# Tuning the electronic and magnetic properties of Heusler alloys: A theoretical and experimental investigation

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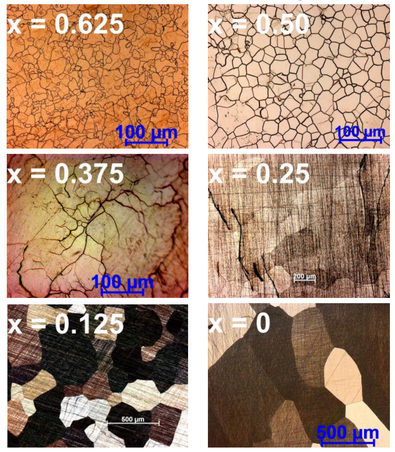
## Experimental Motivation

- Heusler alloys are a large family of interesting materials with applications in information storage and spintronics.
- Some of these alloys exhibit half-metallic characteristics, making them more attractive for device applications.
- Electronic and magnetic properties of these alloys can be extensively tuned through chemical substitution.
- Studied $Fe_{3-x}V_xGe$ (cubic and hexagonal), $Fe_{3-x}Cr_xGe$ (hexagonal), $Co_{2-x}V_xFeGe$ (cubic) and $Co_2Fe_{1-x}V_xGe$ (cubic).

Optical micrograph of cubic $Fe_{3-x}V_xGe$.

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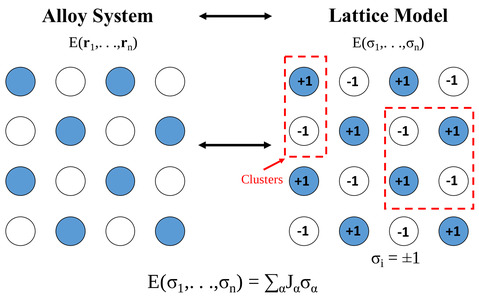
## Methodology




- We used density functional theory (DFT) along with the cluster expansion formalism to study Heusler alloyed structures.

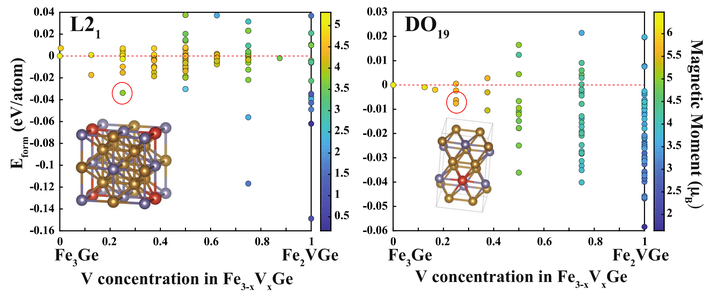
$$E[\rho] = T_s[\rho] + V_{ext}[\rho] + V_H[\rho] + E_{xc}[\rho] + E_U[\rho]$$

- For Co-based alloys, we applied DFT+U correction to treat strongly localized *d* electrons.
- U parameter is the energy bound on a Hubbard-like correction term to the energy functional, determined semi-empirically.

## Acknowledgements

We acknowledge the UMBC High Performance Computing Center and funding from NSF Grant DMR-1726213, DMR-1508680, and DMREF-1235396.

## $Fe_{3-x}V_xGe$

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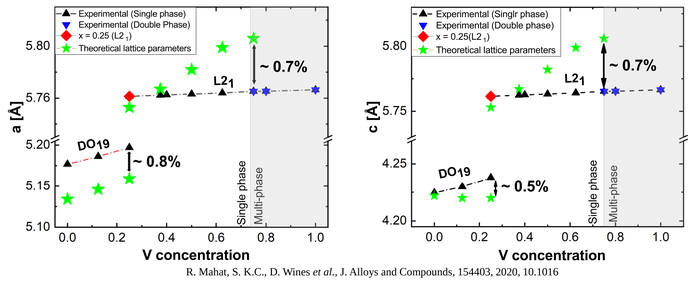
We used DFT as an input to the cluster expansion to search for energetically stable alloys in the cubic and hexagonal phases. Each data point represents the energy of a random structure created.

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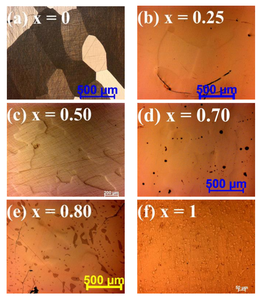
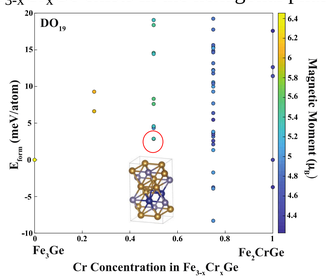
After structural relaxation, our values for lattice constant are in good agreement with measured values.

## $Fe_{3-x}Cr_xGe$



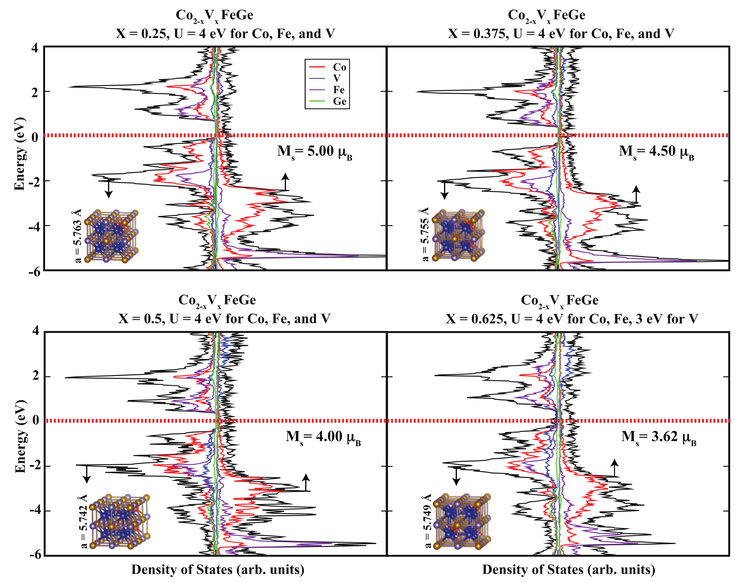

$Fe_{3-x}Cr_xGe$ exists in the hexagonal phase.



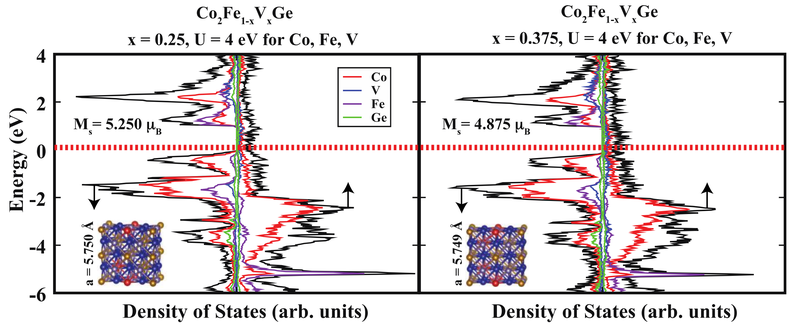

| $x$ | Crystal structure | Experimental lattice (Å) | Theoretical lattice (Å) | Expt. $M_s$ at $T = 5K$ ($\mu_B/f.u.$) | Theor. $M_s$ ($\mu_B/f.u.$) |
|---|---|---|---|---|---|
| 0 | $DO_{19}$† | $a = 5.1768(2)$ | $a = 5.134$ | 6.55(9) | 6.48 |
| | | $c = 4.2246(3)$ | $c = 4.222$ | | |
| 0 | $L1_2$‡* | $a = 3.6667(1)$ | $a = 3.638$ | 6.55(9) | 6.45 |
| 0.25 | $DO_{19}$† | $a = 5.1885(2)$ | $a = 5.144$ | 5.79(5) | 5.98 |
| | | $c = 4.2260(3)$ | $c = 4.202$ | | |
| 0.50 | $DO_{19}$† | $a = 5.1934(2)$ | $a = 5.153$ | 5.11(5) | 5.52 |
| | | $c = 4.2269(2)$ | $c = 4.195$ | | |
| 0.70 | $DO_{19}$† | $a = 5.1958(1)$ | $a = 5.155$ | 4.44(7) | 4.98 |
| | | $c = 4.2275(3)$ | $c = 4.195$ | | |

† annealed at 1000°C for 15 days, ‡ annealed at 650°C for 25 days, * multi-phased

## $Co_{2-x}V_xFeGe$




## $Co_2Fe_{1-x}VGe$




The DFT+U corrected Density of States (DOS) confirm the half-metallic nature of these alloys. Calculated magnetic moments match experimental and Slater-Pauling values.

## Conclusions

Properties of Heusler alloys can effectively be tuned by varying chemical substitution. Our results are in good agreement with experiment and confirm half-metallic properties of $Co_{2-x}V_xFeGe$ and $Co_2Fe_{1-x}V_xGe$.