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Reprint of Multiscale model approaches to the design of advanced permanent magnets☆,☆☆

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ABSTRACT

We describe the process of multiscale modelling of magnetic materials, based on atomistic models coupled parametrically to micromagnetic calculations. At the atomistic lengthscale we use Spin Dynamics (SD) to study switching mechanisms, using structures predicted by Molecular Dynamics. The process is completed using SD to calculate the cell size and temperature dependent parameters for micromagnetic calculations. We demonstrate an unusually weak cell size scaling for $Nd_2Fe_{14}B$ and demonstrate numerically the existence of atomic scale Barkhausen jumps during magnetization switching. Scaling of magnetic properties is shown to be important in micromagnetic calculations of hysteresis, especially considering variation in micromagnetic cell size.

1. Introduction

Micromagnetics is the computational tool of choice for permanent magnet modelling. However, following the proposal of exchange spring magnets by Kneller and Hawig [1] to optimise magnetic properties by coupling a (soft) high moment material with a (hard) low moment material, permanent magnets have been increasingly designed at the nanoscale to achieve the desired functionality. The high remanent magnetization arises due to the high Fe content in the alloy, while the origin of the coercivity combines intrinsic properties of the hard phase (the anisotropy) and extrinsic properties which are intimately tied to the material microstructure [2-5]. Understanding the links between material properties, microstructure and magnetic properties is vital for the optimisation of magnetic properties including the energy product.

Micromagnetic models can be used to investigate the magnetic behaviour of such systems, and remain vital for large scale calculations: for example micromagnetic modelling shows the potential advantages of complex designs such as the 'Battenburg' structure [6]. However, they do not have access to the effect of the detailed interface structure at the atomic level and its effect on intrinsic magnetic properties, which might be expected to have a bearing on the model predictions. A further difficulty is the scaling of magnetic properties with cell size. This was first investigated by Dobrovitski et al. [7] and Grinstein and Koch [8]. The coarse-grained micromagnetic variables tend to lead to an over-estimate of T_c which is shown to be removed by a renormalisation approach [8]. Essentially this means that the anisotropy and saturation magnetization and also the exchange stiffness constant must be taken as cell-size (L) and temperature dependent, i.e., $K = K(T,L)$, $M_s = M_s(T,L)$ and $A = A(T,L)$. Here we show that these variations can be calculated using atomistic model simulations, linking atomistic and micromagnetic models in a multiscale approach. This is especially important for automotive and other applications involving high temperature operation.

In a world with infinite compute resources one could imagine quantum-based materials simulations, however this is unlikely to be achievable in the foreseeable future. The only feasible approach is to

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couple and link the formalisms associated with each lengthscale (an overview of this process is shown in Fig. 1). These are

1. *Ab-initio calculations*. These involve the solution of the quantum mechanical problem of interacting electrons. The most common formalism: Density Functional Theory (DFT), essentially transforms the many-body Schrödinger equation into a problem of determining local electron densities. DFT is highly successful in predicting material properties, at least to the extent of interatomic potentials: a central part of Molecular Dynamics modelling to be described in Section 2. However, the energies associated with important magnetic properties, especially anisotropy energies, are very small. In addition, the determination of anisotropy values by DFT requires relativistic corrections and is a rather difficult and specialised area. Further limitations of DFT are that firstly it is strictly valid at zero Kelvin and secondly CPU requirements limit the calculations to a few hundred atoms.
2. *Atomistic spin dynamics (SD) calculations*. SD models make the (adiabatic) approximation of a classical atomic spin of fixed length able to rotate freely. The exchange interaction between spins is generally taken as being of Heisenberg form. In principle, the first stage of the multiscaling process is to determine the important parameters (spin values, exchange and anisotropy) from the *ab-initio* calculations. Importantly, the atomistic approach allows a thermodynamic treatment of the spin system leading to predictions of the temperature variation of magnetic properties and also of dynamic behaviour at elevated temperatures. In addition to providing a powerful and predictive approach in its own right, the atomistic approach provides the basic input to micromagnetic models in the form of temperature and cell size dependent properties. Atomistic models are currently limited to systems of 10^6 to 10^8 spins and timescales of up to ~100 ns.
3. *Micromagnetic calculations*. These extend the lengthscale and timescale of calculations by orders of magnitude, but lose direct connection to the underlying physics expressed by the *ab-initio* models. The final link in the multiscale chain is to introduce atomistically calculated values of temperature and cell-size dependent properties into the model.
4. *Molecular dynamics (MD)*. While 1–3 represent the magnetic aspects of the multiscale chain, the use of interfaces to provide the functionality of modern magnetic materials requires special consideration. For example, the hard/soft interfaces in exchange spring magnets are likely to influence the magnetic properties in a complicated way due to, for example, stresses arising from lattice mismatch. This is the province of MD, which uses classical equations of motion driven by *ab-initio* parameterised force fields to predict structures of materials and interfaces. Here we use MD calculations of the $Nd_2Fe_{14}B/\alpha$-Fe interfaces to predict interface anisotropy values and use them in atomistic calculations of domain wall propagation across the interface.

The multiscale approach to magnetic materials simulations was first proposed for FePt due to its importance as the material of choice for ultra-high density magnetic storage based on heat assisted magnetic recording (HAMR) [9]. The basic approach, described by Kazantseva et al. [10], is firstly to use *ab-initio* methods to determine the main magnetic parameters [11]. These are used to determine the main parameters for the atomistic spin model. Subsequently, the atomistic model is used to determine the temperature dependent parameters for macrospin models of magnetic recording. Although pictured here as an intermediate model linking *ab-initio* and macroscopic models, atomistic approaches are important in their own right in essentially merging the quantum (*ab-initio*) and thermodynamic regimes, leading to, for example, important insights into HAMR and also the prediction of thermally assisted magnetization switching [12,13]. The multiscale approach is in an early stage of development for permanent magnets. As will be discussed later, this is at least partially due to the difficulty of obtaining reliable *ab-initio* information on the magnetic properties of rare-earths, which necessitates the determination of magnetic parameters by fitting to experiment. Here we outline the overall approach, describing first the molecular dynamic model of interface structures. We then outline the SD model development and the techniques for determination of the important intrinsic atomic parameters, including the calculation of the anisotropy arising from symmetry breaking due to stresses in the soft phase at an interface between $Nd_2Fe_{14}B$ and α-Fe.

2. Molecular dynamics model of interface properties

To be able to understand solids and calculate material parameters such as magnetocrystalline anisotropy it is necessary to use molecular dynamics and first principle studies for the first, in particular molecular force fields to extract physical and chemical properties at bulk and thin film level.

This was shown in our previous work [14,15] where we investigated metallic Nd grain boundary phases and fcc and hcp Nd-rich phases by using Morse potentials. Recent investigations have shown, if there are different Nd-O grain boundary phases depending on the oxygen content in the production process a mixed MD approach has to be used.

The atomistic structures of the systems involved are simulated using energy minimisation codes such as the GULP code [16]. As the systems can be comprised of both metallic (e.g. $Nd_2Fe_{14}B$ fcc-Nd, dhcp-Nd) and oxide (e.g. Nd_2O_3-hP5, Nd_2O_3-cI80 and NdO) phases, one has to employ different empirical potential models for the interatomic interactions for each system. In the metallic systems it is possible to ignore the Coulombic forces in the system as each atom is formally charge neutral. However, in the oxide phases the atomic centres carry a formal charge, as a result there is a transfer of electrons from the metal atom to the oxygen, due to their different electronegativities. Therefore a Coulomb term must be calculated from these interactions. A good candidate to simulate oxide interactions is the Buckingham potential. This potential, that describes the Pauli repulsion energy and the van der Waals energy, has the form

$$\Phi_{sr}(r_{ij}) = A_{ij}\exp\left(\frac{r_{ij}}{\rho_{ij}}\right) - \frac{C_{ij}}{r_{ij}^6} \quad (1)$$

where A_{ij} and ρ_{ij} describe the repulsion interaction, related to the electron number and electron density, while C_{ij} describes the attractive energy. The results of such a simulation are shown in Fig. 2. The parameters in our model were adapted from a La-O potential and fitted to the inter-atomic separation of Nd-O. This model, which incorporates a core-shell separation of the charge [17] on oxygen that accounts for the polarizability of the oxygen atoms, is validated against experimental data for Nd_2O_3 crystal structures.

The metallic phase is modelled with a Morse potential with the form

$$\Phi_{sr}(r_{ij}) = D_{ij}\left[1 - \exp\left(-\beta_{ij}\left\{r_{ij} - r_0\right\}\right)\right]^2 \quad (2)$$

where D_{ij} is the disassociation energy of the bond and β_{ij} is a variable parameter that can be determined from spectroscopic data. This type of model is particularly useful in chemical systems to model bonded covalent interactions. The parameters for these interactions are taken from the work of Chen et al. [18].