Chapter 2

Magnetic excitations and electron scattering

This chapter is divided into two parts. In the first section, a short introduction to different kinds of interactions between magnetic moments in solids is given. These interactions govern the statics as well as the dynamics of magnetic systems. The dynamics is determined by different magnetic excitations. The theoretical description of these will focus on the collective excitations, i.e. spin waves, since these are the subject of this work. The second part of the chapter gives an overview of different existing experimental techniques that allow the investigation of magnetic excitations and a summary of the main results obtained from these studies. The chapter ends with a brief introduction into electron scattering.

2.1 Interactions of spins in solids

In this thesis, we concentrate our study on Co, a 3d-metal in which the orbital contributions to the magnetic moments are strongly quenched by the crystalline field [16]. Thus, the magnetic moments are dominated by the contribution of the electron spin. Therefore, in the following we consider the spin moment, only.

A spin has a projection of its magnetic moment along one direction (chosen as z) of size $\vec{\mu}_S = -g\mu_B S_z$, with $g$ the gyromagnetic ratio, $\mu_B$ the Bohr magneton, and $S_z = \pm \frac{1}{2}$. The magnetic moment feels an external magnetic field $\vec{B}$ via the Zeeman term $H_Z = g\mu_B S_z \vec{B}$. Since a spin itself produces a dipolar magnetic field, two spins couple by a dipole-dipole interaction. These interactions range over long distances, but they are relatively weak. In typical cases, they lead to magnetic order at temperatures below 1 K [17]. Since magnetic order is also observed at much higher temperatures, a stronger interaction between spins has to be present. It was discovered by Heisenberg that the strong interaction between spins is caused by the exchange interaction, which is of quantum mechanical origin [18]. The exchange interaction is a consequence of the Pauli exclusion principle and the fact that electrons are indistinguishable. The combination of these two principles leads to the
necessity that the total wave function of a two electron system has to be antisymmetric under the exchange of the two electrons. Therefore, the symmetry of the wave function in spin space effects the real space and vise versa. Since electrons are charged particles, it might be energetically more favorable to have an antisymmetric wave function in real space. The Coulomb interaction then favors a parallel alignment of the spins. Because the driving force is the strong electrostatic force rather than a weak direct spin-spin interaction, the exchange interaction can lead to relatively high ordering temperatures. The exchange interaction is based on the possibility that two electrons can physically exchange, which requires an overlap of the electron wave functions. Therefore, though the exchange interaction is strong, it is of short range. In a localized moment picture, it can be described by the Heisenberg Hamiltonian:

\[
H = -\sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j.
\]  

(2.1)

Here, \( J_{ij} \) is the exchange coupling constant between the spins \( \vec{S}_i \) and \( \vec{S}_j \). The definition is such that ferromagnetic order is supported for \( J_{ij} > 0 \). This model has the advantage that it explains all kinds of magnetic order in a natural way. It is successfully applied in several magnetic systems for example in rare earth elements. Due to the localized moment description it is, however, at best a crude approximation in the case of magnetic 3d-metals. We come back to this point in section 2.2.2.

Another interaction is the spin-orbit interaction. It leads to a coupling of the spin moment to the crystal lattice. The interaction is a relativistic effect and typically small in 3d-metals. Nevertheless, it gives rise to for example the magnetocrystalline anisotropy, so that magnetic moments in a ferromagnet prefer to arrange in certain crystallographic directions.

## 2.2 Theoretical description of magnetic excitations

Magnetic excitations are either of single particle or collective character. In the localized moment picture, a flip of the direction of a single spin costs the full exchange energy for each surrounding neighbor, as can be seen from equation 2.1. As will be shown later, collective excitations can exist at significantly lower energies. Therefore, these spin waves play an important role for example for the thermodynamic properties of magnets.

Spin waves are quantized. In literature a quantum is often called magnon, however, throughout this work the term spin wave will be used. Spin waves are quasi-particles which carry a wave vector \( \vec{q} \), an energy \( E \) and a magnetic moment of \( 1g\mu_B \). In a simple picture, a spin wave can be considered as a quantum of spin reversal spread coherently over the whole crystal [19]. This can be illustrated in a classical, localized moment picture as sketched in Fig. 2.1. Each spin is slightly canted out of its equilibrium position and precesses around this position. Neighboring spins have a fixed phase relation in the precession, which is determined by the wave vector of the spin wave. Spin waves have a characteristic dispersion, which links their energy to their wave vector. Depending
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Figure 2.1: Side and top view of a spin wave in a simple classical picture. Each spin is slightly tilted out of its equilibrium position and rotates around this position. The wave vector of the spin wave determines the phase relation between neighboring spins. This leads to the wave like modulation visible in the top view.

on the wave vector, different magnetic interactions determine the spin wave energy. As discussed above, the exchange interaction is typically much stronger than other magnetic interactions. Nevertheless, due to its relatively short range other interactions, like the long ranged dipole-interaction, can become important at sufficiently small wave vectors. As a rule of thumb, in 3d-ferromagnets one is in this so called dipole regime for wave vectors below \( q < 10^{-3}\ \text{Å}^{-1} \) [19]. For wave vectors above \( q > 10^{-2}\ \text{Å}^{-1} \), the exchange interaction can be safely assumed to be the only interaction that determines the spin wave energy [19].

For the different regimes, different theoretical approaches are used to describe spin waves. For the dipole or small wave vector regime, a macroscopic continuum model description is sufficient [17]. Due to the macroscopic quantities involved, the microscopic details of the system under investigation are rather irrelevant. Contrary to this, a true microscopical description is needed in the exchange dominated regime, where the wavelength can become comparable to the atomic distances in a crystal. As will be shown later, the spin waves investigated in this work are solely exchange dominated. The following discussion thus concentrates on theoretical descriptions of this regime. Two different approaches have been developed: The Heisenberg model, in which localized magnetic moments are assumed and an itinerant electron model, where the moments are carried by delocalized electrons. These are the two extreme cases since in the former model the electrons are localized in real space, while in the latter they are localized in reciprocal space. Measurements of high wave vector spin waves provide the possibility to test the accuracy of the theoretical models, since these spin waves are sensitive to details of the...
underlying description [20].

2.2.1 Spin waves within the Heisenberg description

For the moment, we stay in the localized magnetic moment description. This picture is not expected to be truly valid for the mobile conduction electrons which carry the magnetic moments in Co, however, it provides a simple description of the underlying physical properties. We will use the Heisenberg model later several times, for example to calculate spin wave dispersions. Therefore, an example for the derivation of the spin wave dispersion within a nearest neighbor Heisenberg model is given in the following.

It was found by Bloch that the Heisenberg Hamiltonian (equation 2.1) allows low energy collective excitations of spins [21, 22]. Considering these collective excitations Bloch could explain the well known $T^3$-law of the variation of the magnetization at low temperatures. A quantum mechanical description of spin waves within the Heisenberg model can be found for example in Ref. [23]. It is, however, rather technical and a classical solution of the problem yields exactly the same result. In the following, we therefore stay within the classical description. To simplify the calculations, we consider only nearest neighbor interactions and an exchange coupling constant that is the same in the entire system. Thus, the sum in equation 2.1 runs only over nearest neighbors and $J_{ij} = J$. For a magnetic moment $\vec{\mu}_i$ at position $i$ one can write the exchange interaction of each of its neighbor $j$ as an effective magnetic field $\vec{B}_{ij}$ of the size

$$\vec{B}_{ij} = -\frac{2J}{g\mu_B} \vec{S}_j$$

acting on $\vec{\mu}_i$ [16]. The sum of the torques $\sum_j \vec{\mu}_i \times \vec{B}_{ij}$ defines the rate of change of the angular momentum $\hbar \vec{S}_i$:

$$\hbar \frac{d\vec{S}_i}{dt} = -g\mu_B \sum_j \vec{S}_i \times \vec{B}_i = 2J \sum_j \vec{S}_i \times \vec{S}_j$$

(2.3)

and thus for each component:

$$\hbar \frac{dS^x_i}{dt} = 2J \sum_j [S^y_i S^z_j - S^z_i S^y_j]$$

(2.4)

and cyclic permutations. We define the direction of the $z$-axis as parallel to the magnetization axis and allow the magnetic moments to deviate only little from this direction. Therefore, $S^x \approx S$ and $S^y, S^z \ll S$, so that the products of $S^x$ and $S^y$ can be neglected, then

$$\hbar \frac{dS^x_i}{dt} = 2JS \sum_j [S^y_i - S^y_j]$$

(2.5)

1The factor of two results from the fact that the sum in equation 2.1 runs twice over the same pair of spins.
\begin{equation}
\frac{\hbar dS_i^y}{dt} = 2JS \sum_j [S_j^x - S_i^x]. \tag{2.6}
\end{equation}

The two equations can be decoupled by choosing \( S^+ = S^x + iS^y \) and \( S^- = S^x - iS^y \). Since both yield the same result, except a time reversal, we treat only \( S^+ \) and obtain

\begin{equation}
i\hbar \frac{dS_i^+}{dt} = 2JS \sum_j [S_j^+ - S_i^+]. \tag{2.7}
\end{equation}

We are later interested in the solution of equation 2.7 in terms of surface spin waves. To derive the dispersion of these spin waves we introduce a surface and make the ansatz

\( S_i^+ = A_i e^{i(\vec{Q}_\parallel \vec{R}_i - \omega t)} \). Here, \( A_i \) is the amplitude of the spin wave at position \( \vec{R}_i \), \( \vec{Q}_\parallel \) is the wave vector parallel to a surface, and \( \omega \) is the angular frequency of the spin wave. Inserting \( S^+ \) into equation 2.7 and dividing the result by \( e^{i(\vec{Q}_\parallel \vec{R}_i - \omega t)} \) we obtain

\begin{equation}
\hbar \omega A_i = 2JS \sum_j [A_i - A_j e^{i(\vec{Q}_\parallel(\vec{R}_j - \vec{R}_i))}]. \tag{2.8}
\end{equation}

Equation 2.8 can be used as a starting point to derive the spin wave dispersion in an arbitrary crystalline structure. Later we will investigate spin waves in thin fcc Co-films. Thus, as a case example, we calculate the spin wave dispersion for a semi-infinite fcc crystal with a (001)-surface. In this crystal, 12 nearest neighbors exist in the bulk at positions \( \left\langle \frac{a_0}{2}, \frac{a_0}{2}, 0 \right\rangle \), where \( a_0 \) is the lattice constant (here, \( a_0 = 3.61 \text{ Å}^{-1} \)). At the surface 4 neighbors are missing. \( \vec{Q}_\parallel \) is chosen to be along the [110]-direction so that \( |\vec{Q}_\parallel| = \sqrt{2}q_x = \sqrt{2}q_y \). The amplitude of the spin waves is constant in each layer and differs only between the layers. Thus, we introduce a layer index \( n \), where \( n = 1 \) stands for the surface layer. In this case, we derive from equation 2.8

\begin{equation}
n = 1 : \hbar \omega A_1 = 2JS[8A_1 - 4A_1 \cos^2(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}}) - 4A_2 \cos(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}})], \tag{2.9}
\end{equation}

\begin{equation}
n > 1 : \hbar \omega A_n = 2JS[12A_n - 4A_n \cos^2(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}}) - 4A_{n-1} \cos(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}}) - 4A_{n+1} \cos(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}})]. \tag{2.10}
\end{equation}

These equations can be summarized in the form:

\[
\begin{pmatrix}
a & b & 0 \\
b & c & b \\
b & c & b \\
0 & \cdots & 0 \\
\end{pmatrix}
\begin{pmatrix}
A_1 \\
A_2 \\
A_3 \\
A_4 \\
\end{pmatrix}
= 0. \tag{2.11}
\]

Here, \( b = 8JS(\cos(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}})) \), \( c = -\hbar \omega + 8JS[3 - \cos^2(\vec{Q}_\parallel \frac{a_0}{2\sqrt{2}})] \) and \( a = c - 8JS \). This system of equations has an infinite set of solutions. One of these solutions is a surface
A spin wave mode, for which the excitation amplitude decays exponentially into the bulk, \( A_{n+1} = A_n e^{-\alpha a_0^2} \). The other solutions are bulk spin wave modes, here \( A_{n+1} = A_n e^{i(q_{\perp} a_0^2)} \), where \( q_{\perp} \) is the wave vector perpendicular to the surface. Let us consider the solution of the bulk modes first, here

\[
\hbar \omega = 8JS[3 - \cos^2\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right) - 2\cos\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right)\cos\left(\frac{q_{\perp} a_0}{2}\right)].
\]

(2.12)

For a semi-infinite system, one thus obtains a continuum of bulk modes for each given \( Q_{\parallel} \). In contrast, for the surface mode one obtains

\[
n = 1 : \hbar \omega = 8JS[2 - \cos^2\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right) - e^{-\alpha a_0^2}\cos\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right)],
\]

(2.13)

\[
n > 1 : \hbar \omega = 8JS[3 - \cos^2\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right) - e^{-\alpha a_0^2}\cos\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right) - e^{\alpha a_0^2}\cos\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right)].
\]

(2.14)

Inserting these two equation into each other, one can determine the decay factor

\[
e^{-\alpha a_0^2} = \cos\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right).
\]

(2.15)

Note that the decay factor depends on \( q_{\parallel} \). Thus, the surface localization increases with \( q_{\parallel} \) and the highest localization is reached at the surface Brillouin zone boundary. The spin wave dispersion for the surface mode is given by

\[
\hbar \omega = 8JS[2 - 2\cos^2\left(\frac{q_{\parallel} a_0}{2\sqrt{2}}\right)] = 8JS[1 - \cos\left(\frac{q_{\parallel} a_0}{\sqrt{2}}\right)].
\]

(2.16)

In the experiments shown later, the films under investigation were only a few atomic layers thick. This can be taken into account in the theoretical description by introducing a second surface. Then, the crystal consists of a slab of \( n \)-layers and the matrix given in equation 2.11 contains \( n \) rows only. In this case both, the upper left and the lower right corner of the matrix have the entry \( a \). From this \((n \times n)\)-matrix \( n \) discrete spin wave modes result, two of which are surface modes. These two modes correspond to the in-phase and out-of-phase precession of the magnetic moments in the two surface layers. An analytical solution of a slab which consists of more than 3 layers is tedious, but a numerical solution is relatively simple. The results of the above calculations for a fcc Co semi-infinite crystal as well as for an 8 ML slab are shown in Fig. 2.2 a). In Fig. 2.2 b) the dispersions obtained from similar calculations for hcp Co are presented, since this system is also under investigation later. The only degree of freedom in these calculations is the value of \( JS \). For the two dispersions shown, we have chosen \( JS = 15 \text{ meV} \). The gray regions in Fig. 2.2 illustrate the continuum of bulk modes allowed for each \( Q_{\parallel} \). The eight dispersions of the spin waves in the slab are represented by solid lines. The two surface modes of the slab calculations are the lowest energy branches of the slab system. The surface mode for the semi-infinite crystal lies almost exactly on top of the surface mode of the slab and is therefore not shown here. Noticeable differences in the dispersion of the surface mode of a semi-infinite crystal and a slab occur only for very thin slabs.
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Figure 2.2: Spin wave dispersions calculated within a nearest neighbor Heisenberg model as a function of $\vec{Q}_\parallel$ for an 8 ML slab and a semi-infinite crystal. In a) calculations have been performed for a fcc crystal with a (001)-surface and in b) for a hcp crystal with a (0001)-surface [24]. In these calculations JS = 15 meV. The symbols on the top axis mark the important points in the surface Brillouin zone. The systems presented in the figure correspond to the systems under investigation in this work (see chapter 4).

Like other surface excitations, surface spin waves can only exist as unperturbed modes, when no bulk mode is present at the same point in wave vector and energy space [17]. In principle, surface spin waves for a given wave vector can lie at higher or lower energies compared to the bulk spin waves, several cases are for example discussed in Ref. [25]. Using the simple argument of the reduced number of neighbors at the surface, one would expect that the surface mode lies below the bulk modes for exchange dominated spin waves, as in the above case.

Several interesting, characteristic properties of spin wave dispersions are included in Fig. 2.2. For example, for one spin wave branch the spin wave energy goes to zero for vanishing wave vector. This branch is called acoustic branch. For all other modes, the magnetic moments in some layers process in an out-of-phase condition compared to the magnetic moments in other layers. Therefore, these modes always have a finite spin wave energy, even at $\vec{Q}_\parallel = 0$. They are referred to as optical modes.

The spin wave energy of the acoustic branch at small wave vectors can be approximated by

$$\hbar \omega = 4J Sa^2 \vec{Q}_\parallel^2 = D \vec{Q}_\parallel^2. \tag{2.17}$$

Here, $a$ is the nearest neighbor distance and $D$ is the so called spin wave stiffness. In many experiments, the wave vector transfer is limited to small values so that equation 2.17 is valid. In these cases, the quantity published in literature is typically the spin wave stiffness.

The Heisenberg model is valuable, especially due to its simplicity. It has already been mentioned that it is not expected to be applicable to an itinerant electron system.
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like Co. Nevertheless, we will see in chapter 4 that several of our results are described surprisingly well by this model. Other findings, however, can only be understood in an itinerant electron description. Therefore in the following, an introduction into magnetic excitations in an itinerant electron model is given, with emphasis on the differences to the above mentioned results.

2.2.2 Magnetic excitations in itinerant electron ferromagnets

In the 3d-magnetic metals, the conduction electrons that are the carriers of magnetic moments have to be considered as freely moving in a periodic potential. This leads to a description of electrons which are arranged in bands. Each electron is located in k-space rather than at a particular position in the crystal. The successful discussion of magnetism in such itinerant electron systems goes back to Stoner [26, 27]. Under certain circumstances, called Stoner criterium, it is energetically favorable to arrange itinerant electrons in exchange split and not in spin degenerated bands. This results in a higher occupation of states for electrons of one spin direction (majority electrons) compared to the other spin direction (minority electrons). Thus, part of the spin moments are uncompensated and the system is magnetic. The interaction responsible for the magnetic order is again the exchange interaction. The Stoner criterium is fulfilled for Fe, Co and Ni, which explains why these elements order ferromagnetically, while other 3d-metals do not. The Stoner theory also explains the non integer value of magnetic moments per atom in these elements in a natural way.

A draw back of this theory is that the ordering temperature is overestimated by at least half an order of magnitude [28]. The magnetic excitations allowed in the model are the so called Stoner excitations. In these excitations, an electron of a given spin hops from an occupied state below the Fermi-energy ($E_F$) into an empty state above $E_F$ with opposite spin, leaving a hole behind. In the Stoner model, no interaction between the excited electron and the hole is taken into account. This leads to an overestimation of the minimum energy for magnetic excitations which results in an underestimation of the drop of the magnetization with temperature. The consideration of low energy collective excitations within the itinerant electron model are needed to describe the experimentally observed Curie temperatures. Collective excitations were introduced by Slater [29] for an itinerant electron insulator, in which all spins in the system are aligned parallel to each other except one. He found that the lowest energy magnetic excitations of such a system are of collective nature. This state can be described by the superposition of single particle states and represents the correlated motion of the spin-reversed electron and the hole it left in the sea of aligned electrons [30]. The correlated electron hole pair has a spin moment of $1g\mu_B$ and a defined wave vector. It was found that its properties are similar to the spin wave excitations in the localized model [29]. The extension of the model derived by Slater to itinerant metals has been performed by Herring and Kittel [31, 32]. The general finding is that collective excitations exist in an itinerant electron system and that they are identical to the well-defined spin waves in a localized model in the limit of low wave vectors and low energies. For example, in this limit a quadratic dispersion relation
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Figure 2.3: a) Pair of exchange split bands with the majority band completely below the Fermi-energy $E_F$ (by the amount $\Delta$) to represent a strong ferromagnet. The exchange splitting parameter $U$ is assumed to be constant in the entire Brillouin zone. In b) the gray region shows the low energy part of the Stoner spectrum calculated for the band shown in a). At $q = 0$ the creation of a Stoner excitation costs an energy which is equal to $U$. The minimum energy of the Stoner excitations is equal to $\Delta$ and is located at some higher wave vector. To give some general idea, a possible spin wave dispersion is also shown in b). This dispersion was not calculated from the bands shown in a).

of these excitations was found [31, 32], as in the Heisenberg model (see equation 2.17). At high wave vectors and energies, Stoner excitations are possible in the system as well. A correlated electron hole pair that is created in the region where Stoner excitations are possible can easily decay into such uncorrelated states. Therefore, in the region in which Stoner excitations are possible, the collective excitations are not well-defined long living spin waves, but they are strongly damped [33].

The general concept of magnetic excitations in itinerant ferromagnets is summarized in Fig. 2.3. In Fig. 2.3 a) a simple representation of one pair of exchange split bands is presented. The exchange splitting is assumed to be identical to $U$ over the entire wave vector range. The majority band lies completely below $E_F$ to represent a strong ferromagnet. The energy and wave vector transfer needed to excite an electron from an occupied majority state to an unoccupied minority state is shown in Fig. 2.3 b) as a gray area. In this gray area single particle Stoner excitations are possible. For strong ferromagnets, the minimum energy for Stoner excitations is given by the distance between the majority band and $E_F$, the Stoner gap $\Delta$. For Co, realistic parameters for $U$ and $\Delta$ are of the order of 1 eV and 0.2 eV, respectively [34, and references therein]. A possible acoustic spin wave branch is also shown in Fig. 2.3 b) as a black line. When the spin wave branch enters the Stoner continuum it is strongly damped. This range of the dispersion is presented as a dotted line.

The real physical situation is only partly described by Fig. 2.3. For example, realistic band structures consist of more than one band and s-bands of both spin characters cross
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the Fermi-edge so that no true overall Stoner gap exists. In our studies, we investigate spin waves at surfaces by electron scattering. In this case, additional effects have to be considered. It has been theoretically predicted that in inelastic electron scattering experiments, the creation of free electron like Stoner excitations is as probable as a creation of d-electron Stoner excitations \[35\]. In addition, at the surface the wave vector perpendicular to the surface is not conserved due to the loss of translational invariance. This leads to drastic changes compared to what has been shown in Fig. 2.3. Even at the small \(\vec{Q}_\parallel\) spin waves can decay into Stoner excitations with \(\vec{q}_\perp > 0\) \[36\].

With the advance of theory it became feasible to calculate the above described magnetic excitation spectrum in bulk itinerant ferromagnets on the basis of ab initio calculations \[37\]. It is, however, computationally too demanding to extent these calculations to surfaces and to thin film systems. Therefore, for thin films two other approaches are used, both starting from an ab initio calculation of the underlying band structure. One frequently used approach is based on the adiabatic approximation, in which the electron motion is decoupled from the spin motion (see for example Ref. [38–44, and references therein]). This means that the damping of spin waves by Stoner excitations is not taken into account. As discussed above, this is a good approximation in itinerant electron systems only in the limit of low wave vector and low energy spin waves. In principle, these calculations map the itinerant ferromagnetism onto a Heisenberg like description. Several publications gave values for the exchange coupling constants derived this way \[42,43,45\].

The other approach goes beyond the adiabatic approximation. This is a non trivial extension of the model using the adiabatic approximation, because one has to take into account the full dynamics of the system \[10,20,36,46–54\]. This description includes the damping of spin waves caused by Stoner excitations and therefore this theory is expected to be valid throughout the Brillouin zone. The quantity derived in these calculations is the linear response function of magnetic excitations in the solid, the transverse magnetic susceptibility \(\chi\). This quantity contains all information, i.e. the dispersion and the spectral weight, of magnetic excitations. In other words, \(\chi\) assigns an intensity to every point in graph 2.3 b). So far, these calculations were only possible using an empirical tight binding description of the underlying band structure \[36,48,50,54\].

In a recent series of publications, Mills and coworkers applied this theory to magnetic thin films of Fe, Ni, and Co \[10,20,36,50–54\]. These calculations showed that the damping of spin waves caused by Stoner excitations is strong in these films. As shown in the last section, within the Heisenberg model one expects as many spin wave modes at a given \(\vec{Q}_\parallel\) as layers are contained in the film (see Fig. 2.2). In the itinerant electron theory that goes beyond the adiabatic approximation, this picture changes drastically. Instead of a number of discrete modes each of zero width, the theoretical calculations show only one single broad feature \[52,54\]. This arises from the strong damping of the different modes. They overlap and can hardly be distinguished from each other \[54\].

In this section, two different theoretical concepts of magnetism have been introduced, in which high wave vector spin waves show a different behavior. In the following, we

\[2\] The calculations have been performed for Fe, however, it can be assumed that similar effects may also occur in Co and Ni.
will concentrate on spin waves in Co, in which the electrons have itinerant character. Nevertheless, we will discuss our results to some extent in a nearest neighbor Heisenberg model. Of course, several questions concerning the validity of such a description arise. As discussed above, high wave vector spin waves in this system are expected to be heavily damped. It is expected that this damping influences not only the spectral shape of the spin waves but also effects the dispersion [20]. In addition, in the Heisenberg model rigid magnetic moments are allocated to each lattice point. Thus, the model will break down when changes within the spin density on the atomic scale are significant. Another questionable assumption is that only nearest neighbor interactions are included in our calculations. As will be shown later, it seems possible to take the influence of the above mentioned criticism into account to some extent by an effective nearest neighbor exchange coupling constant in the Heisenberg model.

2.3 Established experimental methods to study spin wave excitations

In the following section, different experimental techniques will be introduced which are used to study spin wave excitations. The description is limited to three important techniques: inelastic neutron scattering (INS), Brillouin light scattering (BLS), and ferromagnetic resonance (FMR). The focus lies on INS. The accessible wave vector and energy transfer range in these experiments is similar to the range investigated later in our electron scattering experiments. Findings obtained from all three techniques are compared to our results in section 5.4.

In all scattering techniques (INS, BLS, and SPEELS) the approach to detect spin wave signals is similar. A particle with known energy is scattered under defined conditions from a sample and afterwards is analyzed with respect to its wave vector and energy transfer during scattering. Usually, either the wave vector transfer (constant q-scan) or the energy transfer (constant E-scan) is kept fixed, while the other is varied. These measurements correspond to a scan along a vertical or horizontal line in Fig. 2.2 or Fig. 2.3. Due to energy and momentum conservation laws, the intensity caused by spin wave excitations appear in the scan only at points (in q- and E-space) where the scan line intersects with a branch of a spin wave.

A very important and well established method is INS. For the development of this technique half of the shared Noble prize in 1994 was given to B. N. Brockhouse. By INS spin waves can be studied in a large portion of the Brillouin zone. For high wave vector and high energy transfers, however, these experiments become tedious even with advanced neutron sources. For this reason, only a few INS studies of high wave vector spin waves in 3d-ferromagnets have been performed [55–59]. One general property of neutrons is their weak interaction with matter, which becomes a disadvantage when thin films or surfaces are the subject of interest. While static structural (magnetic) information can be obtained in favorable cases with a possible monolayer sensitivity [60], measurements of spin wave excitations in ultrathin films or at surfaces are practically out of range [6].
Figure 2.4: Example of recent neutron scattering measurements of spin wave excitations in hcp Co [59]. The left side shows a time-of-flight spectrum, transformed back into energy space, for details see Ref. [59]. On the right side, the resulting spin wave dispersion is shown. The wave vector is normalized in this graph so that the Brillouin zone boundary is at \( q/a^* = 0.5 \). The solid line is a fit obtained by a nearest neighbor Heisenberg model.

Neutrons scatter from matter mainly because of two interactions of approximately equal strength. Neutrons interact with the atomic nucleus resulting in structural information. This interaction will not be discussed here, since it is not of interest in this work. The second interaction is the dipolar coupling between the spin of the neutron and the magnetic field created by the (uncompensated) electron spins [61]. The creation or annihilation of a spin wave takes place via the coupling of the neutron to the fluctuations of this field [62]. It can be shown that the cross section for magnetic inelastic neutron scattering is directly proportional to the imaginary part of the transverse susceptibility [63–65]. As discussed above, this quantity yields the complete information of the magnetic excitations in the system, the spin wave and Stoner excitations. In neutron experiments, however, the signal obtained from Stoner excitations is weak and due to their broad structure Stoner excitations can typically not be distinguished from the background. The broadening or disappearance of spin waves at higher wave vectors is often interpreted as a sign of the merging of the spin wave branch into the Stoner continuum [55,66].

A recent example of high wave vector and high energy spin wave excitations investigated by INS is shown in Fig. 2.4 [59]. The experiments were performed on hcp Co using a time-of-flight technique. The peaks visible in the spectrum in Fig. 2.4 a) belong to energy losses caused by spin wave excitations with different wave vector transfers in different Brillouin zones. The information measured in different Brillouin zones can be back folded into the first Brillouin zone. The resulting dispersion relation is shown in Fig. 2.4 b). In these measurements, the dispersion could be followed up to about \( 2/5 \) of the Brillouin zone. The solid line is a fit to the data using a dispersion relation calculated within a nearest neighbor Heisenberg model. A good agreement between the measured dispersion and this model was found [59]. The INS studies showed a broadening of the
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spin wave peaks due to itinerant effects. Nevertheless, the spin wave losses were relatively well-defined up to the highest wave vectors investigated.

BLS and FMR are used to study spin waves with long wavelength. Both techniques probe several similar properties of magnetic materials. In FMR the sample is exposed to an alternating magnetic field in the micro wave regime and an additional static magnetic field. Under certain conditions, a coherent procession of the magnetic moments in the system may be possible. This resonant excitation leads to a drop in the reflected or transmitted microwave power. This effect was discovered by Griffiths [67] and explained by Kittel [68]. The spin waves are created by the coupling of the magnetic moments in the solid to the external field [69]. Due to the high index of refraction of metals for micro waves, these waves travel almost perpendicular to the surface and thus, $\vec{Q}_\parallel \approx 0$ [70].

BLS exploits the inelastic scattering of light from matter [71]. A photon couples to a spin wave due to the change of the magneto-optical constants in the solid in the presents of a spin wave [72]. Here, mainly the electrical field of the photon couples to the spin wave via the spin-orbit interaction [4]. The first experimental realization of BLS has been performed by Sandercock and Wettling [73]. In BLS, the possible wave vector transfer parallel to the surface is limited by the wave vector of the incident light which is of the order of $\vec{Q}_\parallel \approx 10^{-3}$Å$^{-1}$.

With the quoted wave vector regime both techniques, BLS and FMR, can determine the macroscopic magnetic quantities of a sample, e.g. the anisotropy. In addition, in both techniques standing spin wave modes perpendicular to the surface have been observed in films with thicknesses of several nanometer or thicker [74, 75]. This leads to the possibility to study spin waves with higher wave vectors, up to $\vec{q}_\perp \approx 10^{-2}$Å$^{-1}$ [4]. Using these standing modes, the spin wave stiffness of the material can be determined by BLS and FMR. The experiments are typically performed in air, but experiments in ultra high vacuum are also possible. Both techniques can be used to study ultrathin films. The draw back of BLS and FMR is the limitation to small wave vectors.

2.4 Electron scattering

This section gives a brief introduction into electron scattering. The discussion is limited to monochromatic low energy ($\approx 10$ eV) electrons scattered from crystalline metallic surfaces. Due to the strong interaction of low energy electrons with electrons in the solid, the mean free path is limited to a few atomic layers. Therefore, low energy electrons have a true surface sensitivity. This is the major reason why electrons have been used extensively in almost all kinds of surface studies [7, 76]. The section is divided into two parts. The first part deals with elastic electron scattering and summarizes the interactions between an electron and the matter. The second part treats inelastic scattering focusing on magnetic excitations.
Figure 2.5: The Ewald construction provides a graphical solution to an elastic scattering event on a two dimensional periodic lattice, for which $\vec{K}_f^\parallel = \vec{K}_i^\parallel + \vec{G}_\parallel$. Here, $\vec{K}_i^\parallel$ and $\vec{K}_f^\parallel$ are the wave vector components parallel to the surface of the incident and the outgoing electron, respectively and $\vec{G}_\parallel$ is a reciprocal lattice vector of the two dimensional reciprocal lattice.

2.4.1 Elastic electron scattering

A free electron with a kinetic energy $E_{\text{kin}}$ has a wave vector which is given by the de Broglie relation $k = \frac{(2mE_{\text{kin}})^{1/2}}{h}$, where $m$ is the electron mass. For example, an electron of a kinetic energy of 10 eV has a wave vector of $k \approx 1.6 \text{ Å}^{-1}$. This corresponds to a wavelength of $\lambda = \frac{2\pi}{k} \approx 3.9 \text{ Å}$, which is of the order of a typical lattice constant. Therefore, scattering of electrons from the periodic structure of a crystalline surface results in interference effects. This interference pattern is determined by the arrangement of the atoms at the surface. Due to the limited penetration depth, the scattering electrons mainly experience the lattice periodicity parallel to the surface. For the moment, we assume that the electrons scatter from a perfect two dimensional periodic structure. The two dimensional translational invariance in real space implicates the same invariance in reciprocal space. The resulting structure in reciprocal space is a two dimensional array of rods standing perpendicular to the surface. The distance between the reciprocal lattice rods is given by the two dimensional reciprocal lattice vector $\vec{G}_\parallel$. In a scattering process, the wave vector component parallel to the surface is conserved except for multiples of the reciprocal lattice vector so that the two dimensional Laue condition $\vec{K}_i^\parallel = \vec{K}_f^\parallel + \vec{G}_\parallel$ is fulfilled. Here, $\vec{K}_i^\parallel$ and $\vec{K}_f^\parallel$ are the wave vector components parallel to the surface for the incident and the outgoing beam, respectively. Due to the missing periodicity perpendicular to the surface, the perpendicular wave vector component $\vec{k}_\perp$ of the scattering electron is not conserved. Graphically, this can be illustrated in the Ewald construction [7] shown in Fig. 2.5. At each position where a sphere of radius $|k|$ cuts one of the reciprocal lattice rods, the diffraction condition is fulfilled. Thus, the positions of the diffraction spots contain information of the underlying lattice. This is for example used in low energy electron diffraction (LEED) measurements.

The translational symmetry in the reciprocal space implies that two points that are
connected by $\vec{G}_\parallel$ have the same properties. One can therefore divide the reciprocal space into unit cells, i.e. surface Brillouin zones, which are the complementary structures to the Wigner-Seitz cells in real space [16]. The complete periodic information of the system is contained in each surface Brillouin zone.

The penetration depth of low energy electrons is not limited to the first atomic layer, but extents deeper into the crystal. The exact penetration depth depends on the kinetic energy and the material scattered from as well as possibly on the spin direction of the incoming electrons. The mean free path for Co is about 7 Å (9 Å) for minority (majority) electrons with energies of several eV above the work function [77, 78]. Therefore, the electrons are exposed to the first few repetitions of the periodic structure perpendicular to the surface. This leads to a weakly defined third Laue condition and to a modulation of the intensities along each reciprocal lattice rod [79].

The discussion above was limited to the kinematic approximation, where each electron is scattered only once before it leaves the crystal. As a consequence of the strong interaction of electrons with matter, electrons are, however, on average scattered several times before they leave the crystal. These multiple scattering processes are taken into account in the dynamical scattering theory. The description can be improved by considering the extended charge and spin distribution around each atom, as well as the work function [79].

The scattering of an electron from a crystal is a complicated many-body problem which cannot be solved without any approximations. A natural and quite successful approach is the reduction to an one-particle problem, where the electron is exposed to an effective potential created by all others. The interactions are then represented by the combination of an effective electrostatic and an effective magnetic potential [76,80].

Typically, by considering only single scattering events, one obtains a rough description of the results of an electron scattering experiment. Of course, this simplified description of the scattering processes cannot provide a quantitative analysis of experimental findings. Nevertheless, in the following our results are mainly discussed within the framework of the kinematic model.

The strong interaction between electrons and matter is mainly caused by the Coulomb interaction which is strong and in principle of long range. When an additional electron is embedded into a metal, it is shielded by the proper arrangement of the surrounding charge. In this case, the Coulomb interaction becomes short ranged. The interaction of the electron spin with the solid consists mainly of two types: the exchange and the spin-orbit interaction. Both interactions have been discussed in a different context in section 2.1. In the scattering terminology these have a slightly different meaning, since here a free electron enters and leaves a crystal. The underlying physical concept, however, stays the same.

The exchange interaction results from the fact that electrons are undistinguishable. The electron detected after a scattering event can either be identical to the incoming electron or this electron can be exchanged with an electron from the solid. In the case that the crystal is a ferromagnet this leads, for example, to polarization effects of the scattered electron beam [76,81,82]. The magnetic scattering processes which result from the exchange interaction obey some characteristic symmetry considerations. For example,
the polarization caused by this interaction changes sign when the magnetization of the sample is reversed.

The spin-orbit coupling leads to different scattering potentials for incoming electrons of different spin character, even for non magnetic crystals. The strength of the spin-orbit interaction increases with the atomic number of the atom scattered from. In our SPEELS-experiments, we will see that the spin-orbit coupling can be neglected for Co, whereas for W it is important. The spin-orbit interaction obeys certain symmetry laws, too. For example, the argument of time reversal requires that the interaction changes sign when incoming and outgoing beams are interchanged.

### 2.4.2 Inelastic electron scattering

When electrons are scattered from a crystal, they may experience inelastic scattering processes. In that case, a transfer of energy and momentum occurred between the incoming electron and the sample. In an inelastic electron scattering event, the total energy and typically also the total wave vector parallel to the surface is conserved in the system, therefore:

\[
\vec{K}_f^\parallel = \vec{K}_i^\parallel - \vec{Q}_\parallel + \vec{G}_\parallel, \tag{2.18}
\]

\[
E_{\text{kin}}^f = E_{\text{kin}}^i - E.
\]

Here, \(\vec{Q}_\parallel\) and \(E\) are the wave vector and the energy transferred to the crystal. This energy and momentum are transferred to an excitation in the sample. The quantities that can be measured in our experimental set-up are the wave vector transfer parallel to the surface \(\Delta \vec{K}_\parallel\) and the energy loss \(E_{\text{loss}}\). Due to the conservation laws given above, these are related to the wave vector and energy transferred to the crystal by \(\Delta \vec{K}_\parallel = \vec{K}_f^\parallel - \vec{K}_i^\parallel = -\vec{Q}_\parallel + \vec{G}_\parallel\)

and \(E_{\text{loss}} = E_{\text{kin}}^i - E_{\text{kin}}^f = E\). The measured scattered intensity as a function of \(\Delta \vec{K}_\parallel\) and \(E_{\text{loss}}\) therefore contains the information of the excitations possible in the sample. This is the general experimental idea, which will be followed throughout the next chapters.

Inelastic electron scattering is divided into the dipolar\(^3\) and the impact scattering regime. In dipolar scattering an electron is scattered from the electric dipolar field which under certain circumstances is created by vibrations of atoms on the surface. Dipolar scattering is typically strong close to the specular condition, in the so called dipolar lobe. The high scattering probability in this region is often used in EELS-experiments [83]. Due to the long range interaction of the dipolar field, the scattering typically takes place long before the electron reached the surface. Therefore, it is sufficient to describe this scattering by macroscopic quantities [83]. All experiments presented in the next chapters were obtained at conditions far away from the dipolar lobe where this type of scattering is much less important. Therefore, this interaction will not be considered in more details. For impact scattering, as the name suggests, the incoming electron penetrates into the crystal and is scattered by impact. Because of the close distance between the incoming

\(^3\)The name dipolar regime might be confusing. Here it refers to electric dipoles not to be mixed with the magnetic dipolar regime in the spin wave description.
2.4 Electron scattering

Figure 2.6: Schema of different inelastic exchange scattering processes possible for an incoming $e^i$-electron. The red electron represents the incoming electron which has an energy $E^i_{\text{kin}}$ before the scattering event. The green electron comes from the crystal. $E_F$ and $E_{\text{loss}}$ refer to the Fermi-energy and the energy loss in the scattering process, respectively. The electron of the energy $E^f_{\text{kin}} = E^i_{\text{kin}} - E_{\text{loss}}$ is the one that is detected after the event. Note that even in the "spin flip"-channel each electron keeps its spin direction.

electron and the electrons in the sample, exchange processes are possible in the impact regime. In contrast, exchange processes will not take place in dipolar scattering, due to the large distances between the electrons [84]. The description of the impact process demands a truly microscopic model [83]. Although several scattering processes seem to be properly described by such a model, the process of electron scattering by the excitation of a spin wave is not yet understood. Therefore, the following description of magnetic excitations by inelastic electron scattering is phenomenological.

Several excitations of a solid are possible by electron scattering. They can be roughly classified by the energies which are necessary for the excitation. At low energies, the possible excitations are phonon excitations, vibrations of adsorbates on the surface, electron hole pair excitations from conduction band electrons and spin wave excitations. The first three excitations have been studied by EELS for decades, see for example Ref. [83]. The surface sensitivity of EELS, for instance, can be used to study the vibrational excitations of tiny amounts of adsorbates on surfaces. This capability often is a handicap in our experiments, because the measurements require a high cleanliness of the sample surface.

Now, we focus on magnetic excitations, the spin waves and the Stoner excitations.
Figure 2.7: a) Illustration of the scattering geometry used in the following. $\theta_0$ defines the angle between the incoming and outgoing electron path. $\theta$ is the angle between the surface normal and the incident beam. b) Schematic picture of an electron scattering process in which a spin wave is excited. The wave vector component parallel to the surface is conserved in the scattering process, the component perpendicular to it is not. The creation of a spin wave reduces the magnetization of the sample by $1g\mu_B$. The conservation of the total magnetic moment requires that for the excitation of a spin wave the incoming electron has to be of minority character and the outgoing electron of majority character. Note that by definition the spin and its magnetic moment point into opposite directions.

Beside the conservation of energy and momentum given in equation 2.18 also the total spin moment is conserved during scattering. It is therefore useful to separate the scattering processes in different spin channels defined with respect to the magnetization of the sample. In the following, $e^\uparrow$ ($e^\downarrow$) refers to an incoming electron with a spin moment parallel (anti-parallel) to the spin moment of a majority electron in the solid. Four possible configurations for the outgoing electron exist. The scattered electron can either have the same spin as the incoming electron, marked by $e^{\uparrow\uparrow}$ ($e^{\downarrow\downarrow}$), or can have a reversed spin, $e^{\uparrow\downarrow}$ ($e^{\downarrow\uparrow}$). Due to similarities to polarized neutron scattering, the former two channels are denoted as non spin flip channels and the latter two as spin flip channels. The terms are a bit awkward in electron scattering because in none of the scattering processes discussed here an electron flips its spin. This is illustrated in Fig. 2.6. The scattering process can be a direct scattering process, in which the incoming (red) electron with energy $E_{\text{kin}}^i$ is the one that leaves the crystal with energy $E_{\text{kin}}^f$ (first two cases in Fig. 2.6). Scattering can also take place in an exchange process where the electron that leaves the crystal is originating from the crystal (green) (third and fourth case in Fig. 2.6). In the latter process, the incoming electron drops into an empty state above $E_F$ and transfers most of its energy to an electron in the crystal. The force acting during this scattering events is the Coulomb force, therefore, each electron keeps its original spin direction. Nevertheless, in the exchange process it is possible that the outgoing electron has the opposite spin compared to the incoming one. From the eye of the beholder, it seems as if the spin of the electron has flipped.
2.4 Electron scattering

Fig. 2.7 shows a sketch of a possible scattering process in which an electron excites a spin wave. From the geometry defined in Fig. 2.7 a) one obtains

$$-\vec{Q}_|| + \vec{G}_|| = \Delta \vec{K}_|| = k_f \sin(\theta_0 - \theta) - k_i \sin(\theta).$$  \hspace{1cm} (2.19)

In the case presented in Fig. 2.7 b), the excited spin wave is a surface spin wave that has only a wave vector component parallel to the surface. Since the wave vector component perpendicular to the surface is not conserved during the scattering, with the same process sketched in Fig. 2.7 b) also a spin wave with $\vec{q}_\perp \neq 0$ could be excited.

Because the total magnetic moment has to be conserved in the scattering process, the excitation of spin waves is only possible in a certain spin channel. In a simple picture, the creation of a spin wave reduces the magnetization of the sample by $1g\mu_B$. To fulfill the conservation law, the creation of a spin wave is thus only possible in the $e^{\uparrow\downarrow}$-channel. Therefore, the incoming electron has to be of $e^{\downarrow}$ (minority) character to be able to excite a spin wave (as indicated in Fig. 2.7 b)). Vice versa, the annihilation of a spin wave is only possible in the $e^{\uparrow\downarrow}$-channel.\(^4\) By this selection rule, spin waves are the only excitations in electron scattering that gives a 100\% spin-polarized loss peak. For this reason, it was even suggested to use these excitations as a source of spin-polarized electrons [8]. Though this application seems not promising, the selection rule itself can be used in the experiment to separate spin wave excitations from other excitations. It is sufficient to have a spin-polarized incoming electron beam and a ferromagnetic sample with a defined magnetization direction parallel to the polarization axis. In this particular case, an energy loss caused by the excitation of spin waves is only possible for one spin direction of the incoming electrons. This circumvents the necessity of a "complete" experiment with a spin analysis of the scattered electrons, which is a difficulty in electron scattering experiments due to the notoriously inefficient detectors [85].

Stoner excitations have been extensively studied by SPEELS. First SPEELS-measurements were performed by Hopster (unpolarized incoming electron beam, but spin detector) [82] and Kirschner (exploiting an incoming spin-polarized beam, but no spin detector) [81]. In these measurements, Stoner excitations appear as a broad feature extending from low energies up to several eV. Only little structure as a function of the loss energy has been observed. In most cases, the maximum intensity of the Stoner excitations was found close to the value of the average exchange splitting. Even complete experiments with an incoming spin-polarized electron beam and a spin analysis of the scattered electrons were performed to investigate Stoner excitations [34, 86, 87]. These experiments showed that Stoner excitations are present in both spin flip channels. The excitations had higher intensity for incoming minority electrons. In addition, the non spin flip excitations (simple electron hole pairs) produced a significant amount of the intensity to the spectra. This intensity was found to differ for incoming electrons with majority or minority spin char-

\(^4\)The above argument is strictly valid only for zero temperatures and has to break down at $T_C$. In the studies presented in this work, the temperature will be of the order of half $T_C$ or lower. At this temperatures, the consideration that the creation of a spin wave is only possible in the $e^{\uparrow\downarrow}$-channel is a good approximation.
Figure 2.8: First signature of spin waves obtained by SPEELS-measurements. The data are taken from Ref. [90]. The measured intensity is shown as dots. The crosses mark the asymmetry $A = \frac{I^\downarrow - I^\uparrow}{I^\downarrow + I^\uparrow}$. The increase of the asymmetry below 0.4 eV is attributed to spin wave excitations, the increase above 0.5 eV is caused by Stoner excitations.

It was shown theoretically that the cross section of Stoner excitations in electron scattering is enhanced compared to neutron scattering [36,50]. It was argued that the response function probed in these two experiments is different because of the different interactions of neutrons and electrons. From the theoretical predictions, both response functions differ mainly in the Stoner contributions and are similar in the spin wave regime [50]. The possible detection of spin waves via inelastic electron scattering was theoretically proposed in the middle of the last century [8]. Several values of the cross section between electrons and spin waves were predicted, see for example Ref. [36,88,89]. The results from these calculations differ significantly. In the latest publication by Mills and colleagues [36], its was estimated that the spin wave intensities in SPEELS should be comparable to the intensity of Stoner excitations. A first experimental signature of spin waves in SPEEL-spectra was found by Kirschner [90]. This result was confirmed by Vernoy and Hopster [91]. In both publications, a spin-polarized incident electron beam was used.\(^5\) The main result of Ref. [90] is shown in Fig. 2.8. A broad maximum, centered around 200 meV, is visible in the asymmetry ($\frac{I^\downarrow - I^\uparrow}{I^\downarrow + I^\uparrow}$) of the two intensities of different incoming electron spin directions. The low energy side of the peak was not resolved due to the limited energy resolution and the study of the spin wave dispersion was not carried out. The possibility to study spin wave excitations by SPEELS, however, was demonstrated in this experiment. In order to explore this possibility in detail, we have undertaken the present study.

\(^5\)To discuss the results, we use the same notation that will be used later in our measurements. $I^\uparrow$($I^\downarrow$) denotes the measured intensity for incoming electrons with majority (minority) spin character.