High Temperature Magnetic Material with Temperature Capability Greater Than 500°C

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NASA Aeronautics Research Mission Directorate (ARMD)

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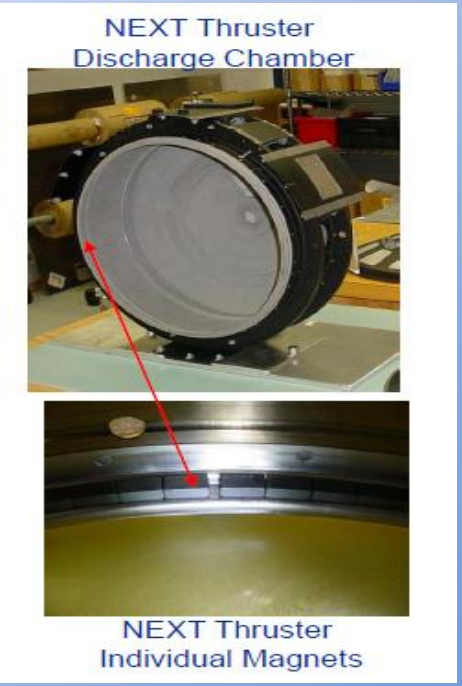
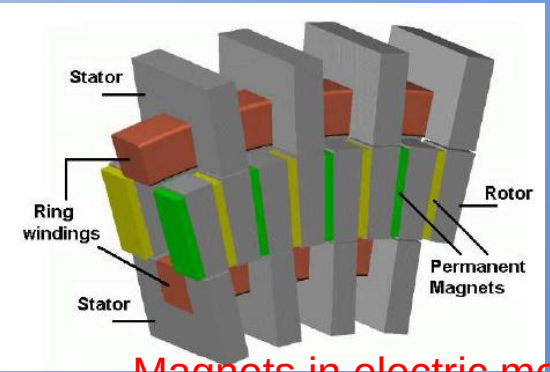
Relevance and Background

- High temperature magnets enable higher temperature operation of electric motors without demagnetization
  - Can lead to higher power density of electric motors for electric aircraft application
- High temperature magnets also essential for increasing power density of ion thrusters used for space electric propulsion
- Need to develop magnets with greater than 500°C temperature capability
- State-of-the-art high temperature Sm-Co magnet has temperature capability of 350-400°C for long durability
  - Does not have high magnetic energy product
- High energy product magnets (NdFeB) used in electric magnets have temperature capability on the order of 150°C





Magnets in electric motor

Scope and Technical Challenge

Innovation:

- Use computational tools to identify candidate high temperature magnet materials
- Fabricate and measure magnetic properties for promising high temperature magnet materials

Technical Challenge:

- Fabricate permanent high temperature magnets which retain strength and maintain stability as a function temperature and time

How to model a high temperature magnet

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Determine
Is the material ferromagnetic?
Magnetic saturation
Magnetization as a function of temperature
Mechanical properties

Curie Temperature

Model domain structure as a function of temperature, stress, and time

Model microstructural stability with respect to time, temperature and stress

Atomic scale

Meso-scale

Macro-scale

Density functional theory

- Density functional theory (DFT) is a quantum mechanical modeling method used in physics and chemistry to investigate the electronic structure (principally the ground state) of many-body systems, in particular atoms, molecules, and the condensed phases.

- With this theory, the properties of a many-electron system can be determined by using functionals, i.e. functions of another function, which in this case is the spatially dependent electron density.

Many types of quantum chemistry and solid-state physics software

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Package	License[†]	Lang.	Basis	Periodic[‡]	Mol. mech.	Semi-emp.	HF	Post-HF	DFT
ABINIT	GPL	Fortran	PW	3d	Yes	No	No	No	Yes
ACES II	GPL	Fortran	GTO	No	No	No	Yes	Yes	Yes
ACES III	GPL	Fortran/C++	GTO	No	No	No	Yes	Yes	No
ADF	Commercial	Fortran	STO	Any	Yes	Yes[4]	Yes	No	Yes
Atomistix ToolKit (ATK)	Commercial	C++/Python	NAO/EHT	3d[9]	Yes	Yes	No	No	Yes
BigDFT	GPL	Fortran	Wavelet	Any	Yes	No	Yes	No	Yes
CADPAC	Academic	Fortran	GTO	No	No	No	Yes	Yes	Yes
CASINO (QMC)	Academic	Fortran 95	GTO / PW / Spline / Grid / STO	Any	No	No	Yes	Yes	No
CASTEP	Academic (UK) / Commercial	Fortran	PW	3d	Yes	No	Yes[5]	Yes	Yes

Approach

- Physics-based computational simulations (based on density functional theory)using the **Cambridge Serial Total Energy Package** (CASTEP) were used to identify candidate materials such based on lattice parameters and its electronic configuration as possible high temperature magnet.

- Promising materials are fabricated by electric arc-melting of the metal alloy components followed by powder metallurgy processing and testing the high temperature magnetic capability.

"First principles methods using CASTEP"
Zeitschrift fuer Kristallographie 220(5-6) pp. 567-570(2005) S. J. Clark, M. D. Segall, C. J. Pickard, P. J. Hasnip, M. J. Probert, K. Refson, M. C. Payne

- Hewlett Packard HP Z820 workstation 2 Intel Xenon processors allowing 32 cores for calculation with 32 GB of random access memory.
- Generalized Gradient Approximation (GGA) using ultra soft potentials, Perdew-Burke-Ernzerhof for Solids (PBEsol)
- Cut-off energy 300 eV
- Spin polarized

Flowchart of the sequence of computational tools

Select the crystal structure and symmetry elements

Populate the crystal with the appropriate atoms

Optimize the geometry of the crystal by minimizing the forces and energies between atoms on the lattice

Things calculated from the models

- Prediction of the lattice parameter
- Mass density
- Interatomic distances
- Magnetic moment per unit cell

- With further computations

  - Full elastic tensor
  - Electronic band structure
  - Phonon dispersion and spectra

Basis for the determination of magnetism

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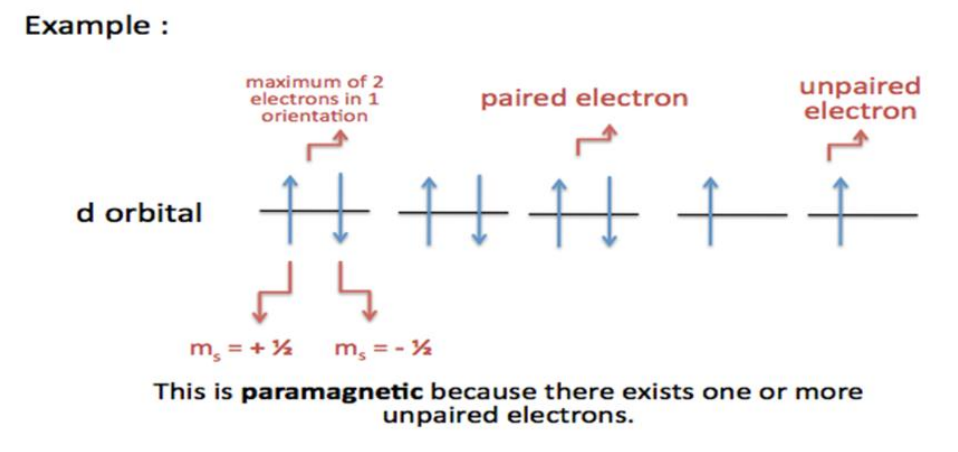
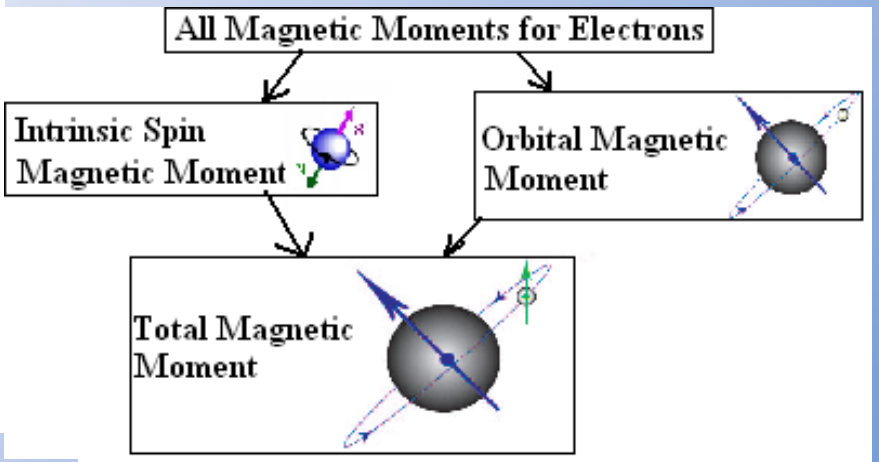




Pseudo atomic calculation performed with the outer electron shells

For example PrGdCoIr

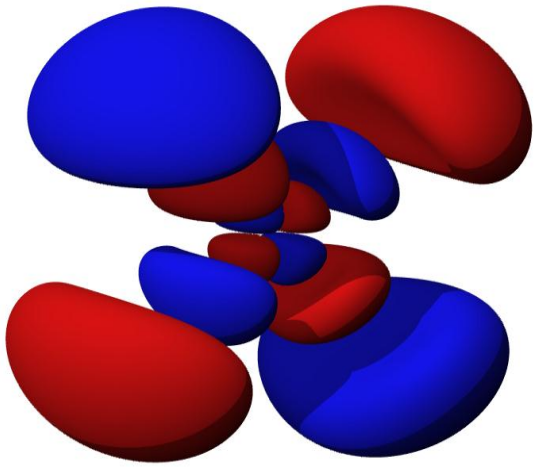
We use

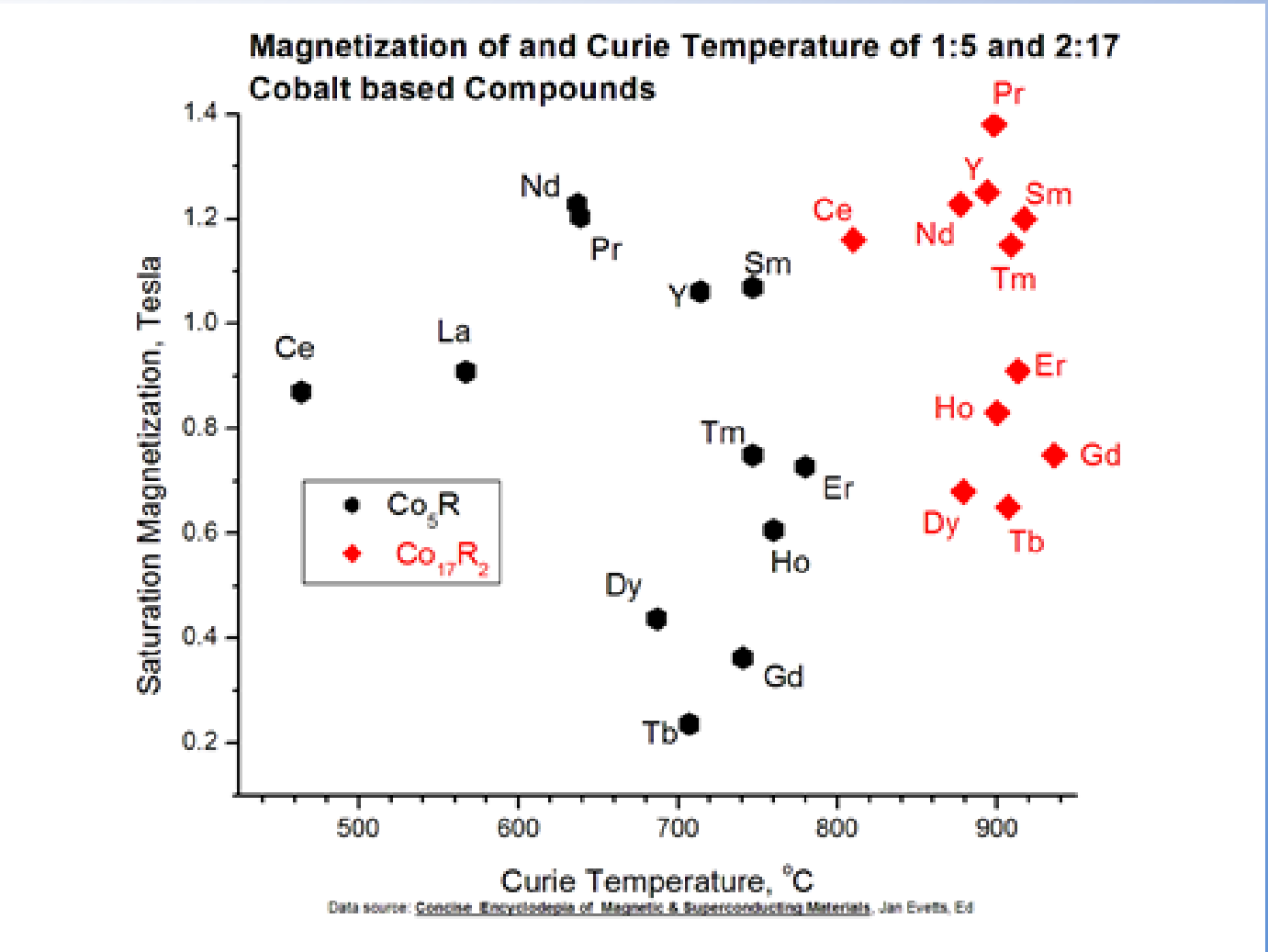
Co $3d^7\ 4s^2$

Pr $4f^3\ 5s^2\ 5p^6\ 6s^2$

Gd $4f^7\ 5s^2\ 5p^6\ 5d^1\ 6s^2$

Ir $5d^7\ 6s^2$




Magnetization of and Curie Temperature of 1:5 and 2:17
Cobalt based Compounds
Saturation Magnetization, Tesla
Curie Temperature, °C
1.4
1.2
1.0
0.8
0.6
0.4
0.2
500
600
700
800
900
Ce
La
Nd
Pr
Y
Sm
Tm
Er
Ho
Dy
Gd
Tb
Co5R
Co17R2
Pr
Y
Sm
Ce
Nd
Tm
Er
Ho
Gd
Dy
Tb
Data source: Concise Encyclopedia of Magnetic & Superconducting Materials, Jan Evetts, Ed

Input Data

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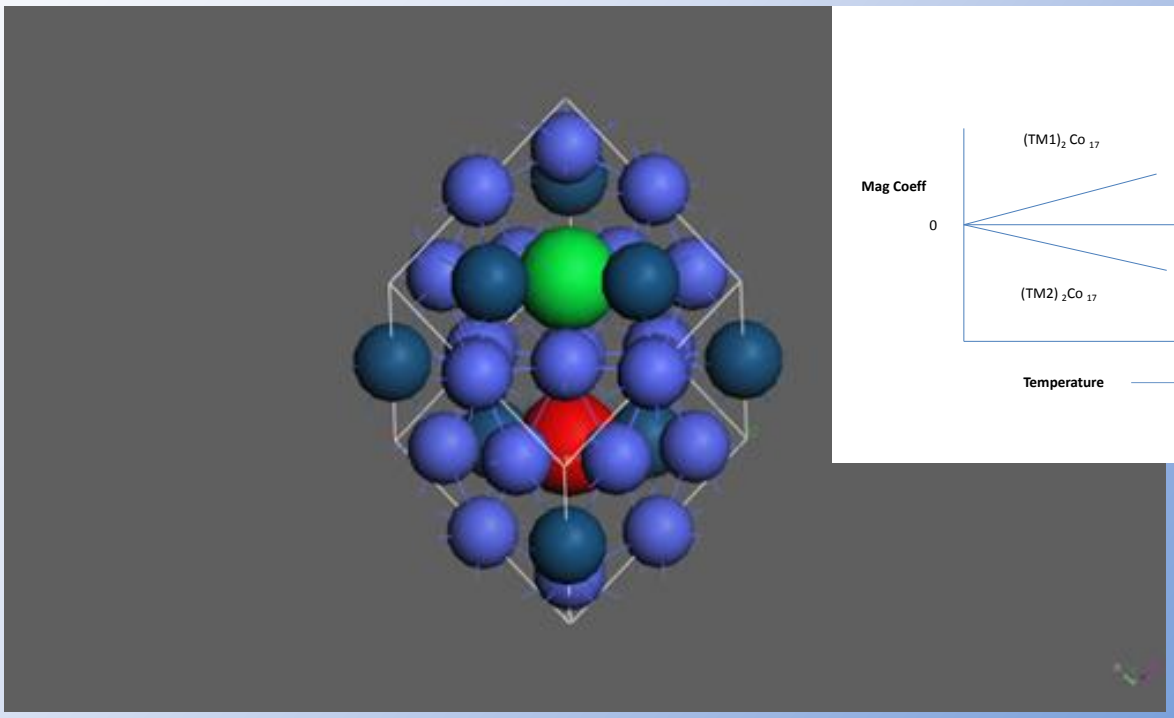
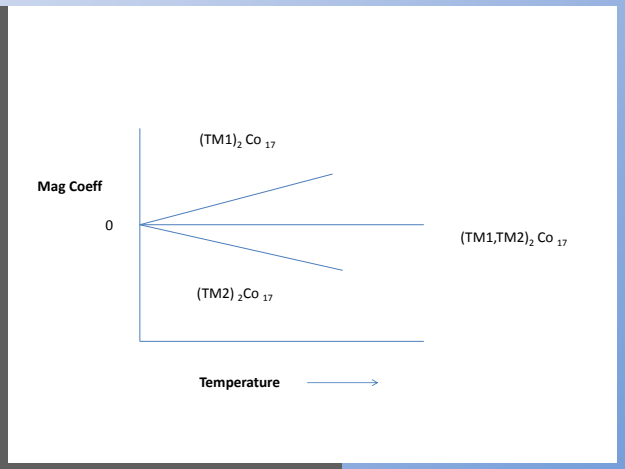
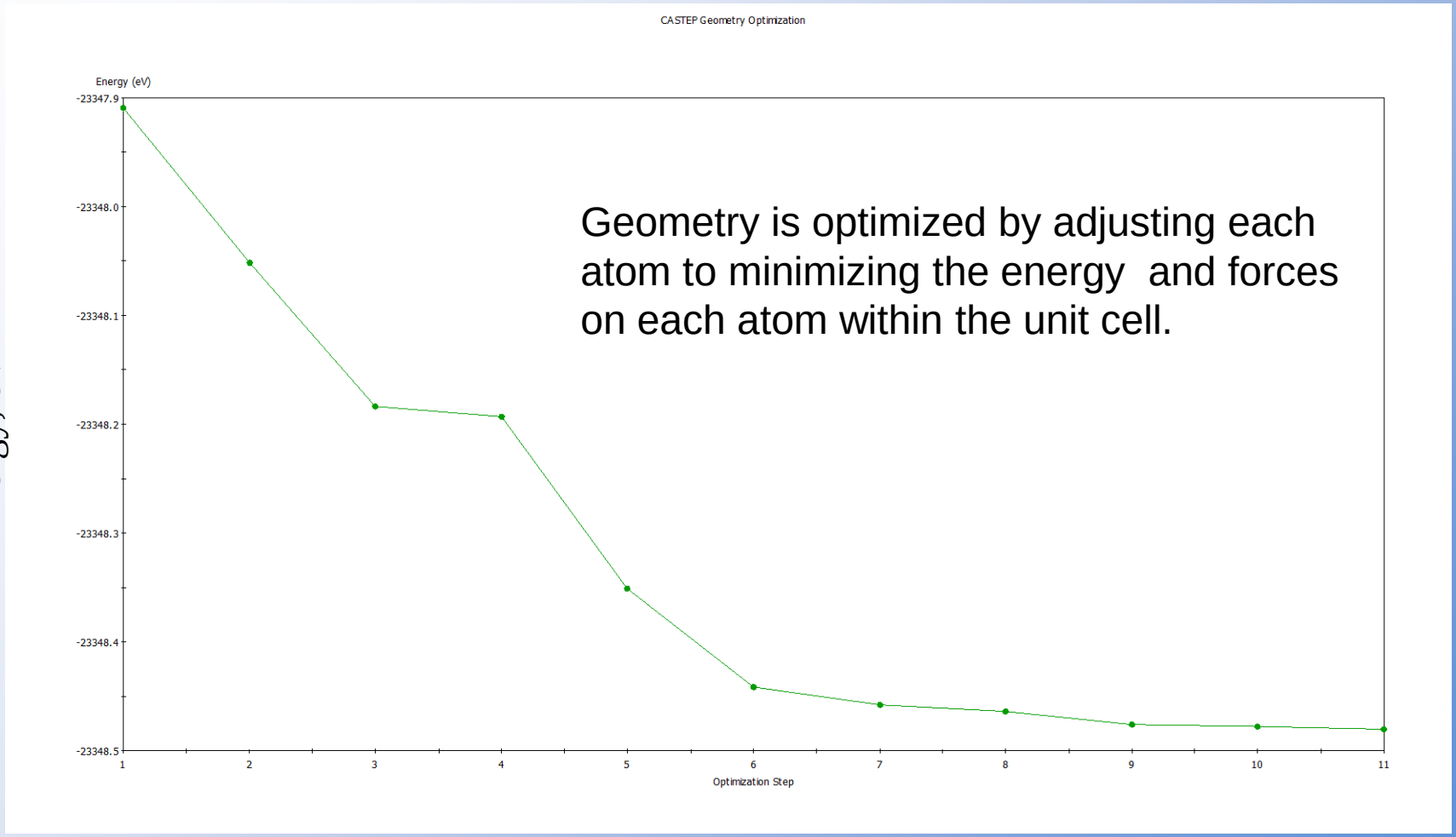




Metallic alloy samarium (red), praseodymium (green), cobalt (light blue), iridium (dark blue) system where atoms are rendered as spheres with radii related to van der Waals' radii of the respective atoms.



Geometry is optimized by adjusting each atom to minimizing the energy and forces on each atom within the unit cell.

Energy, eV

Optimization Step

Calculated density of electron states

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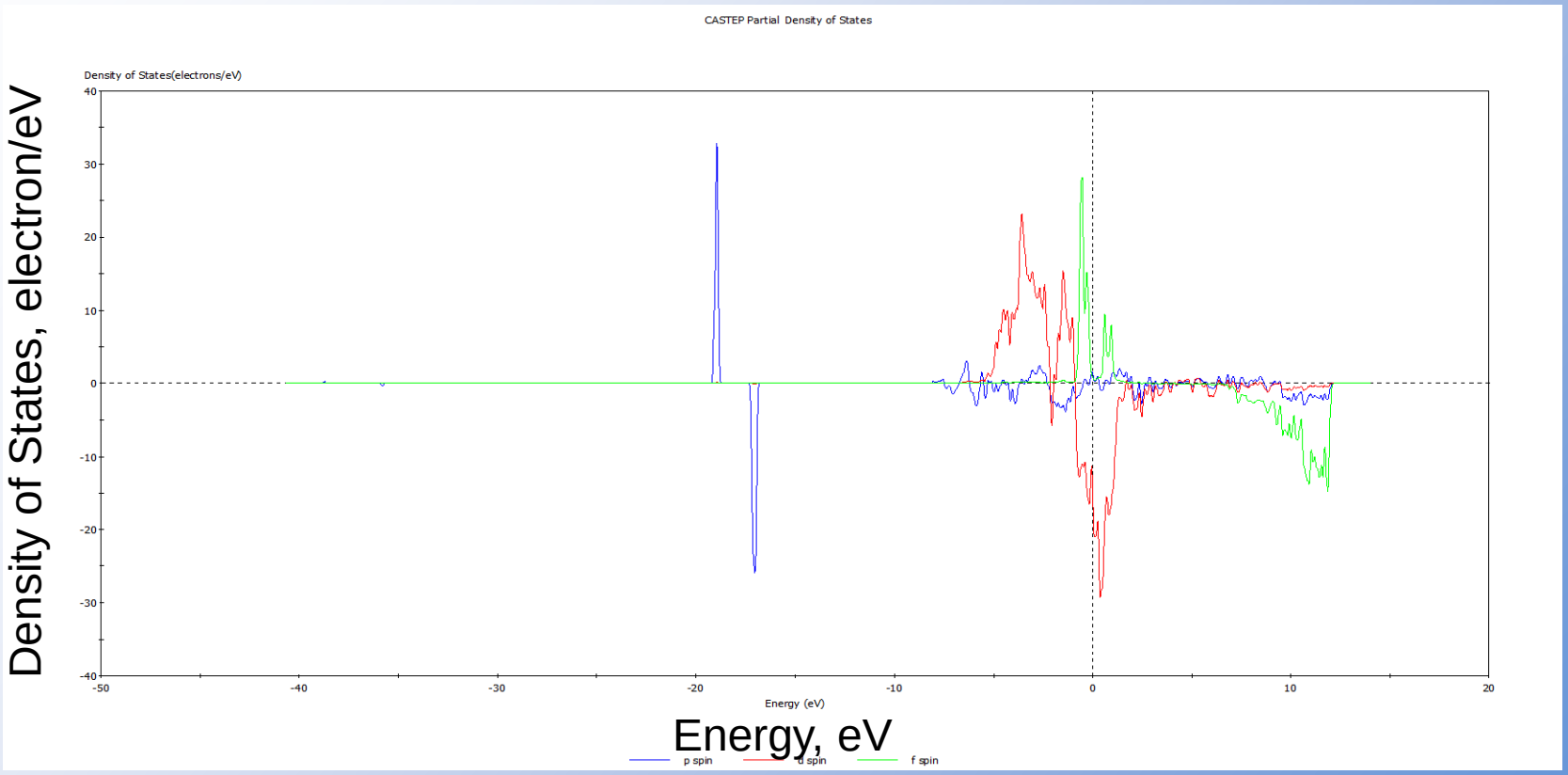

(orbitals: d-red, f-green, p- blue) spin decomposed and symmetry-resolved illustrating the difference between the spin up (positive) and spin down (negative) electron states, which give rise to the ferromagnetism of this alloy.

Property predictions for selected alloys

(Literature experimental values in bold)

Alloys	Bulk Modulus, GPA	Density, g/cm3	Lattice Parameter, Angstrom	Magnetic Moment, Bohr Magneton
Sm_5Ir_3	31	10.3	a=b=11.4 (**11.03**) c= 6.8 (**6.372**)	122
Sm_2Ir_{17}	354	10.69	a=6.47	36.5
Gd_2Co_{17}	239	9.02 (**8.87**)	a=b= 8.275 (**8.349**) c= 12.068 (**12.224**)	76
$GdPrCo_{17}$	260	8.38	a=b= 8.41 C=12.22	36
SmPrCoIr		11.00	a= b=8.3 c= 12.01	32

Tri- Arc furnace for preparing magnetic alloys

University of Utah Facilities

Vibrating Sample Magnetometer

High Vacuum W-resistance Furnace for Heat Treating Magnetic Alloys

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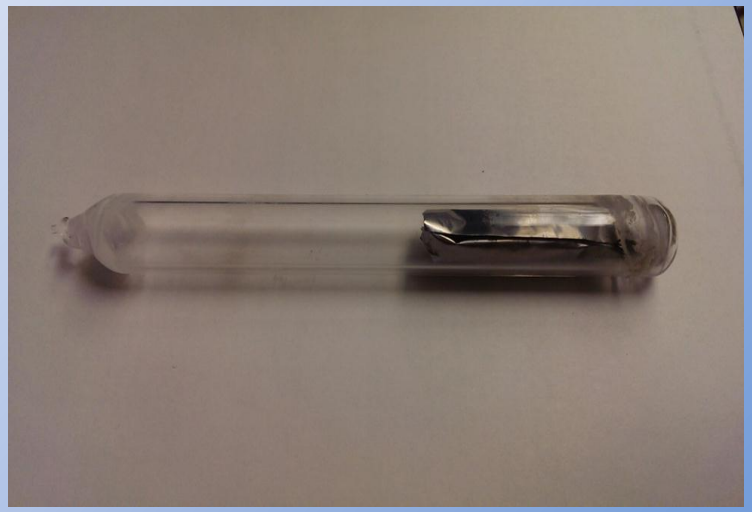
Composition		Samarium	Erbium	Cerium	Aluminum	Manganese	Tungsten	Praseodymium	Gadolinium	Cobalt
(Pr,Gd)Co17	Nominal Composition wt %	*	*	*	*	*	*	10.8	12.1	77.1
13 grams	Analytical Results wt%	*	*	*	*	*	*	11.2	12.7	76.1
(Sm,Gd,Er,Ce) (Co,Al,Mn)	Nominal Composition wt%	4.18	5.93	3.94	0.697	1.545	*	*	5.06	78.6
25.1 grams	Analytical Results wt%	3.7	5.8	4	0.7	1.2	0.13	*	5	79.5
(Ce,Er)Co17	Nominal Composition wt%	*	12.79	10.75	*	*	*	*	*	76.5
15.16 grams	Analytical Results wt%	*	12.3	11	*	*	*	*	*	76.6

Sample Preparation

- **Pellets (6 mm diameter x 6 mm high) were formed using a polyethylene glycol binder**
- **Alloy powders were dissolved initially in acetone as a lubricant/binder (1-1.5 wt. %)**
- **Cold pressed with 1500 kg/cm ^{2}**
- **Heated under argon gas to 325°C to remove the binder**
- **12 or 13 mm Id and thick walled quartz tubes sealed using oxyacetylene torch**
- **Green body pellets were then sealed under argon in a quartz vial and sintered at 1190°C for several hours**



(Sm,Gd,Er,Ce) (Co,Al,Mn)

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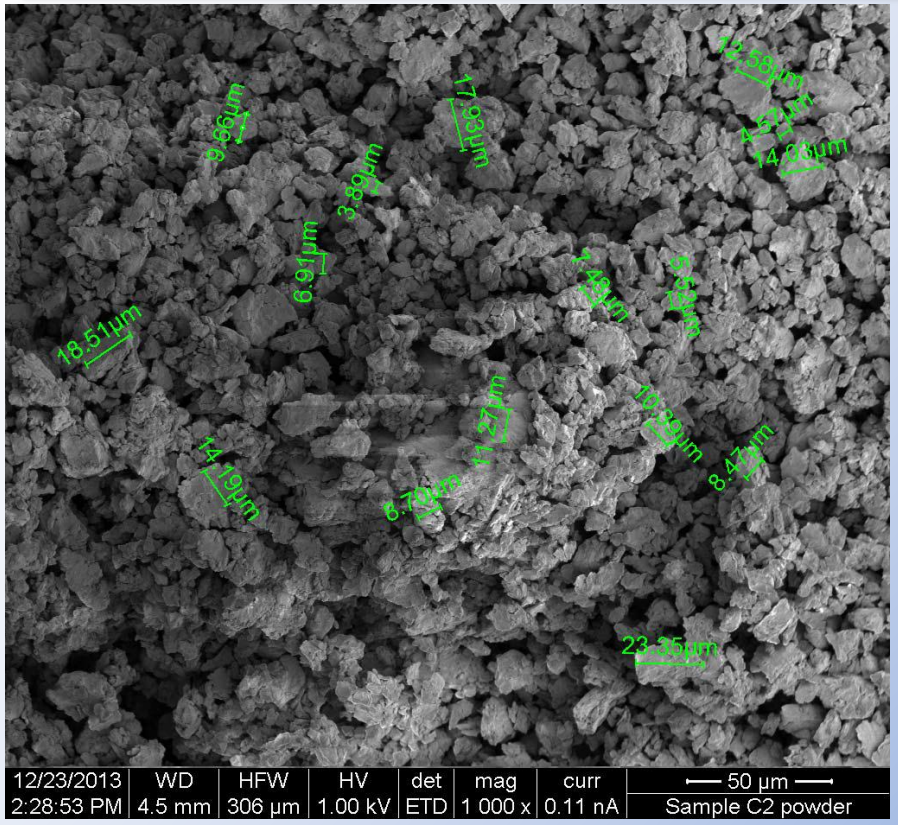

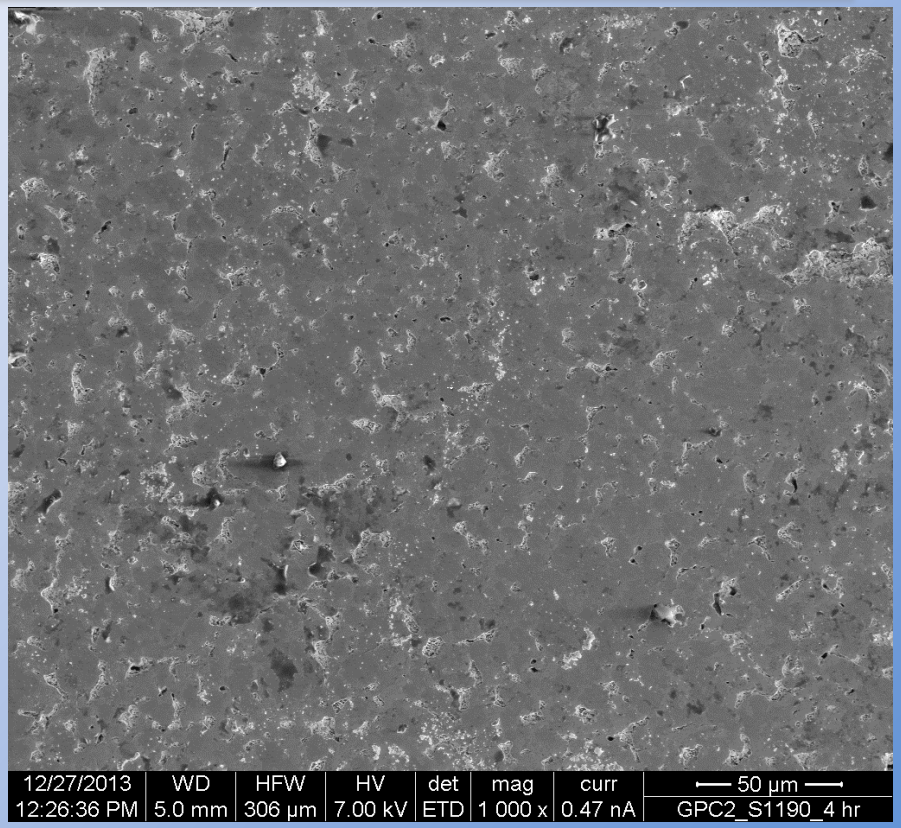

a) Powder milled 90 minutes

b) Sintered 2 hours
Density 6.4 g/cm^3

$(Pr,Gd)Co_{17}$

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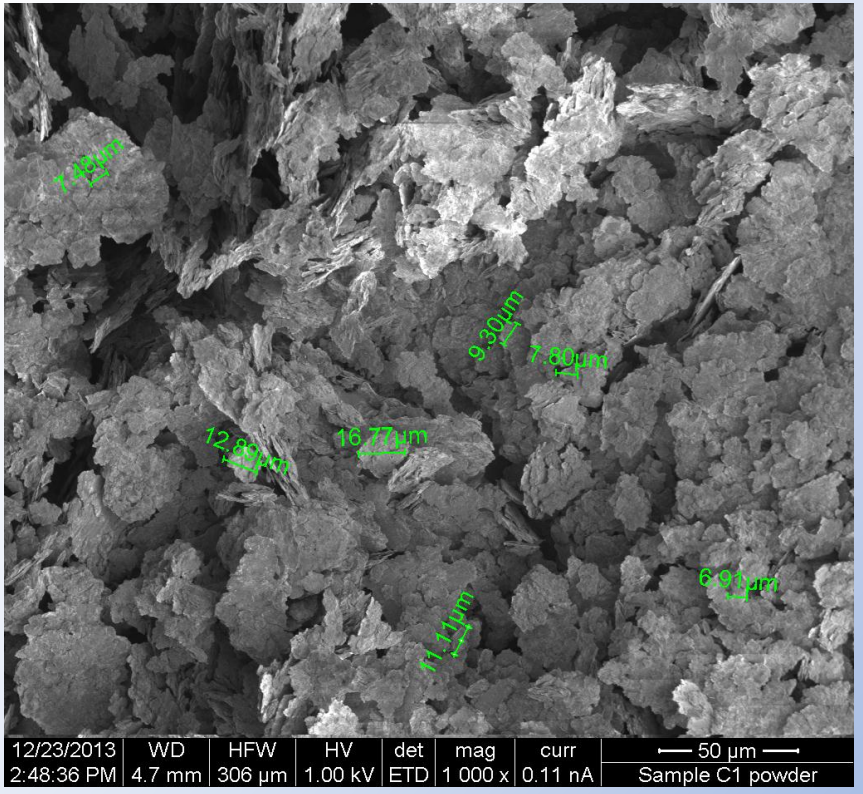
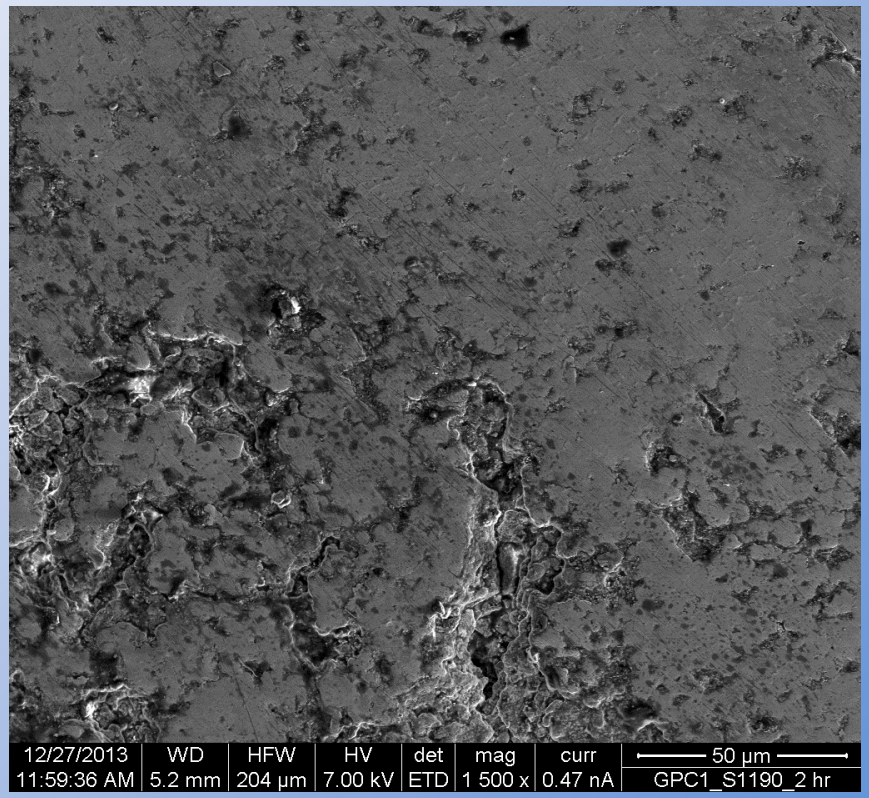


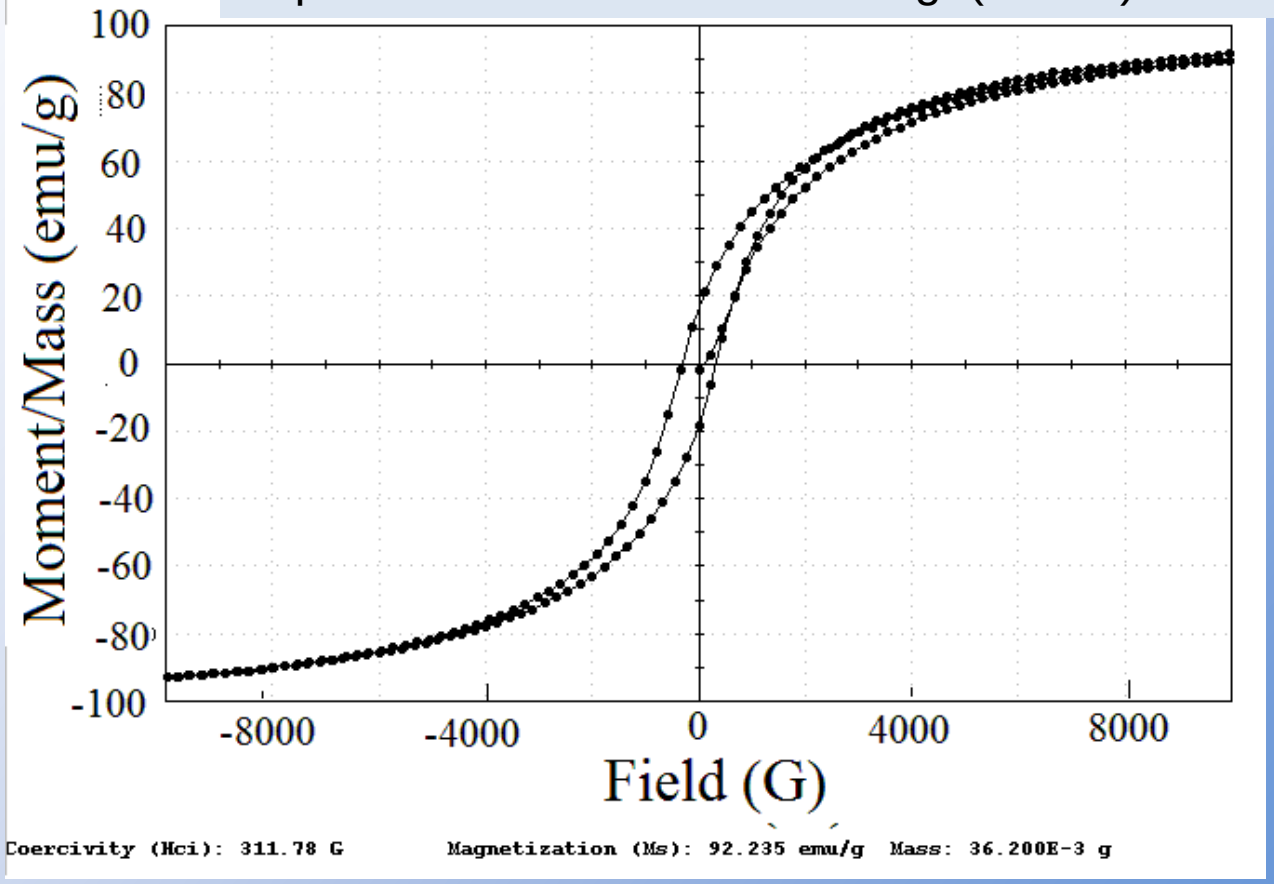
a) Milled powder 90 minutes

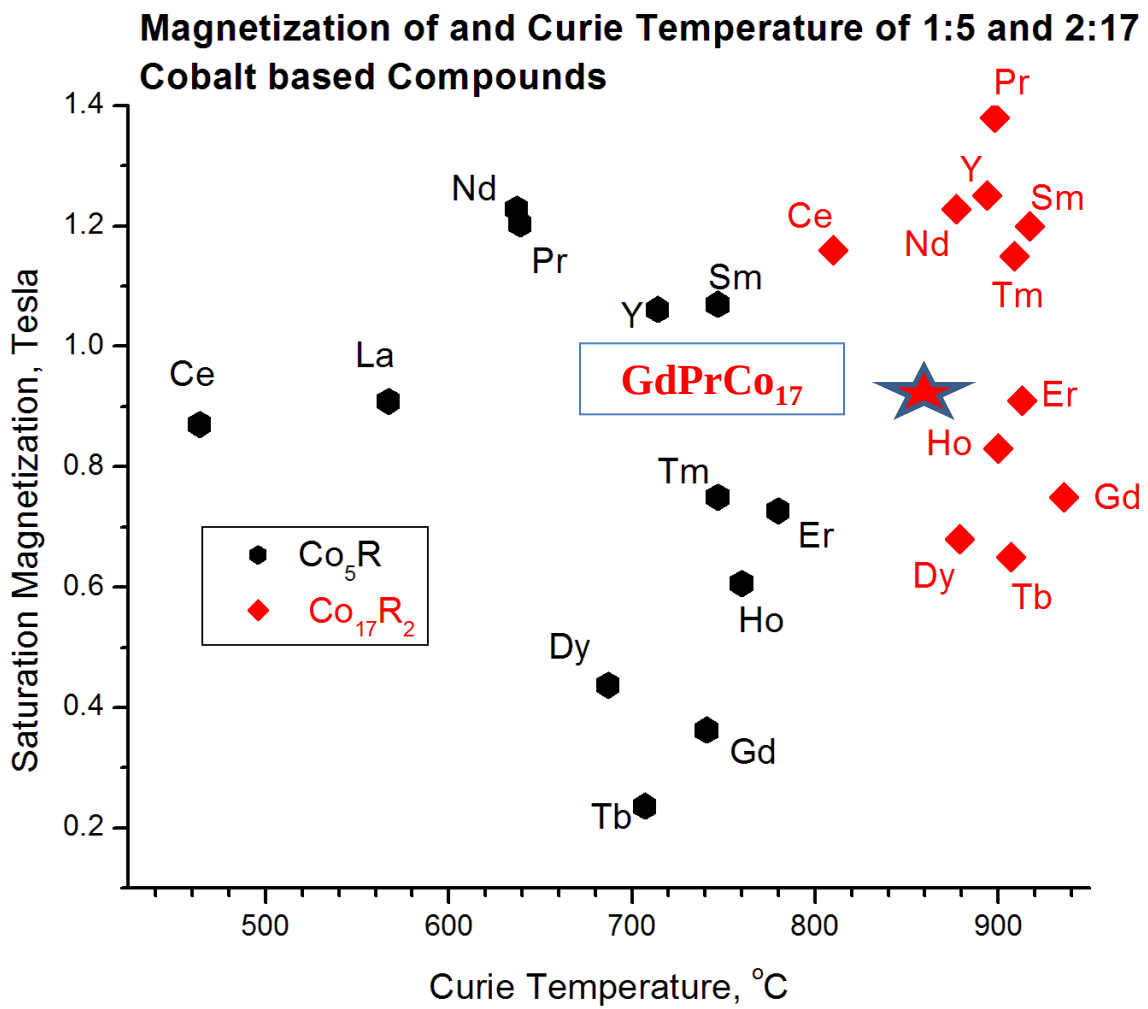


b) Sintered 2 hours density 4.29 g/cm^3

The hysteresis curve for the annealed $GdPrCo_{17}$

Calculated magnetic saturation : 151.0 emu/g (1.59 T)
Experimental value: 92.24 emu/g (0.97 T)





Data source: Concise Encyclodepia of Magnetic & Superconducting Materials, Jan Evetts, Ed

Comparison of temperature dependent magnetic moments

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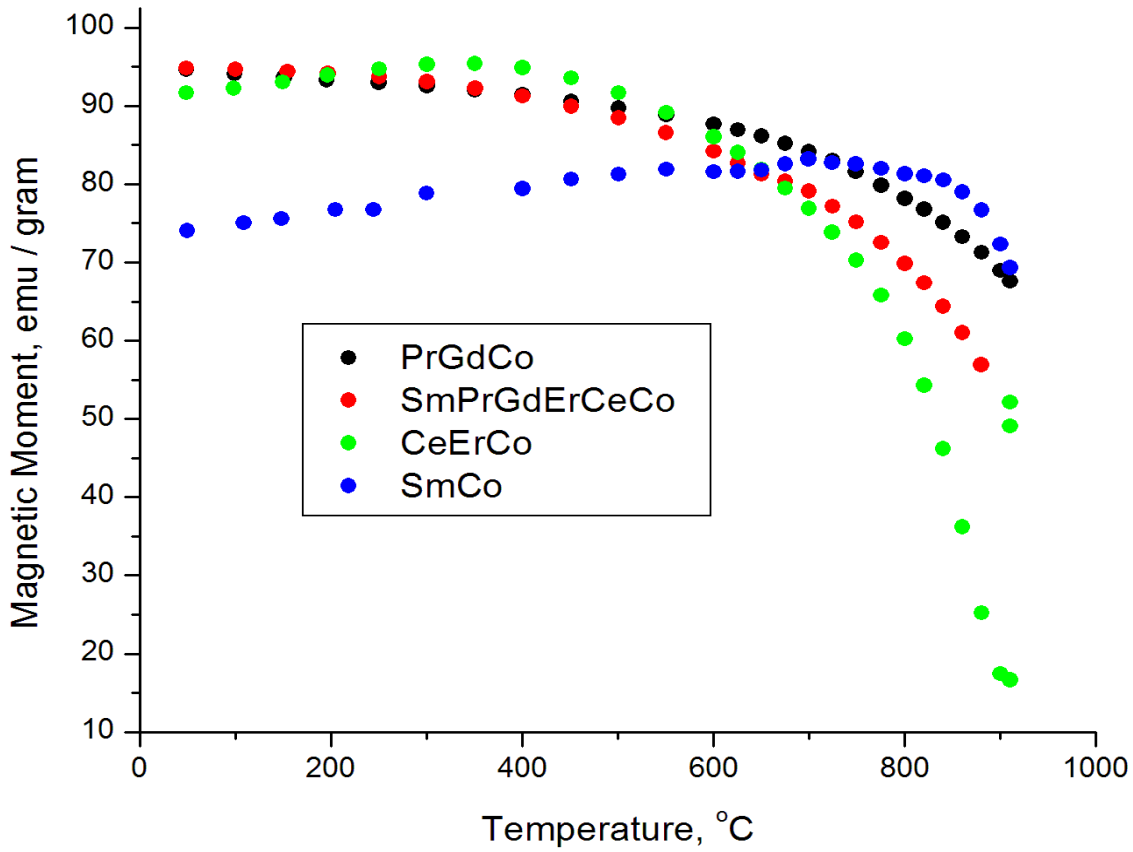

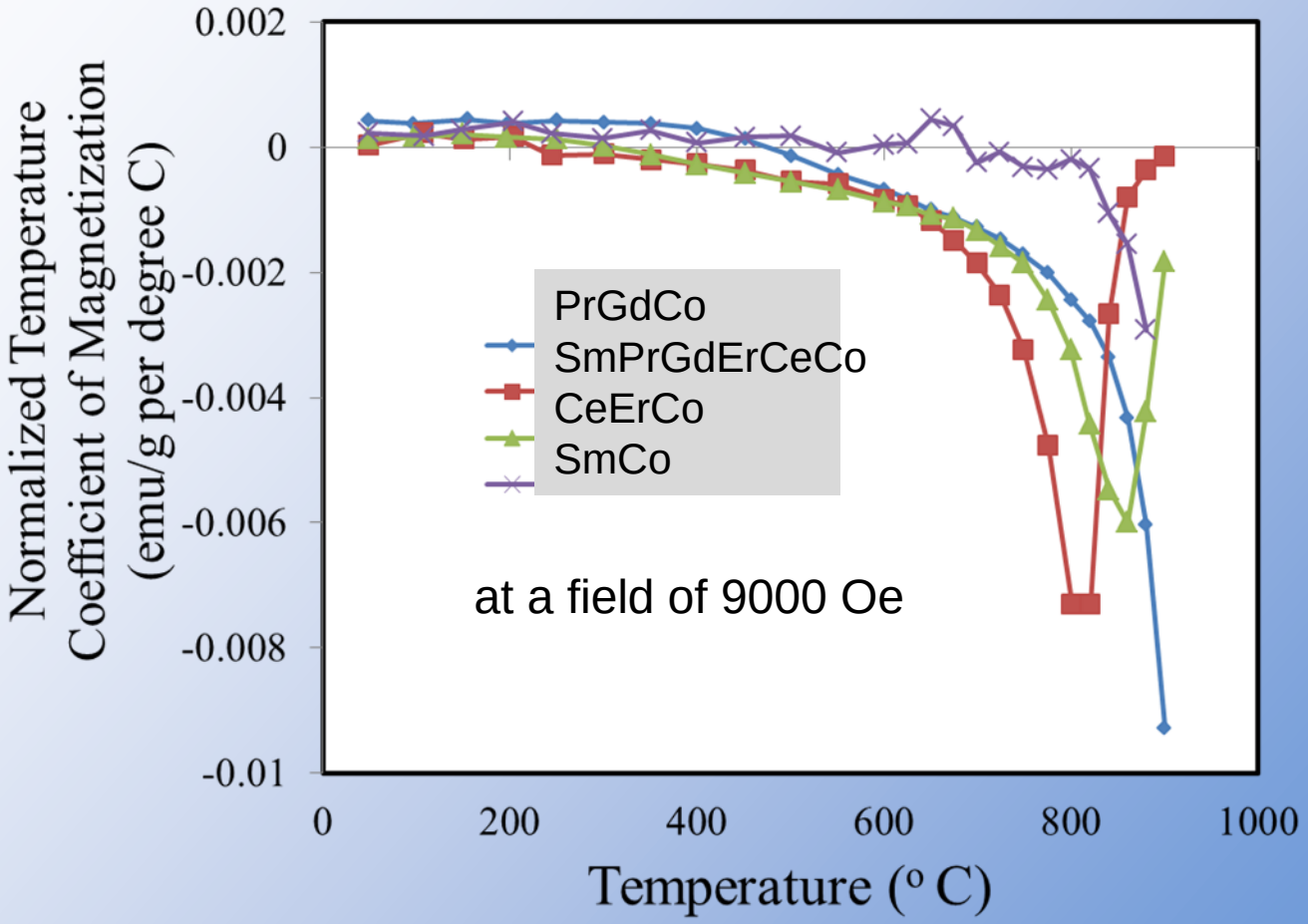
0.002
0
-0.002
-0.004
-0.006
-0.008
-0.01
Normalized Temperature Coefficient of Magnetization (emu/g per degree C)
0
200
400
600
800
1000
Temperature (° C)
PrGdCo
SmPrGdErCeCo
CeErCo
SmCo
at a field of 9000 Oe

Summary

- Increase of 25 % in magnetic moment over similarly processed SmCo
- Excellent magnetic properties to 700°C
- Density functional theory allows prediction of properties for magnetic alloys
- Further development is needed to include finite temperature predictions for more complicated alloy systems.
- Coercivity and temperature insensitivity not as good as SmCo, however better process control is needed for a definitive answer

Next Steps/Dissemination

- Induction melting
  - -More uniform sample preparation
  - -Better temperature control
- Must optimize microstructure
- Modeling code that allows meso-scale simulations
  - -Nano-composites dual phase soft-hard magnets
  - -Domains
  - -Time-stress -temperature effects simulated accelerated test
- Materials and Structure's Division Computer Modeling External Peer Review (September 2013)
- Journal article