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Superconducting spintronics

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Traditional studies that combine spintronics and superconductivity have mainly focused on the injection of spin-polarized quasiparticles into superconducting materials. However, a complete synergy between superconducting and magnetic orders turns out to be possible through the creation of spin-triplet Cooper pairs, which are generated at carefully engineered superconductor interfaces with ferromagnetic materials. Currently, there is intense activity focused on identifying materials combinations that merge superconductivity and spintronics to enhance device functionality and performance. The results look promising: it has been shown, for example, that superconducting order can greatly enhance central effects in spintronics such as spin injection and magnetoresistance. Here, we review the experimental and theoretical advances in this field and provide an outlook for upcoming challenges in superconducting spintronics.

At the interface between materials with radically different properties, new physical phenomena can emerge. A classical example of such an interface is that between a superconductor and a ferromagnet, where the opposing electron orders destructively interfere; however, it turns out that under the right conditions at a superconductor–ferromagnet interface both superconductivity and spin polarization can unite to create a new superconducting state that offers tantalizing possibilities for spin transport in which Joule heating and dissipation are minimized.

Spintronics offers the potential for creating circuits in which logic operations controlled by spin currents can be performed faster and with greater energy efficiency[1] than the charge-based equivalent in semiconductor transistor technologies. Spintronics is one of the most active areas of research and, in addition to offering control of spin and charge at the nanometre scale, it has also found sensory applications in hard disk drive read heads through the giant magnetoresistance effect[2,3]. The idea of combining superconductivity with spintronics has historically focused on the net spin polarization of quasiparticles in superconductors. It is interesting to note that the first spin transport experiments[4–6] involved ferromagnet–superconductor bilayers and pre-dated non-superconducting spin transport experiments[7]. As will be discussed in this review, it is possible to create pseudo-chargeless spin-1/2 excitations in superconductors[8] which have extremely long spin lifetimes.

Recently, a more complete synergy between superconductivity and spintronics has been made possible through the discovery of spin-triplet Cooper pairs at superconductor–ferromagnet interfaces. Non-superconducting spin currents are generated by passing charge currents through ferromagnetic materials. As will be explained in this review, spin currents can also be generated by passing supercurrents through ferromagnetic materials. Charge flow in superconductors is carried by Cooper pairs which consist of interacting pairs of electrons[9]. The idea of combining superconducting and magnetic order was initiated in the late 1950s when Ginzburg[10] demonstrated theoretically that the electrons within a Cooper pair in a conventional superconductor will eventually be torn apart owing to the so-called orbital effect: in the presence of a magnetic field, the Lorentzian force acts differentially on the oppositely aligned electron spins of a pair. Moreover, the Zeeman interaction between spins and a magnetic field favours a parallel alignment, meaning that for a strong enough magnetic field the pairs are energetically unstable as one electron of a pair is required to spin-flip scatter. However, there exists a way to avoid this problem. The two-fermion correlation function f describing Cooper pairs is subject to the Pauli principle, meaning that the spin part does not necessarily have to be in a spin-singlet[9] antisymmetric state $(\uparrow\downarrow - \downarrow\uparrow)$. So long as f is antisymmetric under an overall exchange of fermions $1 \leftrightarrow 2$, which includes the space, spin and time coordinates of the two electrons, the Pauli principle is satisfied. This means that Cooper pairs can reside in a spin-triplet state that is symmetric under fermion exchange—that is, $(\uparrow\downarrow + \downarrow\uparrow)$, $\uparrow\uparrow$, or $\downarrow\downarrow$—as long as f changes sign under an exchange of space and time coordinates as well, allowing odd-in-time (or odd-frequency) pairing[11–13]. Such a spin-triplet state can coexist with a magnetic field as the Zeeman interaction due to the magnetization no longer has a pair-breaking effect on the Cooper pairs so long as the orbital effect is suppressed.

As Cooper pairs can be spin-polarized, it follows therefore that triplet supercurrents can carry a net spin component and so offer the potential to eliminate the heating effects associated with spintronic devices. However, to use such supercurrents in spintronics it is necessary to be able to generate and manipulate triplet pairs in devices. In recent years there has been significant progress in this area, not least on the experimental side, where the generation of triplet pairs in superconductor–ferromagnet (SF) structures is becoming routine.

One of the aims of superconducting spintronics involves identifying ways to enhance central effects in spintronics by introducing superconducting materials and to understand the interactions that arise when superconducting and magnetic order coexist. The results look promising: the existence of spin-polarized supercurrents has been verified; spin-polarized quasiparticles injected into superconductors have been shown to have spin lifetimes that exceed those of spin-polarized quasiparticles in normal metals by several orders of magnitude; and superconducting spin valves offer colossal magnetoresistance effects and can switch on and off superconductivity itself. Even magnetization dynamics have been demonstrated to be strongly influenced by superconducting order, raising the possibility that superconductivity can influence domain wall motion.

The recent experimental and theoretical advances described above serve as a motivation for the present review. First, we will overview the microscopic mechanisms and theoretical framework

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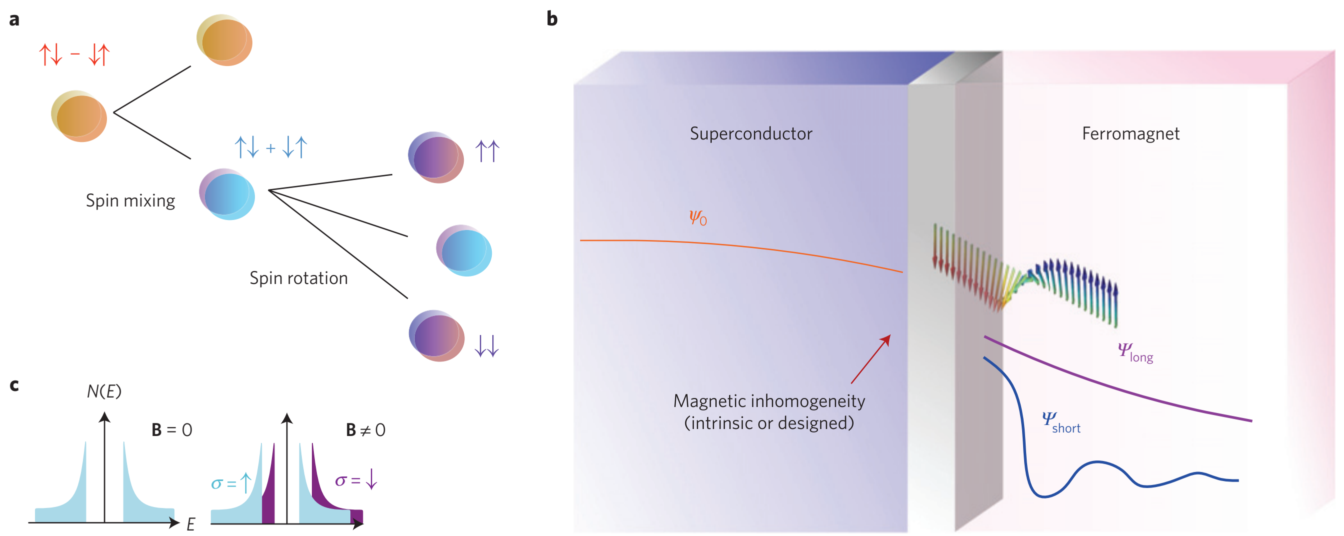

Figure 1 | Cooper pair conversion from a spin-singlet state to a spin-triplet state and spin–charge separation in superconductors. a, Spin mixing generates spin-zero ($S_z = 0$) triplet pair correlations from spin-singlet $S = 0$ superconductivity. If spin rotation occurs owing to a change in the quantization axis, $S_z = \pm 1$ triplet pairs form from the $S_z = 0$ triplets. **b**, Starting out with a conventional s-wave superconductor that proximity couples a homogeneous ferromagnet, the singlet ψ_0 and short-ranged triplet ψ_{short} pair correlations, $S_z = 0$, rapidly decay in an oscillatory way in the ferromagnet. In the presence of magnetic inhomogeneity at the interface, long-range triplet correlations, ψ_{long}, emerge in the ferromagnet. **c**, The relative spin and charge of quasiparticles in a superconductor depends on the energy E of the quasiparticles: near the gap edge, the quasiparticles carry spin but not charge. The density of states $N(E)$ for the spins can be separated by applying an in-plane magnetic field which induces a Zeeman splitting of the superconducting $N(E)$.

which explain how superconducting order and spin polarization can be reconciled and, second, we will discuss a few of the promising proposals which highlight the benefits of superconductivity for spintronics. We also discuss the experimental scene in terms of spin-polarized quasiparticles in superconductors and triplet Cooper pair generation. Finally, we look ahead at promising future directions and outline some of the outstanding issues that need to be addressed to develop the field of superconducting spintronics.

Spin flow in superconductors

A key requirement for spintronics is that the spin degree of freedom relaxes slowly enough for the spin to be manipulated and read out. Spin lifetimes are nevertheless typically fairly short in diffusive materials owing to spin–orbit and spin-flip scattering processes which lead to spin randomization. Another major hurdle relates to the fact that, because electrons carry spin and charge, they are susceptible to processes which cause dissipation and decoherence due to the charge degree of freedom. Finding ways to prolong spin lifetimes in materials is therefore a high priority in spintronics. Superconductors can help resolve this problem. To see why, consider excitations in the superconducting state. Below the energy gap Δ stable excitations do not exist, whereas quasiparticles may be created with energies above the gap. As shown in Box 1, these quasiparticles are always spin-1/2 regardless of their excitation energy, but their effective charge varies strongly with energy E. For large energies $E \gg \Delta$, the excitations in a superconductor are electron- or hole-like in character. For energies close to the gap edge $E \simeq \Delta$, however, the weights of the electron and hole character are almost identical. Consequently, they carry a net spin component in the near absence of charge above the superconducting gap. Furthermore, their average speed is greatly reduced in the same energy range, meaning it takes them longer to scatter through processes involving spin–orbit impurities relative to their scattering rates in the normal state. The net consequence of the above is that the spin lifetime of quasiparticles near the gap edge $E = \Delta$ in a superconductor can be increased by many orders of magnitude relative to in ferromagnetic metals, which is precisely the desirable property sought in spintronics. The realization of spin–charge separation for quasiparticles in superconductors dates back to Kivelson and Rokhsar[8] and the spin injection properties in superconducting spin-valve hybrid structures were later studied theoretically in detail by Takahashi and colleagues[14]. Johnson demonstrated the first experimental evidence of non-equilibrium spin injection in the same geometry[15].

Theoretical investigations of hybrid structures involving superconductors and ferromagnets were pioneered in the late 1970s by Bulaevskii and Buzdin[16]. When a superconductor is placed in good contact with a metal, the tunnelling of electrons across the interface results in a proximity effect: the leakage of superconducting pair correlations into the metal and non-superconducting electrons into the superconductor. If the metal is non-magnetic, the pair correlations decay monotonically on the normal metal layer thickness; however, for a ferromagnet the pair correlations decay in an oscillatory manner[17] superimposed on an exponential decay because the Fermi surfaces for spin-↑ and spin-↓ electrons are no longer degenerate, meaning that the Cooper pairs acquire a finite centre-of-mass momentum.

Owing to spin-dependent scattering at the interface between the superconducting and ferromagnetic regions, triplet pairing correlations are created (Fig. 1) which decay on a length scale of the singlet pair correlations (typically a distance of 1–10 nm from the superconductor–ferromagnet interface). Such triplet pairs carry no net spin projection along the quantization axis and so seem to have no immediate use in spintronics. In 2001 it was demonstrated in a seminal work[18] (see also refs 19,20) that triplet pairs that carry spin in addition to charge could also form by introducing magnetic inhomogeneities at the SF interface. The process of converting a spin-singlet Cooper pair into a spin-triplet pair can be understood by introducing the concepts of spin mixing and spin rotation[21], as described in Box 2 and Fig. 1. The spin-mixing process generates the $S_z = 0$ triplet component from a spin-singlet source through spin-dependent phase shifts that the electrons experience when propagating through a ferromagnetic region or when scattered at a ferromagnetic interface. When the magnetization of the system is textured such that the spin-quantization axis varies spatially, the effect of spin rotation comes into play, thus causing the different

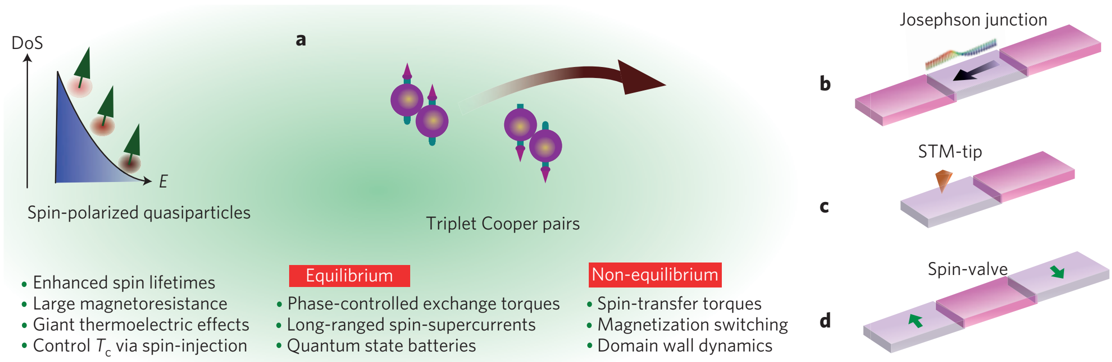

Figure 2 | Applications of superconducting spintronics. a, Schematic overview of different ways to use superconducting spintronics by means of spin-polarized quasiparticles and triplet Cooper pairs, both in equilibrium and non-equilibrium settings. The fading colour of the quasiparticles in the superconducting region represents their loss of effective charge as they approach the gap edge. **b–d**, Schematics for typical experimental set-ups used in superconducting spintronics, including Josephson junctions, bilayers and spin valves.

spin-triplet components to transform into each other. Through this process spin-polarized Cooper pairs form where both electrons of a pair have the same sign of spin. When propagating through a ferromagnet, the Zeeman field no longer has a pair-breaking effect and so triplet Cooper pairs are long-ranged in ferromagnetic materials and have been demonstrated to extend up to hundreds of nanometres even in half-metallic compounds[22]. The history of long-ranged spin-polarized supercurrents has been covered in detail in ref. 23.

There are other ways to generate long-ranged spin-triplet correlations in ferromagnetic structures that are not textured (see examples in Table 1). If a superconducting structure lacks an inversion centre (either owing to its crystal structure or to the geometry of the set-up) it will generally feature antisymmetric spin–orbit coupling, such as Rashba spin–orbit coupling[24]. This leads to a mixing of excitations from the two spin bands in such a fashion that spin is no longer a conserved quantity. Instead, the long-lived excitations now belong to pseudospin bands that may be thought of as momentum-dependent combinations of the original spin species. As a result, the superconducting pairing state in non-centrosymmetric superconductors will intrinsically be a mixture of singlet and triplet pair correlations[25]. When pairing occurs between the quasiparticle excitations of a simple Hamiltonian featuring antisymmetric spin–orbit coupling, such as $\hat{H}=\varepsilon_{\mathbf{k}}+\mathbf{g}_{\mathbf{k}}\cdot\boldsymbol{\sigma}$, where $\varepsilon_{\mathbf{k}}$ is the normal-state dispersion, $\boldsymbol{\sigma}$ is the Pauli matrix vector and $\mathbf{g}_{\mathbf{k}}=-\mathbf{g}_{-\mathbf{k}}$ is a vector characterizing the spin–orbit coupling, the triplet part of the superconducting pairing generally may be described by the relation $\mathbf{d}(\mathbf{k})\parallel\mathbf{g}(\mathbf{k})$, where $\mathbf{d}(\mathbf{k})\equiv[(\Delta_{\downarrow\downarrow}(\mathbf{k})-\Delta_{\uparrow\uparrow}(\mathbf{k}))/2, -\mathrm{i}(\Delta_{\uparrow\uparrow}(\mathbf{k})+\Delta_{\downarrow\downarrow}(\mathbf{k}))/2, \Delta_{\uparrow\downarrow}(\mathbf{k})]$ is the triplet d-vector[26] associated with the spin of the Cooper pair state $\langle\sigma\rangle\propto \mathrm{i}\mathbf{d}(\mathbf{k})\times\mathbf{d}(\mathbf{k})^*$. We emphasize here that the d-vector formalism is very suitable to describe also the proximity-induced triplet correlations in superconductor–ferromagnet structures, where the anomalous Green's functions $f_{\sigma\sigma'}$ take on the role of the gaps $\Delta_{\sigma\sigma'}(\mathbf{k})$ above. One may thus define a 'proximity' triplet vector **f**. As shown in ref. 27, the proximity effect between such a system and a homogeneous ferromagnet will thus produce both short-ranged and long-ranged triplet superconductivity inside the ferromagnetic region, depending on whether the spins of the triplet Cooper pairs are perpendicular to or aligned with the Zeeman field. The generation of long-ranged spin triplets through spin–orbit coupling and homogeneous ferromagnetism has also been expressed in terms of an analogy between D'yakonov–Perel[28] spin relaxation and precession of spins in normal systems and diffusive systems with antisymmetric spin–orbit coupling in contact with s-wave superconductors[29]. More specifically, a comparison between the quasiclassical Usadel equation[30] (which determines the superconducting pairing correlations quantified by the anomalous Green's function **f**) in the presence of such spin–orbit interactions and the spin diffusion equation for normal-state systems (which determines the spin density **S**) shows that the spin–orbit interaction affects the components of **f** and **S** in the same way.

We note in passing that using spin–orbit coupling as a source of singlet–triplet mixing has been a central ingredient in proposals related to the emergence of Majorana fermions in condensed matter systems[31,32].

Although the interaction of conventional spin-singlet superconductors and ferromagnets may result in spin-triplet pairs, they can also be created in bulk spin-triplet superconductors such as Sr_2RuO_4 (ref. 33) and ferromagnetic superconductors such as the uranium-based heavy-fermion compounds[34,35]. This includes the creation of spin currents without resistance[36–40] and spin-valve devices controlling the resistance of the junction by means of the superconducting critical temperature T_c (ref. 41). There are, however, practical problems to overcome in using triplet superconductors rather than conventional superconductors for spintronics, such as the requirement for high pressures or sub-kelvin critical temperatures. Interestingly, the first prototype of a triplet superconductor–ferromagnet bilayer structure (Fig. 2c) was very recently experimentally reported[42], which may be the first step towards investigating the interface between spintronics and bulk triplet superconductors.

Spin-polarized quasiparticles and magnetoresistance

The application of superconducting elements in spintronics necessarily requires non-equilibrium transport driven by means of, for example, voltages or temperature gradients. In this section, we review experimental advances in both equilibrium and non-equilibrium transport and discuss recent theoretical insights which have yet to be realized experimentally.

We begin by discussing effects related to spin-polarized quasiparticles in superconductors. Although early studies of spin imbalance in superconducting spin valves assumed that the spin lifetime in the superconducting state τ_s was unchanged[43] from the normal state τ_n, more recent experiments have demonstrated greatly enhanced quasiparticle spin lifetimes in the superconducting state. For example, Yang *et al.*[44] reported spin lifetimes of a non-equilibrium spin density in superconducting Al that were a million times longer than in the normal state by measuring a considerable tunnel magnetoresistance due to spin imbalance that could only be consistent with a very large spin lifetime. The spin–charge separation and reduced spin–orbit scattering rate near the gap edge for quasiparticles in a superconductor leads to strongly increased spin lifetimes relative to the normal state due to their movement

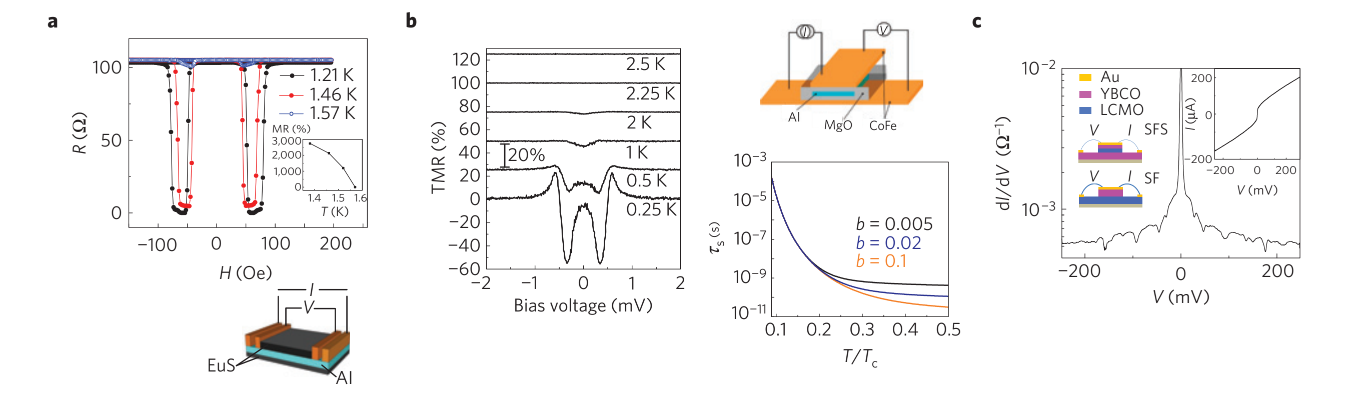

Figure 3 | Recent experimental highlights for superconducting spintronics. a, Infinite magnetoresistance (MR) effect in a superconducting spin valve with a ferromagnetic insulator. Reprinted and adapted figure with permission from ref. 68, APS. **b**, Evidence of an extremely large spin lifetime as probed through tunnel magnetoresistance (TMR) oscillations due to spin imbalance in the superconducting state. Adapted from ref. 44, NPG. **c**, Spectroscopic signature of long-ranged triplet correlations in a half-metal with quasiparticle interference giving rise to conductance oscillations. Adapted from ref. 115, NPG.

slowing down greatly at this energy range (see Box 1 for discussion). Importantly, the enhancement of the spin density lifetime in the superconducting state relative to the normal state becomes much larger when accounting for impurity spin–orbit scattering[44] in the relative spin susceptibility χ_S/χ_N, which in this case remains finite as $T \to 0$ (Fig. 3b). A treatment without spin–orbit effects, on the other hand, provides a much smaller increase of the spin lifetime in the superconducting state relative to the normal state[45]. Using a slightly different set-up where an intrinsic Zeeman splitting was induced in the superconducting region by means of in-plane magnetic fields, Quay *et al.*[46] showed evidence of a nearly chargeless spin imbalance in superconducting Al using a spin-valve set-up with Co ferromagnets. Their measurements of the non-local resistance due to diffusion of the spin imbalance revealed vastly different timescales for spin and charge relaxation (25 ns versus 3 ps). In addition, their results implied a strongly enhanced spin lifetime in the superconducting state, $\tau_s \simeq 500\tau_n$. The intrinsic spin-splitting of the density of states permitted a strong spin accumulation of fully polarized spins when the tunnelling from an F electrode matched the gap edge of one of the spin species. Similar conclusions were also reached by Hübler and colleagues[47].

It is important to note that the change in spin-relaxation length λ_{sf} in the superconducting state relative to the normal state depends on the origin of the spin-flip processes. For spin–orbit scattering due to impurities, λ_{sf} is predicted to be the same both above and below T_c (ref. 45), although Poli *et al.*[48] reported a decrease of λ_{sf} by an order of magnitude in the superconducting state which was attributed to spin-flip scattering from magnetic impurities[49]. Information about the spin-relaxation length was obtained by non-local resistance measurements that could probe the diffusion of the spin imbalance that originated at the spin injection point. We also note that spin absorption by superconductors with strong spin–orbit coupling has very recently been demonstrated by Wakamura *et al.*[50], where the spin-relaxation time was found to be much greater in the superconducting state of Nb than in its normal state.

Another example of how superconducting order can enhance conventional spintronics is through the magnetoresistance effect. In the superconducting analogue of a spin-valve device, the metallic spacer between two ferromagnets is replaced with a superconductor. The magnetization configuration influences the resistance experienced by an injected current just as it does in a non-superconducting device, but here it can also switch on and off the superconducting state, which corresponds to an infinite magnetoresistance. The earliest theoretical investigation of a superconducting spin-valve set-up dates back to de Gennes[51], with experiments[52] soon after confirming his prediction of a higher T_c in the anti-parallel state of the ferromagnets than in the parallel configuration. When the superconductor is sufficiently thin, a proximitized ferromagnet will influence the superconducting state in the following way. Even in the absence of a potential gradient, the superconducting critical temperature T_c is non-monotonic and, in certain cases, re-entrant on ferromagnetic layer thickness d_F (refs 53–56). The strong oscillatory dependence of T_c on d_F may be understood in terms of quasiparticle interference inside the ferromagnetic region[57]. This effect is most pronounced when the superconductor thickness d_S drops below the superconducting coherence length ξ_S, suggesting that the inverse proximity effect (the induction of ferromagnetic order inside the superconductor) is responsible for this phenomenon.

A variation of T_c with d_F requires the measurement of multiple samples, but controlling T_c through the relative orientation of the F layers in an FSF spin valve can be achieved within a single device[60–63]. Generally one expects the AP configuration of the F layers to be more compatible with spin-singlet pairing than the P configuration: when the thickness of the S layer is comparable to the superconducting coherence length ξ_S, the electrons in a singlet pair feel a reduced Zeeman field, whereas in the P state the fields are additive and so T_c is suppressed, as confirmed by Gu *et al.*[64] and Moraru and colleagues[65]. When the magnetizations are non-collinear T_c behaves non-monotonically on the angle between the F layers, exhibiting a minimum at a relative misalignment angle of $\pi/2$ (refs 61,62,66) due to the generation of triplet pairs. Such an effect can be understood qualitatively from the fact that the proximity-induced triplet pairing was theoretically found to be 'anti-correlated' to the change in T_c (ref. 62): with more singlet Cooper pairs leaking into the ferromagnetic side (suppression of T_c), triplet pairing becomes enhanced. Recently, an unusually large change in T_c of the order of 1 K, was reported by using half-metallic ferromagnets in a spin-valve set-up[67]. In ref. 68, the ferromagnetic insulator EuS was used in contrast to a metallic ferromagnet: using a EuS/Al/EuS set-up with layer thicknesses of a few nanometres, a full transition from a superconducting to resistive state (governed by the proximity-induced Zeeman field in the superconductor) was observed on going from an AP to a P configuration, resulting in an infinite magnetoresistance (Fig. 3a). Large changes in T_c have also been reported for V/Fe spin valves[69,70].

The control of T_c of superconducting spin valves is generally achieved without applying an intentional voltage bias and is therefore due to the proximity effect. In non-equilibrium situations where voltages are applied, spin injection or transport measurements can be performed to assess how superconductivity modifies spin transport. Several experiments have considered a superconducting spin-valve set-up in which a bias voltage is

Table 1 | Emergent superconducting correlations in generic hybrid structures.

Material Y	Insulating X	Spin-active X
Normal metal	ψ_0	$\psi_0+\psi_{\rm long}$
Homogeneous F	$\psi_0+\psi_{\rm short}$	$\psi_0+\psi_{\rm short}+\psi_{\rm long}$
Homogen. F + SOC	$\psi_0+\psi_{\rm short}+\psi_{\rm long}$	$\psi_0+\psi_{\rm short}+\psi_{\rm long}$
Inhomogeneous F	$\psi_0+\psi_{\rm short}+\psi_{\rm long}$	$\psi_0+\psi_{\rm short}+\psi_{\rm long}$
Half-metallic F	None	$\psi_{\rm long}$

Consider a $S/X/Y$ structure where S is an s-wave superconductor, X is the layer separating the two materials and Y is a material with certain properties, as tabulated. We allow X to be an insulator that is either non-magnetic or spin-polarized with a misaligned moment relative to the magnetization in Y, denoting the latter as spin active. F stands for ferromagnet, SOC for antisymmetric spin–orbit coupling (such as Rashba type); ψ_0 denotes spin-singlet Cooper pairs and $\psi_{\rm short/long}$ denotes short-ranged and long-ranged triplet Cooper pairs.

applied between metallic ferromagnets[44,64,65,71,72]. In the presence of tunnelling barriers which suppress the proximity effect, the role of the magnetization configuration can be reversed relative to the case when no voltage is applied. In the P state, the injected spin from one ferromagnet provides the output in the second ferromagnet and no net spin imbalance occurs in the superconducting region. The superconducting gap T_c is thus unaffected by the spin injection irrespective of the bias voltage applied. This changes in the AP state: owing to the different density of states for the majority and minority spins in the two ferromagnetic regions, spin injection from one ferromagnet cannot be compensated by an outflow of spin in the other, which results in a net spin imbalance in the superconductor. The superconducting state is therefore weakened and is ultimately destroyed on increasing the voltage V (ref. 14). The spin imbalance can in turn be detected through magnetoresistance measurements.

Triplet Cooper pairs and magnetization dynamics

An interesting prospect that emerges from the combination of magnetic and superconducting order is that of spin supercurrents. If Cooper pairs are spin-polarized they should be able to transport not only charge, but also a net spin component, but without dissipation. A number of proposals have been put forward to explain how spin supercurrents can be created and controlled in hybrid structures, including Josephson junctions (Fig. 2b) with domain walls or textured ferromagnets[73,74], bilayer and trilayer ferromagnetic regions[75], spin injection[76], and via spin-active interfaces[77] where a net interface magnetic moment is misaligned with respect to the bulk magnetization. The first experimental demonstration of long-ranged supercurrents was reported by Keizer *et al.*[22] from the observation of supercurrents through the half-metallic ferromagnet CrO_2. Because spin-singlet superconductivity cannot penetrate a fully spin-polarized material, this result necessarily implied the supercurrents were fully spin-polarized. The results were later repeated by Anwar and colleagues[78]. In 2010, a series of experiments by different groups demonstrated systematic evidence of spin-triplet pairing in SFS Josephson junctions: Khaire *et al.*[79] used ferromagnetic/non-magnetic multilayer spin mixers which were positioned at both superconductor interfaces, Robinson *et al.*[80] used the helical rare-earth antiferromagnet Ho to generate triplet supercurrents in Co, and Sprungmann *et al.*[81] used a Heusler alloy to generate triplet supercurrents. All of these experiments share similarities to the SF′FF′S device proposed by Houzet and Buzdin[82]: where the F′/F interfaces are magnetically coupled non-parallel.

Although it is now established that triplet supercurrents exist, their most interesting property—spin—is only inferred indirectly from supercurrent measurements. In conventional spintronics, it is known that spin currents cause effects such as spin-transfer torque-switching of magnetic elements and magnetization dynamics, so the observation of similar effects due to triplet supercurrents would confirm the net spin of triplet pairs and would therefore pave the way for applications. Several theoretical works have considered such situations and demonstrated that triplet supercurrents can indeed induce spin-transfer torque switching[83,84] and magnetization dynamics in the superconducting state[85–89]. Furthermore, the influence of superconductivity on spin-pumping effects have been theoretically investigated both in Josephson junctions[90] and in SF bilayers[91]. Other works have discussed spin dynamics in Josephson junctions[92] and the possibility of using spin-polarized supercurrents to induce magnetic domain wall motion[93–95]. Magnetic domain wall motion is a major research theme in spintronics as it can offer an alternative way to transmit and store information in a non-volatile way. It has been shown in ref. 95 that domain wall motion in superconducting junction can control whether the system resides in a dissipative or lossless state by locally switching on or off the superconductivity. The enhancement of supercurrents through the generation of triplet Cooper pairs when passing through a magnetic domain wall has been experimentally demonstrated in ref. 96. Another work[97] proposed making use of exchange spring magnetic systems where the magnetization texture is tunable by means of an external field which in turn triggers transitions between 0 and π states. The study of superconducting magnetization dynamics is at an early stage, especially from the experimental side, so there remains much work to be done in this particular area of superconducting spintronics. We note that the current densities required to obtain magnetization switching and domain wall motion in non-superconducting systems can in some cases be achieved with densities as low as 10^5 A cm^{-2}, which is comparable to critical current densities reported in SFS junctions. It is clear that domain wall motion would necessitate a non-equilibrium supercurrent set-up.

The relation between triplet supercurrents and the spin-transfer torque that they can induce is intricate, as they will have a feedback effect on each other[98]. This was explained by Waintal and Brouwer[83]: let F be the free energy of a Josephson junction containing two ferromagnetic layers with magnetization vectors that are misaligned with an angle θ. Denoting the superconducting phase difference as ϕ, the equilibrium charge and spin currents $I_{\rm Q}$ and $I_{\rm S}$ at a finite temperature are given by $I_{\rm Q}=(2e/\hbar)(\partial F/\partial\phi)$ and $I_{\rm S}=\partial F/\partial\theta$. Note that the equilibrium spin current is formally equivalent to a torque τ acting on the magnetizations which is equal in magnitude but opposite in sign for the two layers. On combining these equations, one finds that

$$\frac{\partial I_{\rm Q}}{\partial\theta}=\frac{2e}{\hbar}\frac{\partial\tau}{\partial\phi}$$

Because the charge supercurrent depends sensitively on θ (refs 18, 99,100), the above equation shows that spin-transfer torque is tunable by means of the superconducting phase difference ϕ.

Phase batteries and thermoelectric effects

The combination of superconducting and magnetic order in hybrid structures also produces quantum effects that may find applications in cryogenic spintronics in the form of so-called phase battery junctions or φ-junctions. In a Josephson junction without any magnetic elements, the equilibrium phase difference between the superconductors is zero. Introducing a ferromagnet as the interlayer separating the superconductors opens the possibility of π-coupling in the equilibrium state, as first predicted in ref. 16 and experimentally verified in ref. 101. However, the quantum ground state phase difference φ between two conventional s-wave superconductors separated by a magnetic interlayer is not necessarily 0 or π, but $0\leq\varphi\leq\pi$. Such a state can consist of either an extra phase shift in the first harmonic of the current–phase relation, providing a non-degenerate minimum for the ground

Box 1 | Spin injection and spin imbalance in superconductors.

The quasiparticle excitations in a superconductor can be described by 4×1 spinors when considering both particle–hole and spin space. The excitations are, in general, a mixture of electron and hole states, carrying a weight from each of these branches in their wavefunction. Nevertheless, they are typically characterized as being electron- or hole-like, depending on the asymptotic behaviour of the wavefunction for energies $E \gg \Delta$. For instance, an electron-like quasiparticle with spin-↑ may be written as $\psi = [u, 0, 0, v]^{\mathrm{T}} e^{iq_e x}$, where

$$u(v) = \sqrt{(1 + (-)\sqrt{E^2 - \Delta^2}/E)/2}$$

For $E \gg \Delta$, $u \to 1$ and $v \to 0$. The wavevector of the excitation is

$$q_e = \sqrt{2m(\mu + \sqrt{E^2 - \Delta^2})}$$

for a simple parabolic normal-state dispersion relation $\varepsilon_k = k^2/2m^*$ where m^* is an effective mass. The spin and charge content of this quasiparticle can be evaluated by introducing the operators

$$\hat{\mathbf{S}} = \frac{\hbar}{2}\begin{pmatrix} \boldsymbol{\sigma} & 0 \\ 0 & -\boldsymbol{\sigma}^* \end{pmatrix}, \quad \hat{Q} = -|e|\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

where $|e|$ is the magnitude of the electron charge and $\boldsymbol{\sigma}$ is a vector with the Pauli spin matrices as components. Computing the expectation values for spin and charge using ψ above then yields:

$$\langle \hat{\mathbf{S}} \rangle = (\hbar/2)\hat{\mathbf{z}}, \langle \hat{Q} \rangle = -|e|\sqrt{E^2 - \Delta^2}/E.$$

It is seen that whereas the spin of quasiparticles is constant, the effective charge is strongly dependent on its excitation energy E and vanishes near the gap edge $E \to \Delta$. This is the key property of the excitations that cause spin–charge separation and enhanced spin lifetimes in superconductors. The group velocity $v_{\mathrm{g}} = \partial E/\partial k = (k/m^*)(\varepsilon_k - \mu)/E$ of the excitation $E = \sqrt{(\varepsilon_k - \mu)^2 + \Delta^2}$ is also very small near the gap edge, as $E \to \Delta$ implies $(\varepsilon_k - \mu) \to 0$, causing scattering events to be less frequent and thus the lifetime to increase. With regard to spin current injection into a superconducting spin valve (Fig. 2d), the resulting spin imbalance in the superconductor depends strongly on the magnetization configuration. Following ref. 14, for a superconductor of smaller thickness than the spin diffusion length, the spin-↑ and spin-↓ distribution functions for quasiparticles can be taken as spatially uniform and described by the Fermi–Dirac function $f(E)$, but with shifted chemical potentials. In the P alignment, the spin conductances G_σ are equal at both interfaces due to the symmetric set-up and there is no net shift $\delta\mu$ in the chemical potential for any of the spin species σ. For the AP alignment, the different density of states for spin-↑ and spin-↓ at the two interfaces gives rise to imbalanced spin currents and produces a net shift in chemical potential for spins σ inside the superconductor. One may write $f_\uparrow(E) = f_0(E - \delta\mu)$ and $f_\downarrow(E) = f_0(E + \delta\mu)$. On evaluating the self-consistency equation for the superconducting order parameter, $1 = gN_0 \int_0^{\omega_{\mathrm{D}}} \mathrm{d}\varepsilon E^{-1}(1 - f_\uparrow - f_\downarrow)$, it is seen that the spin-discriminating shift in chemical potential plays an equivalent role to that of a Zeeman splitting $\mu_{\mathrm{B}}H$ due to an external field H, causing a first-order phase transition at the Clogston–Chandrasekhar[58,59] limit $\mu_{\mathrm{B}}H = \Delta_0/\sqrt{2}$. Above, ε is the normal-state dispersion, g is the attractive pairing potential, N_0 is the normal-state DoS at the Fermi level, μ_{B} is the Bohr magneton, and ω_{D} is the Debye cutoff.

state[102–104] or doubly degenerate minima $\pm\varphi$ for the ground state resulting from an interplay between the sign and magnitude of the first two harmonics[105,106]. The merit of creating a φ-junction where the equilibrium phase difference is tunable is that it may serve as a phase battery: a device which provides a constant phase shift between the two superconductors in a quantum circuit. Such a junction then supplies a phase shift φ in analogy to how a voltage is supplied by a battery, with the important difference that the phase does not discharge because the superconducting currents flowing in the system are dissipationless. In junctions that effectively feature three ferromagnetic layers with misaligned magnetizations, the spin chirality χ has been demonstrated[102,103] to be intimately linked to the realization of a φ state: $\chi \equiv \mathbf{M}_1 \cdot (\mathbf{M}_2 \times \mathbf{M}_3)$. However, the φ-junction may also be realized in other geometries and with homogeneous Zeeman fields in the presence of spin–orbit coupling, as predicted in ref. 104. Another example is a magnetic Josephson junction where the interlayer consists of two magnetic regions with different thicknesses and generates a spontaneous fractional vortex state, resulting in a degenerate φ-state, as shown in ref. 106.

Finally, we briefly discuss thermal biasing—thermoelectric—devices for superconducting spintronics. Thermoelectric effects chiefly arise due to the breaking of electron–hole symmetry, a feature most apparent in semiconducting materials where the chemical potential is electrically tunable. In superconductors, electron–hole symmetry is preserved near the Fermi level, and so thermoelectric effects are negligible. However, it is possible to break electron–hole symmetry per spin species σ while maintaining the overall electron–hole symmetry by using ferromagnet–superconductor hybrid structures[107,108], which can lead to large thermopowers and figures of merit. In the presence of spin-selective tunnelling, as may be achieved by tunnelling to a ferromagnetic electrode, one may also achieve large thermoelectric effects because electron–hole symmetry is broken for each spin species[109,110].

Outlook and perspectives

We end the review by offering our perspective on possible directions that may be fruitful to explore in developing superconducting spintronics. Although progress has been most pronounced on the theoretical understanding of SF proximity effects over the past decade, the experimental activity has in the past few years started to catch up. Nevertheless, there remains a plethora of interesting physics to investigate and we speculate that the most valuable experiments in the near future will directly verify (and quantify) the spin polarization of triplet states generated by different SF systems—existing experiments provide compelling evidence for spin-triplet pairing in SF structures, but they are not directly probing or using the spin carried by triplet supercurrents. Experiments which, therefore, demonstrate effects such as magnetization switching, magnetization precession, spin-transfer torque, or domain wall motion due to spin-polarized supercurrents will be pivotal in establishing applications of superconducting spintronics. Another issue that deserves investigation is the injection of spin-triplet pairs into superconductors, akin to the injection of spin-polarized quasiparticles into superconductors. Here, tunnelling experiments will be essential to understand how the density of states in a superconductor is modified due to the formation of a triplet state—in effect, the inverse of what is usually studied. We also mention that it might be interesting to design more comprehensive theories for the treatment of the ferromagnetic order in superconducting

Box 2 | Spin mixing and spin rotation at superconducting interfaces.

The process of generating spin-triplet superconductivity starting out from a spin-singlet Cooper pair can be understood conveniently by drawing on the phenomena of spin mixing and spin rotation[21] (see also ref. 20). The wavefunction for a spin-singlet Cooper pair can be written as

$$\psi_0 = \sqrt{\frac{1}{2}}(|\uparrow, \mathbf{k}\rangle\, |\downarrow, -\mathbf{k}\rangle - |\downarrow, \mathbf{k}\rangle\, |\uparrow, -\mathbf{k}\rangle)$$

where the prefactor ensures proper normalization. When the electrons of a Cooper pair encounter an interface region to a ferromagnetic material, scattering at the interface is accompanied not only by a shift in momentum but also a spin-dependent shift θ_σ, $\sigma = \uparrow, \downarrow$ in the phase of the wavefunction due to the Zeeman field that splits the majority and minority spin carriers. This may be written as

$$|\uparrow, \mathbf{k}\rangle \rightarrow e^{i\theta_\uparrow}\, |\uparrow, -\mathbf{k}\rangle, |\downarrow, \mathbf{k}\rangle \rightarrow e^{i\theta_\downarrow}\, |\downarrow, -\mathbf{k}\rangle$$

Applying these transformations to ψ_0 results in a new wavefunction which is a superposition of a spin-singlet and $S_z = 0$ spin-triplet wavefunction $\psi_{\text{short}} \equiv \sqrt{1/2}(|\uparrow, \mathbf{k}\rangle\, |\downarrow, -\mathbf{k}\rangle + |\downarrow, \mathbf{k}\rangle\, |\uparrow, -\mathbf{k}\rangle)$. The singlet and triplet parts are weighted by $\cos \Delta\theta$ and $\sin \Delta\theta$, respectively, where $\Delta\theta \equiv \theta_\uparrow - \theta_\downarrow$. In the absence of spin-dependent phase shifts ($\Delta\theta = 0$), the triplet component vanishes. The next step is to generate the equal-spin-triplet components $S_z = \pm 1$, which are insensitive to the paramagnetic pair-breaking effect of a Zeeman field, as the spins of the electrons in the Cooper pair are already aligned. The appearance of such long-ranged triplet correlations $\psi_{\text{long}} \equiv |\uparrow, \mathbf{k}\rangle\, |\uparrow, -\mathbf{k}\rangle$ (or $|\downarrow, \mathbf{k}\rangle\, |\downarrow, -\mathbf{k}\rangle$) can be brought about only by rotating/flipping one of the spins in the $S_z = 0$ triplet component. In this sense, the singlet Cooper pairs have served their purpose in terms of generating long-ranged triplets once the short-ranged triplets ψ_{short} have been created and are no longer needed. A magnetic texture serves as a source for spin rotation, which can be seen by letting the quantization axis be aligned with the local magnetization direction. Consider an $S_z = 0$ triplet state in a part of the system where the magnetization (and thus quantization axis) points along the z-direction. In another part of the system where the magnetization points in the x-direction, the same triplet state now looks like a combination of the equal-spin pairing states $S_z = \pm 1$ as seen from the new quantization axis. The combination of spin-mixing and spin-rotation processes then explains how the spin-singlet s-wave component of the bulk superconductor may be converted into a long-range spin-triplet component that is able to survive even in extreme environments, such as half-metallic ferromagnets that are fully spin-polarized[22,78,116].

proximity structures, which is usually simply modelled by a Zeeman field h acting on the spins of the electrons. This could be done by incorporating the effect of spin-bandwidth asymmetry (spin-dependent carrier masses) and also by considering more seriously the role of the magnetic vector potential in the proximity effect. We also note that the electromagnetic effect of stray fields in SF structures have been experimentally shown to offer an interesting way to control superconductivity[111–113].

There is also a need to develop spin-triplet theory to understand better the interactions between superconducting and spin-polarized order, particularly in non-equilibrium devices where spin and charge dynamics are important. The mechanisms required for generating triplet pairs at SF interfaces are generally well understood, just as equilibrium proximity effects are in Josephson junctions and SF multilayers, but to advance superconducting spintronics it is essential to develop a framework for non-equilibrium transport that can account for dynamic interactions involving spin-triplet pairs and ferromagnetic layers[87,88]. Related to this, it is also necessary to clarify the mutual dependence between supercurrent flow and magnetization configuration. The formation of so-called Andreev bound states[114] in textured magnetic Josephson junctions should influence the spin pattern in the ground state of such systems, as they contribute to the effective field $\mathbf{H}_{\text{eff}}$, which in turn determines the equilibrium magnetization profile through the condition $\mathbf{m} \times \mathbf{H}_{\text{eff}} = 0$. Whereas the magnetic profile of a junction is usually considered as being fixed, the Andreev bound state contribution is phase sensitive, suggesting that the magnetization texture could be controlled by means of the superconducting phase difference[89]. Moreover, the sizable thermoelectric effects in superconducting hybrids are of practical interest owing to the possibility of transforming excess heat into electric energy in a highly efficient manner, suggesting applications in cooling of nanoscale systems and thermal sensors/detectors.

In summary, we have provided an overview into past and present activity related to superconducting spintronics, including the associated quantum effects that appear. With advances in experimental fabrication processes and better control of interface properties, there is good reason to be optimistic about further discoveries of novel physics arising due to the synergy between superconductivity and spintronics.

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Competing financial interests
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