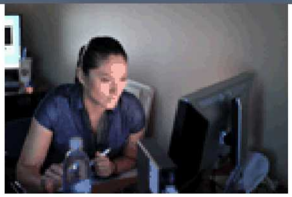
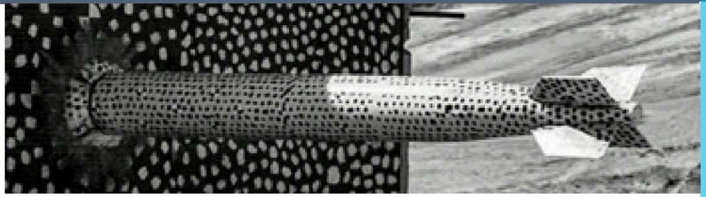
SAND2020-10711C

Electrodeposited Ni-Fe-Co Alloys for Magnetoelastic Resonators





• PRESENTED BY

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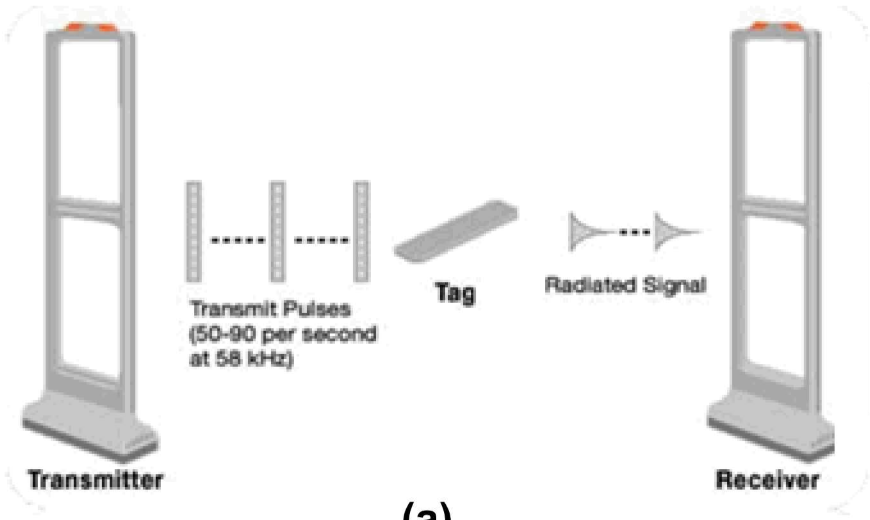
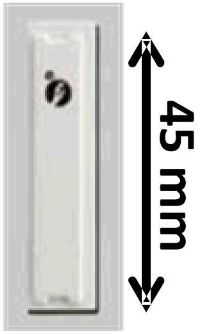
Magnetoelastic Resonator Overview & Joule Magnetostriction



(a)



(b)

a) **magnetic dipole antenna interrogation zone**
b) **magnetoelastic tag in plastic package**

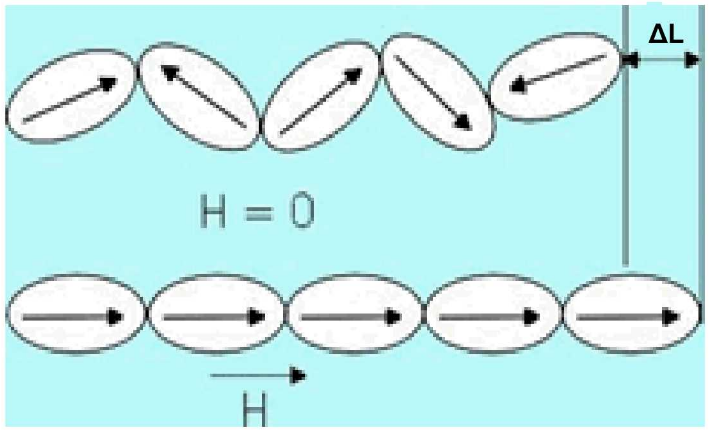
Status Quo

- **article theft deterrent used in Barnes & Noble, etc.**
- **imparts only “on” or “off” single-bit information**

amorphous magnetostrictive material such as METGLAS™ ($Ni_{40\text{-}50}Fe_{40\text{-}50}Mo_{5\text{-}10}B_{1\text{-}5}$) and a semi-permanent magnet ribbon comprise the tag

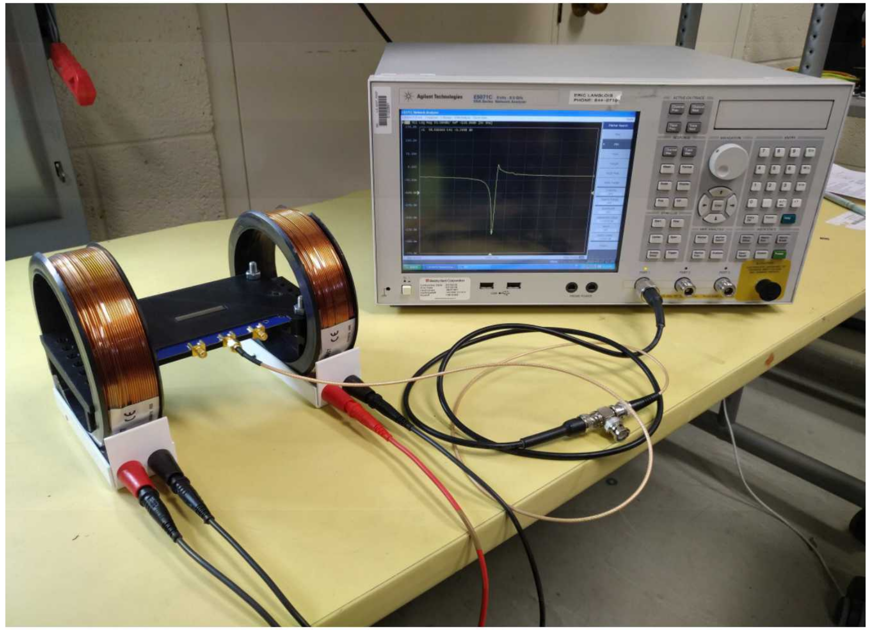
Operation Principle

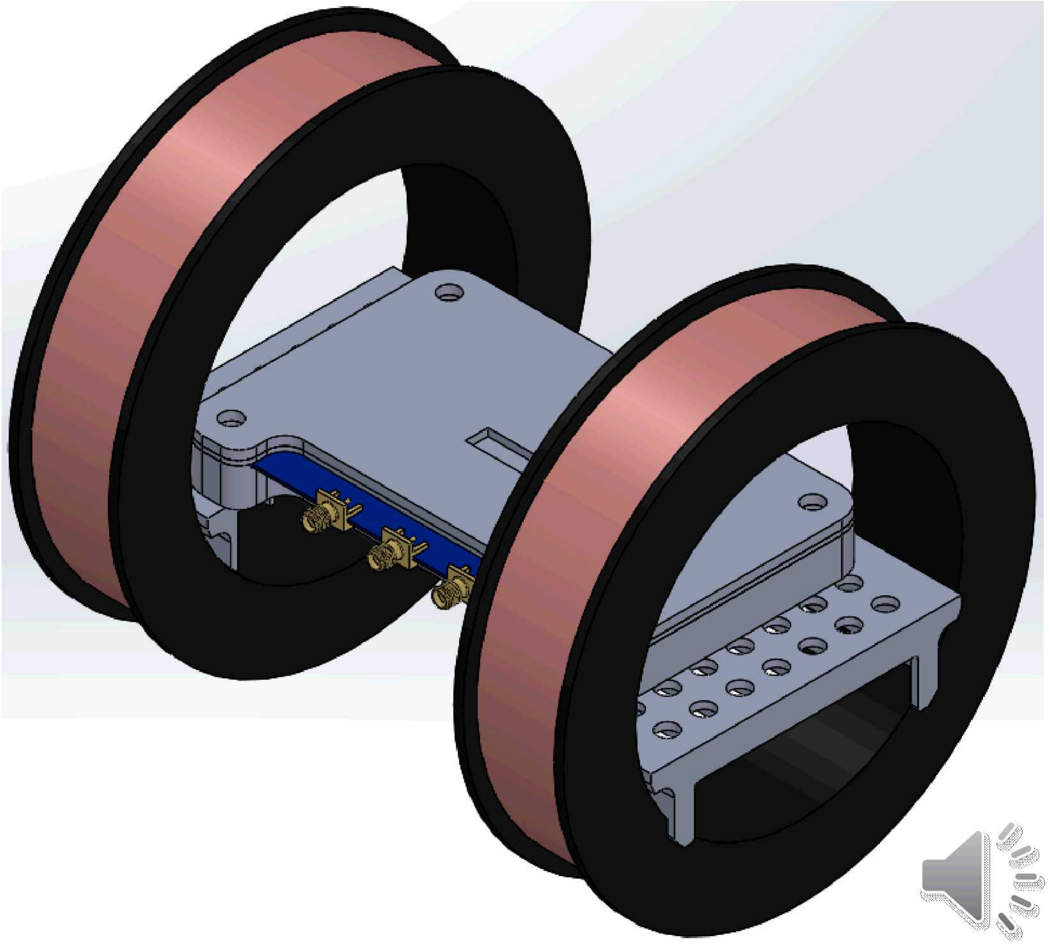
- **Joule Magnetostriction**
- **$\lambda = \Delta L / L$**



Resonator Response Testing

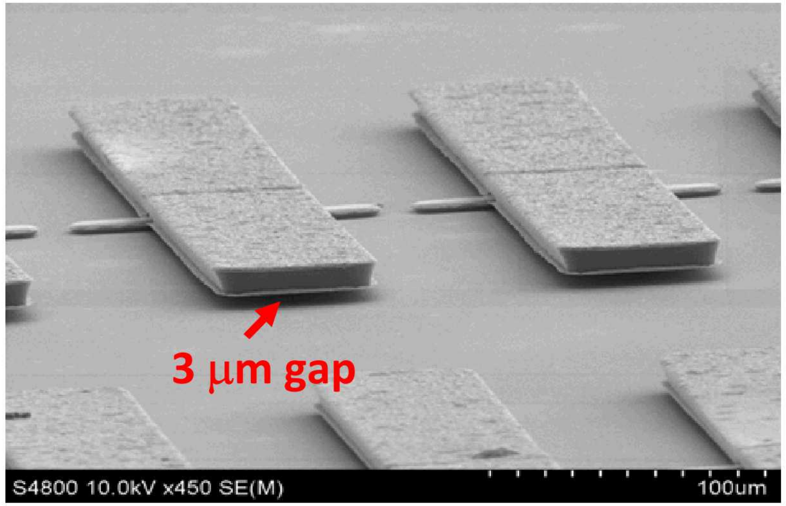
- Universal test fixture designed and produced to locate resonator to known location (while still allowing to freely resonate), and place Helmholtz coils in fixed location
  - Allows for repeatable results throughout project lifetime
- Agilent Technologies E5071C Vector Network Analyzer used to analyzed resonance
- Helmholtz coils used to create DC bias field to excite the resonators

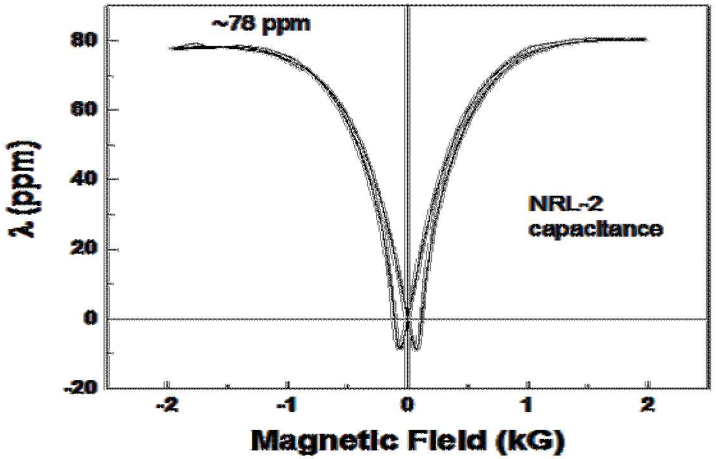




Success and Technical Challenges with Electrodeposited Magnetoelastic Films

- Previous chemistry was Co sulfamate based:
- CoFe films have high magnetostriction with low stress
- Make the film magnetically softer – less hysteresis loss on switching
- Keep magnetostriction high – usually a trade off
- Increase electrical resistivity by addition of boron or phosphorous
- Elemental additions of Ho, Ge, Nb, Si, B, P, and Mn have been investigated
- Films plated on Cu test strips at different stoichiometric additions
- Analyzed in SQUID magnetometer and qualitative measurement of magnetostriction in solenoid with optical probe
- Magnetic "softness": Lower coercivity (H_C) and lower magnetic squareness = more energy absorption and less energy loss





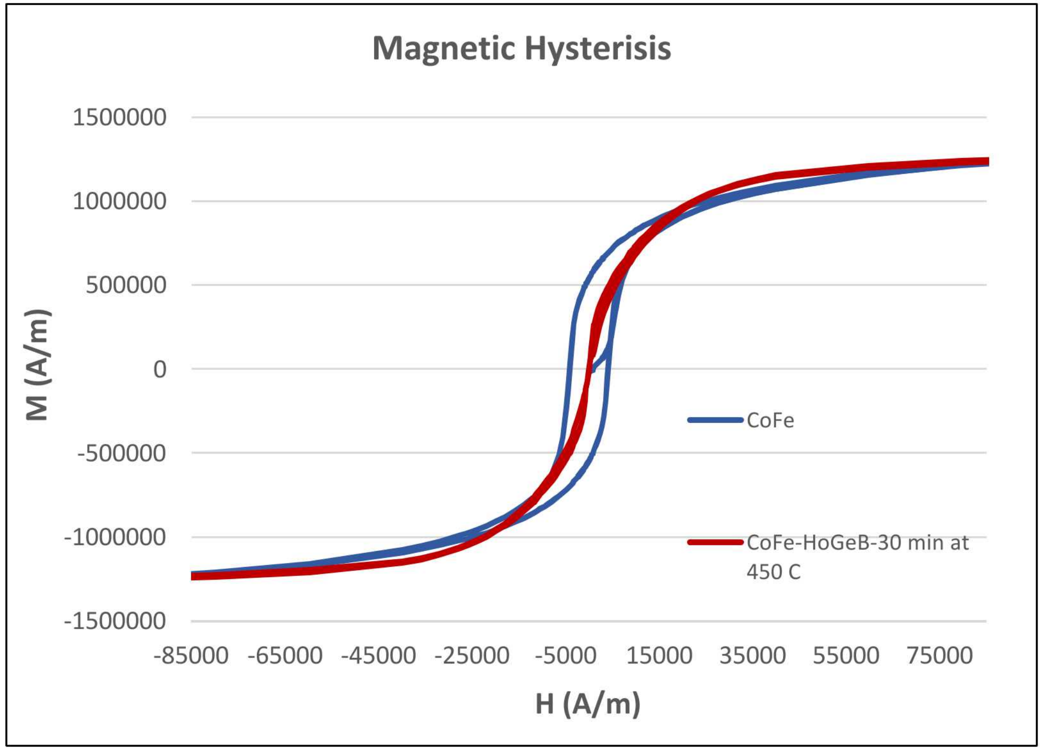
Magnetostriction measurement on original material performed at the NRL.

Elemental Additions to Adjust Magnetic Properties of Alloys

Testing performed to look at addition of a single element and combinations of elements on magnetic properties, magnetostriction, and electrical resistivity

Additions of Ho, Ge, Nb, Si and B increased magnetic softness while maintaining M_s and improving magnetostriction

- Ho has the highest permeability of any element and is used as the poles in permanent magnets
- Coercivity decreased two orders of magnitude and squareness decreased one order of magnitude with elemental additions and annealing step
- Electrical resistivity increased proportional to boron incorporation (amount of B used doubles resistivity)
- Qualitative measurement of magnetostriction doubled (only a parallel field is applied)
- Ge and B showed a decrease in grain size
  - Less anisotropy from crystal structure as film is nanocrystalline to amorphous



M-H Curve Material	M_S(A/m)	H_C(A/m)	Squareness ($S=M_R/M_S$)
Co-Fe	1.62E+06	7.06E+03	1.98E-0
Co-Fe-Ho-Ge-B	1.51E+06	7.08E+01	9.21E-0

NiFeCo Alloy Chemistry Development to Achieve Resonant Signal Measurement

A sulfate salt chemistry was investigated based off of a NiFe permalloy chemistry

- Less corrosive and less vapor

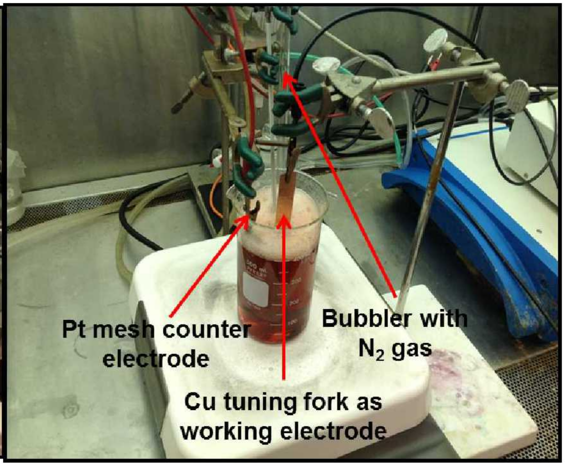
A NiFeCo-HoGeBNbSi (sulfate rather than sulfamate) chemistry was formulated that operates at a temperature of 30°C and a pH of 3.0

- Less corrosion and vapor = less contaminates from fixturing and cabling
- Bath is stable and can be plated out of for weeks without much pH drift or side reaction degradation
- Holmium sulfate salts were synthesized and incorporated with germanium oxide in the chemistry
- Dimethylamine boride (DMAB) was added as a source of boron to increase electrical resistivity
- Niobium silicide is complexed and added in small concentrations

DC plating rather than a pulse with a 50% duty cycle → half the time to plate a given thickness

New process to release material for testing using UV tape method

- Control of intrinsic stress



Controlling Microstructure Through Elemental Additions

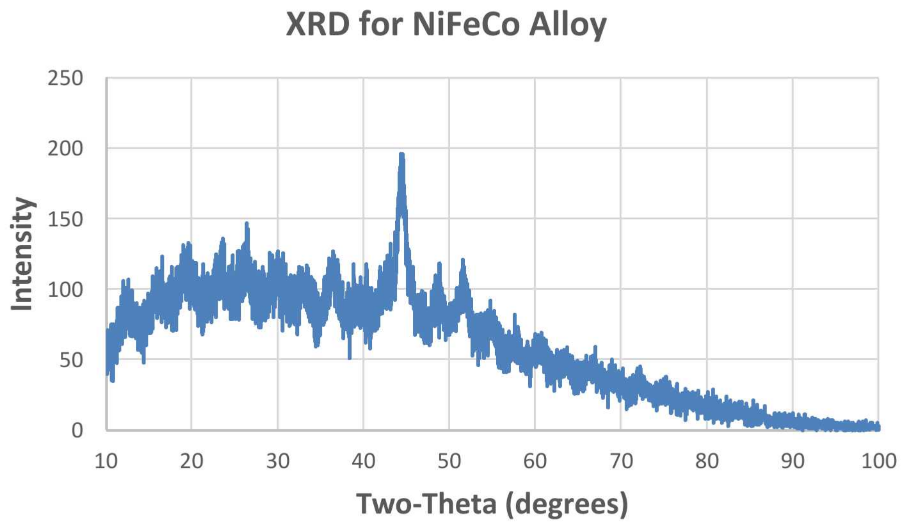
XRD analysis of NiFeCo alloy

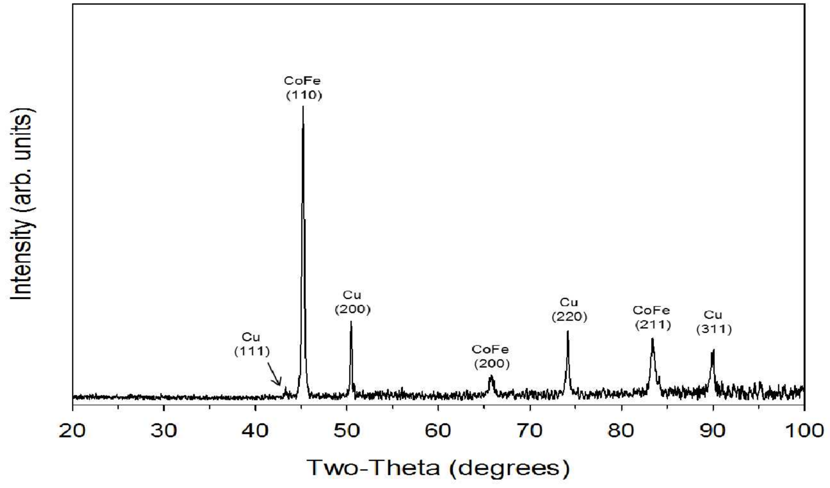
- Low count rate, film is mostly amorphous
- Small peak with crystallite size of approximately 8 nm
- Comparison with original CoFe material
  - CoFe nanocrystalline but ordered

EDS measurements to analyze composition as a function of plating parameters

- Compared with commercial APEX material
- Ho and Ge added later to reduce coercivity and increase permeability

Atomic % Results for Spectrum									
	Si	Cl	Fe	Co	Ni	Cu	Si	Nb	Total
DC	0.66	0.15	20.72	30.19	41.11	2.56	0.04	4.57	100
86	0.47		25.87	29.26	39.74	1.17		3.49	100
90	0.47		28.30	29.23	38.76			3.23	100
APX	2.03		29.5	14.21	54.27				100

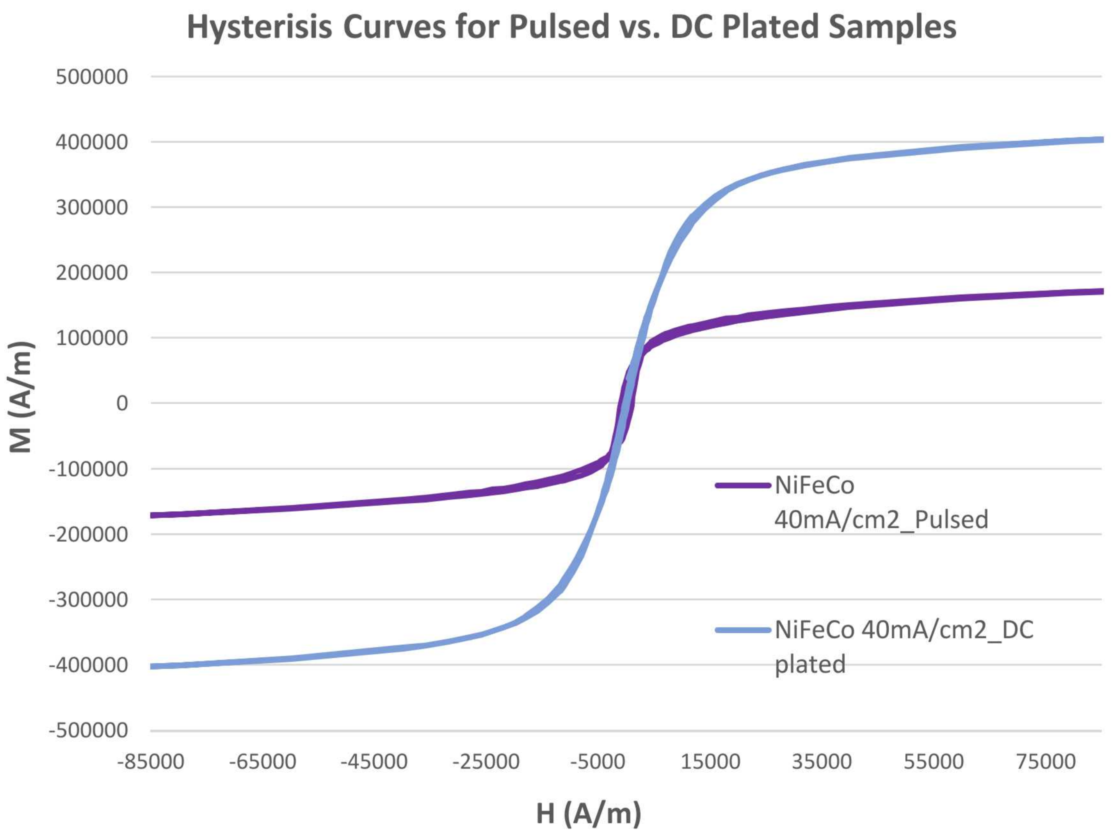




Plating Parameters for Increasing M_{sat} and Decreasing H_c

Magnetometry analysis of new chemistry

- Pulsed plating vs. DC plating
  - Changes in stoichiometry, stress and microstructure
- Lower M_{sat} than CoFe chemistry
  - Trade off with higher Co incorporation- still investigating
- Comparison of NiFeCo alloy to CoFe alloy without post processing treatment
- Characterization of working material is still ongoing
  - With post processing we achieved the first resonating, free standing, electroplated NiFeCo material



M-H Curve Material	M_s(A/m)	H_c(A/m)	Squareness ($S=M_r/M_s$)
Co-Fe	1.62E+06	7.06E+03	1.98E-02
NiFeCo Alloy	4.68E+05	2.65E+02	2.12E-02

Post Processing for Improved Magnetic Performance

Carbolite annealing furnace with N_2 for inert environment
- Oxidation changes magnetic properties

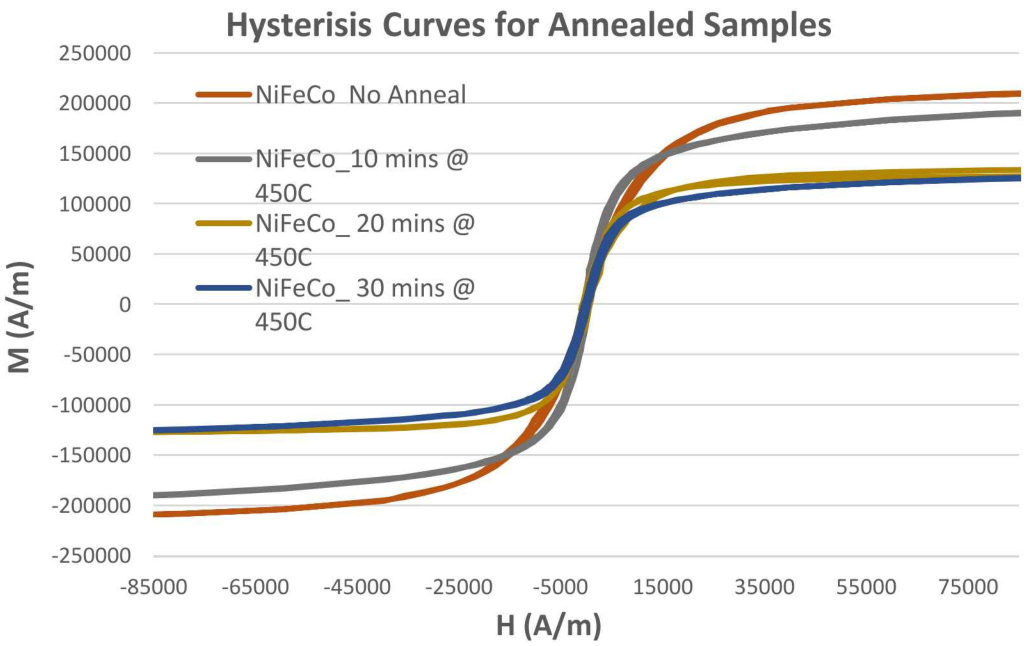
DOE of annealing experiments
- Vary dwell time with set max temperature – no mag field
- Vary dwell time with set max temperature – with 30 mT magnetic field applied
  - Max temperature 450°C
    - From literature review
    - Variability
      - Temperature range
      - 2 stage anneal process

Current annealing setup problems
- Limited magnetic field strength
- Wire metal (Cu) was depositing on samples
- Crucible material and size limitations

Annealing Experiments Demonstrate Magnetic Performance can be Adjusted by Changing Dwell Time

Magnetometry for annealed samples – SQUID magnetometer

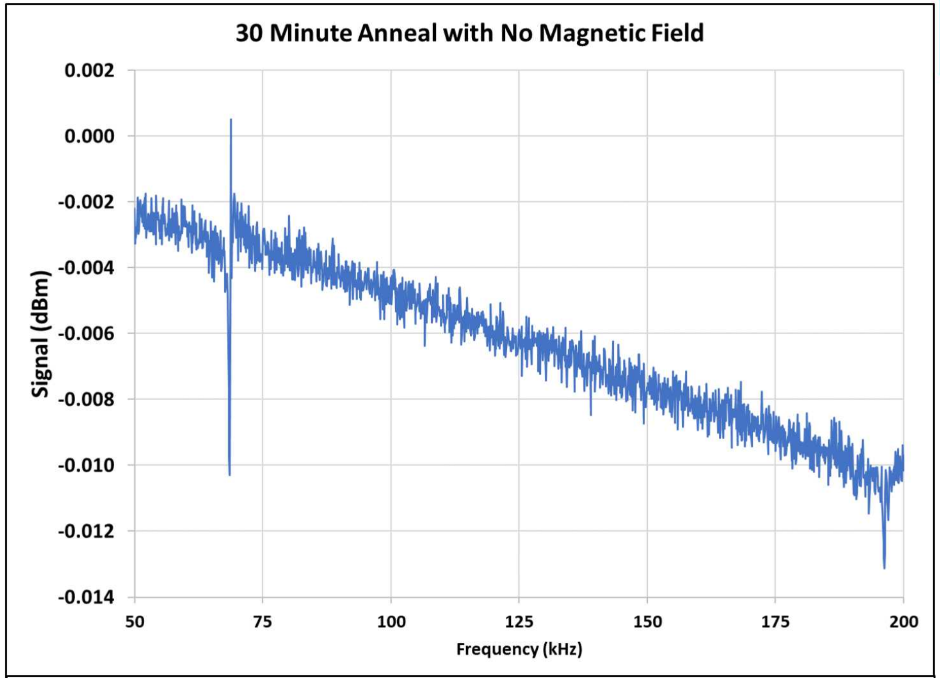
- No magnetic field applied on samples for hysteresis plot
- M_{sat} decreased as time increased
- H_c decreased until the dwell time was 20 minutes, then returned to original value
- Remanence increased with 10 and 20 minute dwell times but decreased to approximately nonannealed value after 30 minutes

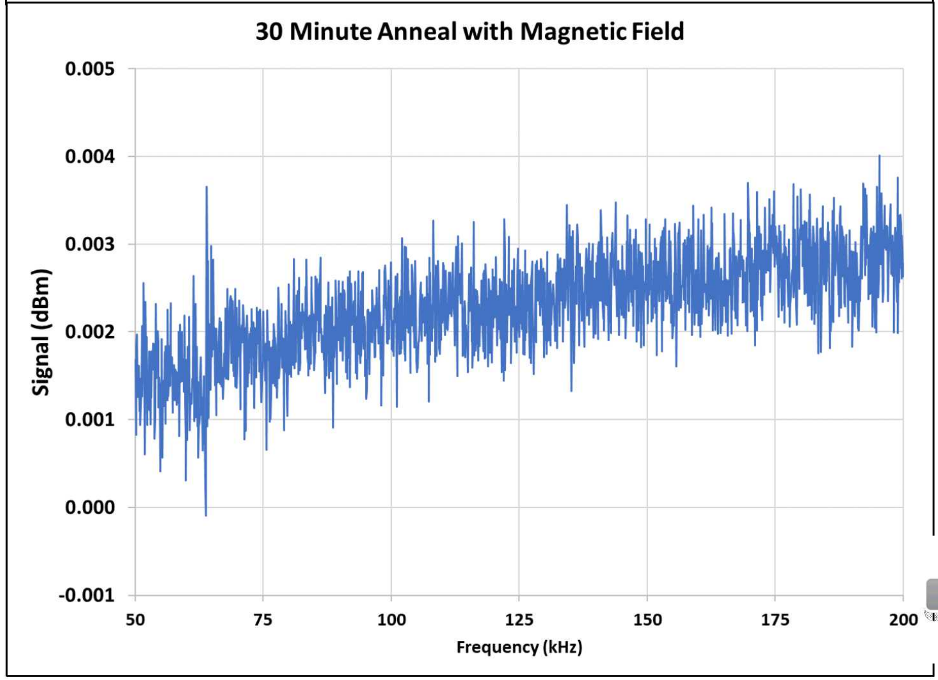


Anneal Time at 450˚C (min)	Magnetic Field Applied	H_c (A/m)
0	-	497
10	-	492
20	-	772
30	-	497
20	Parallel	627
20	Perpendicular	627

NiFeCo Alloy Sample Testing

- The latest chemistry is the first to have an observable resonance
  - Annealing was utilized to assist in achieving resonance
- With most recent chemistry, annealing study was performed
  - Samples were held at temperature for determined length of time with some samples subjected to a magnetic field while annealing
- From initial analysis, an anneal time of approximately 30 minutes appears to allows for the best resonance (top)
  - More samples and testing needed to make conclusive statement
  - Magnetic annealing process possibly deposited copper into samples resulting in skewed results (bottom)





Continuing Experimental Plan to Increase Resonant Signal Amplitude

- **Adjusting the stoichiometry to optimize the performance**
  - Small amount of Si, Nb, B, and Ho have a large impact on magnetic properties
  - Co content of films
- **Electroplating in an applied magnetic field**
  - DOE to determine changes in stoichiometry, microstructure, stress, and then resulting magnetic properties.
  - Tested in conjunction with a post plating anneal.
- **Improved annealing setup and larger power supply to increase magnetic field during annealing**
  - Currently, applied mag field is a little over 30mT - increase that to at least hundreds of mT.
  - More domain alignment and better magnetostriction if the field is stronger during annealing