# First-principles study of voltage-induced switching, optical properties, and heat capacity of antiferromagnetic metals

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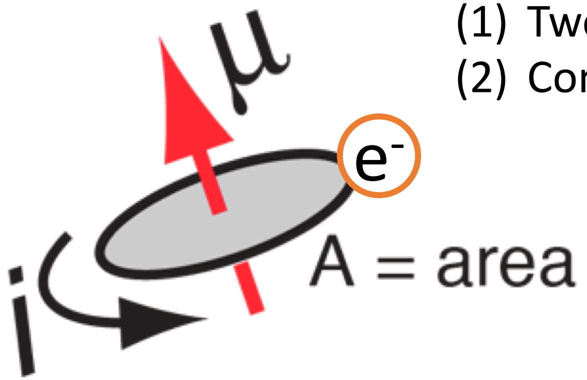
OCI-0725070
ACI-1238993

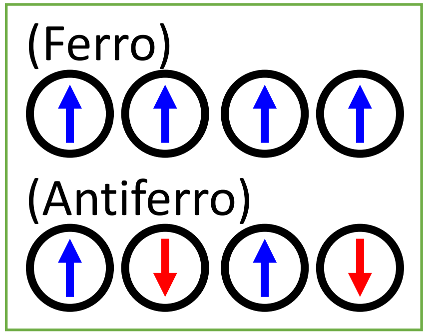
# Introduction

## 1) Motivation

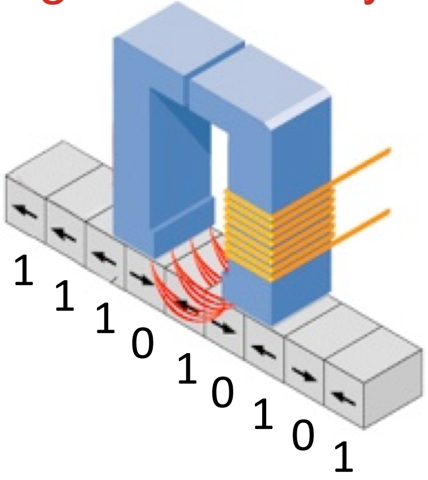
### Magnetic Moment, Spin




(1) Two different directions
(2) Controllable by stimulation




### Magnetic Memory device




Hard Drives

Adapted from: www.igcseict.info, hyperphysics.phy-astr.gsu.edu

### Why Antiferromagnetic Materials?

(1) Robust to external magnetic field


Easily Affected

Robust

(2) Fast Dynamics


GHz Response

THz Response

(3) No Stray Field

Affects

No effect

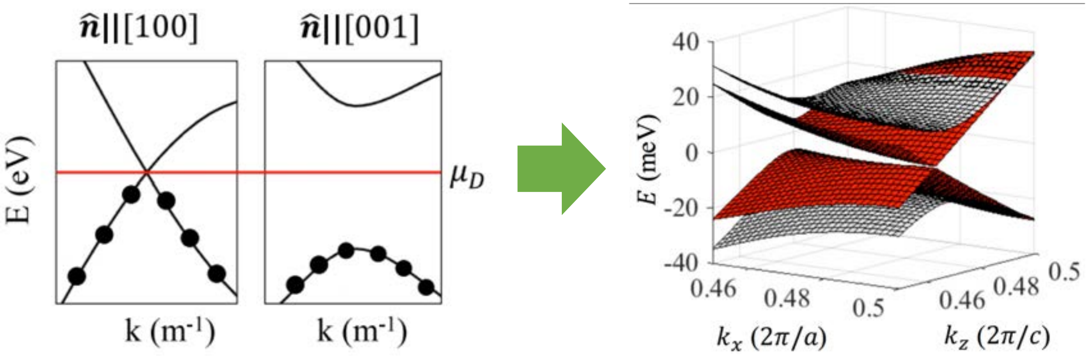
# Introduction

## 2) Projects

### Project 1

Voltage-induced switching of antiferromagnetic semimetal

- Order parameter switching can causes band gap opening
- Model Hamiltonian for symmetry study
- Density functional theory (DFT) to investigate antiferromagnetic semimetals

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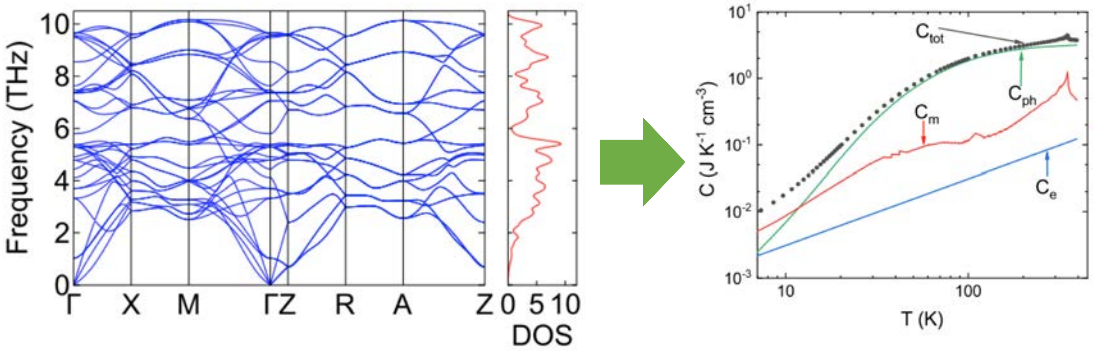
### Project 2

Optical and thermal properties of antiferromagnetic metallic $Fe_2As$

- Linear magneto-optical Kerr effect (MOKE) from antiferromagnetic metal under external magnetic field is predicted
- Relationship between quadratic MOKE and heat capacity is confirmed




*arXiv*:1903.07810

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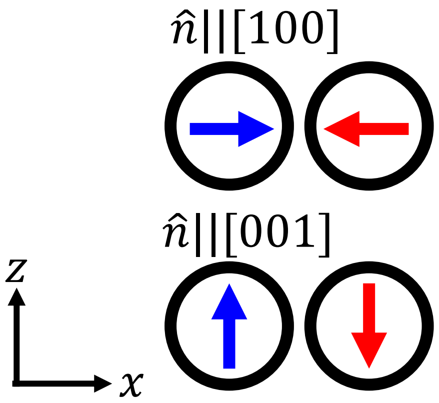
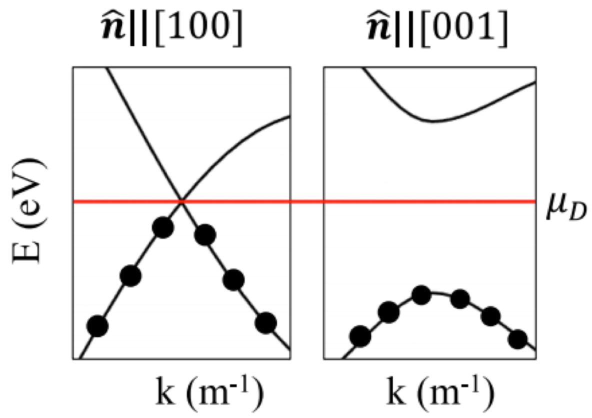
# Voltage-induced Switching

## 1) Background and Theory

### (1) Antiferromagnetic Semimetal (AFS)



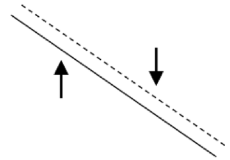




- $\mathcal{PT}$ symmetry may be preserved with an extra nonsymmorphic crystal symmetry
- Under satisfying condition, reorientation of spin configuration may break underlying symmetry and change the gap of Dirac fermion
- This may be detected by electronic transport response of AFS and become potential novel platform for spintronic applications

### (2) (Semi)metal-insulator transition (MIT)

1. Two-fold degeneracy:
   Combined inversion ($\mathcal{P}$) and time-reversal ($\mathcal{T}$) symmetry
   ⇒ **$\mathcal{PT}$ Symmetry**

$$\mathbf{k},\mathbf{s} \xrightarrow{\mathcal{T}} -\mathbf{k},-\mathbf{s} \xrightarrow{\mathcal{P}} \mathbf{k},-\mathbf{s}$$



2. Protected Dirac point:
   Depending on **additional symmetry** and **reciprocal space**

level-repulsion Protected crossing

$$H = \begin{pmatrix} \mathbf{k}\cdot\boldsymbol{\sigma} & m \\ m & -\mathbf{k}\cdot\boldsymbol{\sigma} \end{pmatrix} \qquad H = \begin{pmatrix} \mathbf{k}\cdot\boldsymbol{\sigma} & \cancel{m} \\ \cancel{m} & -\mathbf{k}\cdot\boldsymbol{\sigma} \end{pmatrix}$$

# Voltage-induced Switching

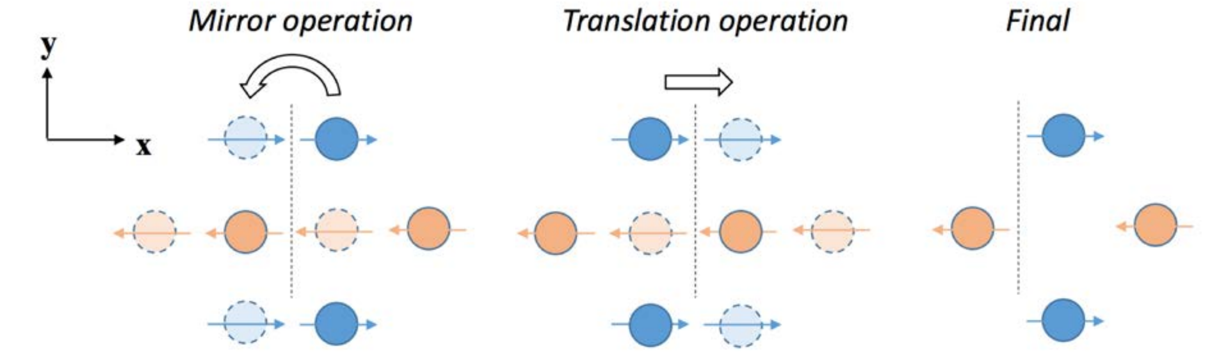
*Phys. Rev. B* **97,** 134415 (2018)

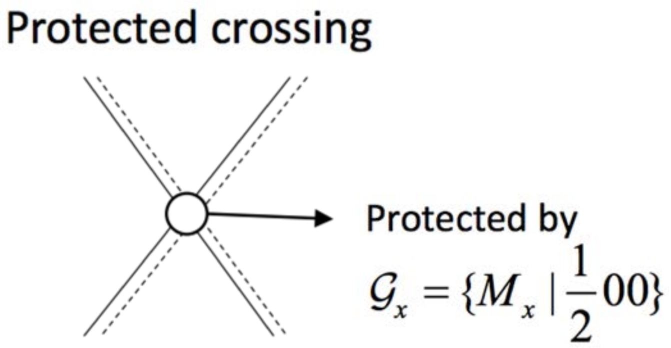
## 2) Example

**PT symmetry + Additional symmetry:**
Non-symmorphic glide-reflectional symmetry

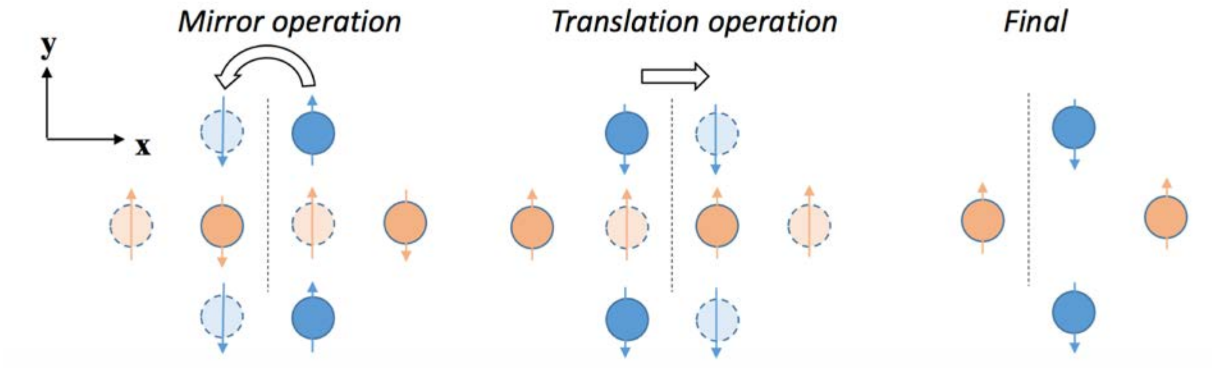
$$(x, y, z) \xrightarrow{\mathcal{M}_x} (-x, y, z) \xrightarrow{T_x} (-x + ½, y, z)$$
$$(s_x, s_y, s_z) \rightarrow (s_x, -s_y, -s_z) \rightarrow (s_x, -s_y, -s_z)$$

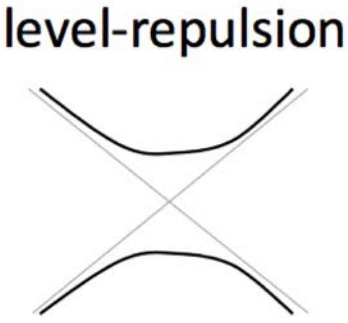
when $\hat{\boldsymbol{n}}$||[100] → preserved




when $\hat{\boldsymbol{n}}$||[010] → broken



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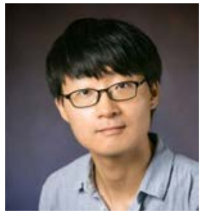
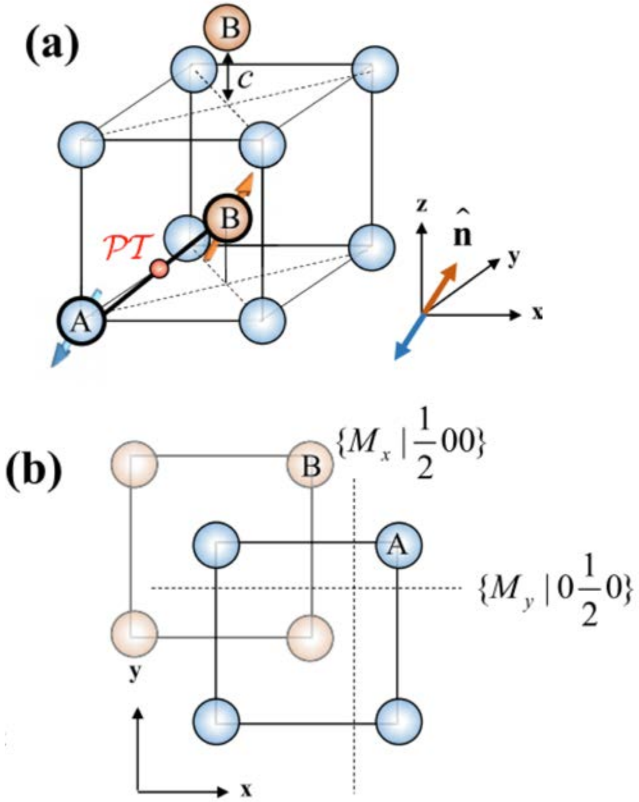
# Voltage-induced Switching

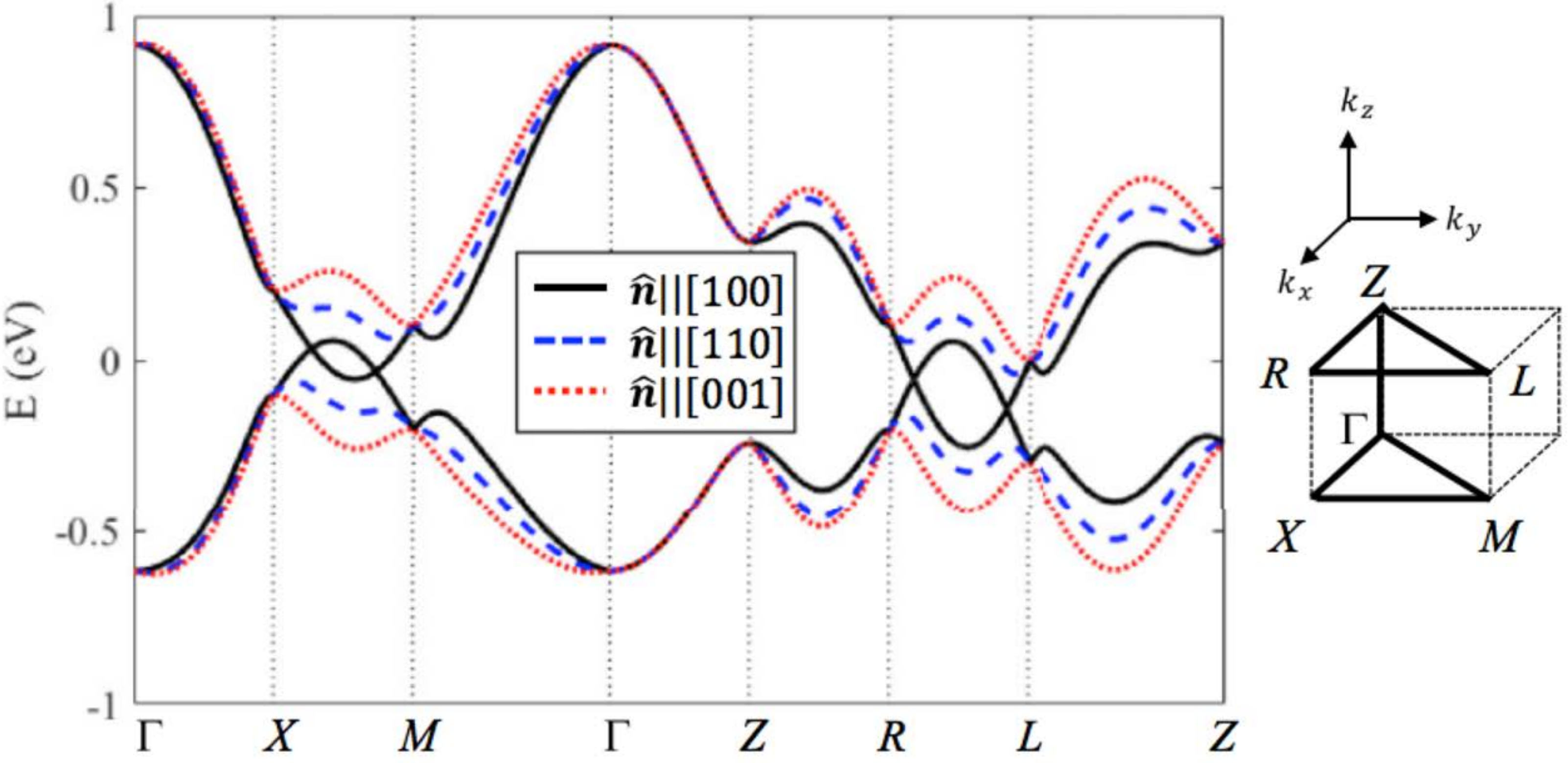
## 3) Model Hamiltonian Study

Kim Gilbert



Tight binding model in momentum space presents that gapped or gapless state is determined by orientation of antiferromagnetic order parameters.



# Voltage-induced Switching

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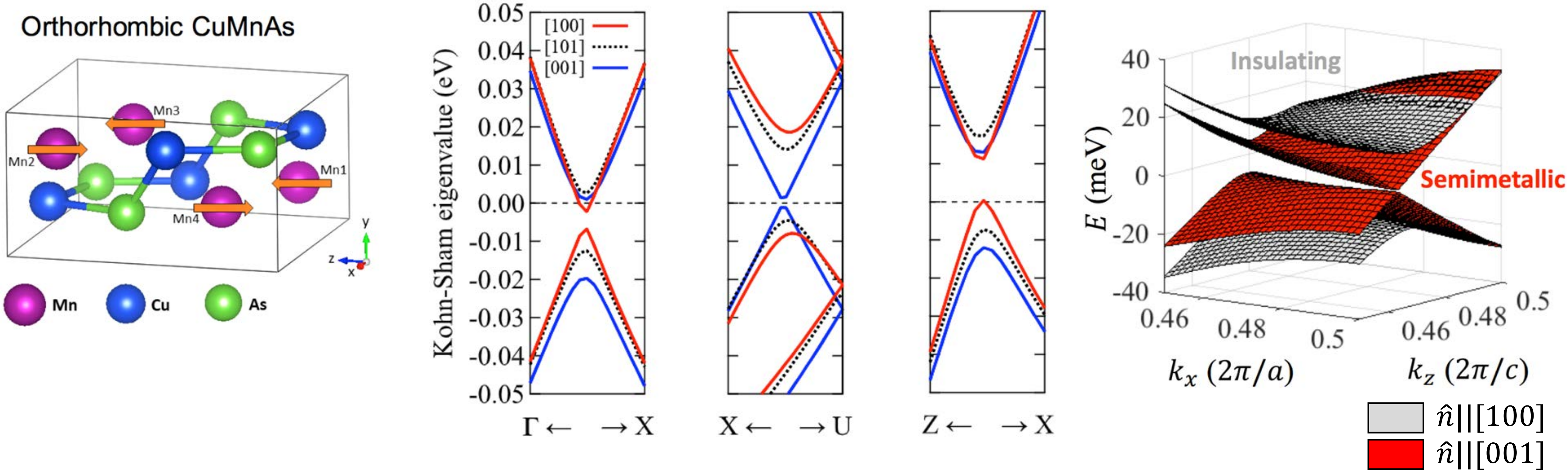
*Comput. Mater. Sci.* **160,** 207 (2019)

## 4) Density Functional Theory (DFT) Study

**Kang** **Schleife**

DFT calculates electronic band structure of orthorhombic CuMnAs and finds the states changes in terms of Néel vector.



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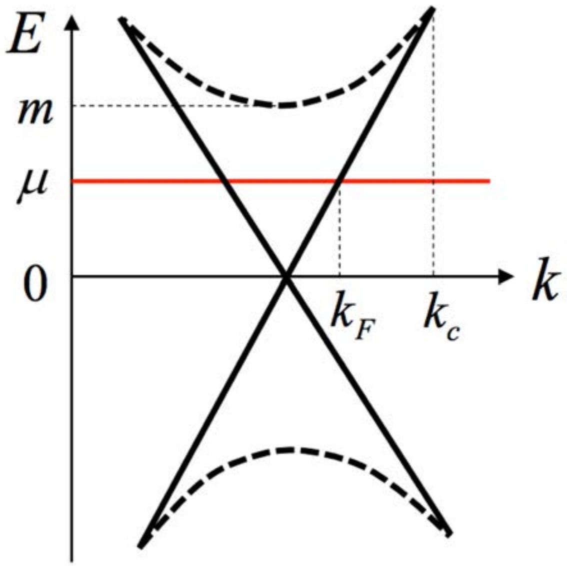
# Voltage-induced Switching

## 5) New Switching Process Prediction

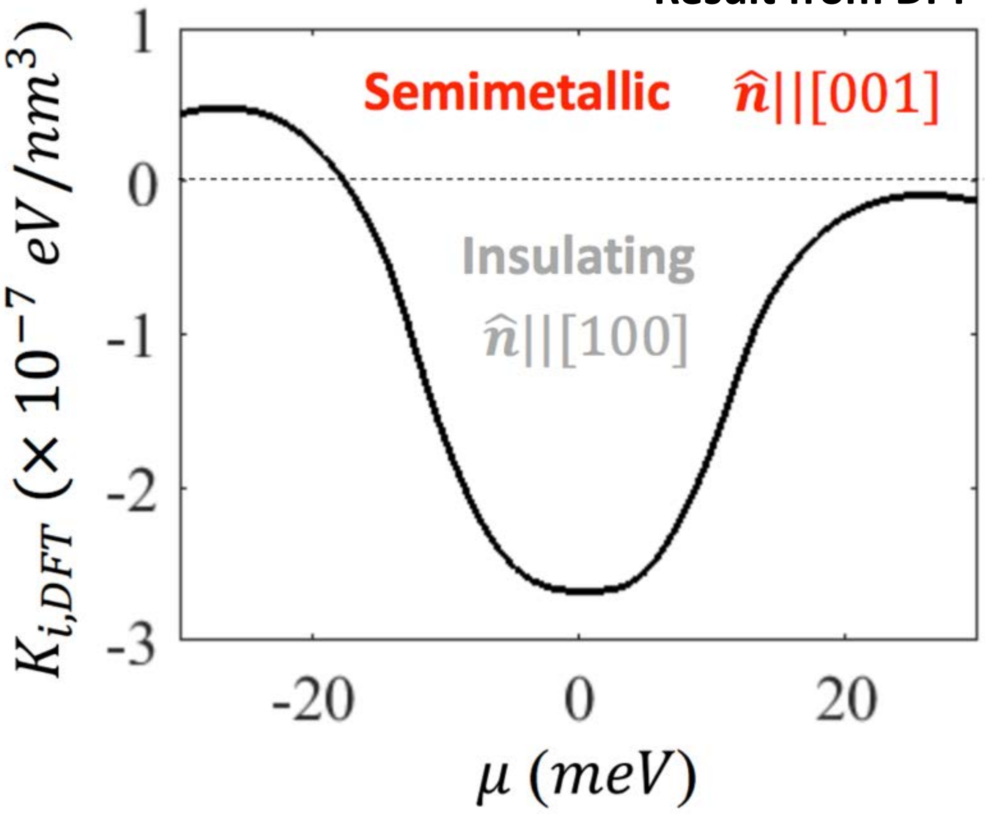
### Voltage-induced switching

- By tuning the chemical potential, anisotropy energy can be changed.
- Thus, MIT can occur by tuning the chemical potential

$$K_i(\mu) = \frac{F_{\rightarrow} - F_{\uparrow}}{V} \propto -\frac{1}{4v_F^3}\left(m^2 (v_F k_C)^2 - \frac{\mu^4}{3}\right)$$




**Result from DFT**



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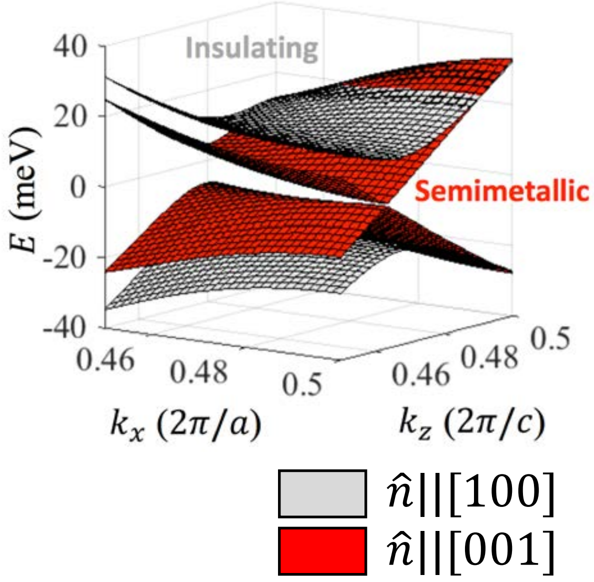
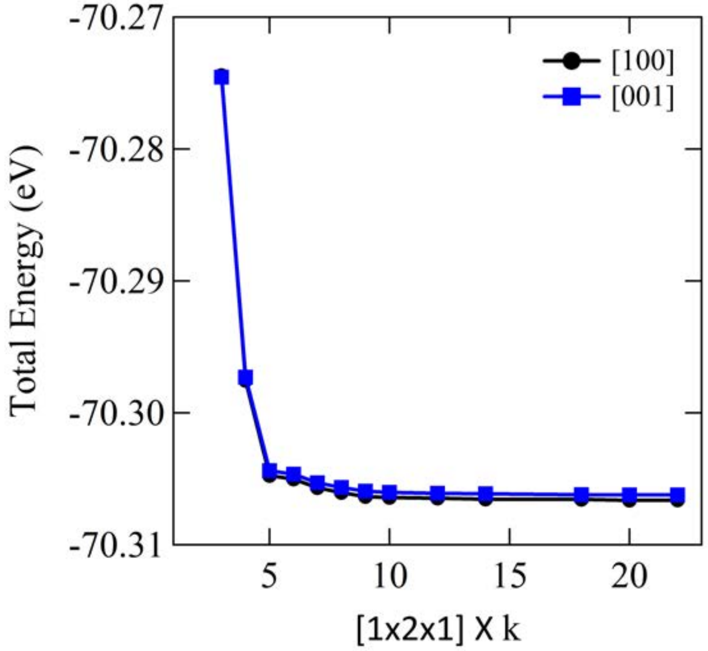
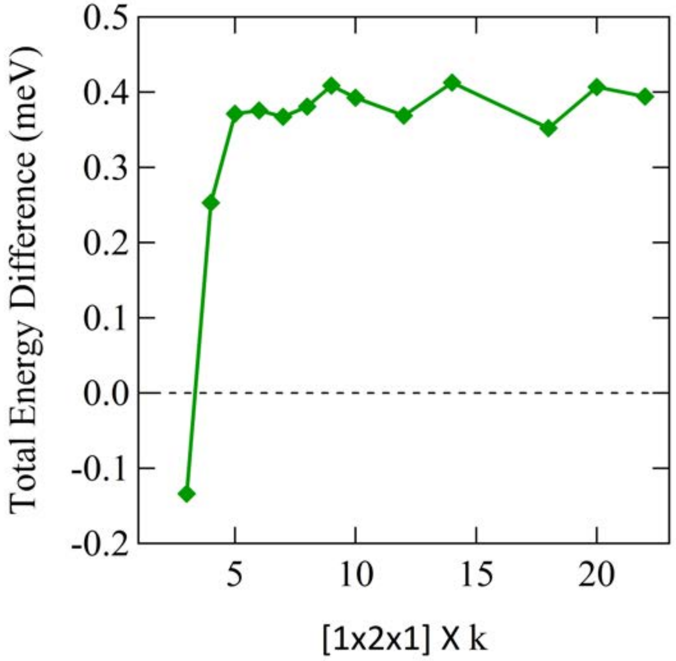
# Voltage-induced Switching

## 6) Why Blue Waters?




### Total Energy Convergence Test




### Computational Details

- Implemented by Vienna *ab initio* simulation package (VASP)
- Generalized-gradient approximation by Perdew, Burke, and Ernzerhof (PBE) for exchange and correlation description with plane kinetic cutoff energy of 600 eV
- *k*-points mesh grid gradually increases up to $22 \times 44 \times 22$ **(total 21296 points)**
- **Noncollinear magnetism** and **spin-orbit coupling effect** are included
- Each calculation requires about **3000 node hours with 140 GB memory**

# Introduction

## 2) Projects

### Project 1

Voltage-induced switching of antiferromagnetic semimetal

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- Model Hamiltonian for symmetry study
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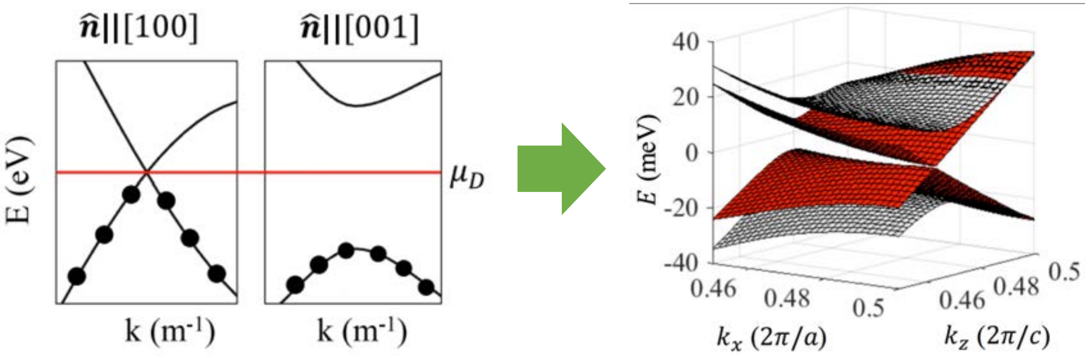


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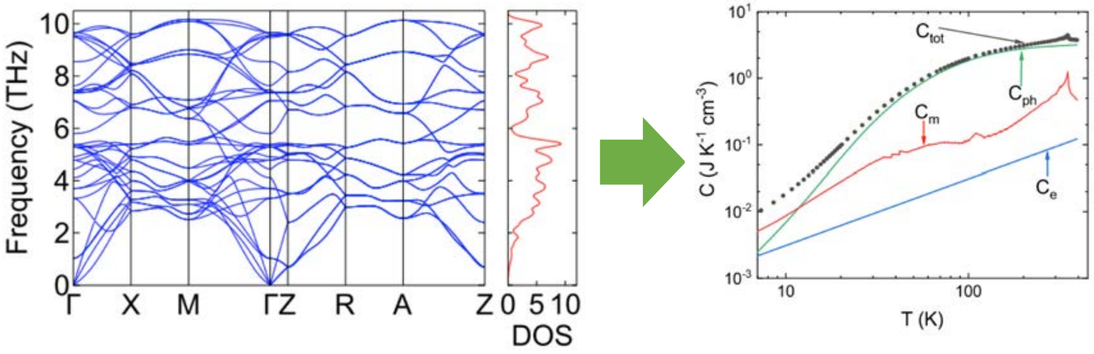
Optical and thermal properties of antiferromagnetic metallic $Fe_2As$

- Linear magneto-optical Kerr effect (MOKE) from antiferromagnetic metal under external magnetic field is predicted
- Relationship between quadratic MOKE and heat capacity is confirmed




*arXiv*:1903.07810

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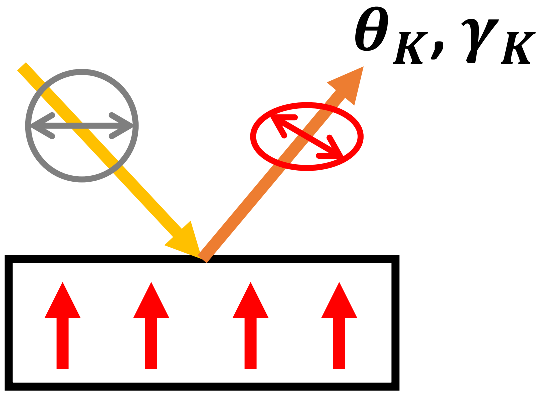
# Optical and Thermal Properties of $Fe_2As$

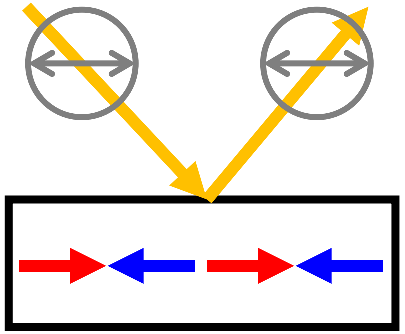
## 1) Background and Theory

**Linear Magneto-Optical Kerr Effect (Linear MOKE)**

[Ferromagnetic Case]




[Antiferromagnetic Case]

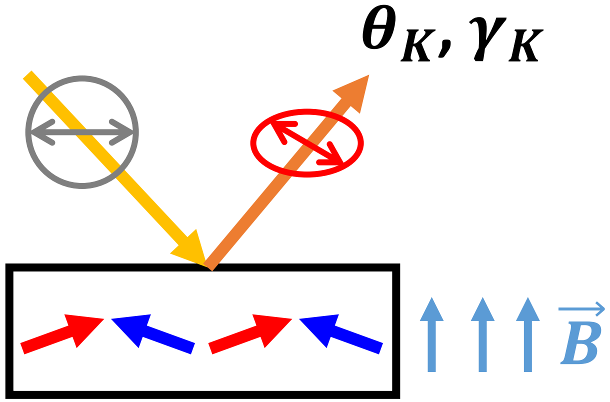


**How to utilize optical detection for antiferromagnets?**

### Linear MOKE under external magnetic field

- Spin-tilted state calculation
- Electronic band structure
- Dielectric function

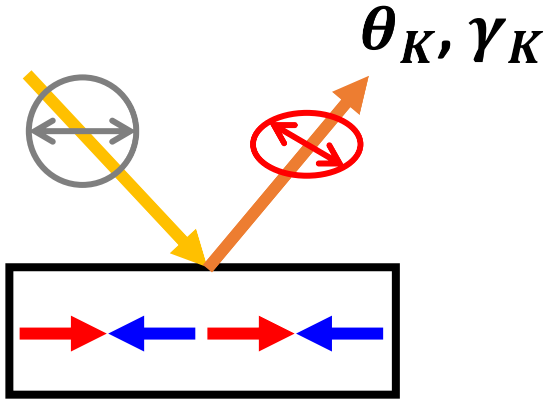
Optical property




### Quadratic MOKE

- Related to magnetic heat capacity
- Experiments cannot decompose heat capacity contribution of electron, phonon and magnon

Thermal property



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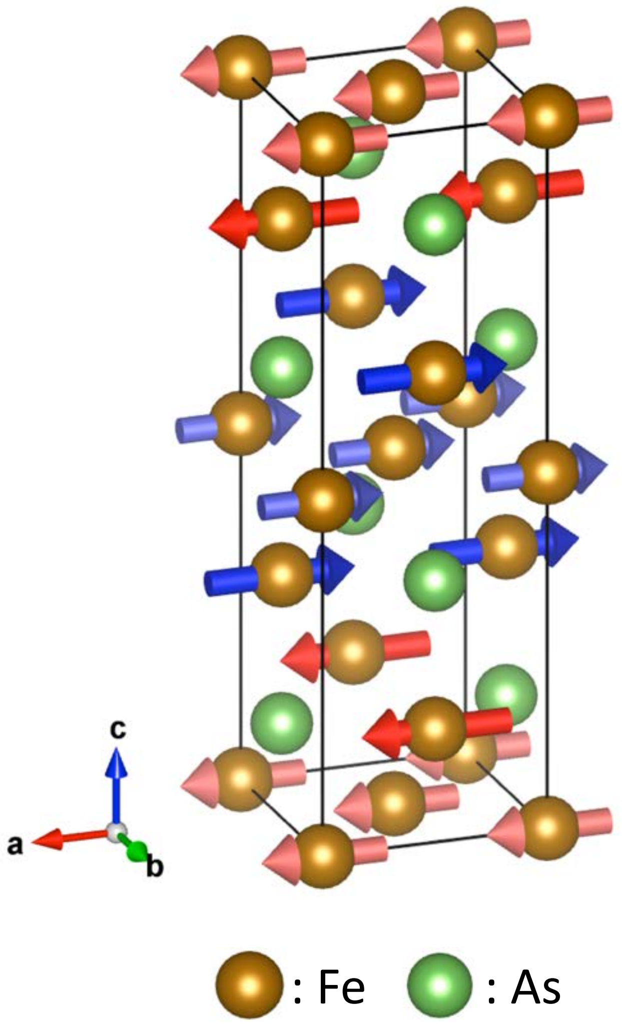
# Optical and Thermal Properties of $Fe_2As$

## 2) Magnetic Ground State




### Lattice Parameters

| $Fe_2As$ | a (Å) | b (Å) | c (Å) |
|---|---|---|---|
| **DFT** | 3.624 | 3.624 | 11.724 |
| **Exp. [1]** | 3.630 | 3.630 | 11.96 |
| **Δ (%)** | -0.17 | -0.17 | -1.97 |

### Magnetic Moments

**[300 K]** → **[0 K]**

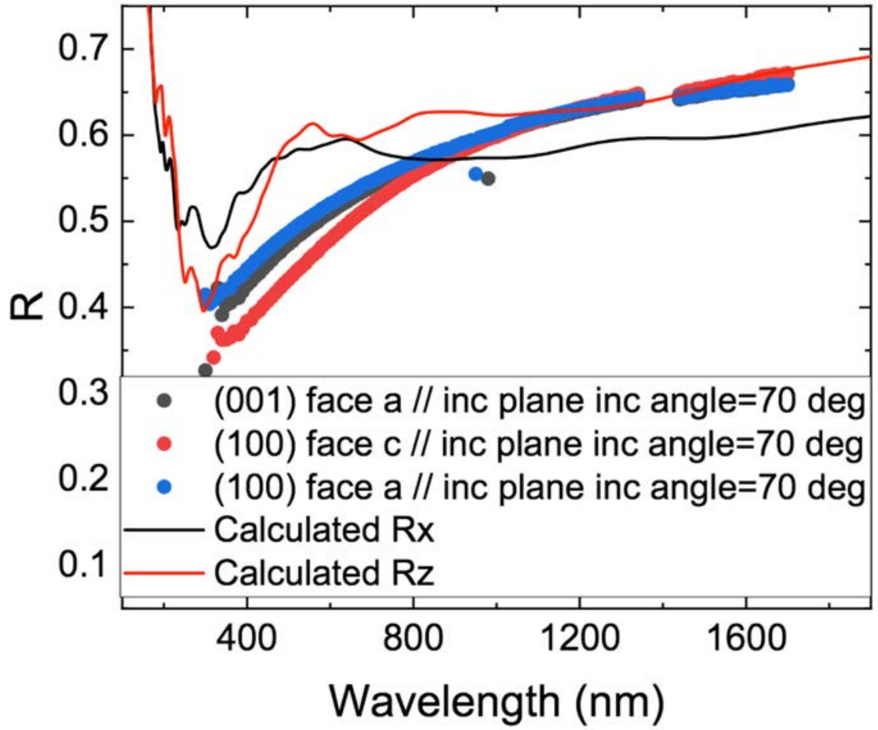
$M1^{exp.} = 0.95\ \mu_B$ → $1.28\ \mu_B$

$M2^{exp.} = 1.52\ \mu_B$ → $2.05\ \mu_B$

$M1^{DFT} = 1.24\ \mu_B$

$M2^{DFT} = 2.25\ \mu_B$

### Reflectivity of $Fe_2As$



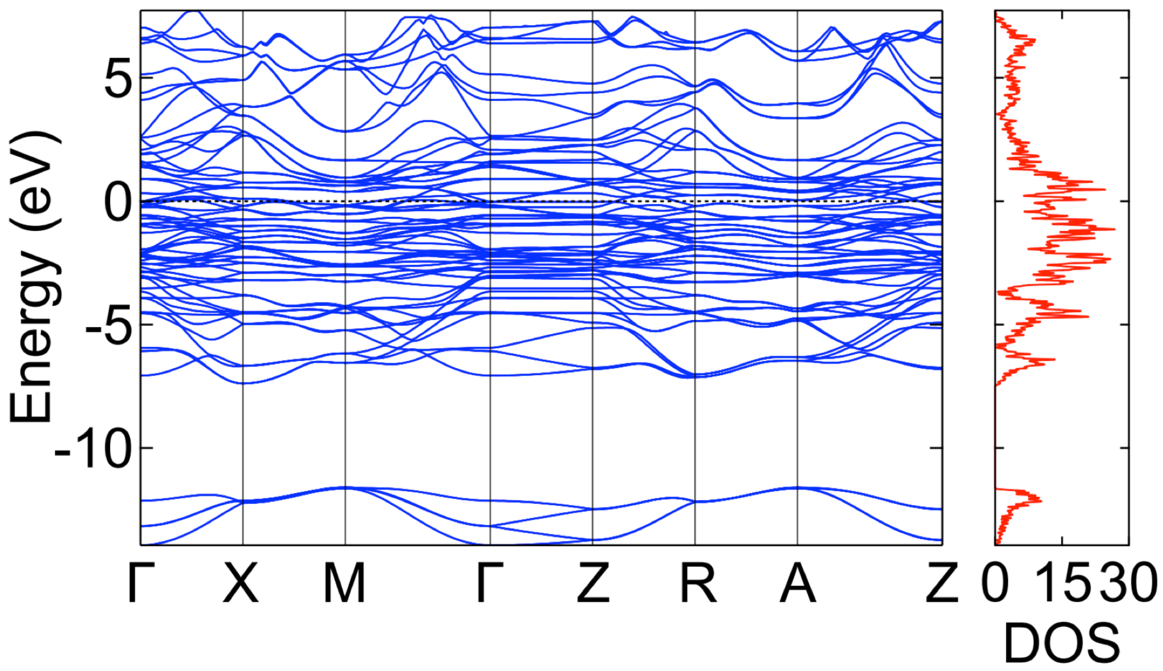

[1] H. Katsuraki *et al. J. Phys. Soc. Jpn.* **21,** 2238 (1966)

arXiv:1903.07810

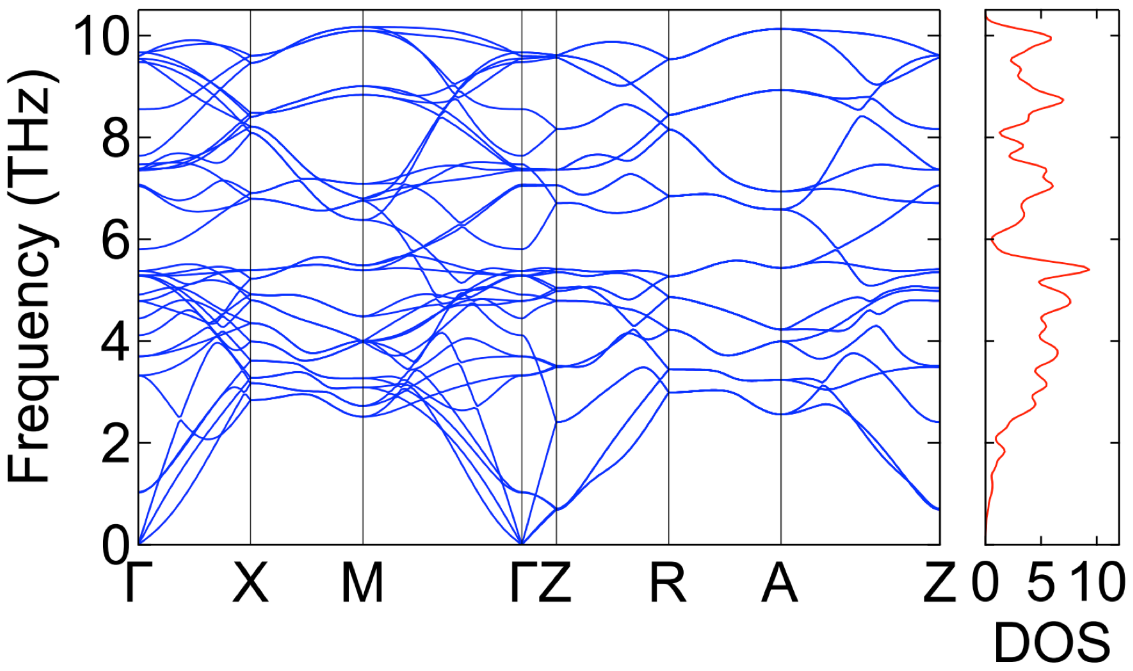
# Optical and Thermal Properties of $Fe_2As$

## 3) Band Dispersion Study

Electronic Band structure




Phonon Band structure




- Dielectric function
- Linear MOKE rotation and ellipticity signals
- Electron heat capacity

- Phonon heat capacity

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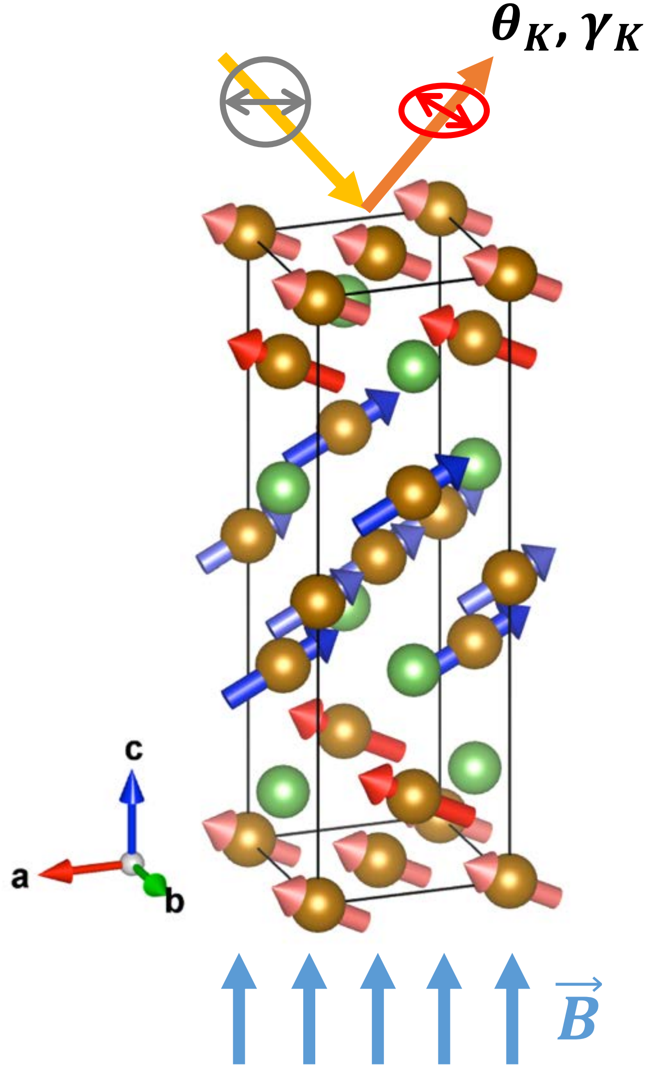
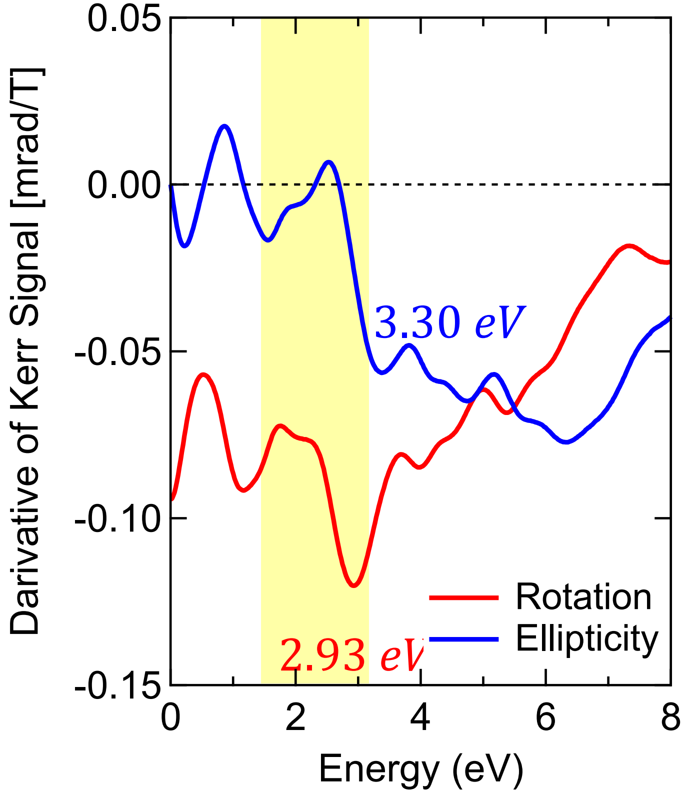
# Optical and Thermal Properties of $Fe_2As$

## 4) Linear MOKE study

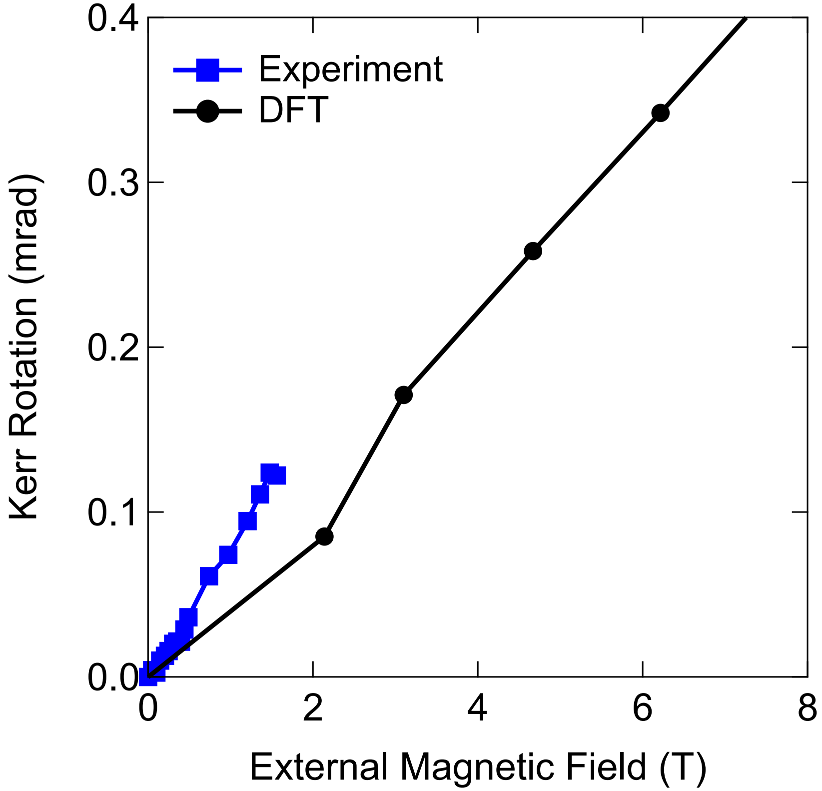
### Spin-tilted State




### Spectral Results




### Kerr Rotation at 793 nm (1.56 eV)




$$\Psi_K(\omega) = \theta_K(\omega) + i\gamma_K(\omega) = \frac{-\varepsilon_{xy}(\omega)}{(\varepsilon_{xx}(\omega) - 1)\sqrt{\varepsilon_{xx}(\omega)}}$$

Rotation | Ellipticity of reflected light

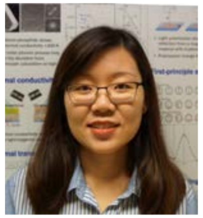
This can be the guidance to experimentalist to find the wavelength to maximum the MOKE response

# Optical and Thermal Properties of $Fe_2As$

## 5) Quadratic MOKE study

$C_{tot}, \frac{\Delta\Theta}{\Delta T}$ from Experiment

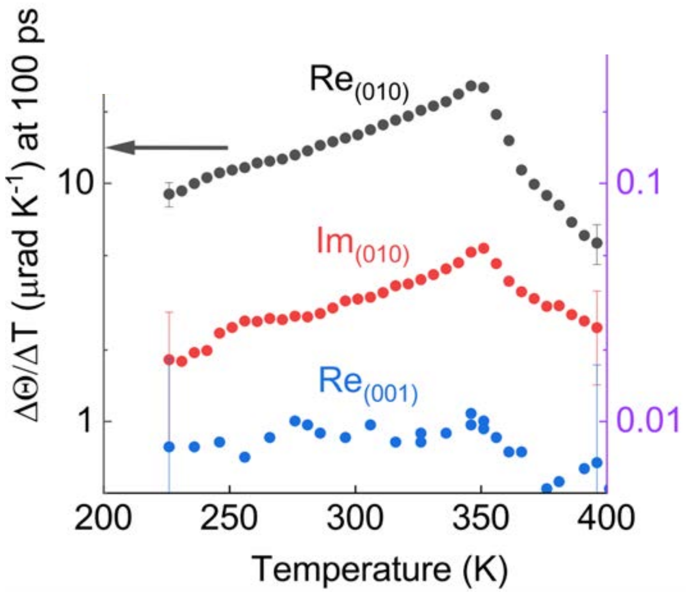
**Yang** **Cahill**



$C_e$ and $C_{ph}$ from DFT

**Kang** **Schleife**

Quadratic MOKE response



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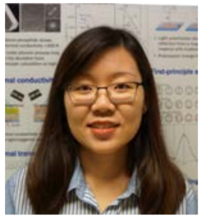
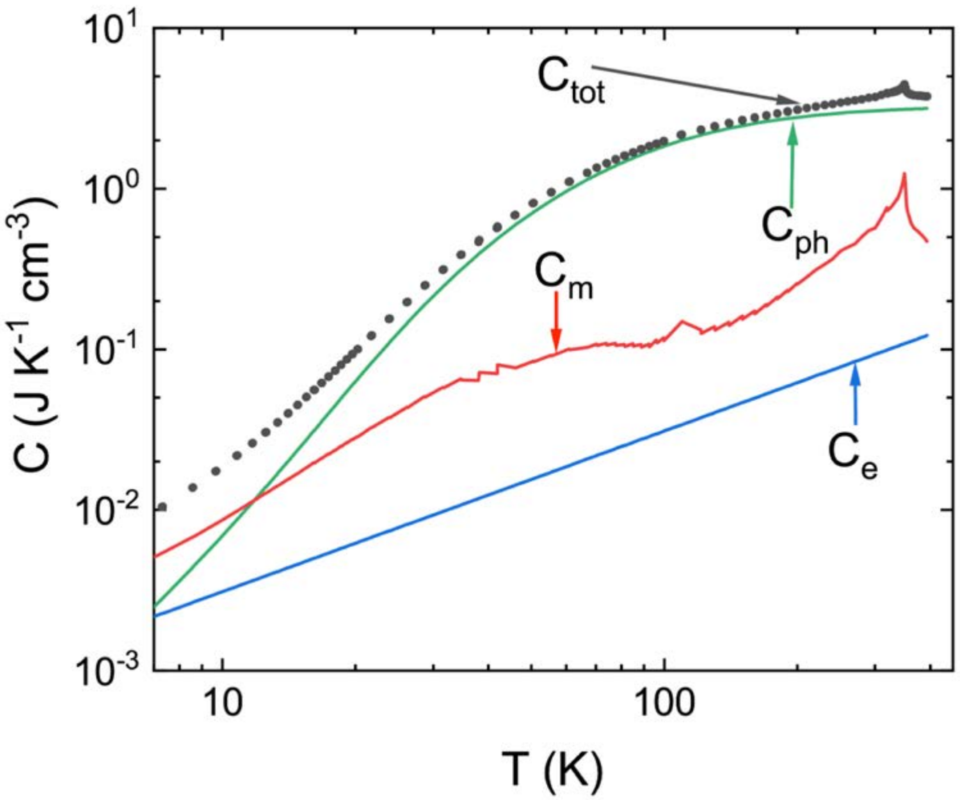



$C_e$ and $C_{ph}$ from DFT

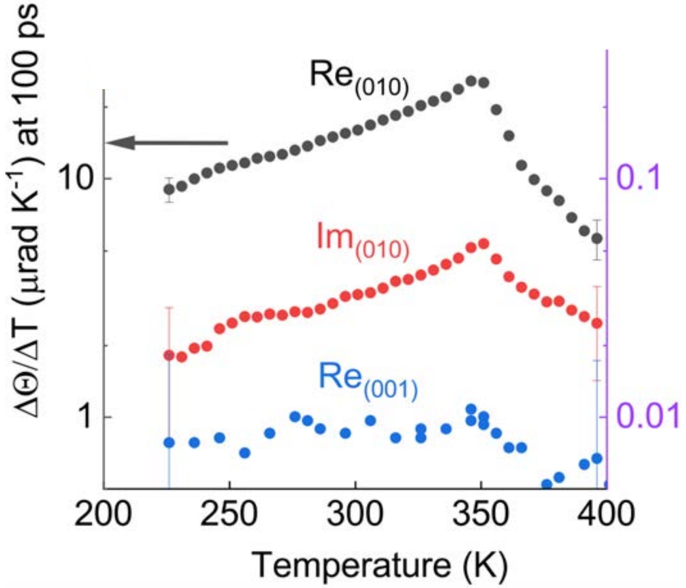
**Kang** **Schleife**


### Heat Capacity Analysis




$C_m = C_{tot} - C_e - C_{ph}$
Magnetic heat capacity can be extracted.

### Quadratic MOKE response



# Optical and Thermal Properties of $Fe_2As$

*arXiv*:1903.07810

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$C_{tot}, \frac{\Delta\Theta}{\Delta T}$ from Experiment

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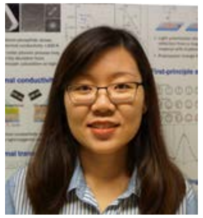
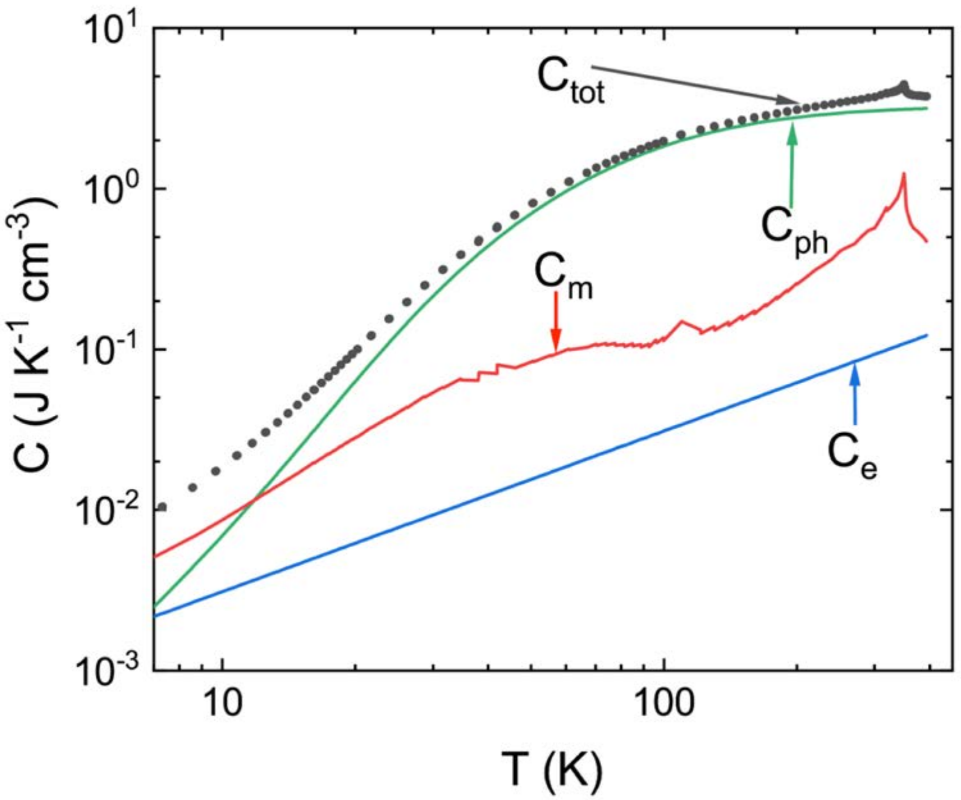



$C_e$ and $C_{ph}$ from DFT

**Kang** **Schleife**


### Heat Capacity Analysis

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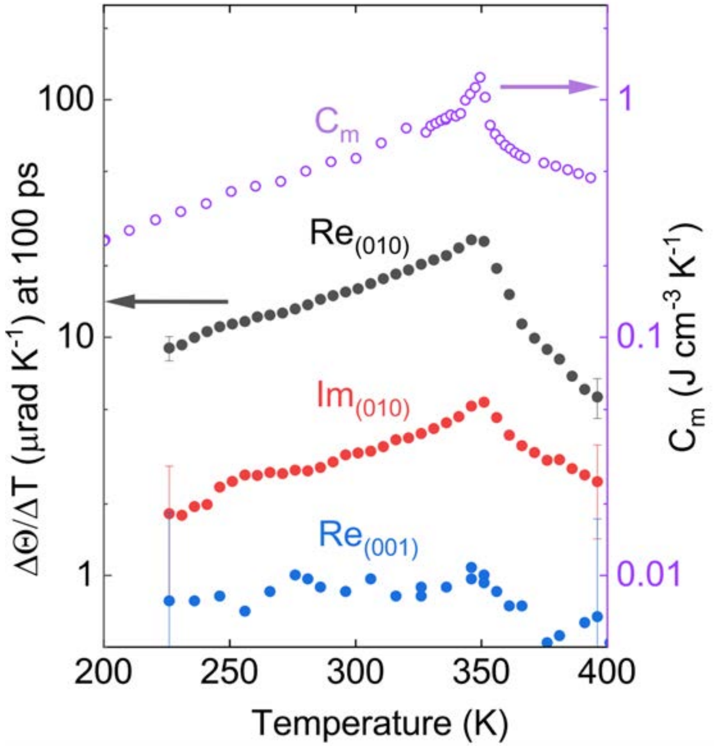
### Quadratic MOKE response




Quadratic MOKE response is dominantly related to magnetic heat capacity ($C_m$)

*arXiv*:1903.07810

# Optical and Thermal Properties of $Fe_2As$

## 6) Why Blue Waters?

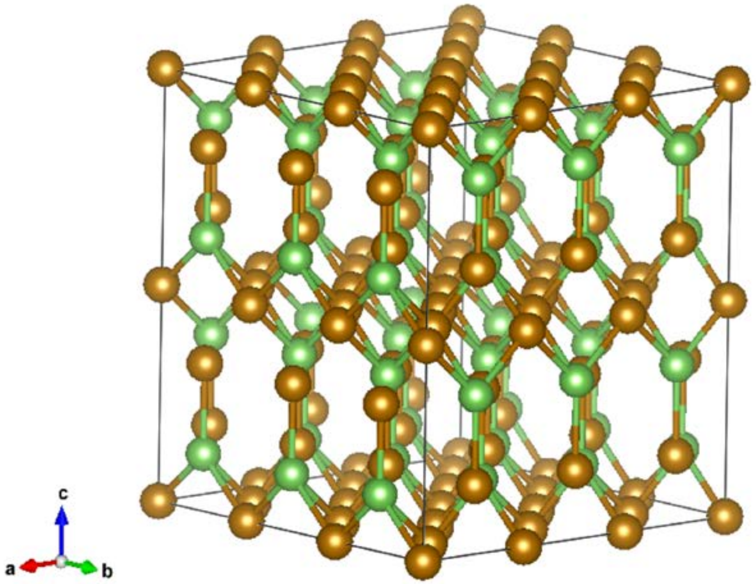
### Computational Details

- Implemented by Vienna *ab initio* simulation package (VASP)
- Generalized-gradient approximation by Perdew, Burke, and Ernzerhof (PBE) for exchange and correlation description with plane kinetic cutoff energy of 500 eV

- For phonon calculation, **supercell** $\mathbf{3 \times 3 \times 2}$ from chemical structures is used
- Total atoms are **108 atoms** (72 Fe atoms and 36 As atoms)
- **Noncollinear magnetism** and **spin-orbit coupling effect** are included
- *k*-points in Brillouin zone is sampled by $\mathbf{4 \times 4 \times 4}$ mesh grid

- Instead of one long calculation in Density Functional Perturbation Theory, Phonopy provides **6 displacements** calculations which is suitable with in wall time.
- Each calculation requires around **2700 node hours with 53 GB memory**.
- Total wavefunctions occupy **8.4 TB** storage space.

Phonopy

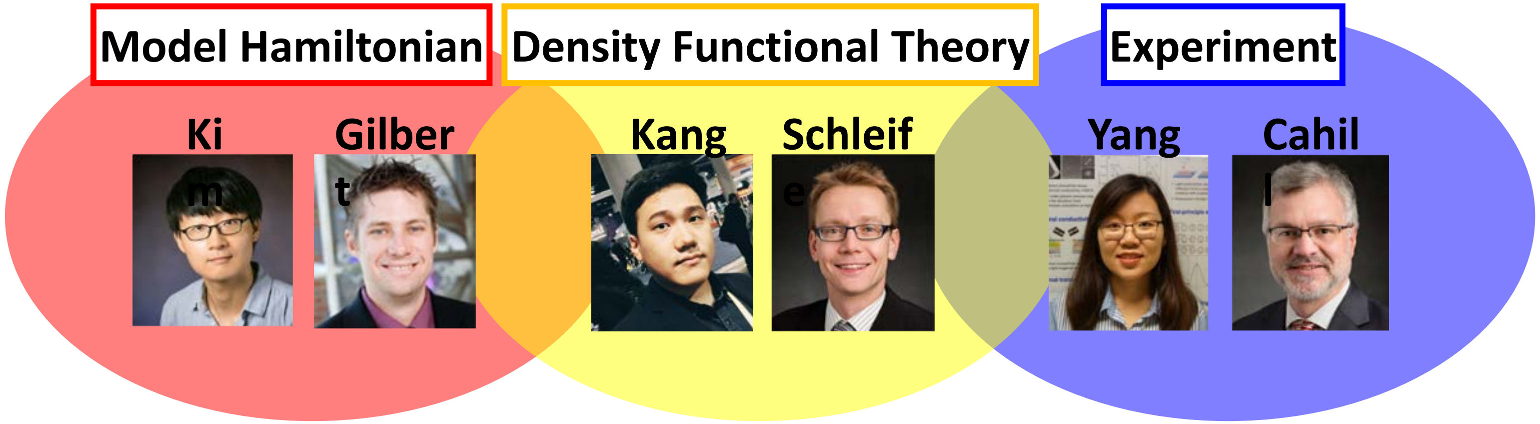


*Phys. Rev. B* **97,** 134415 (2018)
*arXiv*:1903.07810

# Conclusion

## 1) Summary

- **(Semi)metal-insulator transition** in specific symmetry condition can happen through reorientation of antiferromagnetic order parameter
- **Voltage-induced switching** is predicted by **model Hamiltonian study** and confirmed by **DFT** in orthorhombic CuMnAs
- **Linear MOKE** signal generation from antiferromagnetic $Fe_2As$ under external magnetic field is predicted by **DFT** and confirmed by **experiment**
- **Magnetic heat capacity** extracted by combination of measurement and calculation presents close relationship with **quadratic MOKE signal**



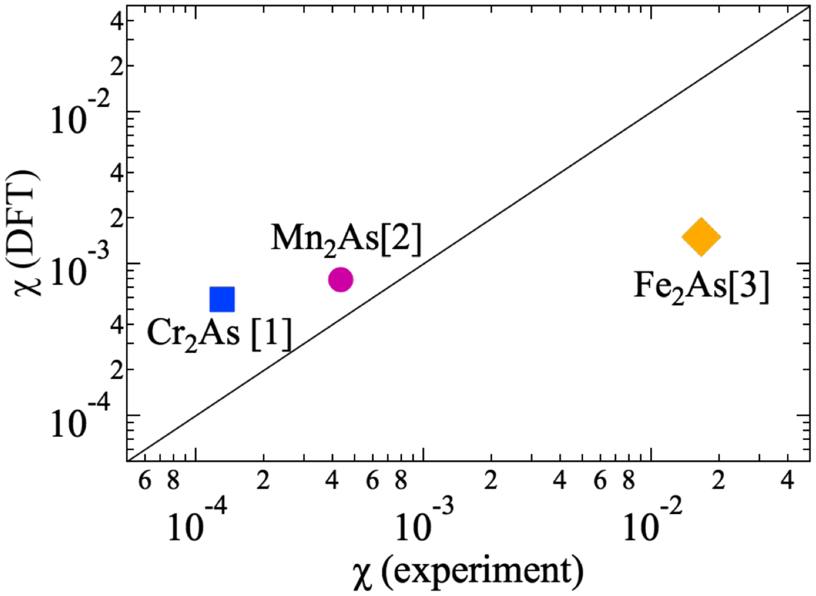

I-MRSEC

BLUE WATERS
SUSTAINED PETASCALE COMPUTING

Kisung Kang

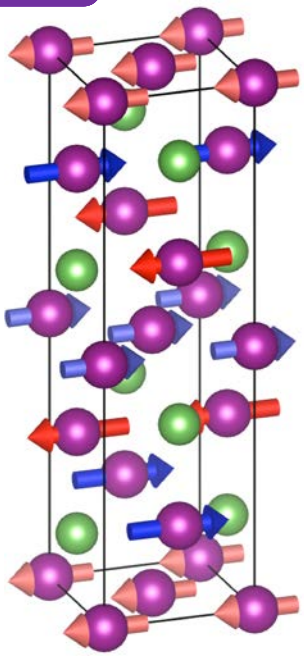
# Spin Tilting Calculation: $M_2As$ (M= Cr, Mn, Fe)

## 3) Magnetic Susceptibility

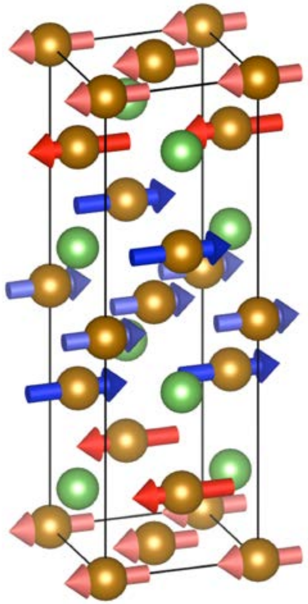



[1] M. Yuzuri, *J. Phys. Soc. Jpn.* **15,** 2007 (1960)
[2] M. Yuzuri *et al. J. Phys. Soc. Jpn.* **15,** 1845 (1960)
[3] H. Katsuraki *et al. J. Phys. Soc. Jpn.* **21,** 2238 (1966)

**$Mn_2As$**



**$Fe_2As$**

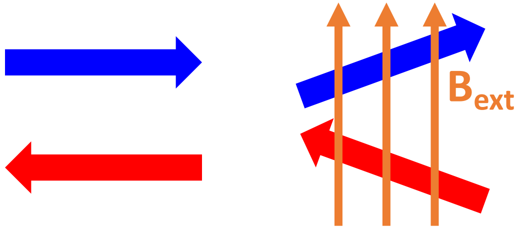


### Effective Exchange Parameters [4]

[4] Y. Zhang *et al. Inorg. Chem.* **52,** 3013 (2013)

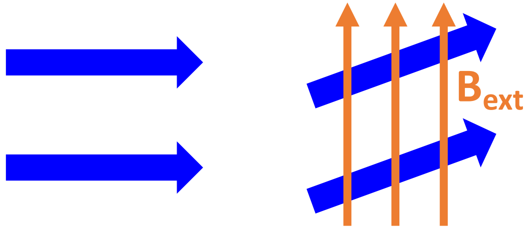
| (meV) | $Cr_2As$ | $Mn_2As$ | $Fe_2As$ |
|---|---|---|---|
| $J_{M1-M1}$ | $-14.1$ | $-1.68$ | $+25.4$ |
| $J_{M1-M2}$ | $-7.85, -12.8$ | $-14.5$ | $+6.52$ |
| $J'^{\,a}_{M2-M2}$ | $+1.83$ | $-19.6$ | $-3.52$ |
| $J'^{\,b}_{M2-M2}$ | $-6.02$ | $-0.70$ | $+8.52$ |

**Antiferromagnetic Coupling**




Energetically large change

**Ferromagnetic Coupling**

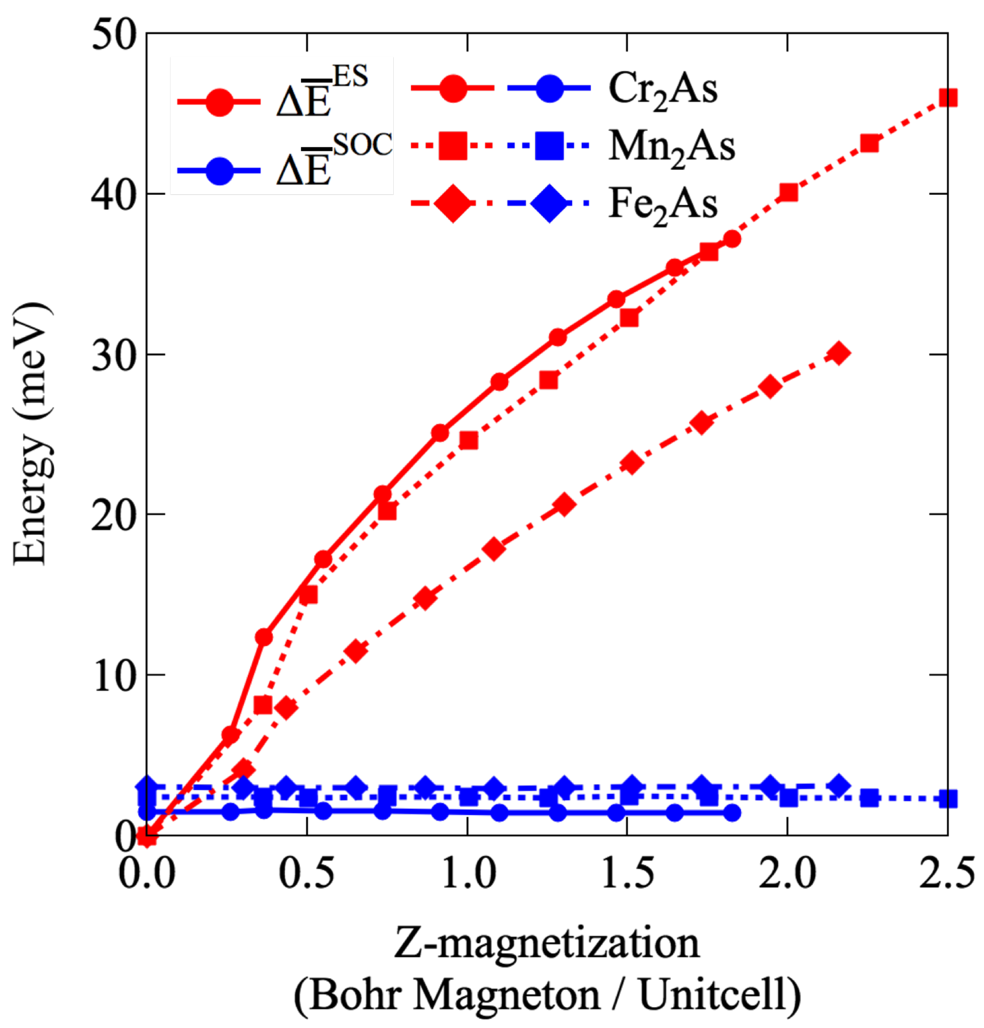



Energetically small change

Kisung Kang

# Polar Magneto-Optical Kerr Effect (PMOKE) : $M_2As$

## 5) Tilting Angle Dependence of ES and SOC




$$\Delta\bar{E}^{ES} = \sum_{\vec{k},i} \frac{\left|E^{maj.}(\vec{k},i) - E^{min.}(\vec{k},i)\right|}{N_{\vec{k}}N_i}, \quad \Delta\bar{E}^{SOC} = \sum_{\vec{k},i} \frac{\left|E^{SOC}(\vec{k},i) - E^{woSOC}(\vec{k},i)\right|}{N_{\vec{k}}N_i}$$

where $\vec{k}$ is $k$-point in first Brillouin Zone, $i$ is band index
$N_{\vec{k}}$ is number of k-point in first BZ, $N_i$ is number of bands
$E^{maj.}(\vec{k},i)$ is majority spin energy of i$^{th}$ band at $\vec{k}$
$E^{min.}(\vec{k},i)$ is minority spin energy of i$^{th}$ band at $\vec{k}$
$E^{SOC}(\vec{k},i)$ is energy of i$^{th}$ band at $\vec{k}$ with SOC
$E^{woSOC}(\vec{k},i)$ is energy of i$^{th}$ band at $\vec{k}$ w/o SOC

As net magnetization arises,
(1) average energy of exchange splitting increases and
(2) average energy of spin-orbit coupling does not change.

PMOKE signal change from AFM $M_2As$ under external magnetic field is mostly originated from exchange splitting effect change.