parallel \hat{\mathbf{z}}$, the proximity effect suppresses the p-wave component normal to the interface, and so we expect the ABS contribution to $F_{\rm el}$ to be more similar to that in the p_y-p_y junction than the p_z-p_z junction [17]; this should remain valid for small misalignment angles η. A possible material for the interface is a two-unit-cell-thick layer of $SrRuO_3$, which is both metallic (and hence has a low Z value appropriate to the p_y-p_y junction) and is close to a FM instability [18].

Conclusions.—We have shown that a TRS breaking configuration of two TSCs in a Josephson junction can cause the tunneling barrier to develop a spontaneous magnetization. This realizes an exotic Josephson state with stable phase difference $0 < \phi_{\min} < \pi$. The orbital part of the TSC pairing state was demonstrated to control the magnetic instability. The existence of fractional flux quanta at the barrier, and a large increase in the critical current beneath the magnetic transition temperature, are key experimental signatures of this state.

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