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Spin-polarized supercurrents for spintronics

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A marriage between superconductivity and ferromagnetism is opening the door for new spin-based applications.

The year 1911 was remarkable for physics in several respects. Ernest Rutherford postulated the existence of the atomic nucleus, which led him to his atom model; Max Planck introduced the quantum concept of zero-point motion; and Heike Kamerlingh Onnes announced the discovery of superconductivity in mercury below a critical transition temperature (see the article by Dirk van Delft and Peter Kes in PHYSICS TODAY, September 2010, page 38). All three discoveries are equally revolutionary, and all helped pave the way for the transition from a classical to a quantum mechanical picture of our world.

The superconducting phase is only one of the possible phases a metal can enter when cooled down. Nature preferred to acquaint humans first with another possible metallic phase, the ferromagnetic one. Both phases are fundamental and are direct, macroscopic manifestations of microscopic many-body quantum physics. But the two phases are largely incompatible.

The past decade, however, has seen dramatic advances in designing and building structures that combine the zero-resistance supercurrents of superconductors with the spin alignment of ferromagnets. The resulting spin-polarized supercurrents can propagate over long distances, and as a controllable source of spin states, they hold tantalizing promise for spintronics, an emerging field that exploits electrons’ spin degrees of freedom for electronic devices.

The coexistence problem

It was Vitaly Ginzburg who in 1956 first formulated the problem of coexistence of ferromagnetism and superconductivity. In 1950 he and Lev Landau had introduced the then mind-boggling notion of a macroscopic quantum wavefunction. The 1957 microscopic theory of superconductivity by John Bardeen, Leon Cooper, and Robert Schrieffer showed that the Ginzburg–Landau macroscopic wavefunction corresponded to a pair wavefunction: Electrons at the highest-energy filled states—the Fermi surface—build Cooper pairs with zero center-of-mass momentum, zero total spin (a spin singlet), and charge $2e$ that constitute the superconducting condensate. Excitations above the condensate need a minimum finite energy $\Delta$, the so-called excitation gap.

In looking at the coexistence problem, Ginzburg focused on the so-called orbital mechanism, in which the interaction between the charged Cooper pairs and the vector potential of the internal magnetic field suppresses the superconductivity. In 1958 Bernd Matthias, Harry Suhl, and Ernst Corenzwit suggested an additional mechanism, the quantum mechanical exchange interaction. That interaction tries to align the electron spins in a ferromagnet, whereas the spins in a Cooper pair—in the usual case of singlet superconductors—are antiparallel. Those antagonistic tendencies lead to the so-called paramagnetic effect of pair breaking. A similar situation arises when an external magnetic field is applied to Cooper pairs; the role of the exchange interaction is played by the Zeeman interaction. In both cases the interactions spin-polarize the electronic system and split the energy levels for excitations with spin parallel or antiparallel to the magnetization vector, by an amount given by twice the exchange or Zeeman energy.

When the paramagnetic effect is sufficiently large, a Cooper pair has two options for survival. It can become an equal-spin pair, in which the two spins point in the same direction with respect to the magnetization vector. Or it can keep its spins pointing in opposite directions with respect to the magnetization vector and acquire instead a nonzero center-of-mass momentum. That latter possibility was independently discovered in 1964 on both sides of the iron curtain: by Peter Fulde and Richard Ferrell at the University of Maryland and by Anatoly Larkin and Yurii Ovchinnikov at the Moscow Physico-Technical Institute. It is known in the Western literature as the FFLO state and in the Eastern as the LOFF state. (Fulde and Ferrell in fact submitted and published slightly earlier.)

The FFLO state exhibits an inhomogeneous pair wavefunction that oscillates periodically in space, as shown in box 1. The experimental search for the FFLO state in a superconductor is ongoing, and few candidates are known, among them the heavy-fermion material CeCoIn$_5$ and the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. However, the existence of FFLO-type states was established without doubt in the early 2000s in ferromagnetic metals in contact with a superconductor—so-called FS proximity structures. (For more on proximity structures, see box 2.) That experimental realization was one of the igniting events for the intense current developments in the field of spin-triplet supercurrents.

The FFLO mechanism is an intrinsic, bulk effect that, as box 1 describes, leads to mixing between singlet pairs ($\uparrow \downarrow - \downarrow \uparrow$)—where the left arrow in each term represents the spin orientation of one electron, and the right arrow, that of the...
Due to spin–spin interactions in a ferromagnet, the electronic bands for up spins (blue) and down spins (red) are shifted with respect to each other by an amount $2\hbar\omega_{\text{ex}}$, as shown in the left figure. That exchange splitting shifts the momenta at the Fermi energy $E_F$ from $k_i$ to the new positions $k_i = k_i + Q/2$ and $k_F = k_F - Q/2$. Two electrons at the Fermi energy with opposite spin and almost opposite momentum form a Cooper pair with center-of-mass momentum $\pm Q$.

Without exchange splitting, Cooper pairs in most superconductors prefer the singlet state $(\uparrow\downarrow - \downarrow\uparrow)$. With exchange splitting, the two spin contributions to the pair amplitude are proportional to $\exp\left(\pm (k_F - k_i) \cdot R\right)$, thus leading to a modulation of the pair amplitude with position $R$. The resulting state is a mixture of singlet and triplet spin states with zero spin projection on the direction of magnetization—that is, opposite spins:

$$(\uparrow\downarrow - \downarrow\uparrow) \sim (\uparrow\downarrow + \downarrow\uparrow)\cos(Q \cdot R) + i(\uparrow\downarrow - \downarrow\uparrow)\sin(Q \cdot R).$$

Such a state is called an FFLO phase after its discoverers, Peter Fulde and Richard Ferrell and also Anatoly Larkin and Yuriy Ovchinnikov.1 In a bulk FFLO phase, the direction of the vector $Q$ is determined by the crystal-field anisotropy in the ferromagnet, and not into the ferromagnet. At the same time, the internal structure of Cooper pairs in the superconductor is strongly modified near the interface.

That modification is due to phase shifts that electrons acquire when quantum mechanically penetrating into the interface barrier regions.2 Because the barrier is spin polarized, the phase shifts are spin dependent, as indicated by the offset between the up-spin (blue) and down-spin (red) states in the right-hand figure. The net phase difference $\theta$ acquired during reflection leads to singlet–triplet mixing

$$(\uparrow\downarrow - \downarrow\uparrow) \sim (\uparrow\downarrow + \downarrow\uparrow)\cos(\theta) + i(\uparrow\downarrow - \downarrow\uparrow)\sin(\theta)$$

of Cooper pairs in a layer roughly 15–150 nm thick next to the interface, depending on the material.
Most importantly, those proximity amplitudes decay much faster than in a normal metal and are progressively suppressed with increasing spin splitting. For sufficiently strong splitting, the proximity amplitudes are expected to be immeasurably small.

The oscillatory character has important implications; combined with the Josephson effect it leads to the possibility of a so-called $\pi$-junction, in which the Cooper pair wavefunctions in the superconductors on either side have a phase difference of $\pi$ (see figure 1). Bulaevskii and coworkers in 1977 had proposed the possibility of Josephson junctions with a $\pi$ phase difference when a magnetic impurity is inserted in the junction.4

Although the $\pi$-junction predictions were made decades ago, the experimental breakthrough came only in 2001–02 with the verification of the switching between 0- and $\pi$-junctions in SFS structures. That pioneering work was done by Valery Ryazanov and coworkers from the Institute of Solid State Physics in Chernogolovka, Russia, in collaboration with Alexander Golubov from the University of Twente and Jan Aarts from the University of Leiden;5 by Takis Kontos and coworkers at the Université Paris–Sud;6 and by the group led by Alexander Palevski at Tel Aviv University.6 Figure 1c shows some classic experimental results.

Despite the success from a fundamental perspective, one problem for practical applications became quickly evident: For a clear and controllable effect, weakly spin-polarized systems, like ferromagnetic copper–nickel or palladium–nickel alloys, must be used; otherwise, the proximity amplitudes become too short ranged. But the rapidly developing field of spintronics had provided strong motivation to search for long-range proximity effects in ferromagnets. Such long-range spintronics had provided strong motivation to search for long-range supercurrents in ferromagnets.

A renewed theoretical effort in the beginning of the 2000s. Early experiments in the groups of Nicholas Giordano at Purdue University; Bernard Pannetier at the CNRS in Grenoble, France; Victor Petrashev at Royal Holloway, University of London; and Venkat Chandrasekhar at Northwestern University spurred vivid discussions about the existence of long-range effects.

The lack of a proper understanding of those effects led to a renewed theoretical effort in the beginning of the 2000s. It was clear very early that for pairs composed of two equal spins ($\uparrow\uparrow$ or $\downarrow\downarrow$), the arguments of box 1 do not apply. Two $\uparrow$ spins at the Fermi energy can pair with equal and opposite momenta, $k_F$ and $-k_F$, without introducing a finite center-of-mass momentum, and likewise for two $\downarrow$ spins. Thus no oscillations will occur for equal-spin proximity amplitudes, and the penetration is long range; the penetration length behaves as it does in a normal metal, increasing to microns with decreasing temperature. There was a big obstacle, however: how to create an appreciable amount of such equal-spin pairs, which are triplet states, in the first place.

The solution can be understood by starting with the ($\uparrow\downarrow$)-triplet amplitudes that are created by either the FFLO or the spin-mixing effect and considering them from a different angle in spin space. Singlet states are rotationally invariant: They’re the same regardless of the quantization direction. But the three triplet spin states transform into each other when the quantization direction changes. In particular, the ($\uparrow\downarrow$ + $\downarrow\uparrow$)-triplet state in the $y$-basis is the equal-spin $\downarrow\downarrow$ triplet state in the $z$-basis, as highlighted in figure 2.

In a series of papers starting in 2001, Sébastien Bergeret, Anatoly Volkov, and Konstantin Efetov at Ruhr University Bochum considered an SF structure in which the direction of the ferromagnet’s magnetization rotates in a spiral in a region near the interface7 (see figure 3a). The spin quantization followed the rotating magnetization, and the resulting triplet mixing produced an equal-spin pair amplitude with a long-range penetration into the ferromagnet. The theorists also found surprising symmetry implications. When the electron transport in the ferromagnet is diffusive, the pair amplitude has to be isotropic in space, and consequently unchanged—that is, even—under the interchange of the spatial coordinates of the two electrons. And since the pair is in a triplet state, it is even with respect to interchanging the electron spins. Thus, to obey the Pauli principle, the pair wavefunction necessarily must be odd with respect to interchanging the time coordinates of the two electrons. Such “odd frequency” pairing is novel—no known material has that symmetry. Slightly after Bergeret and colleagues’ first paper, Anatoli Kadigrobov, Robert Shekhter, and Mats Jonson at Chalmers University of Technology and the University of Gothenburg found similar long-range equal-spin penetration when there is a region of magnetic inhomogeneity near the SF interface.7 The triplet-mixing effect has inspired much of the subsequent experimental and theoretical work.

In those inhomogeneity studies, the mechanism for creating the long-range triplet amplitudes requires that the short-range pair amplitudes and the magnetic inhomogene-

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Box 2. The proximity and Josephson effects

When a normal metal is adjacent to a superconductor, Cooper pairs in the superconductor will diffuse into the metal over a distance that depends on the disorder in the metal and on temperature. That behavior is known as the proximity effect. The theory behind it was developed largely in the 1960s by Pierre-Gilles de Gennes at the Université Paris–Sud in Orsay. A closely related phenomenon is Andreev reflection, discovered by Alexandr Andreev in 1964 at the Kapitza Institute for Physical Problems in Moscow. It describes the correlations between electrons and holes at the normal side of the interface due to penetration of pairs. Andreev reflection and the proximity effect are two sides of the same coin. The so-called proximity pair amplitude—closely related to the complex pair wavefunction in the normal metal—is proportional to the pair potential in the superconductor (which determines the superconducting energy gap $|\Delta|$) and the product of the transmission amplitudes through the interface of the two particles that compose the pair. The penetration depth of the proximity effect increases with lowering temperature and can reach the micron scale.

The proximity effect can be used to create superconductor-normal metal-superconductor (SNS) junctions that exhibit the Josephson effect. Discovered by Brian Josephson in 1962 in Cambridge, UK (see the PHYSICS TODAY articles by Philip W. Anderson, November 1970, page 23, and by Donald McDonald, July 2001, page 46), the effect originally described the zero-resistance tunneling of Cooper pairs between two superconductors through an insulating barrier (SIS junctions), but with the help of the proximity effect it can also occur through a long normal metallic region. The Josephson effect is a hallmark of macroscopic quantum coherence and a revolutionary discovery. It makes the pair wavefunction visible and proves its macroscopic character.
ity vary on a scale much longer than the Fermi wavelength. But in a strongly spin-polarized ferromagnet, the short-range components are restricted to atomically short distances at the interface, and the above theories are not applicable.

When I started to build a group in 2001 in the Institute for Theoretical Solid-State Physics led by Gerd Schön at Karlsruhe University, one goal was to tackle that problem. In 2003 we studied the extreme case in which one spin band is insulating and one is metallic, a so-called half metal (see the article by Warren Pickett and Jagadeesh Moorad in PHYSICS TODAY, May 2001, page 39). In our theory, the main physics takes place at atomically thin interfaces. The crucial idea was to generalize the Tokuyasu-Sauls-Rainer spin-mixing effect at an SF interface to a spin-dependent interface barrier (see box 1). And it demonstrated the crucial role of the misalignment from the early experiments that used Ni or Pd–Ni and Cu–Ni alloys, and it demonstrated the crucial role of the misalignment of the phases at the interfaces. In 2004, we confirmed the existence of triplet supercurrents under various conditions in half-metallic or strongly spin-polarized ferromagnets or suggesting new geometries for finding such components.

Mikhail Kupriyanov at Moscow State University and coworkers are pursuing a promising, out-of-the-mainstream idea. They propose connecting an NF bilayer or an FNF trilayer structure to two superconducting electrodes such that the current flows along the layers, instead of perpendicularly through them. Thanks to the interactions between the ferromagnetic and normal layers, the normal metal should be able to support long-range, equal-spin pair amplitudes. To date, however, such structures have not yet been experimentally studied.

It was realized quite early that the needed inhomogeneous magnetization does not necessarily have to be intrinsic but could simply be introduced by a multilayer geometry. In one promising arrangement, suggested in 2007 by Manuel Houzet and Buzdin, the interfaces are replaced by two magnetized layers that are misaligned with respect to the center ferromagnetic region. The advantage of such an arrangement became clear with the experimental results published last April by Trupti Khaire and coworkers in the group of Norman Birge at Michigan State University. With a layer of misaligned ferromagnetic alloy next to each superconducting electrode to create equal-spin states, a strongly spin-polarized ferromagnet between the layers carried the triplet supercurrent over long distances. The experiment incorporated into the quest for long-range triplet currents the knowledge from the early experiments that used Ni or Pd–Ni and Cu–Ni alloys, and it demonstrated the crucial role of the mis-

**Figure 1. Ferromagnets and Josephson junctions.**

(a) When a Josephson junction is formed by sandwiching a normal metal between two conventional superconductors in which the Cooper pairs are in a spin-singlet state, the pair-wavefunction amplitudes penetrate from the superconductors into the normal metal due to the proximity effect (see box 2). In the ground state, the amplitudes equilibrate and the superconducting wavefunctions have the same phase on both sides. (b) If the normal metal is replaced by a weakly spin-polarized ferromagnet, the pair amplitude will oscillate in the ferromagnetic region. Depending on the length of the junction, the ground state is either a 0-junction with equal superconducting phases, or a π-junction in which the phases differ by π. (c) Experimental data show the transitions between the two ground states. The left plot shows the so-called characteristic voltage, given by the product of the junction's maximum supercurrent (the critical current) I, and normal-state resistance $R_n$ through junctions with different ferromagnetic lengths $d_F$. Because the oscillation length scale is, in general, temperature dependent, varying the temperature $T$ can also cause a switch, as shown on the right in the plot of the critical current density. (Left plot adapted from ref. 6, Kontos et al.; right plot adapted from ref. 5, Ryazanov et al.)
alignment of the magnetizations. With that breakthrough, the researchers had a simple and reliable way to produce long-range triplet supercurrents. It initiated a new generation of experiments that focus on the controlled manipulation of triplet supercurrents by using a refined multilayer structure made of various materials each serving a particular function (see figure 4a).

New experiments by the Birge group show that, indeed, the triplet supercurrent can be strongly enhanced by maximizing the misalignment angle between the magnetizations of the outer layers and the center layer. Theory predicts that the current would additionally depend on the relative orientation between the outer magnetization vectors themselves; that dependence would give additional control over the equal-spin supercurrent and allow switching between 0- and π-junctions.5

In an intriguing 2006 experiment using holmium, Igor Sosnin, Hsiuchi Cho, and Petashov realized the earlier ideas by Bergeret, Volkov, and Efetov. At low temperatures, Ho is a conical ferromagnet. The atoms’ magnetic moments process in a spiral from layer to layer, tracing out the surface of a cone. That intrinsic magnetic inhomogeneity generated equal-spin pairs and a long-range proximity effect. Following that work, Jason Robinson, James Witt, and Mark Blamire from the University of Cambridge last year found a triplet supercurrent when they sandwiched a cobalt layer between two Ho layers.11 Those works have opened the door to a second type of new experiment that utilizes intrinsically inhomogeneous magnets to controllably create long-range triplet supercurrents (see figure 4b).

The spell seems broken, and further announcements of long-range triplet supercurrents keep appearing. For example, in 2010 Jian Wang and coworkers in the group of Moses Chan from the Pennsylvania State University found zero-ple, in 2010 Jian Wang and coworkers in the group of Moses long-range triplet supercurrents keep appearing. For exam-

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Figure 2. Making equal-spin pairs. (a) When the interface between a strongly spin-polarized ferromagnet and a superconductor is magnetized in the same direction as the ferromagnet (here, the z-axis), the ferromagnet induces opposite-spin triplet mixing in the superconductor (red curve), but the proximity-induced superconducting pair amplitudes do not survive into the ferromagnet. (b) When the interface magnetization is in a different direction, the spin-singlet state, which is rotationally invariant, still doesn’t survive. But the three triplet spin states mix in the different quantization bases. In the case shown here, an interface magnetized in the y-direction generates opposite-spin pairs with respect to the y-axis. But such a state is equivalent to a combination of equal-spin pairs when viewed with respect to the z-axis, and that equal-spin state penetrates over a long distance into a ferromagnet magnetized in the z-direction.

of experiments concentrates on artificially creating different types of noncollinear magnetic structures that can be interfaced with superconductors. Jiyeong Gu and Jeery Kusnadi from California State University, Long Beach, and Chun-Yeol You from Inha University, South Korea, have developed one such structure,14 as have Robinson, Gábor Halász, Buzdin, and Blamire.11 And Dirk Sprungmann and coworkers from the group of Hartmut Zabel at Bochum reported evidence for triplet supercurrents through ferromagnetic barriers made of Cu,MnAl, a so-called Heusler material.15

Figure 3. Toward real-world equal-spin pairs. (a) In certain ferromagnetic domain walls, known as Bloch domain walls, the magnetization direction rotates in a spiral fashion, into and out of the page. Such domain walls in a weakly spin-polarized ferromagnet will create and mix all triplet pair amplitudes, thus allowing equal-spin components to penetrate a homogeneously magnetized region over a long distance.7 (b) In a strongly spin-polarized system, the triplet amplitudes are created in the interface barrier. If the interface magnetic moments are misaligned with the bulk magnetization, the triplet amplitudes have equal-spin components with respect to the bulk magnetization axis and penetrate over long distances. Depending on the relative magnetic orientations of the two interface regions, a 0-junction or a π-junction can be prepared.
Future developments and applications

The recent developments allow, for the first time, devices with the controlled creation of long-range, equal-spin triplet supercurrents. Such a capability has implications both for basic research and for applications.

New and exotic pairing amplitudes—like the odd-frequency pairing—will be the subject of much attention. Future studies will concentrate on the physical properties of such amplitudes and the possibility of triggering a phase transition in the entire heterostructure to an odd-frequency superconducting order parameter. Such an order parameter has never been observed in nature before. Triplet order parameters are rare, and the known systems with triplet pair amplitudes have never been observed in nature before.

In addition to enabling such exciting fundamental investigations, the controlled production of triplet supercurrents will open several directions for possible applications. With the availability of fully polarized triplet supercurrents, spin-dependent quantum-coherence phenomena will enter the realm of spintronics devices. Superconducting spintronics devices are appealing since they introduce in a natural way the elements of nonlocality, entanglement, and quantum coherence, all of which are crucial for quantum computing.

Nonlocality enters in two ways. First, since the Cooper pairs are typically larger than 50 nm in size, a nanometer-scale superconducting device with several electrical contacts will exhibit nonlocal transport phenomena: The current through one contact will influence the current through others. Nonlocality thus allows for control in nanoelectronics. In addition, the nonlocal phenomena in superconducting devices are quantum coherent. As Guy Deutscher from Tel Aviv and Denis Feinberg from Grenoble pointed out in 2000, a Cooper pair can give one of its electrons to one contact, and the other to a second contact, and the two electrons stay quantum coherently coupled. Quantum entanglement is based on the presence of such coherence. Other exciting applications of quantum coherence are so-called Andreev interferometers, introduced in 1994 by Petrashov and Vladimir Antonov at the Institute of Microelectronics Technology in Chernogolovka and Per Delsing and Tord Claeson in Gothenburg. In such a device, a superconducting wire is attached at two points to a normal wire to form a loop; the conductance of the normal wire oscillates as a function of the magnetic flux through the loop, due to the quantum coherence established in the normal wire.

The second way nonlocality enters is illustrated by a variation on the superconducting quantum interference device called the π-SQUID. That ingenious SNS device is based on a pioneering idea of Volkov’s in 1995 and was first made in 1998–99 by Alberto Morpurgo and Jochem Baselmans in the groups of Bart van Wees and Klapwijk at the University of Groningen. In such devices, applying a control voltage directly to the normal metal modifies its electron distribution. The nonequilibrium distribution spreads to the superconducting wire, which in turn modifies the electron distribution. The result is direct control of the supercurrent through the device, even allowing the switching between a 0-junction and a π-junction.

The next step is to combine such ideas with the new availability of triplet supercurrents, which in the near future will be routinely produced. That marriage will unite the hitherto separate fields of mesoscopic superconductivity and spintronics. When such devices as spin transistors, spin filters, spin pumps, spin valves, and spin switches are combined with superconductivity, a new spin electronics may
emerge, and some of the future devices might well turn out to be as transformational as SQUIDs were 45 years ago.

References


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- The enlightenment of physicists and the public regarding the history of physics or other cultural aspects of physics
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