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15 Superconductor/ferromagnet hybrids

Abstract: Superconductivity and magnetism have very different underlying order parameters and so it is to be expected that the two phenomena can combine only over very short lengthscales. However, at nanometer lengthscales a rich range of phenomena have been predicted, many of which have now been experimentally observed. In this chapter, the range of such phenomena is reviewed, together with a forward view of potential applications.

15.1 Introduction

Conventional superconductivity is mediated by the formation of Cooper pairs of electrons. These singlet pairs consist of electrons with antiparallel spins so that a supercurrent carries a charge but cannot carry a net spin. The pairing process is driven by a condensation energy which lowers the overall electron energy; this energy per electron is in the meV range for most superconductors and so much smaller than the typical exchange energies associated with magnetism (eV range per electron). This means that magnetic impurities, even isolated magnetic ions, strongly suppress superconductivity via a tendency to align electron spins and so break Cooper pairs.

Despite this, it is possible to create hybrid materials and devices in which superconductivity and magnetism can co-exist and, indeed, can cooperate to create novel behavior. However, the lengthscales over which the cooperation can exist are frequently very short (nanometers) meaning that sophisticated heterostructure growth processes are required to create structures in which this can be studied. For this reason, much of the early study of hybrid systems was theoretical and only in the past couple of decades has it become possible to perform detailed experimental studies of this behavior.

Rather than present a chronological perspective of this development, this chapter will explain the underlying factors which determine the interaction between superconductivity and magnetism and demonstrate ways in which these can be exploited to create interesting and potentially exploitable devices.

The primary coupling mechanisms between a ferromagnet and a superconductor involve magnetic fields – either real or virtual exchange fields within the materials themselves. The former is relatively simple and well understood: the critical field (H_c) of a (Type I) superconductor is reached when the Zeeman energy associated with the switch from antiparallel to parallel alignment of the electrons within the Cooper pair is equal to the condensation energy. Because of the formation of Abrikosov vortices

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which screen a proportion of a Type II superconductor from an external field, the upper critical field (H_{c2}) of these materials can be much larger but, even then, laboratory-scale fields $\mu_0 H$ of the order of 10 T will strongly suppress superconductivity in most materials apart from those specifically developed for high-field performance. As well as any field externally applied, stray magnetic fields can also be generated in hybrid structures through sample geometry (for example magnetic nanoparticles or the edges of patterned magnetic films), film roughness, and the presence of domain walls or vortices in the magnetic layers [1]. Several mechanisms have been proposed by which magnetic hybrid structures can be used to enhance vortex pinning and so increase the critical current density; these are discussed in Section 15.5. Although it may be possible to exploit such stray fields, for example by generating fixed local magnetic fields using patterned magnetic layers on a superconductor and so changing the overall dependence of the superconducting properties on applied field [2], in most experiments care is taken to minimize them so that intrinsic effects can be studied in isolation.

The internal exchange field within a ferromagnet can be considerably larger than any field that could be externally applied and hence the effective suppression of superconductivity is very strong. The simplest experimental geometry to study this is a superconductor / ferromagnet (S/F) bilayer. If the ferromagnet is metallic there are actually two effects at work: the first is the conventional proximity effect which occurs at any superconductor / normal metal (S/N) interface and the second is the additional pair-breaking interaction of the exchange field experienced when the pairs enter the ferromagnet.

At very short length-scales, the interaction between Cooper pairs and exchange fields can be understood in terms of a loss of phase coherence between the electrons. Where other scattering effects are comparatively weak – for example in the clean limit – the phase of the pair wavefunction can oscillate while remaining finite. The phase can therefore be reversed and so create a so-called π-state, which can be experimentally observed in several types of experiment.

The effects introduced above represent the standard response of singlet pairs to fields and are discussed further in Section 15.2. More complex behavior can be observed if the ferromagnet, instead of being homogeneous, contains noncolinear elements. Here a spin-mixing effect can be generated that results in triplet pairing where it is possible for a pair (strictly, a pair correlation because there is no condensation energy) to be formed of spin-aligned electrons. Such pairs have a net spin and so can potentially enable a supercurrent to carry a spin; this is discussed further in Section 15.4.

The proximity effect within a bilayer can be eliminated if the ferromagnet is insulating; the electrons then experience an effective exchange field *within the superconductor* as a consequence of scattering from the spin-active interface with the ferromagnet [3]. In addition to a direct pair-breaking effect, a field within a superconductor (whether real or virtual) leads to the splitting of the quasiparticle density of states (DoS) which can be experimentally measured. This is discussed in more detail in Section 15.3.

The aim of this chapter is to give an overview of the range of effects possible and their potential applications. There are a number of more specialist reviews covering aspects of the material presented here [1, 4–8] and the reader is referred to these for further information.

15.2 Singlet proximity coupling

The standard S/N proximity effect can be understood in terms of the dilution of the pairs intrinsic to the superconductor by the unpaired electrons in the normal metal. Thus, for a superconductor thinner than the coherence length, as the thickness of the normal metal is increased, the critical temperature (T_c) progressively decreases towards zero. For thicker superconductors, there are pairs that do not interact with the normal metal and so the bulk T_c is maintained; this is illustrated schematically in Figure 15.1.

This suppression of T_c is enhanced in the case of an S/F bilayer. Here, the exchange field within the ferromagnet decreases the amplitude of the pair potential in the ferromagnet much faster than for the equivalent normal metal, meaning that for, a given thickness, the S/F T_c is lower than that for an S/N bilayer. Superimposed on this suppression, there is the potential for the Larkin–Ovchinnikov–Ferrell–Fulde (LOFF) [9, 10] phase oscillation effect discussed in the introduction. This can give rise to a weak oscillation of T_c as a function of ferromagnet thickness [11], although little more than the appearance of a nonmonotonic suppression is usually observed experimentally (Figure 15.1).

If a second ferromagnetic layer is added – to create either a F/S/F (Figure 15.2) or S/F/F‘ superconducting spin valve – a rather more dramatic effect can be observed. In

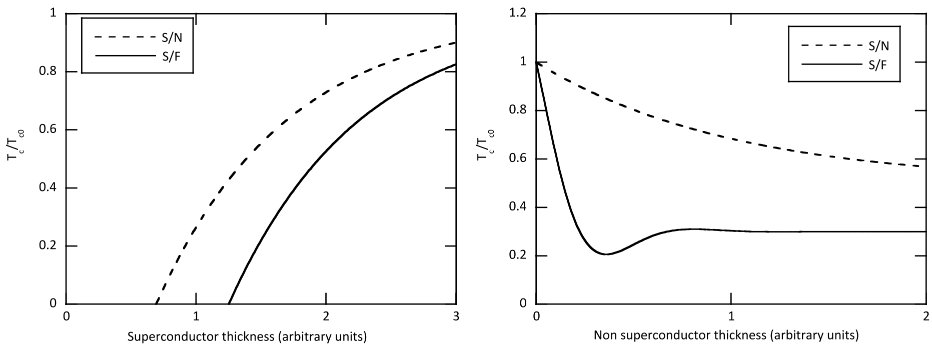

Fig. 15.1: Left: schematic dependence of the normalized critical temperature on the superconductor thickness for superconductor / normal metal (S/N) and superconductor / ferromagnet (S/F) bilayers. Right: schematic dependence of the normalized critical temperature on the nonsuperconductor thickness for superconductor / normal metal (S/N) and superconductor / ferromagnet (S/F) bilayers.

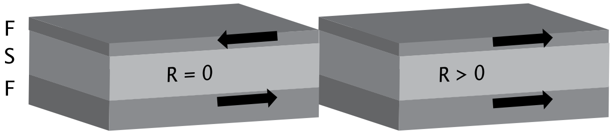

Fig. 15.2: The two magnetic states of a superconducting spin valve. Left, antiparallel, superconducting state and, right, parallel nonsuperconducting state.

the original predictions for the behavior of such devices [12, 13], which predated their experimental realization, the antiparallel (AP) magnetic alignment of the two F layers should cancel the phase oscillation effect meaning that the T_c for this alignment should always be higher than the parallel (P) state in which the effects add and so in principle it should be possible to switch between a zero resistance, superconducting state and the normal state (infinite magnetoresistance). In practice, the effects seen in spin valves containing standard transition-metal ferromagnets are rather weak (typically the change in T_c (ΔT_c) is only a few mK [14–16]). The primary reason for this is the direct pair-breaking effect of the exchange field within the ferromagnets acting in conjunction with standard scattering and proximity effects in them which are present regardless of the layers' relative alignment.

Several systems have recently shown considerably larger values of ΔT_c. The first of these involves epitaxial films of the rare-earth ferromagnets Ho and Dy. As-cooled through their Curie temperatures, Ho and Dy thin films show an antiferromagnetic spin spiral structure, but this can be irreversibly converted into a linear ferromagnetic state by the action of moderate magnetic fields [17, 18]. Such devices then show superconducting spin-valve behavior with $\Delta T_c \sim 0.5$ K [19] together with infinite resistance. The underlying reason for these large values is currently unclear, but it may be linked to a resistance or density of states mismatch between the superconductor (Nb) and the rare earth so that the devices have a functional similarity to the ferromagnetic insulator devices discussed in Section 15.3. Even larger values of ΔT_c have been reported in devices that generate triplet pairing as discussed in Section 15.4.

The decay and LOFF oscillations can be detected much more strongly, and the induced phase difference measured directly, in proximity-coupled S/F/S or S/F/I/S Josephson junctions. Such devices were first created using weak ferromagnetic alloys as the barrier which enabled thicker layers to be grown [20, 21], but since then all the transition-metal ferromagnets have been used so that the underlying theories can be fully probed [22–24]. As with superconducting spin valves, the LOFF oscillations are superimposed on a general decay associated with scattering, but this can be minimized more effectively in S/F/S junctions through the use of materials which can approach the clean limit [23]. The most striking experimental outcome is the measurement of multiple oscillations of the critical current (I_c) as a function of barrier thickness [23, 24]; these arise as a result of the phase shifts acquired by the pairs under the exchange field of the ferromagnet which translate directly to a periodic switching

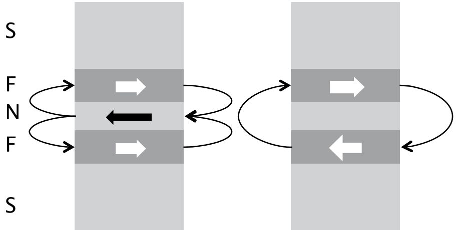

Fig. 15.3: The two magnetic states of a spin-valve Josephson junction. Left: parallel, illustrating the flux injection into the junction which generates a corresponding phase difference; right: antiparallel, illustrating the cancelation of the net flux.

of the ground-state phase difference of the junction between zero and π. It is possible to measure this phase shift directly using a phase-sensitive circuit [25] and junctions maintaining a π phase-shift can be directly applied in quantum bit (qubit) structures [26].

As with superconducting spin valves, a barrier consisting of two ferromagnetic layers has the potential to cancel the LOFF-induced phase-shifts. Such spin-valve Josephson junctions [27] were first demonstrated before detailed tests of the underlying theories had been performed, but nevertheless showed large changes in the critical current (ΔI_c). Somewhat later, it was appreciated that two effects contribute to ΔI_c in such devices: in addition to the phase-shift discussed above, the magnetic flux associated with the barrier magnetization also depends strongly on the magnetic alignment of the spin-valve barrier [28] and so directly controls I_c through the standard Josephson relation (Figure 15.3). By designing devices in which the two effects can act in conjunction it has been possible to create spin-valve Josephson junctions that can act as cryogenic memory elements [29, 30].

15.3 Exchange fields and DoS splitting in superconductors

The pioneering experiment of Meservey and Tedrow demonstrated that the quasiparticle DoS in ultra-thin Al could be significantly split by an applied magnetic field [31]. As illustrated in Figure 15.4, this splitting enables selective tunneling from the exchange-split bands of a ferromagnet in an S/I/F tunnel junction and thus provides a direct method of measuring the tunneling spin polarization of ferromagnets [32]. These experiments were extended to measuring the properties of Al in contact with the ferromagnetic insulator EuS in F I/S/I/N junctions [3, 33]. Here it was shown that the Al presented a strong exchange splitting of the quasiparticle density of states even at low applied fields. The effective exchange field responsible for the splitting is believed to be acquired during scattering at the S/FI interface but, as with a physical field, can also lead to a direct suppression of superconductivity via spin-orbit scattering. Al is therefore the material on which most experiments have been performed, but splitting has also been observed in NbN [34] and Ga [35].

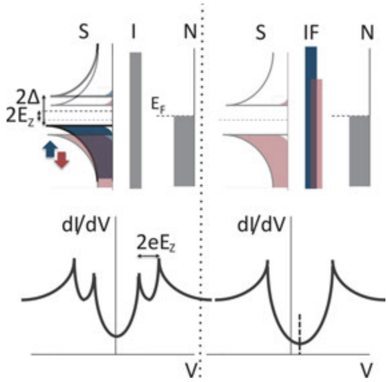

Fig. 15.4: Zeeman splitting of the superconductor quasiparticle density of states (upper row) and the corresponding conductance vs voltage characteristics (lower row), in the case of a standard tunnel barrier (left column) and a ferromagnetic insulator tunnel barrier (right column). In the ferromagnetic insulator case, the spin filtering of the tunnel barrier eliminates tunneling from one of the spin-split bands of the superconductor.

As well as providing a means of detecting the splitting, exchange-split S/I/N tunnel junctions can also be configured to inject spin-polarized quasiparticle currents into the superconductor [36]. This is illustrated schematically in Figure 15.4: alignment of the spin-split DoS with the N electrode Fermi energy results in a strongly spin-selective tunneling which can be controlled via the junction bias voltage.

Certain ferromagnetic insulators, such as EuS [37] and GdN [38] can also be grown as tunnel barriers which means that the tunneling DoS of one or both superconducting electrodes is directly split by the exchange field from the barrier. In this case the conductance spectra are also affected by the spin-filtering effect of the tunnel barrier, which presents a different barrier height for the two electron spin directions and generally leads to large intrinsic spin polarization [39]. Thus, although four conductance peaks should arise from the spin-splitting of the quasiparticle DoS of both electrodes, in the case of a strongly spin-polarizing barrier, only the two corresponding to the allowed tunneling spin are observed [34] (Figure 15.4).

S/FI/S junctions can also show a Josephson supercurrent [38]. Singlet pair tunneling should be strongly suppressed by a spin-filtering barrier [40] and the presence of a finite critical current even for very high spin-filter efficiencies suggests the potential role of triplet pairs in the tunneling process. Although the theories for such devices are still being developed, evidence for unconventional superconductivity which is probably linked to triplet pairing comes from a pure 2nd harmonic in the current-phase relation of such devices [41].

The exchange splitting within FI/S/FI ferromagnetic insulator superconducting spin valves is responsible for much larger values of ΔT_c than so far measured in metallic devices. The underlying reason is simply that the proximity effect which strongly suppresses superconductivity regardless of the magnetic configuration in metallic devices is absent in ferromagnetic insulator structures. Indeed the basic concept, proposed by de Gennes in 1966 [42] is largely valid in explaining the behavior so that the critical temperature depends on the net exchange field in the superconductor which, for superconductor thicknesses less than the coherence length, is effectively canceled in the AP configuration. EuS/Al/EuS [43] and GdN/Nb/GdN spin valves both show ΔT_c ~ 1 K in combination with infinite magnetoresistance (i.e., full switching between superconducting and normal states at fixed temperature) extending over a significant temperature range.

15.4 Triplet pairing in hybrid systems

Various modes of triplet pairing in superconductors are theoretically possible. Intrinsic triplet pairing superconductors, such as Sr_2RuO_4 [44] are very rare, and because of their even-frequency p-wave nature, are highly susceptible to defects. The potential for odd-frequency s-wave triplet pairing in superconductor ferromagnet hybrid systems was first proposed in 2001 [45] in the form of a proximity effect mediated by an inhomogeneous magnetic interface which could "mix" the singlet pairs into the superconductor in the various triplet combinations. The formation of spin-aligned triplet pairs as part of this process gives the potential for a long-range proximity effect in a (homogeneous) ferromagnet attached to the mixer layer.

A landmark experiment in 2006 provided the first evidence that this process was possible [46]; here an S/F/S junction was created in which the barrier was CrO_2, a material generally accepted to be intrinsically half-metallic – i.e., a ferromagnetic material in which there is a band gap in the density of states for one spin direction [47]. Although this first experiment provided no information about the nature of the mixing interface between the NbTiN superconductor and the CrO_2, a singlet pair cannot exist in a half-metallic material because only electrons of one spin sign are present at the Fermi energy; the supercurrent that was measured therefore had to consist of spin-aligned pairs.

More direct confirmation of the underlying theories were provided by a series of experiments in which engineered artificial spin-mixer layers were inserted at the S/F interfaces. These were Nb/Ho/Co/Ho/Nb junctions in which the spin-spiral antiferromagnetism of the Ho provides an intrinsically inhomogeneous interface [48], and Nb/PdNi/Co/PdNi/Nb structures in which the thin interfacial PdNi layers could be noncollinear with the thick Co barrier structure [49]; Finally, it was shown that the MgO/CrO_2 interface could be deliberately engineered to increase the critical current of junctions [50]. A series of further experiments have demonstrated that the misorientation angle of the F' mixer layers in S/F'/F/F'/S devices changes the induced triplet critical current in quantitative agreement with theory [51, 52].

A triplet supercurrent, provided it contains unequal numbers of up-up and down-down spin-aligned pairs should therefore carry a dissipation-less spin current. This has to be the case if the barrier is half-metallic, but so far no experiment based on conventional ferromagnets has been able to determine directly the induced spin polarization of the supercurrent. Despite this, the discovery of a controllable triplet state has raised serious prospects for a superconducting version of spin electronics or "superspintronics". The potential applications are discussed further in Section 15.6.

Evidence for triplet pairing has also been obtained from passive proximity effect spin-valve structures. The generic structures explored in such experiments are S/F'/F or F/M/S/M/F heterostructures (where M is an intrinsic spin-mixing interface). An S/F'/F structure in the P or AP configuration should not enable any singlet-triplet conversion at the interface and so the critical temperature is determined by a combination of the singlet proximity effect and exchange-field driven pair-breaking as discussed in Section 15.2. Singlet pair propagation is suppressed by the polarization of the Fermi surface in the F layer, and is obviously zero if a half-metal is used. If the F' and F layers are not collinear, triplet pairs should be generated at the interface and such pairs (at least those parallel to the majority states at the F layer Fermi energy) should be able to enter the F layer and should be immune from LOFF dephasing and pair-breaking effects. Thus, the proximity effect should be stronger than the collinear case and the T_c correspondingly lower. This effect has been observed experimentally in both conventional ferromagnetic spin valves [53, 54], and in CrO_2-based spin valves for which very large values of ΔT_c were observed because of the effective blocking of conventional proximity coupling in the collinear configuration [55].

The F/M/S/M/F structure is a development of the standard superconducting spin valve discussed in Section 15.2 for which P alignment should give a lower T_c. Identical structures, but with Ho spin-mixer layers inserted at the interface gave the opposite effect – i.e., the AP state had the lowest T_c [56]. This can be understood in terms of the creation of both up-up and down-down triplet pairs at both interfaces; if the F layers are AP then each pair direction is parallel to the magnetization direction of one of the F layers and so can induce a strong proximity coupling. In contrast, for P alignment, one spin sign is prevented from entering either F layer and thus weakening the proximity effect and raising the T_c in accordance with the experimental results. Since this model is dependent on the conventional ferromagnetic layer being spin selective, this result also provides indirect evidence that the polarization of the triplet pairs can be controlled by the magnetic state of a device.

15.5 Abrikosov vortex pinning in hybrid systems

Conventional vortex pinning processes depend on the sample microstructure to break the translation invariance of the superconducting properties. Such processes can be broadly separated into those mediated by the condensation energy ("vortex core pinning") and by the disruption of the vortex screening currents ("magnetic pinning") [57]. The former is generated by nonsuperconducting, ideally insulating, inclusions or voids within the superconducting matrix; the pinning energy is then the difference between the core of the vortex passing through the superconducting matrix (where the condensation energy is lost within the core) and passing through the pinning center (in which there is no loss of condensation energy). The latter, magnetic pinning, is mediated by extended defects, such as grain boundaries, which disrupt the flow of the screening currents surrounding a vortex, thus altering the total energy. An optimized combination of these effects is used to create high critical current conductors such as Nb_3Sn.

A range of experiments have been performed to try to demonstrate that magnetic inclusions can provide more effective pinning, particularly in high T_c materials in which pinning effects associated with microstructural defects are ineffective because of the direct suppression of the superconductivity inherent in materials with very short coherence lengths. A number of model experimental systems have been explored, such as Hg/Fe [58] and Nb/Gd [59] in which the insolubility of the magnetic species in the superconducting matrix can generate a dispersion of ferromagnetic nanoparticles.

The simplest model for the interaction between a magnetic particle and a vortex assumes that the magnetization of the particle is constant [58] and this generates an interaction force via the induced changes in the circulating screening currents surrounding the particle and vortex as their separation changes. Thus, this is an enhanced version of the conventional magnetic pinning discussed above and requires that the particle spacing is larger than the magnetic penetration depth, otherwise the pinning energy associated with an assembly of particles averages to zero. The large penetration depths of technological superconductors means that this effect is likely to be relevant only at the lowest fields.

The behavior is more complex if the ferromagnetic particles are magnetically soft enough to respond to the fields associated with vortices. Various models for pinning in these circumstances have been proposed. Two examples are: one which is based on hysteresis losses in the particles as vortices pass over them seems to adequately explain experimental data from Nb/Gd nanocomposites [59]; a second is based on the effective capturing of flux by extended high susceptibility defects which effectively lowers the mobile flux within a vortex hence reducing the Lorentz forces acting on it – in other words decreasing the driving force for displacement from conventional pinning centers rather than directly increasing the pinning force [60].

15.6 Potential applications

Conventional spintronics emerged from the discovery of giant magnetoresistance (GMR) in the 1980s. It rapidly achieved enormous technological success in the data storage field: initially as a means of improving magnetic field sensors for reading data from hard discs and more recently as the data storage element in magnetic random access memory (MRAM). More broadly, spintronics has been promoted as an eventual low-power replacement for charge-based semiconductor (CMOS) logic in which information is carried by spin currents and controlled and sensed by magnetic elements within a circuit [61].

An idea of the potential energy savings that can be gained by taking the minimum energy required for switching a magnetic element (for example the free layer in a magnetic tunnel junction) as the anisotropy energy barrier required to prevent thermally activated reversal: for memory applications this is typically taken to be $50k_BT$, where k_B is Boltzmann's constant and T is the operating temperature. At 4 K this corresponds to only 3×10^{-21} J and so is orders of magnitude below the single bit write energy for currently used random access memories. Even though this argument ignores dynamic effects it is clear that the low switching energy of a magnetic memory element has the potential to massively lower energy consumption in computing systems. Such operations can be most directly achieved via spin-transfer torque (STT) in which a spin current can switch a ferromagnetic element [62]. However, the intrinsic inefficiency of STT means that large charge currents, with correspondingly large ohmic losses, are required for switching and have so far eliminated the potential gains over semiconductor electronics.

Combining superconductivity with spintronics within superspintronics [63, 64] brings in phenomena that do not exist in the normal state, such as quantum coherence and spin-polarized supercurrents, potentially enabling much lower energy spin-transfer and magnetic switching. Indeed, preliminary steps have already been taken to develop superspintronic technology: the cryogenic memory elements discussed in Section 15.2 have already been shown to switch between P and AP states via STT, albeit still with large current densities which take the device into the normal state [30].

The potential for superconducting spin currents has already been discussed in Section 15.4 and there is the possibility for such currents to be able to modify the magnetic state of an element. In addition, although the pair condensate in a singlet superconductor has zero net spin, this is not necessarily true for the population of quasiparticle excitations (see Section 15.3). Indeed, there are circumstances in which the quasiparticle spin-decay length in the superconducting state is much longer than in the normal state and a very large effective spin polarization can be induced even by unpolarized current injection [36] and quasiparticle spin currents can be detected via the inverse spin Hall effect [65]. However, in the superconducting state any quasiparticle spin currents must be diffusive and independent of the (zero-spin) charge supercurrent meaning that many of the familiar concepts of conventional spintronics such as giant magnetoresistance do not have a direct quasiparticle spin equivalent.

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