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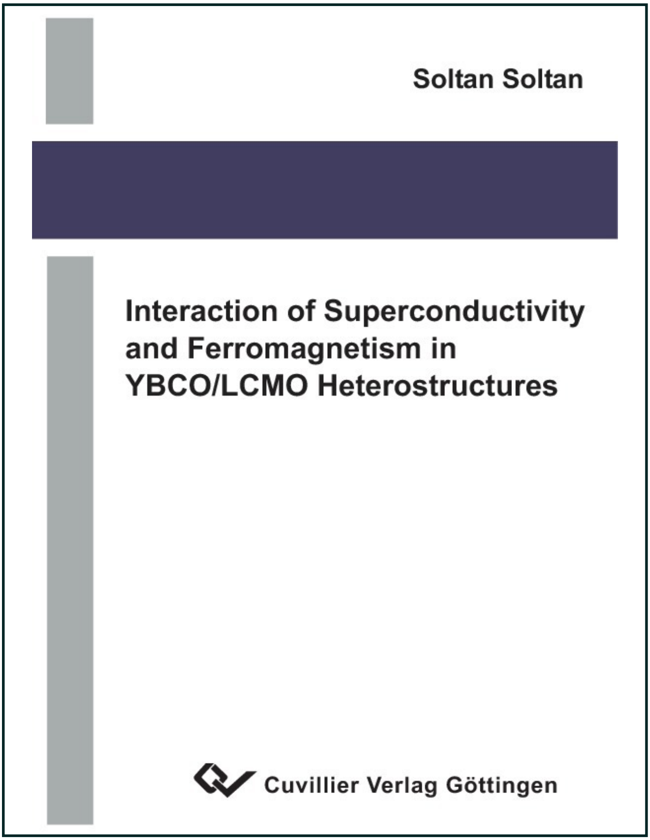
Interaction of Superconductivity and Ferromagnetism in YBCO/LCMO Heterostructures



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Chapter 1

Introduction and Motivation

1.1 History

Superconductivity was discovered by HEIKE KAMERLINGH ONNES [1] in Holland in 1911 as a result of his investigations leading to the liquefaction of helium gas. Two years later he got the Nobel prize 1913. In Onnes‘ time superconductors were simple metals like mercury, lead, bismuth etc. [1]. These elements become superconductors only at the very low temperatures of liquid helium. During the 75 years that followed, great studies were made in the understanding of how superconductors work. Over that time, various alloys were found that show superconductivity at somewhat higher temperatures. Unfortunately, none of these alloy superconductors worked at temperatures much more than 23 K. Thus, liquid helium remained the only convenient refrigerant that could be employed with these superconductors.

The transition of a normal metal into the superconducting state is revealed by the total disappearance of the electrical resistance at low temperatures. Indeed, the current in a closed superconducting circuit can circulate forever without damping.

Another fundamental property of the superconducting state was discovered in 1933 when Walther Meissner and his Ph.D. student Robert Ochsenfeld demonstrated that superconductors expel any residual magnetic field [3]. Similarly, superconductivity can be destroyed by applying a magnetic field that exceeds the critical value B_c. Superconductivity and magnetism usually try to avoid each other this feature can be exploited to, for example, levitate a magnet above a superconductor.

The recent discovery of compounds that are both ferromagnetic and superconducting at the same time came as a surprise to experimental and theoretical, condensed matter physicists.

The microscopic theory of superconductivity was created by JOHN BARDEEN,

[1] From now on, the names of researchers that were awarded the Nobel Prize will be displayed with capital letters.

LEON COOPER and ROBERT SCHRIEFFER[1] in 1957 [2]. According to this so-called *BCS–theory*, the electrons form pairs, known as Cooper-pairs , due to interactions with the crystal lattice at low temperatures. Electrons in these Cooper-pairs have opposite values of momentum, meaning that the pairs themselves generally have zero orbital angular momentum. Additionally, the angular momenta add up to zero. The formation of Cooper-pairs leads to a superconducting energy gap, which means that single electrons cannot occupy states near the Fermi surface. Such energy gaps which are essentially equal to the energy needed to break up the Cooper-pairs show up clearly as jumps in the specific heat and thermal conductivity at what is known as the critical temperature T_{c}.

Another significant theoretical advancement came in 1962 when BRIAN JOSEPHSON[2], a graduate student at Cambridge University, predicted that electrical current would flow between two superconducting materials, even when they are separated by a non-superconductor or an insulator. His prediction was later confirmed and won him a shared of the 1973 Nobel Prize in Physics with LEO ESAKI and IVAR GIAEVER. This tunnelling phenomenon is today known as the "Josephson effect" and has been applied to electronic devices such as the Superconducting Quantum Interference Device (SQUID), an instrument capabable of detecting even the weakest magnetic fields.

Then, in 1986, a truly breakthrough discovery was made in the field of superconductivity. GEORG BEDNORZ and ALEXANDER MÜLLER [3] researchers at the IBM Research Laboratory in Rüschlikon, Switzerland, created a brittle ceramic compound that showed superconductivity at the highest then temperature known 30 K. What made this discovery so remarkable was that ceramics are normally insulators. They do not conduct electricity well at all. So, researchers had not considered them as possible high-temperature superconductor candidates. The Lanthanum, Barium, Copper and Oxygen ($La_{1.85}Ba_{0.15}CuO_4$) compound that MÜLLER and BEDNORZ synthesized, behaved in a not-as-yet-understood way. The discovery of this first of the superconducting copper-oxides (cuprates) won the 2 men a Nobel Prize the following year. It was later found that tiny amounts of this material were actually superconducting at 58 K, due to a small amount of lead having been added as a calibration standard making the discovery even more noteworthy.

The BCS theory is quite successful at explaining the properties of most classical superconducting materials. But the discovery in 1986 of a new class of materials that are superconducting at high temperatures remains a challenge to the theoreticians, and there is still no unambiguous theoretical explanation for this phenomenon.

The observation of superconductivity in organic conductors, heavy-fermion systems, the ruthenates and, most recently, the new ferromagnetic superconductors provides strong

[1] Nobel prize 1972
[2] Nobel prize 1973
[3] Nobel prize 1987

arguments for the existence of more exotic types of superconductivity. Indeed, pairing in ferromagnets must result from a different type of electron-pairing. In these materials, electrons with spins of the same direction are paired up with each other to form Cooper-pairs with one unit of spin, resulting in so-called ***triplet superconductivity***. In contrast, conventional superconductivity, also known as ***s*–wave** singlet superconductivity, occurs when electrons with opposite spins bind together to form Cooper-pairs with zero momentum and spin.

A magnetic field can destroy singlet superconductivity in two ways. The first of those effects is known as the *orbital effect* and is simply a manifestation of the Lorentz force. Since the electrons in the Cooper-pair have opposite momenta, the Lorentz force acts in opposite directions and the pairs break up. The second phenomenon, known as the *paramagnetic effect*, occurs when a strong magnetic field attempts to align the spins of both the electrons along the field direction. Such fields, however, do not wreck triplet superconductivity because the spins of both electrons may point in the same direction as the field. This means that triplet superconductivity can only be destroyed by the orbital effect.

Ferromagnetism arises when a large number of atoms or electrons align their spins in the same direction. There are actually two sources of magnetism in metals, localized magnetic moments and the “sea” of conduction electrons. Local magnetism occurs in rare-earth metals (such as gadolinium) due to the incomplete filling of electrons in the inner atomic shells. This leads to a well defined magnetic moment at every fixed atomic site, which in turn produces long-range magnetic coupling due to the exchange of conduction electrons.

The second type of magnetism known as band magnetism, (such as ruthenium) arises from the magnetic moments of the conduction electrons themselves. In a metal, the electrons are “itinerant”, that is they are free to move from one atomic site to another, and they tend to align their magnetic moments in the direction of an applied field.

Ferromagnets only have a net magnetic moment at low temperatures; the internal magnetic field spontaneously appears at the so called *Curie temperature*, which is typically in the range 10–1000 K. At higher temperatures, however, the magnetic moments of the atoms continually change their direction so that the net moment is zero. A similar magnetic transition occurs in antiferromagnetic materials in which the spins of neighboring atoms point in opposite directions. This transition takes place at the *Néel temperature* and leads to the disappearance of the internal magnetic field.

Although superconductivity and magnetism seem to be antagonistic phenomena, could they co-exist in the same compound? This question was first posed by the Russian theorist VITALY GINZBURG[4] in 1957, but early experiments in 1959 by Bernd Matthias, demonstrated that a very small concentration of magnetic rare-earth impu-

[4]Nobel prize 2003

rities, even a few percent, was enough to completely destroy superconductivity when ferromagnetic ordering was present.

The origin of this destructive phenomenon is a quantum mechanical interaction between the spins of the electrons and the atomic magnetic moments. Below the superconducting transition temperature, this "exchange interaction" attempts to align the Cooper-pairs . Exchange interactions therefore place stringent limits on the existence of superconductivity.

- ***But can superconductivity and ferromagnetism co-exist?***

The answer to this question is much more fascinating. Finally, the discovery of ferromagnetism ($T_{\mathrm{Curie}} \approx$ 135 K) and superconductivity ($T_{\mathrm{c}} \approx$ 40 K) in the $RuSr_2GdCu_2O_8$ (Ru1212) compound opens a lot of questions. Such as the coupling of the ferromagnetic layers between the superconducting layers without killing the superconductivity. Also, the existence of superconductivity and ferromagnetism in the same unit cell. It was demonstrated that, superconductivity in these ferromagnet materials, could be explained by the Larkin-Ovchinnikov-Fulde-Farrell–(LOFF) theory.

When a superconductor is in contact to a normal metal , a number of phenomena occurs such as the *proximity effect*. The two materials influence each other on a spatial scale of the order of the coherence length (ξ_{sc}) in the vicinity of the interface. In particular, the correlations between quasiparticles of the superconducting state are induced into the normal metal, Cooper-pairs penetrate into the normal metal with a finite life time. Until they decay into two independent electrons, they preserve the superconducting properties. Alternatively, the proximity effect can be viewed as resulting from a fundamental process known as Andreev-reflection. Imagine a low energy electron propagating from the normal metal onto the interface with the superconductor. A single electron can penetrate the superconductor only if its energy is larger than the superconducting energy gap ($E_{\mathrm{el-nm}} > \Delta_{\mathrm{sc}}$). Below this energy only Cooper-pairs can exist. Thus low energy electrons can not penetrate into the superconductor and are reflected back as holes, while Cooper-pairs are transferred into the superconductor.

When a superconductor is in contact to a ferromagnet another important phenomenon occurs, the *inverse*–proximity effect (or the so called spin diffusion length ξ_{fm}). The exchange energy (J_{spin}) of the ferromagnet quenches the Andreev-reflections, due to the absence of available states for the reflected holes with different spin. This prevents the Cooper-pairs to diffuse deeply into the ferromagnet layer. In addition, the quasiparticles using the exchange energy as a driving force to break the Cooper-pairs in the superconductor within the inverse proximity effect scale (ξ_{fm}). ξ_{fm} can strongly depend on the type of the ferromagnet , it is rather small for classical ferromagnets, much larger in materials that show the colossal magnetoresistance (CMR) effect.

1.2 Motivation

After the more fundamental aspects of superconductivity now the particular issues are discussed that became subject of the thesis.

Although the investigation of the proximity effect in superconductor /normal metal (SN) systems was started about 40 years ago, the technology allowing to produce and measure experimental samples of mesoscopic dimensions was achieved much more recently. In particular, it became possible to study superconductor /normal metal structures consisting of thin layers (having thicknesses smaller than the coherence length). Such structures behave as a single superconductor with nontrivial properties. Many of them have already been studied for the case of ideally transparent interfaces. At the same time, the experimental progress requires the corresponding advances in theory, especially taking into account arbitrary interface transparency. This crucial parameter determines the strength of the proximity effect and at the same time is not directly measurable. From the practical point of view, the superconductor /normal metal proximity structures can be used as superconductors with relatively easily adjustable parameters, in particular, the energy gap and the critical temperature. The parameters of the proximity structures can be tuned, e.g., by varying the thicknesses of the layers. This method has already found its application in superconducting transition edge bolometers and photon detectors for astrophysics.

The physics of superconductor /ferromagnet systems is even richer. In contrast to the superconductor /normal metal case, the superconducting order parameter does not simply decay into the *non*–superconducting metal but it can also oscillate. This behavior is due to the exchange energy J_{spin} of the conduction quasiparticles . In the classical ferromagnet (J_{spin} ≈1 eV) it acts as a potential of different signs for two electrons in Cooper-pairs and leads to a finite momentum of the pair. These oscillations reveal themselves in a *non*–monotonic dependence of the critical temperature T_{c} of superconductor /ferromagnet systems as a function of the thickness of the ferromagnet layers, both in the cases of superconductor /ferromagnet superlattices and bilayers. At the same time, in most of the papers investigating this effect, the methods to calculate T_{c} were approximate.

So far, the oscillation of T_{c} in the superconductor /colossal magnetoresistance bilayers or superlattices was not found. This can be explained due to the high-exchange energy of colossal magnetoresistance materials (J_{spin} ≈3 eV). This high exchange energy J_{spin} not only quenches the oscillation of T_{c} but also the Andreev-reflection as mentioned before. This high exchange energy can also act as driving force for the quasiparticles to tunnel into the superconducting layer, called *spin-polarized* quasiparticles self injection. Although such effects have been studied before, it was often done in the simplest models and simplest assumptions about the system parameters. To achieve a better understanding of these phenomena, one should study them at various conditions and determine the physical mechanisms behind the effect.

A possible practical application of ferromagnet /superconductor /ferromagnet heterostructures uses spin-dependent properties of high-T_c superconductors that can lead to the design of new superconducting devices such as “spintronic devices”, like transistors with high gain current and high speed. “Spintronics” means the exploitation of the spins of the electrons rather than their charge. Spin controlled solid state devices based on the giant magnetoresistance (GMR) effect are already realized in read-out heads of hard disks. Further challenges in the field of spintronics that are addressed by experiments and theory include the optimization of electron–spin life times and the detection of the spin coherence length in nanoscale structures. Although the superconducting spintronics are not yet experimentally realized, the work in this direction has already started. Using the classical ferromagnet as source for spin–polarized quasiparticle injection into *s*-wave superconductors in the superconducting state was started more than 30 years ago. To achieve better understanding of this phenomenon, colossal magnetoresistance electrodes with full spin polarization are used to inject spin-polarized quasiparticles injection into *d*-wave superconductor . This experiment is now done under various conditions to study the physical mechanisms both in the superconducting and in the normal state of the superconductor.

For a complete description of these heterostructures also the physics of the flux line lattice in the superconductor has to be considered. High-temperature superconductors are extreme type-II superconductors containing Abrikosov flux lines in a large range of applied fields between B_{c1}= 0.01 T and $B_{c2} > 100$ T. Apart from their high-transition temperatures of T_c = 90 K to 133 K, HTSC differ from conventional superconductors by their short coherence length ξ_{sc}, a large magnetic penetration depth λ, a pronounced material anisotropy and a layered structure. These four properties drastically enhance the thermally activated depinning of flux lines. Small ξ_{sc} reduces the pinning energy. Large λ softens the flux-line lattice. The layered structure of HTSC causes fascinating novel phenomena, a flux line is now a string of 2D “pancake vortices” in the superconducting CuO_2 layers. In case of heterostructures of HTSC and ferromagnets now an additional interaction between the flux line lattice and the domain structure of the ferromagnet has to be taken into account. This will lead to new effects that are also discussed in this thesis.

1.3 Outline of the thesis

In Chapter (2) a comprehensive presentation of the theoretical background used in the work will be given. This addresses topics such as Ginzburg-Landau theory (GL-theory) and BCS–theory for classical superconductors , also the theoretical approximations that can be used to deal with high–T_c materials. Especially the role of flux line pinning in presence of ferromagnetic layer is discussed.

In Chapter (3) a comprehensive presentation of the theoretical background for colossal magnetoresistance is given.

A brief summary of the fundamentals of proximity–effect and spin–diffusion length estimation in normal-metal/superconductor and ferromanet/superconductor bilayers will also be included in Chapter (4).

- As already mentioned, several experimental techniques are applied in this work. Their underlying theory and the experimental apparatuses will be described in Chapter (5). More specifically, the instrumentation and theory of magneto–optical (MOP) Faraday effect will be explained.

- The structural analysis of all of the heterostructures is the main part of Chapter (6). The roughness of layers after the growth is analyzed by atomic–force–microscopy (AFM). The results allow statements of the growth mechanisms. Detailed informations about the lattice parameters and oxygen stoichiometry are found in x-ray diffraction investigation. One important issue is the morphology of the interface. This addressed by high–resolution transmission electron microscopy.

- Spin–diffusion length determination will be one of the topics of Chapter (7), Section (7.1), where two series of experimental data on CMR/HTSC bilayers grown on two different single crystalline substrates STO and LSGO will be presented and analyzed with the help of the theory described in Chapter (2). The role of the diffusion of the spin–polarized quasiparticles will allow to extract some interesting relations between the temperature dependence of the quasiparticle density n_{qp}(T) and the order parameter band gap in the cuprates. Additionally, the spin diffusion length ξ_{fm} is derived from the results.

- Section (7.2) concerns injection of spin–polarized quasiparticles (SPQI) in the normal and superconducting state of YBCO. As mentioned in the previous section, the spin–polarized quasiparticles of the magnetic layer can be self–injected into a superconducting layer. In case of an injection the quasiparticles have additional energy to penetrate into the YBCO layer. This method can be used to measure the effect of a polarization enhancement in the CuO_2–plane around the so called pseudogap temperature T^* in the normal state of the superconducting layer. Also, the effects of SPQI in the superconducting state will be shown. As counter experiment several junctions will be shown using different sources for quasiparticle injection. Non–polarized and lower–polarized quasiparticles will be injected into the YBCO in the normal and superconducting states.

- In Section (7.3) the critical current density j_{c} in superconductor /ferromagnet heterostructures is investigated. From quantitative magneto–optical measurements and SQUID magnetization data it is found that the critical current density in a superconducting film is strongly affected by the presence of the ferromagnet . First,