Ab-initio, Analytical and Micromagnetic Study of Low-dimensional Materials and Intermetallic Alloys

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by

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DECLARATION

I hereby declare that the entire work embodied in the thesis entitled ***Ab-initio*, Analytical and Micromagnetic Study of Low-dimensional Materials and Intermetallic Alloys** has been carried out by me. The work presented in the thesis is a bonafide record of research carried out by me under the supervision of **Dr. Arti Kashyap** and is not submitted elsewhere for any degree or diploma. As a general practice, all due acknowledgements have been made wherever the work described is based on the findings of other investigators.

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THESIS CERTIFICATE

I hereby certify that the entire work embodied in the thesis entitled ***Ab-initio*, Analytical and Micromagnetic Study of Low-dimensional Materials and Intermetallic Alloys** has been carried out under my supervision in the Indian Institute of Technology Mandi. Further, no part of it is submitted elsewhere for any degree or diploma.

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ABSTRACT

Low-dimensional magnetism has long fascinated the scientific community because of the new and exotic properties exhibited by materials at this scale. With the demand of increasing data storage and speed of communication, the researchers are trying to invent new types of structures and materials. The competition between various electrostatic interactions responsible for various exotic properties happen at atomic scale which give rise to various zero and finite temperature phenomena governing the static and dynamic behavior of structures. But fabricating and characterizing such materials are time consuming and expensive and demands a good understanding of the material. The analytical and first principle or *ab-initio* calculations in this regard are very useful. They not only reproduce the earlier experimental findings, but also act as a tool to predict the new geometries of alloys and nanostructures. Using these methods, we can look into the properties of numerous configurations which otherwise is not possible with the experiments. The present thesis is devoted to the *ab-initio*, analytical and micromagnetic simulations of the various properties of bulk and low dimensional materials and intermetallic alloys. In chapter 1, we have introduced the types of systems and the properties which we have studied in this thesis. The chapter 2, sheds some light on the theory behind the calculations and the numerical details used for calculating the material properties.

Spin moment, orbital moment and anisotropy are the three essential properties of magnetic structures both in bulk and at nanoscale. Undoubtedly, the relation between spin and orbital moment affect the orientation of the anisotropy axis or the easy axis of magnetization. But surprisingly, very little effort has been put to their noncollinear orientations. Therefore, in chapter 3 and 4, we investigated the effect of orientation of the spin axis on the other magnetic properties and calculated the anisotropy coefficients using least square fitting. For this purpose, we employ three configurations of nanochains (of Rh, Ir, Pd and Pt) namely: linear chains, ladders and zigzag belts. In the ladders and zigzag belts of Pd, the maximum angle between spin and orbital moment is about 23^{o}, but for the monatomic chain, we find an unexpected continuous change in the angle,

covering the whole range between 0^{o} and 180^{o}.

In the zigzag belts of Rh and Ir, we found a strong non-collinearity in the direction of spin and orbital moments in the plane perpendicular to the axis of the chains. The maximum orbital moments for the chains are along the axis of the chains. A ferromagnetic coupling has been observed in the zigzag chains of Rh as well as Ir. A key feature of the study is to observe the alteration of the occupation states on changing the direction of magnetization vector which results in switching the direction of MAE of Ir. This research shows how nanostructuring can be used to control and tune the anisotropic properties which will be really helpful for magnetic and spintronic applications. Magnetic anisotropy is a crucial parameter for the permanent magnets. As we know that, most of the permanent magnets are rare-earths which are limited and expensive. Hence, new materials and the ways has to be formed to make the cheap magnets.

To increase the magnetic properties of the materials, there are two ways: First, improving the intrinsic properties by changing the chemical composition and crystal structure. Doping of Fe and Co with 4*d* and 5*d* transition-metal atoms is one approach towards new cost effective and rare-earth free permanent magnetic materials. The rationale is to combine the high spin-orbit coupling and anisotropy of the heavy elements with the high magnetization of Fe and Co. Using First-principle methods, in chapter 5, we investigated the intrinsic magnetic properties of intermetallic alloys of the type XM_n, where *X* is a 4*d* or 5*d* element and *M* is Fe or Co. Emphasis is on the hexagonal C14 Laves-phase 1:2 and 1:5 alloys, the latter crystallizes in the $CaCu_5$ structure. These series are of interest in permanent magnetism from fundamental and practical viewpoints, respectively. In the former case, the unit cells form a prototypical motif where a heavy atom with high spin-orbit coupling and magnetocrystalline anisotropy is surrounded by many somewhat smaller *M* atoms with high magnetization. The latter are Laves-phase derivatives of renewed interest in permanent magnetism. In this chapter, we predicted magnetic moments, magnetizations, and anisotropies, as well as the calculation of formation energies. The results are analyzed across the 4*d* and 5*d* series, especially with respect to hybridization effects between 3*d* and 4*d*/5*d* bands.

The another way to increase the magnetic properties is by improving the extrinsic properties through nanostructuring. But it is essential to have an understanding of size and geometrical dependence of main figure of merits (e.g. energy product, hysteresis loop etc.) for hard magnetic nanostructures. As mentioned in [3], it is possible to improve the energy product by 1000 kJ/m^3 through nanostructring. In chapter 6, we investigated analytically and through micromagnetic simulations, the various figure of merits and the geometries of hard soft nanocomposites down to small feature sizes.

In chapter 7, we investigated the effect of strain on the various electronic properties of organic two dimensional layer of Carbon (graphene) and found an increase in the effective mass due to which the mobility of the electrons in graphene sheet decreases.

Chapter 8 concludes the work done in this thesis along with the limitations and the future directions.

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