Summary of the dissertation

Ab initio description of transverse transport due to impurity scattering in transition-metals

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The dream of spintronics is to use the electron's spin degree of freedom to store, transport and process information, with the ultimate goal to increase the performance and reduce the power consumption of electronic devices. This work sheds light on some of the most important phenomena in spintronics, namely spin-relaxation, the anomalous Hall effect (AHE) as well as the Spin-Hall effect (SHE). Spin-relaxation commonly represents a major drawback, since the information encoded in spin is lost after the so-called spin-relaxation time. It is essentially enabled by the coupling of the electron's spin- and orbital degrees of freedom (spin-orbit coupling, SOC). On the other hand, SOC also leads to the generation of pure spin currents (via the SHE) or spin-polarized currents (via the AHE) from a non spin-polarized charge current. However, the up-to-date low efficiency of the AHE and SHE hinders the realization of practically applicable spin-based electronic devices.

The solution lies in the specific design of new materials, where SOC effects can be tuned by microscopic means, e.g. through the interplay with magnetism, through doping with impurities or by introducing other kinds of disorder. We investigate trends and formulate design guidelines by calculations from first principles. We pay special attention to transition-metal compounds, because of their technological relevance and possibility to support magnetism.

In order to study complex transition-metal compounds, a tetrahedron method for the precise calculation of the Fermi surface of complicated shape in the framework of the Korringa-Kohn-Rostoker Green function method was developed in *Chapter 4*. Furthermore, an efficiently parallelized and thus highly scalable implementation of the accurate calculation of scattering properties off impurities and finite clusters of disorder was achieved. As it turns out, the AHE and SHE are very sensitive to details of the electronic structure. Therefore, we emphasize that within the chosen and developed method, no shape approximations to the potential are made (i.e. the full potential is used) and SOC is included self-consistently in all steps of the calculation.

In Chapter 5, the Elliott-Yafet spin-mixing parameter of 5*d* and 6*sp* transition metals was investigated, which displays the host-crystal contribution to spin-relaxation processes in metals. Special focus was addressed to a yet unexplored dependence of the Elliott-Yafet parameter on the electron's spin-polarization direction (also called spin-quantization axis, SQA). This anisotropy was found to reach gigantic values in uniaxial hcp transition metals due to the emergence of large spin-flip hot-areas and hot-loops on the Fermi surface, as the direction of the SQA is tilted away from the *c*-axis of the hcp crystal. The origin of the effect lies in the high anisotropy of the

spin-flip part of SOC itself, which is amplified by a peculiar orbital character of the degenerate electronic states in an hcp crystal, superimposed by non-symmorphic symmetries. In turn, the combination of those aspects leads to gigantic values, as high as 800% for hcp Hf as our explicit calculations show. There exists no theoretical upper limit, and the race for materials with an even higher anisotropy of the Elliott-Yafet parameter is open. Finally, the newly discovered anisotropy offers the invaluable possibility for experimentalists to tune the spin-relaxation time by simple means, e.g. the direction of an external magnetic field or spin injector, instead of changing the sample.

Chapter 6 deals with the analysis of the anomalous and spin Hall effect in various dilute alloys. We performed *ab-initio* calculations of the skew-scattering contribution in ferromagnetic hosts for the first time, and addressed a vast number of materials ranging from simple dilute alloys to complicated compounds. This included the calculation of substitutional impurities in a ferromagnetic bcc Fe host, where we identified a sign change in the anomalous Hall angle when the impurity atom is changed systematically across the 3*d* series of the periodic table. We delved into the effect by determining the Fermi-surface resolved contributions to the anomalous Hall conductivity, and discovered strongly peaked contributions at small ``hot spots'' around spin-orbit lifted degeneracies. However, these contributions appear in pairs of different sign and constitute an overall tiny anomalous Hall angle in bcc Fe. Larger Hall angles are achieved by considering alloys with stronger intrinsic spin-orbit coupling, and we address different kinds of disorder in the $L1_0$-ordered FePt alloy. Finally, the power of our newly developed method was showcased at the example of the very complicated compound MnSi. Here, the unusual space group of the B20 crystal-structure leads to fascinating manifestations of spin-orbit broken symmetries, best seen by the Fermi-surface topology, anisotropy of electron lifetimes and the unusual symmetry of the conductivity tensors.

Doping of heavy, non-magnetic hosts with magnetic impurities presents an alternative to enhance the AHE, as was shown in *Chapter 7* by the calculation of 3*d* impurities in Pd, Pt and Au hosts. An analysis of the spin-resolved conductivities in terms of a two-current model reveals a strong suppression of skew-scattering in one spin channel for nearly every host-impurity combination. A close relation between transverse charge and spin currents follows, which is interesting from an application and experimental point of view, since it serves as a source of strongly spin-polarized currents.

For the generation of pure spin-currents, the skew-scattering mechanism has been invesigated in *Chapter 8* for various 4*d*/5*sp* and 5*d*/6*sp* impurities in an fcc Ir and hcp Re host. A vast range of spin-Hall angles can be obtained by either changing the dopant, or — and this has been shown for the first time — via a strong anisotropy of the skew-scattering contribution to the SHE in hcp Re as function of the direction of the SQA, which can be tuned by e.g. an external magnetic field. It is surprising, that there exists also a strong anisotropy within the *ab* plane of the hcp crystal, which is absent for the intrinsic contribution to the SHE.