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Proximity effects in superconducting spin-valve structures

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Chapter 1

Introduction

1.1 Superconductivity

Absolutely nothing... is all that is left of the resistance of a superconductor. This stunning result was first seen by Heike Kamerlingh Onnes in 1911 at the university of Leiden when cooling down a piece of metallic Mercury (Hg) to below 4.2 K [1]. Normally for metallic substances the electrical resistance goes down with decreasing temperature and saturates at low temperatures meaning that a finite resistance remains, caused by imperfections to the (infinite) crystal lattice. The unique property of a superconductor is that the resistance suddenly vanishes at the moment that the temperature falls below the critical temperature (T_c) of the material (see Fig. 1.1). Several well-known elemental superconductors are Aluminum (Al), Tin (Sn), Lead (Pb) and Niobium (Nb) with respective T_c's of 1.2, 3.7, 7.2 and 9.3 K. The latter being the highest of elemental bulk superconductors. Higher T_c's are found in a variety of alloys and compounds. Widely used are the compounds of Niobium with Nitrogen (NbN) and with Titanium ($Nb_{0.6}Ti_{0.4}$), which have respective T_c's of 16.1 and 9.8 K. In 1986, a new class of superconductors was discovered by Bednorz and Müller [2], the superconducting copper-oxides (cuprates). These are synthesized ceramic compounds with a perovskite crystal structure. A (at that time) staggering T_c as high as 92 K was found in the cuprate $YBa_2Cu_3O_7$, now know as YBCO (Yttrium-Barium-Copper-Oxide). This meant superconductivity at temperatures above the liquid Nitrogen (LN_2) temperature of 77 K, which is the standard coolant to get far below room temperature. Currently the highest observed T_c for a bulk material is found in a cuprate and reaches 138 K (the cuprate being $(Hg_{0.8}Tl_{0.2})Ba_2Ca_2Cu_3O_{8.33}$). The complete disappearance of electrical resistance is not the only unique property of a superconductor. A second important property is the expulsion of magnetic flux from the interior of the superconductor (Meissner effect [3]) which also starts when the temperature gets below T_c. This is very different from ordinary, non-superconducting matter, where magnetic flux can (almost) penetrate straight through. Two main types of superconductors are distinguished based on their ability to expel magnetic flux. In type-I superconductors the expulsion is

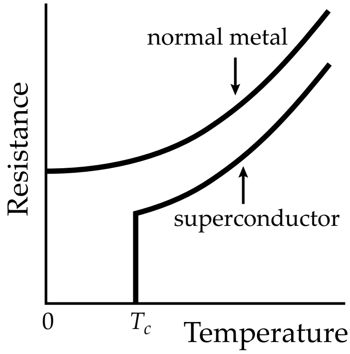

Figure 1.1: Typical resistance versus temperature curves for a normal metal and superconductor with transition temperature T_c.

complete up to a certain maximum or critical field H_{c1}, above which the superconductor returns to the normal state. Type-II superconductors also show complete expulsion up to a field H_{c1}, but now for higher fields magnetic flux starts to enter the superconductor in the form of small flux bundles, so-called vortices, crossing the interior of the superconductor (see Fig. 1.2). These

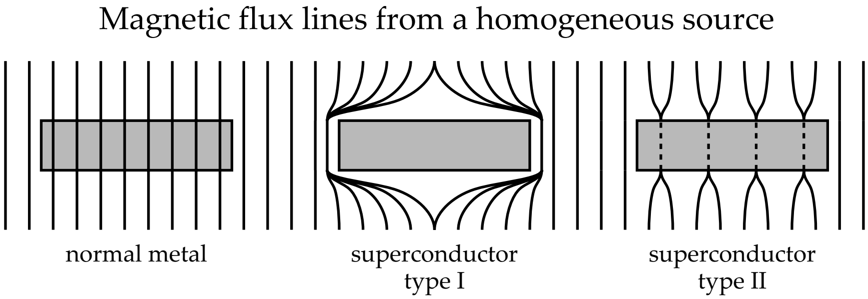

Figure 1.2: Magnetic flux lines through a normal metal, a type-I superconductor and a type-II superconductor. Dashed lines represent the vortices.

vortices are like tubes having a non-superconducting core and carry a single quantum of flux $\phi_0 = h/(2e)$. With increasing field the density of vortices increases until at a field H_{c2} the superconductor becomes normal. Almost all elemental superconductors are type-I (exceptions are Niobium, Vanadium (V) and Technetium (Tc)), while the cuprates and the other superconducting alloys and compounds are all type-II.

But what exactly *is* superconductivity? This question remained unanswered for almost 50 years. The microscopic mechanism and description of this phenomenon got shape in 1957, when it was recognized that an attractive interaction between electrons is needed. For the classical superconductors this is mediated by electron-phonon coupling, but for the high-T_c superconductors the interaction mechanism is not fully unravelled yet even at present day. In short, superconductivity is a condensed state (or condensate) of specially paired electrons isolated from the normal electrons by an energy gap Δ. The binding energy of the pairs is weak (order of meV) and easily destroyed by temperature, so only at very low temperatures superconductivity will appear. It is a macroscopic quantum phenomena which can be described by a single wave function characterized by an amplitude and phase (both in general functions of position and time). This condensate of electron pairs thus moves/acts as a single entity and is not hindered by the obstacles that cause dissipation for the normal electrons. One could say, the mechanism for dissipation in normal metals (such as inelastic collisions with other electrons or the crystal lattice) are just not powerful enough to break these electron pairs apart. For temperatures *above* T_c the superconductor act as a normal metal. The Fermi sphere (the collection of electronic states) is filled up to the Fermi energy (E_F) with the exception of an energy shell of order k_BT around E_F which contains excited states. Furthermore, around E_F the density of states is continuous and more or less constant. For temperatures *below* T_c the superconducting condensate emerges with a peculiar density of states around the Fermi energy; the condensate itself is located *at* E_F, but the continuum of states closely around E_F have disappeared. The condensate is isolated from the normal electron states by an an energy of Δ, both to the states below and above E_F. The full size of the energy gap is $2\Delta \sim 3.5k_BT_c$ (for weak coupling) and thus directly relates to T_c. The electron pairs in the condensate are called Cooper pairs, named after Leon Cooper who in 1956 came up with the idea of these pairs. The pairing goes in a special way: only electrons with opposite momentum (k-vector) and opposite spin can form a Cooper pair and condense into the condensate (in case of a conventional superconductor). The conventional Cooper pair thus contains an electron with spin up and an electron with spin down. This makes superconductivity intrinsically incompatible with magnetic fields, which tend to align all the electron spins. A direct result of the fact that the condensate can move freely, is that it can create circular currents (screening currents) to generate a magnetic field. This is what happens when a superconductor is exposed to magnetic flux; it generates and equal but opposite field to expel the magnetic flux from its interior. Can a superconductor expel just any field? No, because the supercurrent density, and thus the generated field strength, is limited. Above this maximum the Cooper pairs become unstable and the condensate collapses, forcing the superconductor back to the normal state. The field at which this happens is called the (upper) critical field, and the maximum current it can carry is the critical current I_c. Whether a superconductor is type-I or type-II depends on the ratio between two important parameters of the superconductor. These two are the superconducting coherence length ξ_S and the magnetic penetration depth λ. The first one (although a variety of definitions exists) is roughly the characteristic length over which Δ can change its amplitude significantly. It is also roughly the average size of a Cooper pair, which is a very non-local object. The second one is the distance over which magnetic flux can penetrate the superconductor from the sides/surfaces (a normal metal has infinite λ). If the ratio $\lambda/\xi_S < 1/\sqrt{2}$ the superconductor is type-I, else it is type-II, where for ξ_S the Ginzburg-Landau coherence length should be used. In other words, only when the magnetic field screening is strong enough to reduce λ to about

ξ_S, the superconductor can completely free its interior from magnetic flux. Otherwise, the superconductor gives up certain areas, in the form of vortices, to permit flux passing through its interior.

1.2 S/F junctions

Whenever (a layer of) one material is connected to another (layer of) material, an interface is created. At the interface the electronic states from both sides are glued together and the electronic properties of both materials become mixed in a small region near the interface. The quality of the interface determines the resistance, or better, the transmission and reflection probabilities for an incoming electron. In general these probabilities depend on the wave vector (or energy) and spin of the incoming electron. The latter becomes important when magnetic materials are used. The resulting interface region may have new properties (i.e. properties that don't appear in the bulk materials) and transport characteristics can change drastically. This makes it very interesting for both fundamental research and for applied / device-oriented research. A good example is the diode (or pn-junction), which is a very commonly used device and can be formed by connecting a p-type to an n-type doped semiconductor. It shows a highly non-linear (non-ohmic) current-voltage characteristic: the interface allows electrical current to pass in one direction, while blocking the other direction. Whenever a magnetic material is used the interface will have a spin dependent transmission/reflection. Usually the transmission for one spin direction is (much) better than for the opposite spin direction. This difference originates from the availability of electron states near the Fermi energy, which for magnetic materials are generally unequal for the two spin directions. A particular convenience of the electron spin is that its direction can be externally manipulated by applying a magnetic field. This possible manipulation combined with spin dependent interface properties has led to many spin based devices. Perhaps most important is the Giant-Magneto-Resistance (GMR) effect. In GMR devices, the electrical resistance can be changed significantly by a relative small external field. This effect forms the basis for modern spintronics, also called spin transport electronics. These are devices where the electron spin is exploited to manipulate the transport of its electrical charge, thus manipulating the transport characteristics of the device. Although superconductors can carry electrical current without energy loss, they are not well fitted to integrate in standard electronics due to the very low temperature of operation. Yet they have found their way into medical instruments where macroscopically large superconducting coils

Bloch states (plane waves) are defined by wave vector $\mathbf{k}$ and spin σ

are used to generate high magnetic fields (necessary for the Magnetic Resonance Imaging (MRI) scanners) Next to this application of bulky "power" sources, they are also integrated into space technology electronics, usually as highly sensitive measurement/detection electronics (sensors), as in outer space the natural environment temperature is only a few Kelvin.

A superconductor (S) and ferromagnet (F) both have a global, but mutually antagonistic, ordering for electron spins. This makes that interfaces between the two materials (proximity systems) are expected to be very rich in physics as multiple energy and length scales are competing with each other. One of the main questions in these systems is: how does the induced superconductivity (in F) behave and over what distances can it survive. The main competitors are the superconducting gap energy Δ, the ferromagnetic exchange energy E_{ex}, the superconducting (ξ_S) and ferromagnetic (ξ_F) coherence length, and the size of the system (thicknesses of the layers). The energies Δ and E_{ex} are respectively coupled to the lengths ξ_S and ξ_F. Here, ξ_F is the typical distance over which Cooper pairs dephase in the F layer, and E_{ex} is related to the potential energy difference between the (Fermi levels of the) two spin bands. These S/F proximity systems are in general most interesting from a fundamental and theoretical point of view. The first reports on junctions between a superconductor and non-superconducting material date from 1970-1973. Meservey *et al.* [4] and Tedrow *et al.* [5, 6] examined S/F junctions and showed the existence of a spin-polarized current across Al/AlO_x/F junctions. From this an estimate for the degree of polarization of the ferromagnetic layer was obtained. The Aluminum-Oxide (AlO_x) is a (thin) insulator layer, and as ferromagnetic material they used Iron (Fe), Cobalt (Co), Nickel (Ni) and Gadolinium (Gd). In these experiments the superconductor was used for its distinctive quasiparticle density of state (quasiparticles are the elementary excitations of a system), which is zero for sub-gap energies and sharply peaked near the gap edge. During the same period, Tinkham and Clarke [7, 8] studied non-equilibrium superconductivity (which is an imbalance in the quasiparticle density of states) in S/N junctions. They found that the conversion of a normal (quasiparticle) current into a supercurrent leads to an imbalance in the quasiparticle spectrum inside the superconductor. The quasiparticles become distributed over the available states in a way similar to what is expected from an increase in temperature. Later it turned out that the effect of non-equilibrium superconductivity is fundamentally not very different from the effect of a ferromagnetic exchange field (or energy) on a superconductor.

The technological advancements in micro-structuring have boosted, and made it possible to investigate proximity systems on a mesoscopic scale. This means down to the scale where the various characteristic lengths of the systems (like ξ_s) are competing (or: become visible). In S/F proximity systems, superconductivity is induced into a natural hostile environment, where it is broken down by the exchange energy E_{ex} roughly over a distance ξ_F. In a normal metal superconductivity is also broken down (roughly over a distance ξ_N), but the important energy is then the "temperature" k_BT which is usually much smaller than E_{ex}, leading to $\xi_N \gg \xi_F$, and hence the Cooper pair dephasing in N is much weaker. Apart from the much smaller distance over which superconductivity survives in F, it also behaves in a rather different way. Instead of a clean monotonic decay, it oscillates (see Fig. 1.3). But the

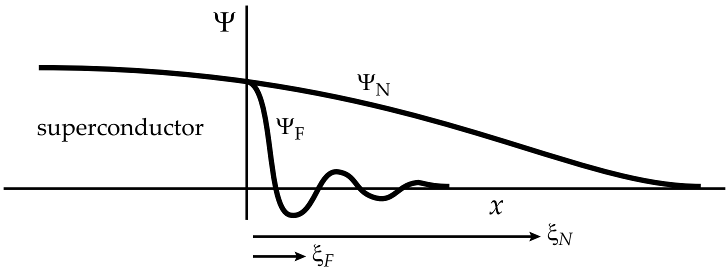

Figure 1.3: Amplitude of the (induced) superconducting wave function Ψ as function of distance x, in a normal metal Ψ_N and a ferromagnet Ψ_F, with ξ_N and ξ_F the respective coherence lengths.

interesting part does not stop with the oscillation, in fact, it is where it starts! The oscillations are not just simple amplitude oscillations. Instead, it is the nature of the pairing itself that is changing which causes this oscillation to appear. The conventional Cooper pair is in a spin singlet state, but now, under the influence of a homogeneous exchange field, spin triplet correlations appear (due to spin rotation of the conventional Cooper pairs) and the Cooper pair becomes a mixture of the two. Under the condition of inhomogeneous exchange fields, it is even possible to create all of the three spin triplet (instead of only the $m_z = 0$ component), which include the "spin equal" components ($m_z = \pm 1$). The ferromagnet has succeeded in changing a part of the normal Cooper pairs, which cannot live long in the ferromagnet, into other types of pairs for which the ferromagnet is no longer a specially hostile environment. This triplet pairing, or, more general, the effect of inhomogeneous exchange fields on the superconducting state, is currently a burning question and a serious research topic in the field of S/F proximity systems. Moreover, there is also interest in the behavior at the S-side of the interface where it is possible that ferromagnetism is induced.

1.3 Motivation & Outline

The oscillation of the Cooper pair density appears whenever the Cooper pairs experience a homogeneous exchange field. However, for the generation of the ”spin equal” triplet components the Cooper pairs need to experience a non-homogeneous exchange field. Such non-homogeneous exchange fields are found in magnetic domains and domain walls. About half of this thesis relates to this problem: what is the effect of inhomogeneous magnetism (in the form of domains and domain walls) on the superconducting state. Thus, rather than examining the induced superconductivity in the ferromagnet, we examine the changes to the superconducting state as caused by the magnetic domains. This we do in a special type of structure that gained much attention: the superconducting spin-valve. Also in such a spin-valve structure we search for traces of induced magnetism in the superconductor, and by replacing the ferromagnet for a normal metal under non-equilibrium conditions we (theoretically) examine non-equilibrium effects on the superconducting state. The latter two form the other half of this thesis.

The superconducting spin-valve consist of a superconductor sandwiched between two ferromagnetic layers (see Fig. 1.4). Calculations show that for such a device T_c is always higher if the magnetization, and therefore the exchange fields, form a anti-parallel (AP) configuration than if they form a parallel (P) configuration [9, 10]. In effect it is a organized inhomogeneous device, which can be switched from inhomogeneous (AP) to homogeneous (P). When the ferromagnetic layers are thin enough, they are also subject to the oscillatory nature of the induced superconductivity and can show full re-entrant behavior, which is the most interesting feature of such a device: the possibility to switch on and off superconductivity by a small field manipulation, and hence, controlling the supercurrent through the device. To gain control of the exchange field, soft magnetic materials are favorable since they can be switched by small external fields such to not disturb the condensate. To achieve separate switching, materials (or layers) with different switching fields can be used, or one could pin one of the layers by using an anti-ferromagnet. The latter one is the one proposed in the first proposal of the spin-valve and is most popular.

Theory describing the superconducting spin-valve (or more general, S/F proximity) is well developed for the case of weak ferromagnetism ($E_{\mathrm{ex}} \ll E_F$)

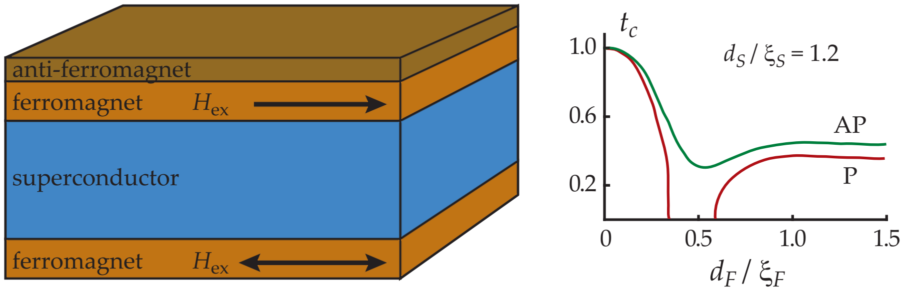

Figure 1.4: Schematic of the proposed superconducting spin-valve, where the direction of the exchange field H_{ex} of one of the layers is pinned by the anti-ferromagnet, while the other can be manipulated by a (small) external field. The calculations are taken from [9] (with interface transparency parameter $T_F = 25$) and show that the reduced transition temperature t_c in anti-parallel (AP) configuration is always higher compared to parallel (P) configuration of the ferromagnetic exchange fields. Here, d is the layer thickness, ξ the coherence length, and subscripts S and F denote the superconductor and ferromagnet.

with homogeneous exchange fields. For strong ferromagnetic materials, or for inhomogeneities in the exchange field (in particular from domains or domain walls), the framework is either intrinsically not suitable or poses severe difficulties. From the experimental side, measurements on spin-valves based on weak ferromagnets (and almost zero spin polarization) seem to coincide with the theoretical prediction, although the effects are generally weaker than what might be expected from theory. However, it is doubtful that the pre-assumed conditions of homogeneous exchange fields are always realized. This makes the interpretation of the results at least "open for discussion", as the effects of magnetic domains and domain walls might be the actual dominating mechanism for the observed effect. When using strong ferromagnets, where E_{ex} is no longer much smaller than E_F and there will be a non-zero spin polarization, contradictory looking results are obtained. Apart from differences in the sample geometry and/or used materials, dipolar fields coming from domains (or domain walls) are often mentioned as being the source for these results. As these main difficulties are all related to domains, it is only natural to examine these effects in more detail, which is the main part of this thesis. *Chapter two* describes the basics of the theoretical concepts encountered in the following chapters. *Chapter three* gives a brief description of the sample fabrication and measurements setup. *Chapter four and five* focus on the effect of domain structure in the ferromagnetic layers on the working of the superconducting spin-valve. This is examined for both the weak ferromagnetic based CuNi spin-valve (chapter four) as for the strong ferromagnetic based Py spin-valve (chapter five). *Chapter six* is a rather different type of experiment on the superconducting spin-valve, where an attempt is made to detect inverse proximity (induced magnetism in the superconductor) by the usage of muon spin rotation experiments. *Chapter seven* is a theoretical work on non-equilibrium superconductivity in a mesoscopic superconducting wire, connected to normal metallic reservoirs.