PD_16-120917 - Final report

Budapest University of Technology and Economics

Theoretical study of complex magnetic states in thin films

The main purpose of this proposal was to understand the mechanism of complex magnetic states in thin film structures relying on first principles calculations. During the project, different thin film structure was investigated using systematic numerical studies. For the systematic investigations, a previously developed methods were used based on the screened Korringa-Kohn-Rostoker (SKKR) method. Beyond these calculations, a non-relativistic and relativistic spin-spiral method was implemented into the SKKR program, which gives an other proper tool for investigating the magnetic structure of thin film systems.

In the present final report the main results of this proposal is presented for each year:

1st Year:

We examined localized metastable spin configurations in the field-polarized state of $(Pt_{0.95}Ir_{0.05})$/Fe bilayer on Pd(111) surface by using spin dynamics calculations. We demonstrated that the frustration of the isotropic exchange interactions is responsible for the creation of various types of skyrmionic structures, while the Dzyaloshinsky-Moriya interaction selects skyrmions with -1 topological charge which is the most favoruable configurations. The results were reported in Physical Review B **95**, 094423 (Ref. [2] in the publications list).

In the $(Pt_{1-x}Ir_x)$Fe/Pd(111) ultrathin magnetic film, we investigated metastable skyrmionic spin structures with various topological charges. Based on the calculated SP-STM images, we conclude that an out-of-plane magnetized tip already results in distinguished SP-STM contrasts for the different skyrmionic structures corresponding to their symmetries. Moreover we have demonstrated that the magnitude of the topological charge can be determined from the image contrast of a single SP-STM measurement using an in-plane magnetized tip. The results were reported in Physical Review B **96**, 024410 (Ref. [1] in the publications list).

Based on multiscale calculations combining ab initio methods with spin dynamics simulations, we performed a detailed study of the magnetic behavior of Ni_2MnAl/Fe bilayers. Our simulations showed that such a bilayer exhibits a small exchange bias effect when the Ni_2MnAl Heusler alloy is in a disordered B2 phase. Additionally, we presented an effective way to control the magnetic structure of the Ni_2MnAl antiferromagnet, in the pseudo-ordered B2-I as well as the disordered B2 phases, via a spin-flop coupling to the Fe layer. The results were reported in Physical Review B **96**, 064435 (Ref. [3] in the publications list).

We started the implementation of the non-relativistic spin-spiral code and tested this new development for some systems. From our test calculations we concluded that for Mn monolayer on W(001), the calculated non-relativistic spin-spiral dispersion relation well reproduces other theoretical and experimental results. From these results a publication was not written in this year.

2nd Year:

We continued the self consistent spin-spiral calculations for some selected systems. We found that in case of bulk structures the spin spiral calculations well reproduced the known magnetic ground states. For FeRh we obtained antiferromagnetic ordering, while Fe, Ni_2MnAl and FePt the magnetic ground states were ferromagnetic. These results were not published.

Motivated by previous experimental and theoretical investigations, we studied the magnetic properties of a Co monolayer on a Pt(111) surface with a capping monolayer of selected 5d elements (Re, Os, Ir,

Pt, and Au) using first principles calculations. We focused on the investigation of how the electronic hybridization with heavy metal capping layers possessing different numbers of valence electrons and different strengths of the spin-orbit interaction influences the magnetic properties of the Co layer. We found a close relationship between the magnetic moment of the Co atoms and the nearest-neighbor isotropic exchange interaction, which is attributed to the electronic hybridization between the Co and the capping layers. The Dzyaloshinskii-Moriya interaction is decreased for all overlayers compared to the uncapped Co/Pt(111) system, while even the sign of the Dzyaloshinskii-Moriya interaction changes in the case of the Ir overlayer. We concluded that the variation of the Dzyaloshinskii-Moriya interaction is well correlated with the change of the magnetic anisotropy energy and of the orbital moment anisotropy as Fig. 1 illustrates. The sign of Dzyaloshinskii-Moriya interaction can be defined based on whether the Dzyaloshinskii-Moriya vectors prefer clockwise or counterclockwise rotation of the spins. The unique influence of the Ir overlayer on the Dzyaloshinskii-Moriya interaction was traced by scaling the strength of the spin-orbit coupling of the Ir atoms in Ir/Co/Pt(111) and by changing the Ir concentration in the $Au_{1-x}Ir_x$/Co/Pt(111) system. Our spin dynamics simulations indicated that the magnetic ground state of Re/Co/Pt(111) thin film is a spin spiral with a tilted normal vector, while the other systems are ferromagnetic. The results were reported in Physical Review B **97**, 134405 (Ref. [5] in the publications list).

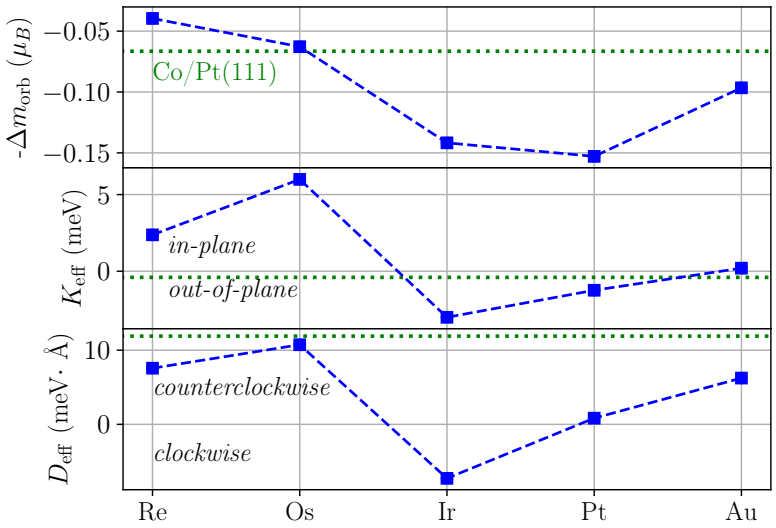

Figure 1: Calculated values of orbital moment anisotropy in the Co layer with negative sign $-\Delta m_{\rm orb}$, magnetic anisotropy, $K_{\rm eff}$, and effective Dzyaloshinskii-Moriya interaction, $D_{\rm eff}$ for X/Co/Pt(111) thin films (X=Re, Os, Ir, Pt, Au). Picture was taken from Phy. Rev. B **97**, 134405.

Motivated by the experimental study for MnN/Fe interface, we investigated the magnetic properties of bulk MnN and the MnN/Fe interface at zero and finite temperatures based on ab initio calculations and spin dynamics simulations. We determined the spin model parameters for bulk MnN, and we found that the competition between the nearest and the next-nearest-neighbor interactions leads to antiferromagnetic ordering of the Mn spins, in agreement with previous theoretical and experimental results. At the MnN/Fe interface, a sizable Dzyaloshinskii-Moriya interaction appears leading to a stable exchange-bias effect. We studied the dependences of the exchange-bias effect on the thicknesses of the ferromagnetic and the antiferromagnetic layers, and we compared them to experimentally obtained result. Hysteresis loops of the MnN/Fe bilayer were evaluated after a field-cooling process, and a large exchange-bias effect was found due to sizeable Dzyaloshinskii-Moriya interactions across the MnN/Fe interface. The variations of the exchange-bias effect as a function of the thickness of the FM and AF layers were also investigated. The results were reported in Physical Review B **98**, 094415 (Ref. [4] in the publications list).

3rd Year:

We finished the implementation of the relativistic and non-relativistic spin-spiral technique into the SKKR code. For layered magnetic systems, the propagation vector of the spin-spiral, **q** lies in the plane of the layers and the rotational axis **n** is an arbitrary unit vector. Various types of spin spirals are distinguished according to the relative direction of **q** and **n** as Fig. 2 represents. The nonrelativis-

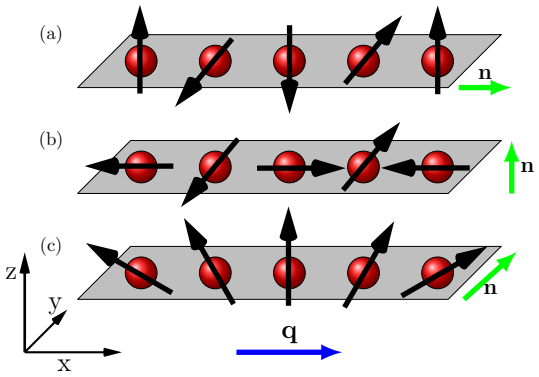

Figure 2: Schematic representation of spin spirals with propagation vector **q** along the x axis and with different rotational axes, **n** . In case of (a), **q** $\parallel$ **n** a Bloch-type spin spiral, while in case of **q** $\perp$ **n** (b) an in-plane or (c) an out-of-plane Néel-type spin spiral are formed. Picture was taken from Phy. Rev. B **100**, 134428.

tic method allows for self-consistent calculations, from which the total energy of the system can be obtained as a function of the spin-wave vector while a relativistic extension of the formalism in first order of the spin-orbit coupling gives an access to the effect of the Dzyaloshinskii-Moriya interactions. A particular advantage of this approach is that the energy related to the Dzyaloshinskii-Moriya interactions can be resolved into layerwise contributions. We demonstrated that the newly developed method properly describes the magnetic ground state of a Mn monolayer on W(001) and that of a Co monolayer on Pt(111). In case of the Co/Pt(111) system, we found that the isotropic interaction between the Co atoms is reduced and the Dzyaloshinskii-Moriya interaction is increased when capped by a Ru layer. In addition, we performed spin-spiral calculations on Ir/Fe/Co/Pt and Ir/Co/Fe/Pt multilayer systems and found a spin-spiral ground state with very long wavelength due to the frustrated isotropic couplings between the Fe atoms, whereas the Dzyaloshinskii-Moriya interaction strongly depends on the sequence of the Fe and Co layers. The results were published in Physical Review B **100**, 134428 (Ref. [7] in the publications list).

We investigated the magnetic interaction of Co layers for $Ru/Co_n/Cu_6/Ru$ and $Pt/Co_n/Cu_6/Ru$ systems, where the subscript means the number of atomic layers, $n = 1 \ldots 6$. We found that the nearest neighbor FM isotropic couplings are increasing in absolute value with the increased number of Co layers for Ru and Pt interface, while the DM couplings are decreasing in absolute value, and the sign of the DM interaction is the same (negative) for all cases, in agreement with the experimental findings. We also examine the magnetic anisotropy of theses systems and found same trend as in the measurements. Moreover, we provided insights into the composition of intralayer and interlayer magnetic interaction contributions and highlighted the effect of the dipole-dipole magnetic interaction on the magnetic anisotropy for thick Co layers. The theoretical results with the experimental findings were summarized in a manuscript. The corresponding manuscript is submitted to the Physical Review Applied and under review, see manuscript: arXiv preprint arXiv:1911.02467 (Ref. [8] in the publications list).

Based on first principles and spin-dynamics simulations we investigated the magnetic properties of Ru_2MnZ (Z=Sn, Sb, Ge, Si) chemically ordered full Heusler compounds for zero as well as finite temperatures. We found frustrated isotropic couplings for all compounds and in case of Z = Si and Sb

a nearest-neighbor biquadratic coupling that favors perpendicular alignment between the Mn spins. From the spin-dynamics simulations we concluded that the biquadratic coupling, in combination with the frustrated isotropic interactions, leads to non-collinear magnetic ground states in the Ru_2MnSi and Ru_2MnSb compounds. In particular, for these alloys we find two distinct, non-collinear ground states which are energetically equivalent and can be identified as $3-q$ and $4-q$ states on a frustrated fcc lattice. Investigating the thermal stability of the non-collinear phase we found that in case of Ru_2MnSi the multiple$-q$ phase undergoes a transition to the single$-q$ phase, while in case of Ru_2MnSb the corresponding transition is not obtained due to the larger magnitude of the nearest-neighbor biquadratic coupling. From the results, a manuscript is submitted to Physical Review Materials and under review, see manuscript: arXiv preprint arXiv:1912.10299 (Ref. [6] in the publications list).

Summary

In summary, the research studies correspond to the plan that was described in the proposal. From the results 6 papers were published and 2 manuscripts were submitted for publication, in case of 4 publications, I am the first author. Budget was not overloaded.