SPIN-FILTERED PHOTOEMISSION AND POLARIMETRY EFFORTS AT EUCLID

ERIC MONTGOMERY

EUCLID TECHLABS / EUCLID BEAMLABS

P3 WORKSHOP - NOVEMBER 2021

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Outline

1. Half Metal Spin Filters

GaAs Spin Polarized Photocathodes

State of the Art

Proposal: Half metals on GaAs

2. DFT Studies

Crystal & Electronic Structure of CrMnSb

Mechanical strain

Sb -> P substitution

Biaxial strain in thin films of $CrMnSb_{0.5}P_{0.5}$

DFT Conclusions

3. Half Heusler QE Experiments

Deposition and Transport

Preliminary data

4. Side Topic: Diamond Polarimetry

Motivation

Apparatus

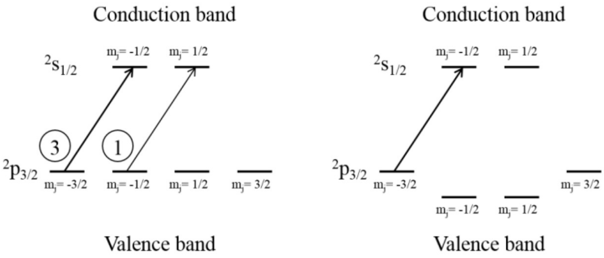
Initial CEBAF data

GaAs Spin-Polarized Photoemission

GaAs chosen because it forms a negative electron affinity surface (for high quantum efficiency). Spin polarized emission is possible from bulk (typ. ~30%, theory 50%), on the left. Axial strain, on the right, breaks degeneracy and improves spin polarization (typ. ~80%, theory 100%).

Other methods such as multi-photon emission and OAM light have yielded poor results.[1]

[1]McCarter et al., “Novel Approaches for Electron Beam Polarization”, Proc. PSTP (2013).



State of the Art Spin-Polarized Photocathodes

Distributed Bragg reflector strained superlattice GaAs/GaAsP photocathode with Cs-F negative electron affinity activation layer, used at CEBAF (JLab):

- QE 6%[1],
- polarization 84%[1]

[1]Liu et al., Appl. Phys. Lett 109, 252104, 2016.

GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	p=5 $\cdot 10^{17}$ cm^{-3}
$GaAsP_{0.35}$	750 nm	p=5 $\times 10^{18}$ cm^{-3}
$GaAsP_{0.35}$/ $AlAsP_{0.4}$ DBR	(54/64 nm) ×12	p=5 $\times 10^{18}$ cm^{-3}
$GaAsP_{0.35}$	2000 nm	p=5 $\times 10^{18}$ cm^{-3}
Graded $GaAsP_x$ ($x = 0 \sim 0.35$)	5000 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	p=2 $\times 10^{18}$ cm^{-3}
p-GaAs substrate (p>10^{18} cm^{-3})		

Standard strained superlattice GaAs/GaAsP with Cs-F[3] :

- QE up to 1.6%[2],
- polarization up to 92%[2]

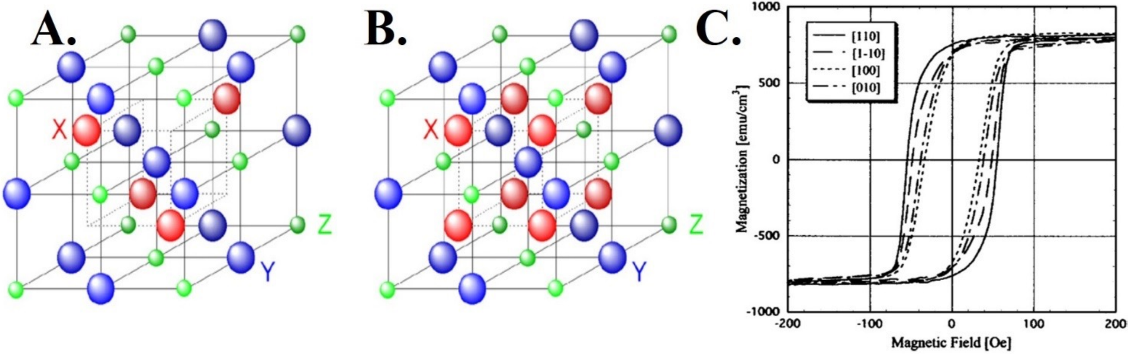
[2]Jin et al., Appl. Phys. Lett. 105, 203509, 2014

[3]Maruyama et al., Appl. Phys. Lett. 85, 2640, 2004.

GaAs	5 nm	p=5 $\times 10^{19}$ cm^{-3}
GaAs/GaAsP SL	(3.8/2.8 nm) ×14	p=5 $\times 10^{17}$ cm^{-3}
$GaAsP_{0.35}$	2750 nm	p=5 $\times 10^{18}$ cm^{-3}
Graded $GaAsP_x$ ($x = 0 \sim 0.35$)	5000 nm	p=5 $\times 10^{18}$ cm^{-3}
GaAs buffer	200 nm	p=2 $\times 10^{18}$ cm^{-3}
p-GaAs substrate (p>10^{18} cm^{-3})		

Heusler Alloys

- Intermetallic: XYZ or X_2YZ
- X ,Y are transition metals; Z is p-band element
- High Curie T (1100 °C) ferromagnet
- Lattice matched with III-V semiconductors
- Transition metals - multiple oxidation states; prone to oxidation
- Require controlled environment (U/XHV)



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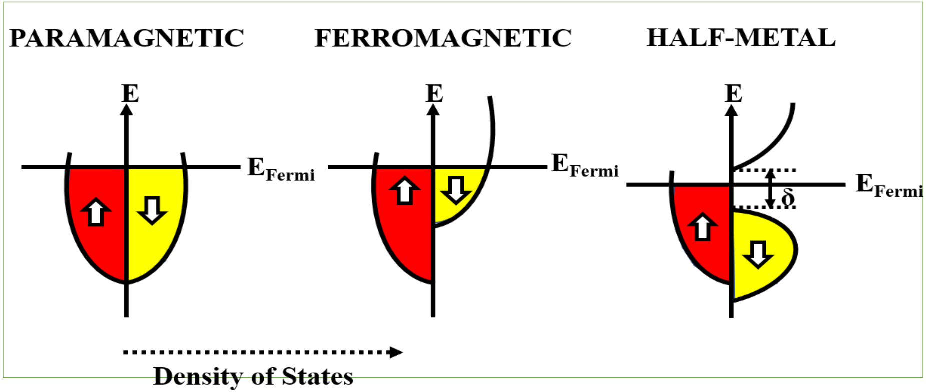
Half-Metallicity

- Common metals like copper (Cu) or aluminum (Al) have no net P : half the electrons are ↑ and half are ↓ at the Fermi Level (E_F).
- Ferromagnets, e.g., iron (Fe) and cobalt (Co) have small imbalance of ↑ and ↓ electrons at E_F.
- Half-metals (HM) the majority spin channel is metallic while the minority channel is insulating (See Fig. below).
- The majority spin band (denoted by upwards arrow) has no band gap at E_F while the minority band possess a finite band gap δ.
- Can result in a population of filled conduction band states with 100 % spin polarization.



Proposal: Half metal spin filters

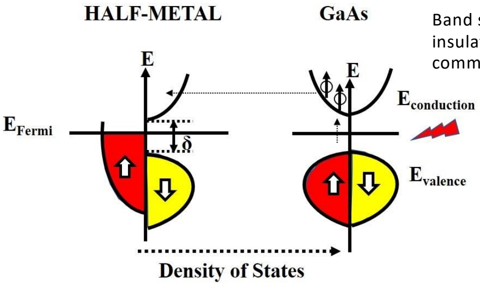
Half metals, discovered in the early 1980s, are untested filters for spin polarized photoemission

We focused on the “Half Heusler” family

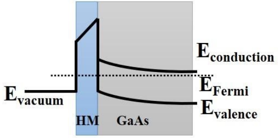
Benefits: III-V lattice match, up to 100% spin polarization in bulk, low coercivity

DFT search for new spin filter candidates yielded $CrMnSb_{0.5}P_{0.5}$ (JAP, 2020, below), other non-phosphorus options under study

Intro first, then a bit of DFT…



Band structure metallic for one spin and insulating for opposite. In-plane spin is most common; out-of-plane is under study.



Chemical substitution induced half-metallicity in $CrMnSb_{(1-x)}P_x$

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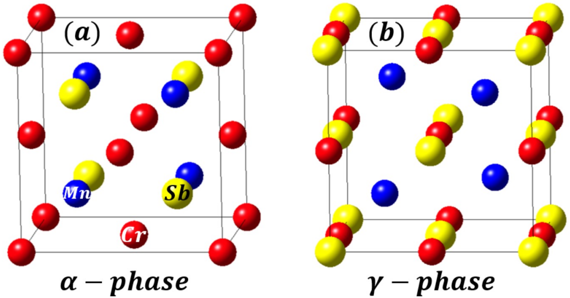
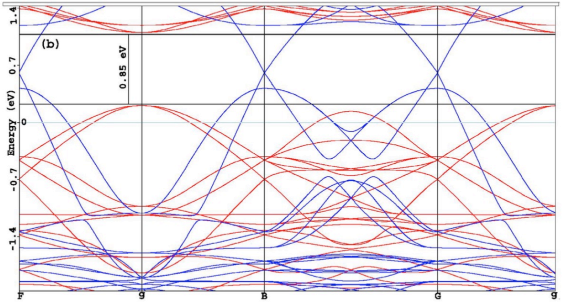
DFT Studies

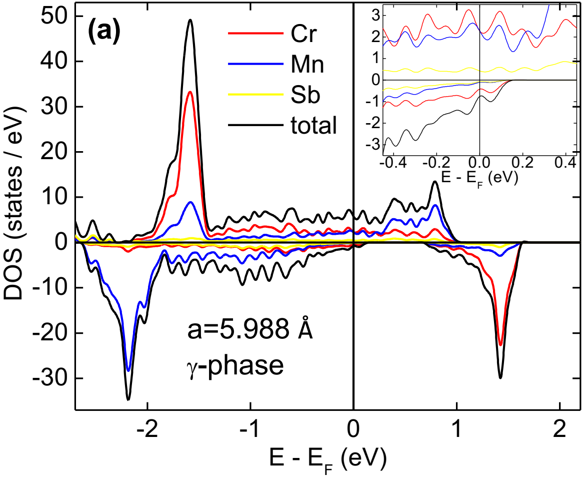
- Software:
  - Density Functional calculations
  - Vienna *Ab-Initio* Simulation package (VASP)
- Hardware:
  - Local: UNI Physics computer cluster
  - External: Pittsburgh Supercomputing Center – Bridges

J. Appl. Phys. **128**, 113906 (2020)

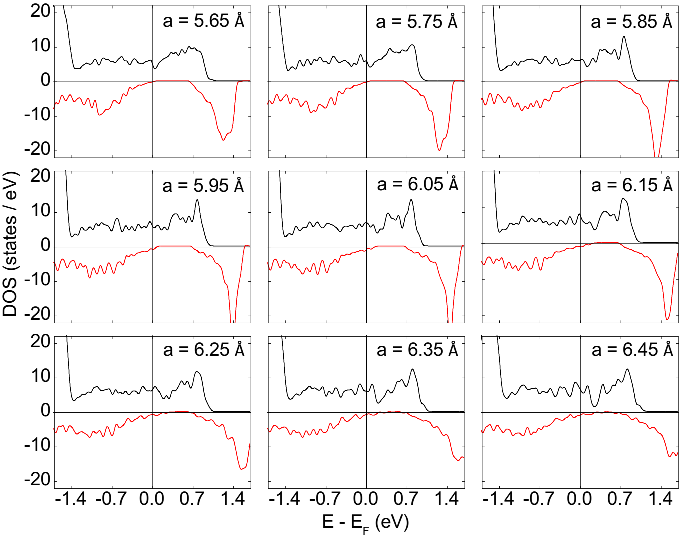
CrMnSb crystal and electronic structure



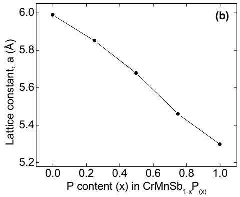


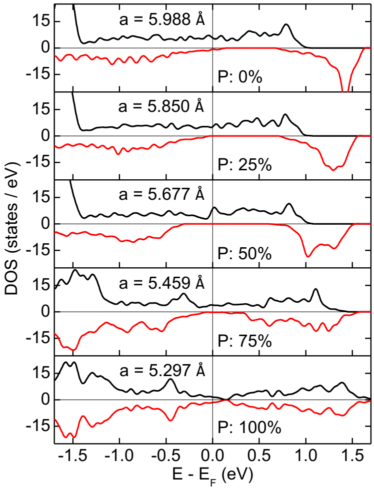


CrMnSb and P substitution



biaxial strain





DFT Summary

- CrMnSb: *nearly* half-metallic electronic structure in ground state.
- Half-metallic transition under uniform pressure.
- Biaxial strain (thin film) – no HM transition.
- Chemical substitution induced half-metallicity: Sb→P.
- $CrMnSb_{0.5}P_{0.5}$ – a new proposed material which is a fully compensated half-metallic ferrimagnet suitable for spin filtering.
- Complementary to known fully compensated half-metallic ferrimagnets, such as the full Heusler alloys Co_2FeSi and Co_2MnSi.

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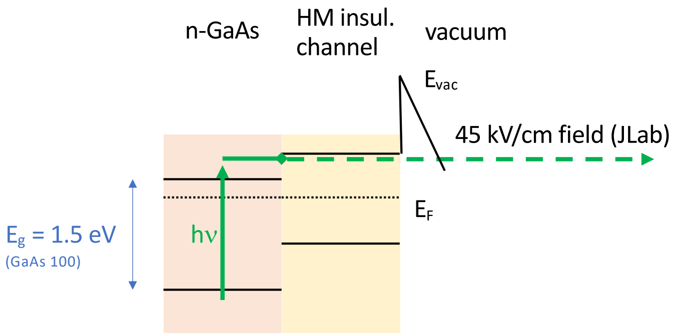
Half metal spin filter band diagram

Band diagram suggests half metal growth on heavily doped n-type GaAs.



Vacuum emission for one spin state possible with NEA condition and/or tunneling through field-lowered barrier

- A large Slater-Pauling gap (delta) in the half metal and a low emission barrier are desirable
- Cesiation and high field will be required for good QE

Half metal spin filter "wish list"

High emitted spin polarization if…

1. 100% polarized at E_F
2. P-type substrate doped so conduction band below that of half metal minority channel
3. Properly terminated
4. Lattice matched so low disorder

High quantum efficiency if…

1. Large minority band gap
2. Cs-F compatible (no alloying @r.t.)
3. Thin half metal – 1-3 formula units

(optional) practically, easier to use if…

1. High Curie temperature
2. Moderate magnetic coercivity
3. Fully compensated magnetic moment so no intrinsic angular momentum of photoemitted electrons (otherwise may need a bucking coil)

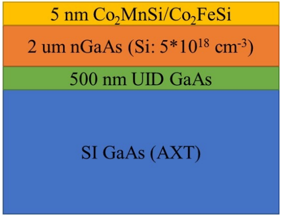
Half metal spin filter selection

Co_2FeSi and Co_2MnSi selected

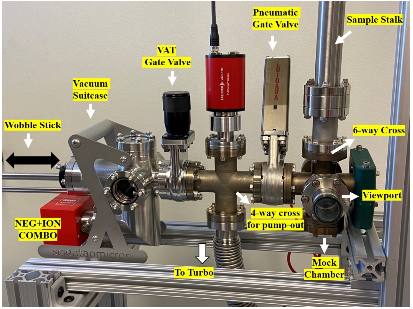
- Ease of growth
- close lattice matching with GaAs
- highest magnetic moments.

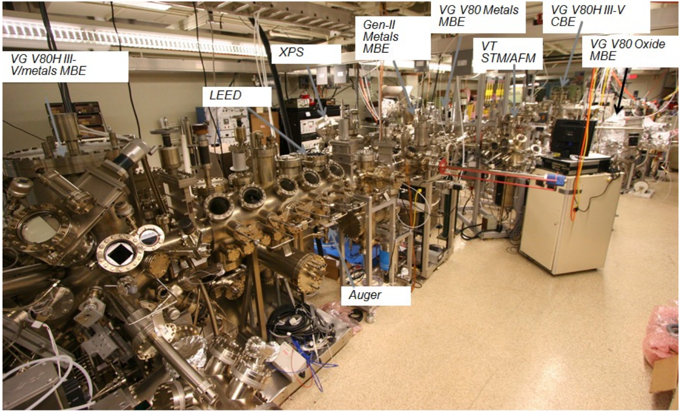
Univ. of California-Santa Barbara (Palmstrøm Lab) grew both on heavily n-doped GaAs.

Heusler Alloy	Curie Temperature (Kelvin)	Magnetic Moment (μB/formula unit)	Lattice mismatch with GaAs (%)
Co_2FeSi	1100	5.46	0.08
Co_2FeAl	1000	4.98	1.3
Co_2MnSi	985	5.0	0.3
Co_2MnGe	905	5.0	1.6
Co_2MnAl	726	4.0	1.8



Transfer to JLab at 10^{-11} Torr via custom Omicron vacuum suitcase to cesiate, measure QE and attempt Mott polarimetry





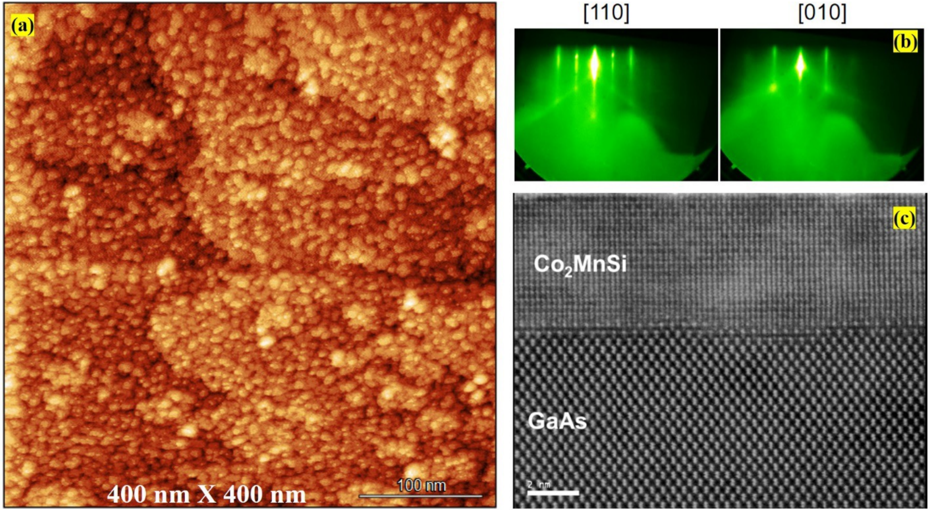
Characterization after growth at UCSB

(a) STM surface scan (scale 100 nm)

(b) RHEED pattern

(c) HAADF-TEM scan for the Co2MnSi on GaAs system (scale 2 nm)

High angle annular dark-field (HAADF) detects Rutherford-scattered electrons as opposed to Bragg-scattered: dark-field contrast improves for higher Z. HAADF-TEM is uniquely suited for imaging of strain relaxation in cross-sections of epitaxial interfaces.



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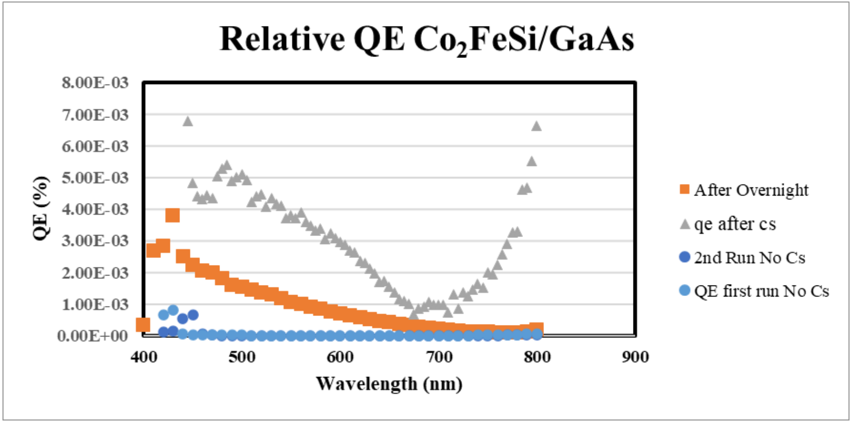
Cesiation and Quantum Efficiency

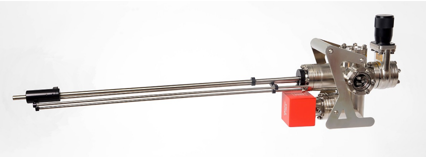
Transfer in UHV suitcase from UCSB to JLab's modified polarimeter for flag-style Omicron samples was successful. (!!)

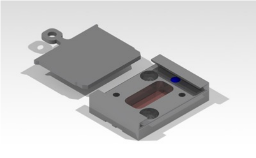
Heat clean and Cs/NF3 yo-yo immediately produced photocurrent, suggesting Cs-F sub-monolayer adsorbtion as desired, the Cs not alloying with the bulk.

QE in the visible (gray curve) is an order of magnitude reduced from GaAs. Assessment of spin filtering requires polarimetry (next!)

Lifetime in the polarimeter is a few hours to 1/e - the QE falls overnight (orange curve). Residual NF_3 is a possible suspect.







Mott Polarimetry

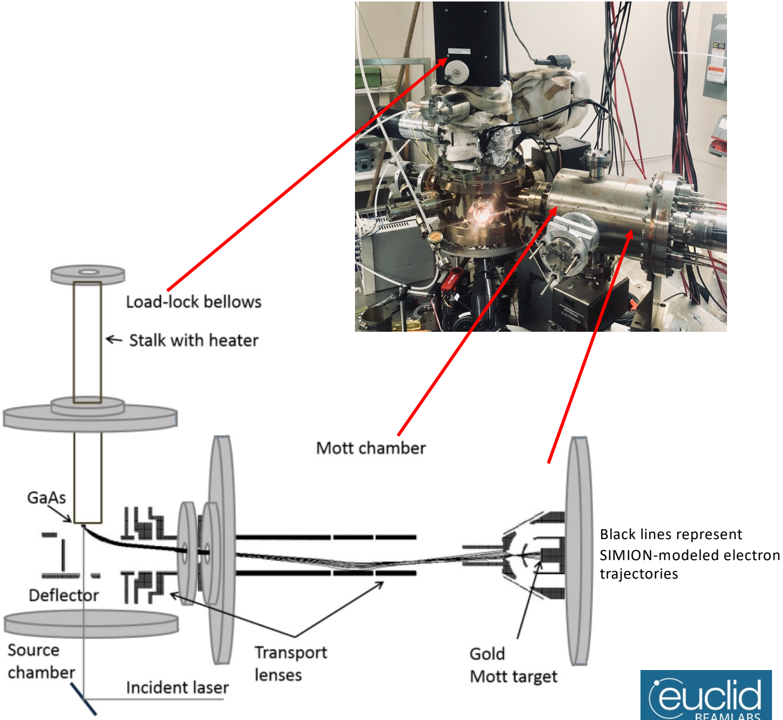
Mott polarimetry exploits elastic scattering asymmetry of spin polarized electrons from nuclear Coulomb field.

$$\sigma(\theta) = I(\theta)\left[1 + S(\theta)\mathbf{P}\cdot\mathbf{n}\right]$$

$$P = \frac{N_{\uparrow} - N_{\downarrow}}{N_{\uparrow} + N_{\downarrow}}$$

Requires known Sherman function S

Illumination is with circularly polarized light



Polarimetry – $Co_2FeSi/GaAs$

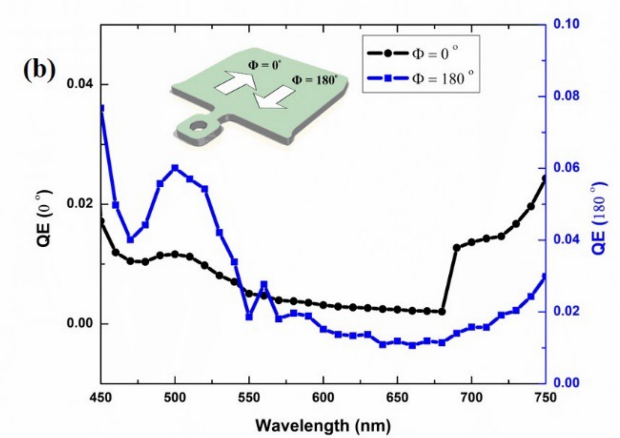
Linearly polarized normally incident illumination.

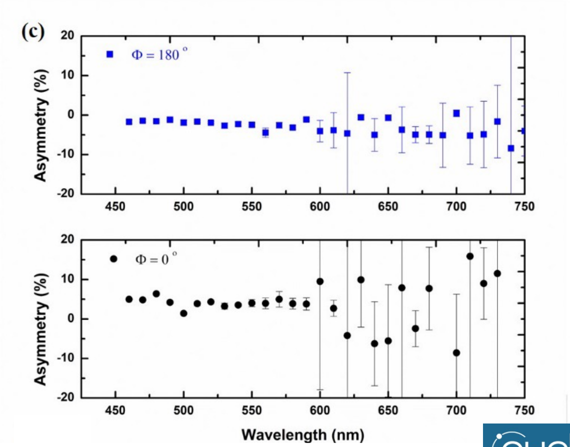
(b) Average QE after in-plane magnetic poling at 0 and 180.

(c) Total asymmetry* via Mott polarimetry for the two cases.

Asymmetry switches sign, indicating spin-filtering. Estimated polarization could be ~60-70% though statistics were not ideal due to time limitations*.

Unclear why spin filtering persists with excess photon energy greater than the half-metal minority spin channel band gap ~0.5 eV.





*Very challenging experiment. Not repeated; systematic errors possible.

Polarimetry – $Co_2MnSi/GaAs$

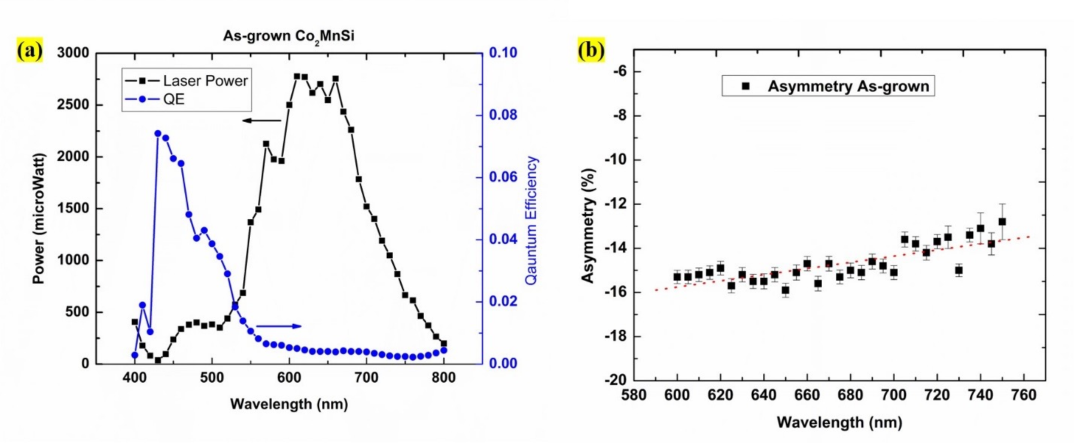
Linearly polarized normally incident illumination.

Only one poled in-plane field direction tested.

Similar QE (blue curve), 10x lower than NEA GaAs.

Asymmetry magnitude may be much higher than for Co_2FeSi*.

Inconclusive; Covid restrictions and experimental issues precluded a field flip to check sign change of asymmetry.



*Very challenging experiment. Not repeated; systematic errors possible.

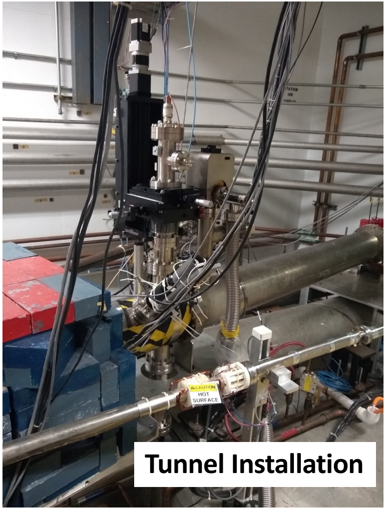
Half Metal Spin Filter Conclusions

- Two half-Heusler half metals were grown on heavily n-doped GaAs: Co_2FeSi and Co_2MnSi.
- Cesium appears not to alloy with the spin filter.
- Both cesiated half-metals on GaAs photoemit. ~10x QE reduction vs bulk NEA GaAs.
- Note: normally-incident, linearly polarized illumination cannot produce spin polarized emission in GaAs without the action of the spin filter, so any asymmetry beyond experimental uncertainty is a sign of spin filtering.
- It works! Spin polarization suggested by asymmetry which flips with magnetic poling direction.
- Quantification of polarization was not possible; rough estimates of 60-70% leave much room for improvement towards the theoretical 100%.
- Near-XHV conditions during handling were not easy; a more stable filter would be preferable.
- In-plane polarization is not ideal; out-of-plane is preferred for longitudinal spin and ease of poling with a solenoid.
- We are continuing to investigate other options for out-of-plane spin filtering with proposals pending. Stay tuned!

Diamond polarimetry – bonus topic!

Diamond Mott Target holders

Target Ladder Build



Tunnel Installation

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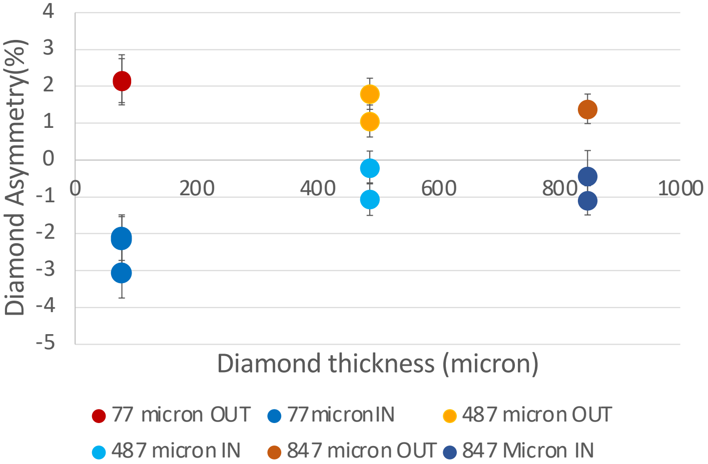
Diamond polarimetry

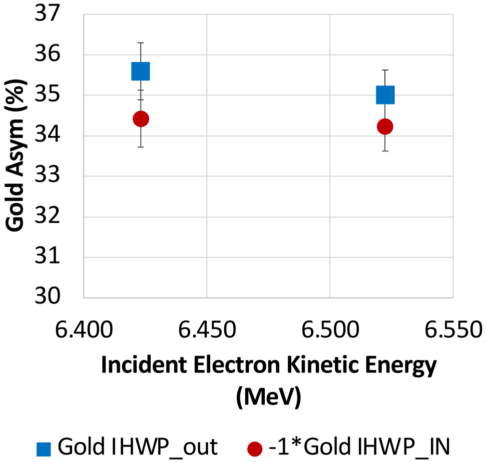
Three CEBAF runs!

	Energy	Targets
Aug 3 8:00-14:30	6.3 MeV Pockles cell off	Open poly 77 µm 487 µm
Aug 5 15:30-19:00	6.3 MeV	Gold 77 µm 487 µm 847 µm Heating test
Aug 13 8:00-16:00	6.3 MeV	Gold 77 µm, steering 487 µm 847 µm
	4 MeV	77 µm 487 µm 847 µm
	4.5 MeV	77 µm
	5 MeV	77 µm

6.3 MeV asymmetry - Diamond vs Gold

Could replace gold as Mott target for high current beams and high precision foils





Trend toward decreased asymmetry with thickness; minimal heating

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Thank you to all P3 attendees
and our organizing committee!