Ph.D. Thesis booklet

Local probing of electronic transport with point contact Andreev reflection measurements

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Introduction

The new era of nanoelectronics relies on physical processes observable only on the nanometer scale. This approach has led to many novel schemes, which can eventually replace today's semiconductor-based computer architectures.

A widely investigated field of nanoelectronics is *spintronics*, which relies on the coherent propagation of the electronic spin. The spin valve has been a prototype model system of spintronics since the discovery of the giant magnetoresistance (GMR) effect in ferromagnetic-nonmagnetic-ferromagnetic layered systems by A. Fert and P. Grünberg in 1988 [Baibich88, Binasch89]. The figure of merit of such devices, nowadays widely adopted for digital data storage, is strongly influenced by the decay of the spin information in the nonmagnetic spacer layer. Hence, the spin relaxation processes in metallic systems are subject of ongoing research.

Another intriguing concept is the utilization of the permanent influence of charge current on certain materials. The study of *memristors* (memory resistors) started with the theoretical proposal of L. Chua in 1971 [Chua71], however the first unambiguous experimental demonstration lacked until 2008 [Strukov08]. The framework of memristive system has been successfully applied to describe a wide variety of material systems. Among these systems, Ag_2S-based cells have widely been investigated [Terabe05] and the memristive behavior is attributed to the formation and retraction of metallic Ag filaments inside the Ag_2S layer. Recently, *ab initio* calculations revealed that very thin ($\lesssim 10\,\mathrm{nm}$) films of Ag_2S enclosed between bulk Ag electrodes undergo a structural transition resulting in metallic conductivity in contrast to the bulk semiconducting behavior [Wang07]. This regime, previously not investigated experimentally in the literature, could provide faster memory cells operating in a wider temperature range.

To gain insight of the electronic transport properties in nanoelectronic devices, superconductivity can be utilized as a local probe based on a special charge conversion process, first described by A. F. Andreev in 1964 [Andreev64]. This process, called the Andreev reflection, is the simultaneous transmission of two charge quanta, which can be expressed as a reflected hole-like quasiparticle in place of the incoming electron. Andreev reflection has become an important tool of nanophysics because of the spin constraint between the incoming electron and the reflected hole. That is, in order to have a spin singlet Cooper pair condense at the superconductor side, the reflected hole has to possess opposite spin orientation as the incoming electron. Based on this property, the degree of spin ordering can be deduced in the normal metallic side of the junction, as it was demonstrated by Soulen et al. in 1998 [Soulen Jr.98].

Objectives

During my Ph.D. work at Department of Physics of the Budapest University of Technology and Economics, my research was focused on the utilization of Andreev reflection for the detection of local spin polarization and the investigation of the memristive material systems.

Reliable determination of the spin polarization demands careful characterization of the properties of the SN point contact. Investigating the role of the size of the junction was an important objective of my Ph.D. work, as larger junctions in the diffusive limit can exhibit fundamentally different phenomena than smaller, ballistic contacts. In order to further test if experimental data provides proper values for the spin polarization, various theoretical models have been applied.

The mesoscopic interference effects arising for diffusive junctions are of considerable interest in the field of nanophysics. I have investigated the contact size dependence of such effects in order to demonstrate the role of the diffusive transport corrections in the corrections for the differential conductance near zero bias.

Another basic length scale is the spin diffusion length, l_s, over which the spin direction of an electron is preserved. Directly investigating the decay of the local spin polarization on top of a nonmagnetic layer deposited on a ferromagnetic metal is possible via Andreev reflection spectroscopy and could provide insight to the physical processes governing the spin relaxation.

The detailed investigation of the switching behavior of Ag_2S-based memory cells in a point contact geometry is an intriguing method to gain knowledge of the details of the resistive switching processes. Characterizing many individual memory cells provides the possibility to statistically analyse the switching properties, and reliability of such devices. Furthermore, by approaching the ultimate limit of a single atomic junction, the interplay of the quantized conductance of individual atoms could be observed. By utilizing Andreev reflection, one can distinguish between the change of the number of the conducting channels and the average transmission for each resistive transition. Detailed evaluation of such data could ease further understanding the resistive switching phenomenon in these devices.

Experimental techniques

The experimental setup, employing the tip-sample approach, has been exclusively developed at the Department of Physics of the Budapest University of Technology and Economics. The careful mechanical and electronic design and construction were essential to obtain the experimental results discussed in my thesis, therefore setting up the measurement system was an essential part of my Ph.D. work.

The sophisticated mechanical design utilizing a twofold actuation scheme made it possible to investigate nanometer scale junctions. The low electronic noise level – characterized by superconductor-insulator-superconductor (SIS) tunneling – is essential for observing the nonlinearities in the $I - V$ curves of superconductor – normal metal (SN) heterojunctions. In addition, utilizing a high bandwidth circuitry, the response of the junction to bias pulses as short as 10 ns was investigated for testing novel nanoscale solid state memory concepts. Most of the measurements were performed at cryogenic temperatures down to 1.5 K, however, occasionally room temperature experiments were made as well.

The dilute magnetic semiconductor compounds (In,Mn)Sb and (In,Be)Sb have been supplied by the group of prof. J. K. Furdyna at the Department of Physics, University of Notre Dame. The metallic thin film samples have been prepared by Dr. F. Tanczikó in the KFKI Research Institute for Particle and Nuclear Physics. The RBS/ERDA analysis in conjunction with my point contact measurements on the Ag_2S-covered samples has been performed by Dr. E. Szilágyi at KFKI Research Institute for Particle and Nuclear Physics.

Summary

The main conclusions of my Ph.D. work are summarized in the following thesis points:

1. I have developed a low temperature point contact measurement system utilizing the tip-sample approach. This work included the designing of the sample holder, as well as assembling different instrumental setups, suitable to measure the non-linear $I - V$ curves of nanoscale junctions, and to investigate memory effects under very short voltage pulses, down to the 10 ns time scale. The measurement system was carefully checked for mechanical stability and was found to properly stabilize point contacts with diameters ranging in the $1 \ldots 100$ nanometer scale. I have shown, that the nonlinearities of the $I - V$ curves can be investigated with a voltage resolution down to $250\,\mu$V, which is required to observe features appearing in superconductor – normal metal (SN) junctions. I have investigated the spin polarization of the current, P_c, in various material systems. I have evaluated measurement data both using the modified Blonder–Tinkham–Klapwijk (BTK) theory and the Hamiltonian transport model and have shown that the obtained spin polarization values agree each other within the uncertainties of the fitting procedure. Specifically, I obtained $P_c = 0.60 \pm 0.1$ for the dilute magnetic semiconductor (In,Mn)Sb, $P_c = 0.65 \pm 0.05$ for Fe and $P_c = 0.41 \pm 0.04$ for Co. The Fe and Co films were covered with a thin nonmagnetic metal layer, hence – in contrast to earlier studies – my measurements were not influenced by spurious surface scatterings [1,2].

2. I have investigated the influence of the junction diameter, d, on the electronic transport in superconductor – normal metal heterojunctions. Detailed experiments on the dilute magnetic semiconductor (In,Mn)Sb and its nonmagnetic counterpart (In,Be)Sb have revealed the importance of utilizing point contacts in the ballistic regime ($d \lesssim 15$ nm) for reliable analysis of the spin polarization. For diffusive junctions ($d \gtrsim 15$ nm), however, the larger junction diameter gives rise to mesocopic interference phenomena resulting in a characteristic zero bias feature in the differential conductance. I have obtained a $d \approx 15$ nm crossover diameter for this regime which is in good agreement with the mean free path in the (In,Mn)Sb, and (In,Be)Sb samples. In addition, for the (In,Be)Sb, I have found a transition to another regime characterized by a zero bias peak sharper than the thermal smearing k_BT and higher than the principal limit of $G(V = 0) = 2G_N$

of SN junctions. This feature resembles to Josephson current between two superconductors and I have related it to proximity induced superconductivity in the normal metallic lead [1,3].

3. Adopting a geometry similar to that of typical spin valve experiments, where the current flows perpendicularly to the layers, I have investigated the spin relaxation through nonmagnetic Pt and Au layers. By measuring the decay of the spin polarization on top of the thin nonmagnetic layer deposited on a ferromagnetic metal with increasing thickness, the spin diffusion length could be determined. My measurements on Fe/Au bilayers demonstrated the expected exponential decay of the spin polarization as a function of the Au layer thickness, yielding a $l_s = 53\,\mathrm{nm} \pm 6\,\mathrm{nm}$ at $T = 4.2\,\mathrm{K}$. In addition, I have performed detailed studies to determine the temperature dependence of the spin diffusion length in Pt deposited on Co. Above $T \approx 3.5\,\mathrm{K}$, a power law dependence $l_s \propto T^{-2.6}$ and a saturation in the low temperature limit at $l_s = 67\,\mathrm{nm} \pm 5\,\mathrm{nm}$ was obtained. These findings are consistent with the predictions of the Elliott-Yafet law, which attributes the spin relaxation to the momentum scatterings mediated by the spin-orbit coupling. Based on the temperature dependence of l_s, I have separated two contributions to the spin relaxation, the electron-phonon coupling and the temperature-independent background of boundary and impurity scatterings [2].

4. I have investigated the memristive memory cells based on the Ag_2S mixed ionic and electronic compound. Thin Ag_2S layers were prepared on a silver thin film by exposing it to sulfur atmosphere at an elevated $T = 60\,°\mathrm{C}$ temperature. By characterizing the samples in the point contact geometry, I have demonstrated the impact of the sulfurization time on the electronic behavior. I have separated semiconducting and metallic samples, the latter exhibiting resistive switching and cryogenic temperatures as well. My results define a lower size limit of 3 nm for the reproducible switching behavior, below which a new regime emerges governed by stochastic atomic migration [4,5].

5. I have utilized Andreev reflection to characterize the average transmission and the areal contribution of the conducting channels inside the Ag_2S thin layer. My analysis within the Landauer formalism have demonstrated that the dominant effect is formation and retraction of conducting channels. The obtained average transmission values being in the $T = 0.4 \ldots 0.7$ range is the first experimental confirmation of earlier *ab initio* calculations for thin Ag_2S layers. In order to directly demonstrate the nonvolatile memory operation of the junctions, I applied narrow bias pulses on the cell after which the permanent change in resistance was evaluated. My results are the first experimental confirmation of the memory operation of this material system with pulse widths as narrow as 10 ns, even down to cryogenic temperatures [these results have not been published yet].

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Miscellaneous publications

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