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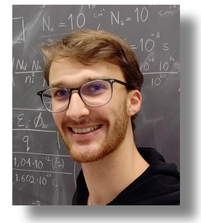
Finding New Two Dimensional Magnets by Evolutionary Structure Search Algorithms and Ab Initio Theory



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Summary

The structure search software USPEX is used to find stable states of two-dimensional iron, cobalt and nickel. The aim is to find single or double atomic layered magnets. Structures of interest are examined individually with greater accuracy in the material modelling program VASP. All possible linear magnetic configurations are studied in order to find the lowest energy state. The final results describe the most stable species' compositions along with their density of states, leaving some other material properties to be calculated in the future.

Ab Initio

Whilst the two primary programs used in this project are mostly regarded as black boxes, requiring only a few input files and then producing visualisable output files, it can be said that the underlying algorithms are based on so called *ab initio* concepts. This latin term means “calculated from first principles”. Instead of extrapolating results from empirical data the programs are built solely upon fundamental laws of physics, primarily the density-functional theory.

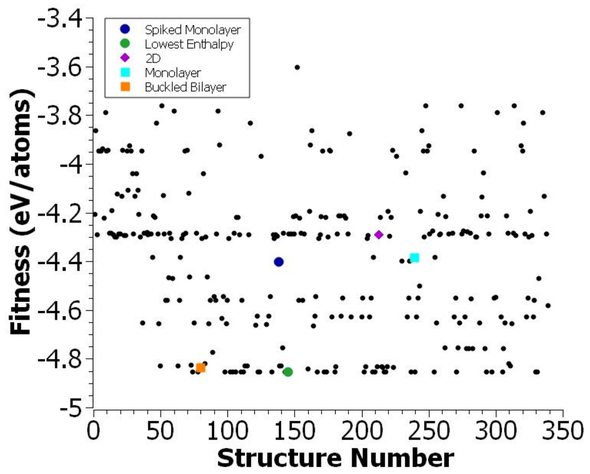
USPEX

USPEX Computational Materials Discovery

Universal Structure Predictor: Evolutionary Xtallography is a software for predicting structures of atoms.

USPEX performs a structure search i.e. calculates larger possible structures from an initial building block of atoms. Our unit cells consisted purely of iron, cobalt , and nickel, respectively. Within these structures USPEX also calculates what were the main quantities of interest here: the energies of the atoms, as well as their magnetic moment and as such the spin configurations of their electrons.

Eventually, a single USPEX run per element yielded all data for us to visualize and interpret. Further processing of these structures was however necessary.



The energy per atom in each structure of nickel; a lower energy corresponds to a more stable structure. While the structure number is assigned arbitrarily by USPEX, the graph shows that there are some energy levels that are more common, indicating a locally more stable state.

VASP

The Vienna Ab initio Simulation Package is used to evaluate material properties. It has a plethora of functions but of interest for this project is the program’s ability to perform ionic relaxations, enthalpy calculations and approximations of linear magnetic properties. A key equation in these calculations is a simplification of the Schrödinger equation for multiple electrons:

$$\hat{H}\Psi = \left[\hat{T} + \hat{V} + \hat{U}\right]\Psi = \left[\sum_{i=1}^{N}\left(-\frac{\hbar^2}{2m_i}\nabla_i^2\right) + \sum_{i=1}^{N} V(\mathbf{r}_i) + \sum_{i<j}^{N} U(\mathbf{r}_i, \mathbf{r}_j)\right]\Psi = E\Psi$$

USPEX is interfaced with VASP in order to perform its material property calculations during the structure search. However, after running USPEX to find our structures of interest, we proceeded with pure VASP calculations on our structures, for greater accuracy of some material properties, as well as additional properties for each species.

Results

17 different iron, nickel and cobalt structures of interest from the USPEX evolutionary search were selected to be investigated further in VASP. For these structures all 552 possible magnetic configurations were examined. These structures were ionically relaxed and then enthalpies and density of states (DOS) were calculated in two separate stages. An example output from the structure relaxation can be seen in the figure below and the corresponding DOS result in the graph to the right.

Conclusions

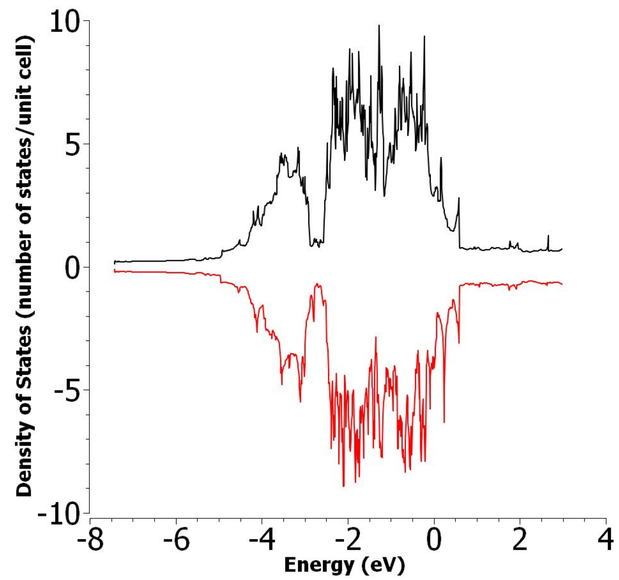
Theoretically, our USPEX calculations predict that purely two dimensional structures of the three metals are indeed possible, however these structures are not among the most stable of the structures found. The refined VASP calculations bring some uncertainty to the results seeing some ionic relaxations caused bulking of previously mono or bilayered structures, suggesting that they may in fact not have been stable states. With this said, there still exists some magnetic configurations that yielded 2-dimensional structures that may very well be candidates for actual synthesisation.

Future Work

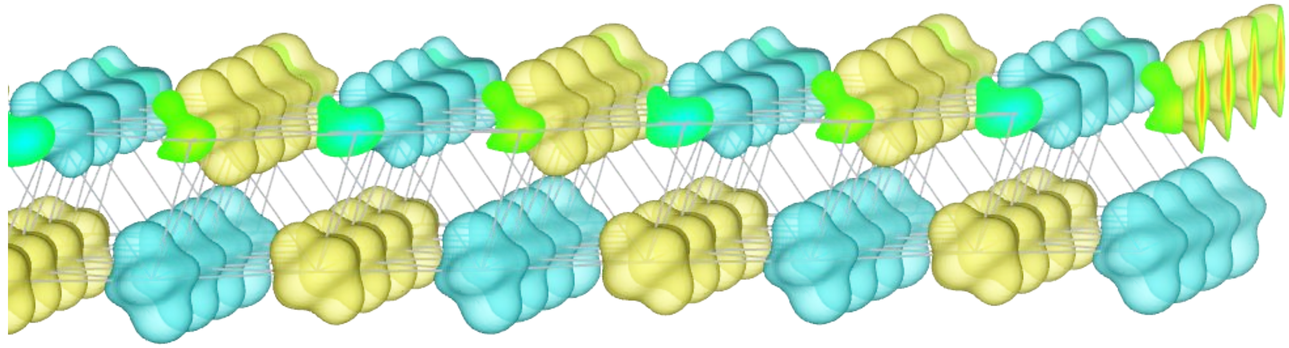
An unsolved problem we encountered was the fact that after relaxation with VASP of the USPEX structures, they often changed into a new structure.

As a continuation of the project, one may delve deeper into the different calculations VASP is able to perform on the structures found by USPEX. These include e.g. band structure calculation, magnetic anisotropic calculation, and Curie temperature.

Hopefully our findings as well as continued work into topics above can be used in future studies where the compounds might even be synthesised.



Total density of states for spin up (black) and spin down (red) states at varying energies in a bilayered nickel sample.



Magnetic configuration of a nickel sample. In this case the result was an antiferromagnetic substance. The interatomic wire frame shows the metallic bonds. The irregular shapes shown corresponds to the isosurfaces of the charge density.

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