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Chapter 1

Introduction

The first descriptions of magnets, around 2500 years ago, show that they were discovered when a magnet attracted a piece of metal [1]. Now, we know that the magnetic field lines (Ørsted field) cause an interaction with the spins in the metal. This interaction is determined by the characteristics of both materials and the resulting force allows to make use of them, enabling a few of the greatest technological revolutions in history:

The first revolution aided by magnetism was performed over two millennia ago by simply making a thin needle out of ferromagnetic (FM) material, in which a positive exchange interaction between the magnetic moments aligns their direction parallel to each other. A rotation degree of freedom allows the magnetization to point towards the magnetic North of the earth by reducing the energy via the reduced field lines. Subsequently, the needle acts as the compass as we know it and travelling became a much less hazardous undertaking, a major step towards globalization.

The second revolution took place in West-Europe by the coupling of magnetism and electricity as described by Ørsted [2] [3] right after the initial industrial revolution. This led to the Maxwell equations and turned coal powered heat into mechanical energy via steam resulting in the electrification and further industrialization.

A more recent revolution in which magnets were involved took place in the information technology. Data can be stored by adjusting the magnetization direction of a magnetic layer that can be read out later [4]. An optimized version of this technique is the giant magnetoresistance (GMR) [5, 6]. Here, the resistance of a metallic magnetic multilayer depends on the relative directions of magnetization in two FM layers. The magnetization of one of the FM layers is easily adjustable and thereby information can be stored in the system. The second FM layer, however, is more difficult to change as it experiences magnetic pinning by an antiferromagnetic (AFM) layer. The AFM is a class of magnets that behaves differently than the FM in the sense that it does not produce any Ørsted field. This was exactly the characteristic needed in GMR as it allows to pin the direction of one of these FM layers irrespectively of the applied magnetic field. The pinning occurs from the exchange bias which is the direct interaction of adjacent magnetic moments at the interface. The lack of spontaneous magnetization makes these AFMs having little interaction with external probes, resulting in their ’stubborn’ nature.

Although AFMs do not repel or attract a piece of metal and neither one could switch its magnetization nor polarize electron spins, this stubborn material was known to posses some magnetic properties. A magnetic field could induce some magnetization in AFMs depending on the strength and direction of the magnetic field [7]. A consequence is that these systems cannot be described by an unexcited macrospin model nor validated by the magnetization. It was Louis Néel who first came up with an explanation for this behaviour in 1937 [8]. He reasoned the existance of two interacting sublattices which would cancel out their Ørsted field by having the two sublattice polarizations in opposite direction with respect to each other at the lowest energetic state. In the previous years, Néel tried to prove the hypothesis that the sign of the exchange interaction, for which the positive and negative exchange interaction results in either parallel or antiparallel alignment of the magnetic moments respectively, depends on the distance between the respective ions [9]. The idea originated from successful research on a local molecular field acting in ferrimagnets resulting in multiple sublattices, albeit with a net magnetic field. It turned out to be an oversimplified representation as negative interactions usually result from superexchange as proposed by Kramer [10]. This framework of theory allowed to interpret the small susceptibility response, identifying the AFM ordering [11]. In the light of applications, AFMs have been assessed in the past as ’interesting but useless’ since its AFM order makes it difficult to interact with it [12]. For his contribution to the field of AFM, Néel was awarded the Nobel prize in 1970.

Halfway the 20^{th} century, however, scientists proved the possibility to interact with magnets with alternating magnetic fields [7, 13] and neutron scattering [14]. AFMs responded to these field, showing that there were excitations in the magnetic lattices, or magnons, which carry spin, explained by the two-sublattice theory of Néel [11] and Keffer and Kittel [15]. Further, AFMs responded to strain by domain movement. The different magnetic domains can be imaged with techniques such as inelastic neutron scattering [16, 17], second harmonic generation [18] and birefringent effects [19]. Nonetheless, the magnetic order of AFMs remained difficult to be read out and this did not result in any active applications other than its non-active role in GMR. An active role of an AFM layer could replace the current electronic systems by spintronics. Instead of transporting electrons, one would be transporting spins.

With the discovery of the possibility to interact with a magnetic sublattice in FMs by making use of the adjacent paramagnetic heavy metal Pt [20–22], AFMs rapidly became of major interest by the spintronic community. In the Pt, a spin current is induced by the spin Hall effect (SHE) [23], creating a spin accumulation which interacts with the magnet. In combination with the so-called inverse SHE (ISHE) [24] this technique is called spin Hall magnetoresistance (SMR) which is quadratic in magnetization. Now, there was room for AFMs to come into play, allowing interaction with their magnetic sublattices. However, these claims were also met with scepticism, with same arguments as before; one would not be able to interact with the AFM as the length scale of the interaction was not small enough to enable the interaction with each sublattice individually.

1.1 Motivation and outline

AFMs are candidates for an active role in spintronics. The lack of an Ørsted field diminishes interaction between magnetic device components, allows for their miniaturization and can thereby result in a device density of a factor 100 larger than established with FM devices [25]. Furthermore, the eigenfrequencies are typically in the order of THz [13, 14], about 3 orders of magnitude higher than the eigenfrequencies of FMs. This allows for high information processing speeds of possible AFM devices.

AFMs are crystalline materials whose basic unit cells (the atomic arrangements that repeats periodically to form a crystal) consist of at least two sublattices whose magnetization vectors (caused by the unpaired electron spins of the atoms present) are equal in magnitude but of opposite direction to each other, therefore giving rise to vanishing net magnetization.

Before chapters in this work were pubblished, SMR, the method to interact with and read out the magnetic order, had been employed on $SrMnO_3$ [26]. No AFM order was observed in this article. This raised the question whether the length scale of the interaction of the electron spin in the Pt with the magnetic moments is greater than the magnetic unit cell. If that is be the case, it is not expected that the electron interacts with individual magnetic moments. Later, the SMR technique has been employed on AFM|FM bilayers [27–29]. For certain thicknesses and temperatures, the signal shifted by 90°. As Ref. [27] points out, it was not fully clear how the spin transfer torque could be exerted on the FM, but Ref. [28] attributed it to the difference in magnetic moment directions in the AFM and FM materials. SMR on Cu_2OSeO_3, showed that individual magnetic lattices could be read out [30], which opened op the possibility to investigate AFMs with SMR.

The ability to interact solely with AFM order might lead to further understanding of AFM systems. SMR would allow us to track the average rotation of all magnetic moments in the system as a function of parameters such as the magnetic field and the temperature. This will reveal characteristics such as the magnetic anisotropy and exchange interactions. It might even allow AFMs to play a more active role as a reading and writing device by detecting or changing the direction of both of these sublattices in future studies.

Magnons are the magnetic excitation quasiparticle representation of the magnetic sublattices. The magnon modes of AFMs are typically degenerate and posses opposite spins, cancelling out in the absence of (weak) magnetization. When all the magnon modes are degenerate, magnon spin currents would vanish in AFMs [31]. However, a magnetic field can bring an imbalance in the magnon mode populations. With this imbalance, a temperature gradient results in finite spin currents. This could be one step towards spintronics, ideally to go to a full spin system without conversion of spin to charge current.

The goal of this thesis is therefore to measure spin currents through AFMs. The application of the ISHE in thin heavy metal films opens up the opportunity to investigate magnetic properties of AFMs. Firstly, this allows to monitor exchange interactions, magnetic anisotropies and to distinguish FM from AFM responses or even both responses within one sample. Secondly, spin currents are sent through the sample in non-local geometries, studying the long distance spin transport through AFMs.

This thesis is organized as follows:

- *Chapter 2 provides an overview of the theoretical background necessary for understanding AFM spintronics. The knowledge of various concepts and techniques are the starting point for the studied effects in this thesis. Some of the discussed techniques are the SMR, spin Seebeck effect SSE and electrical injection and detection of magnons. It further covers the origins of AFM, how to model the equilibrium state and the characteristics of the excited states, or magnons.*

- *Chapter 3 reveals the experimental methods employed in the thesis. For the creation of spin currents, thin films of Pt are deposited on the various studied magnets. The required device fabrication techniques, including polishing and the different steps used for patterning the device structures by electron-beam lithography, followed by the description of the used deposition techniques: electron-beam evaporation and sputtering* *are described. Further, some basics regarding the measurement setup and the harmonics of the obtained signals are explained.*

- *Chapter 4 discusses the observation of resistance changes in a Pt$|$AFM bilayer under influence of a magnetic field. Changes in the Néel vector in the bulk easy-plane AFM NiO are responsible for this observed SMR. The size of the signal reveals the role of magnetic anisotropy and moving domain walls in bulk NiO. The temperature dependence of the signal follows the size of the magnetic order, influenced by magnetic fluctuations.*

- *Chapter 5 explores the complex AFM* $DyFeO_3$*. The magnet has a wide range of appealing characteristics such as two different magnetic ions with large magnetic moments, a weak ferromagnetic moment, spin-reorientation transitions, strong magnetostriction and multiferroicity including a large linear magnetoelectric effect. Further, the Dy magnetic moments with large anisotropy and a large magnetic moment influence both SMR and SSE of the material directly and indirectly via the exchange interaction with the four AFM Fe sublattices. Although the AFM Fe sublattices govern the shape and the sign of the SMR, the Dy determines the magnitude of the signal. At low temperatures, the SSE field dependence originates from their interaction.*

- *Chapter 6 explores long distance transport of spin currents using a non-local geometry (with two Pt strips acting as injector and detector) on the previously studied NiO. SSE signals are obtained at low temperatures and its magnetic field dependence is modelled considering the magnon density of various magnon modes and the influence of the field on their dispersion. This captures the splitting of the magnon modes by the magnetic field and highlights the importance of dipole-dipole interactions and cubic magnetic anisotropy at low temperatures.*

- *Chapter 7 further investigates the non-local geometry on various thicknesses of thin NiO films, grown on conventional ferrimagnetic* $Y_3Fe_5O_12$ *(YIG). The NiO is capable of electrical and thermal spin transport, which is observed both in the local and the non-local geometry. The transmissivity of NiO for all spin transport declines below the NiO Néel temperature. However, the SSE signal shows a remarkable upturn at low temperatures. This behaviour is also obtained non-locally similar to the observed SSE in bulk NiO discussed in chapter 6. Moreover, the thermal signal does not show a dependence on the NiO thickness as opposed to the electrically injected magnons which are damped out at these temperatures. This shows that the low temperature signals originate from the NiO thin film.*

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