특론: 가속기 실험실습 I

(NUCE719P-01/PHYS715P-01, 정모세)

Soft X-ray Spectroscopy for Next-Generation Devices: Applications to AI Semiconductors and Rechargeable Batteries

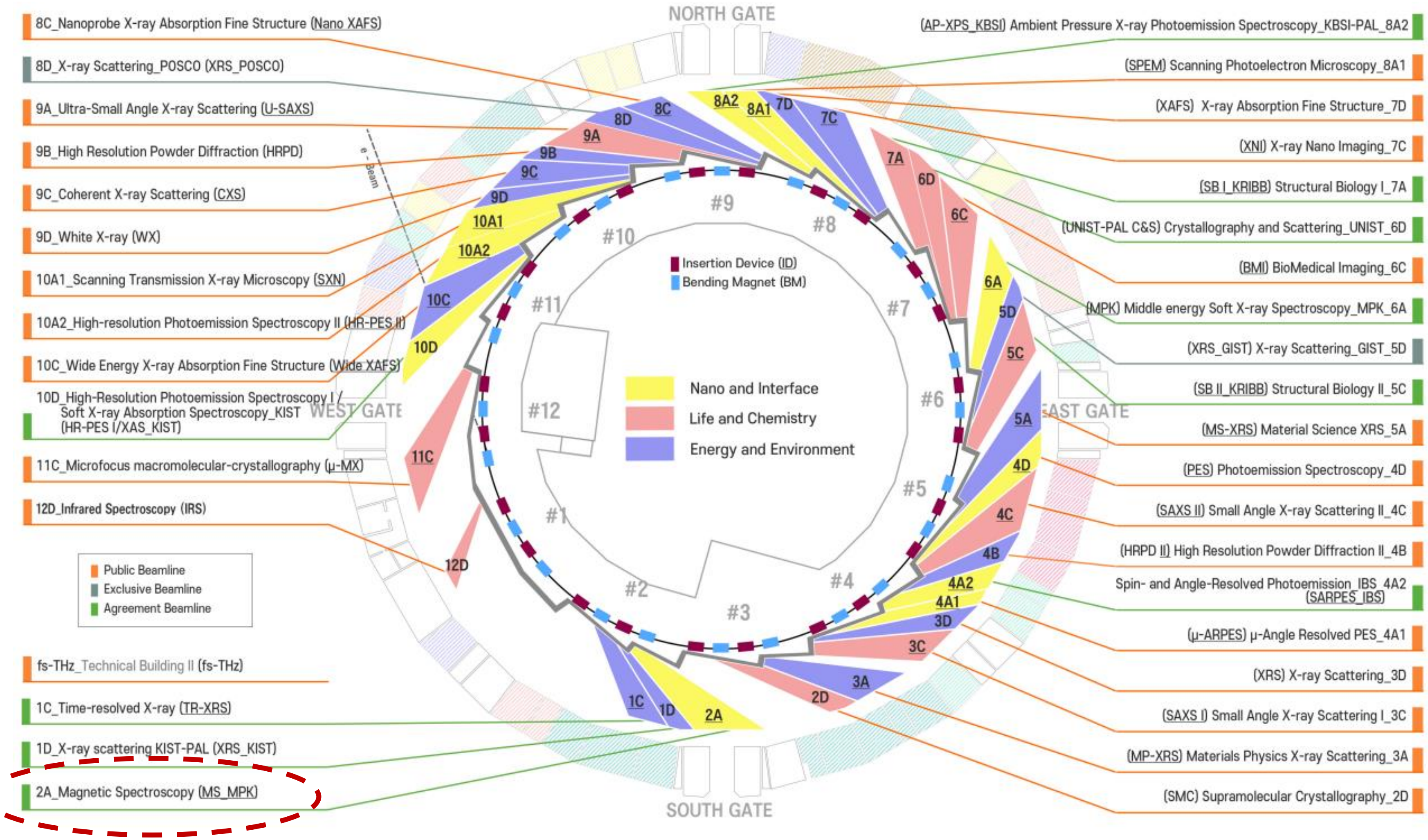
김 영 학

첨단원자력공학부

iyhkim@postech.ac.kr, PAL 저장링동 252호

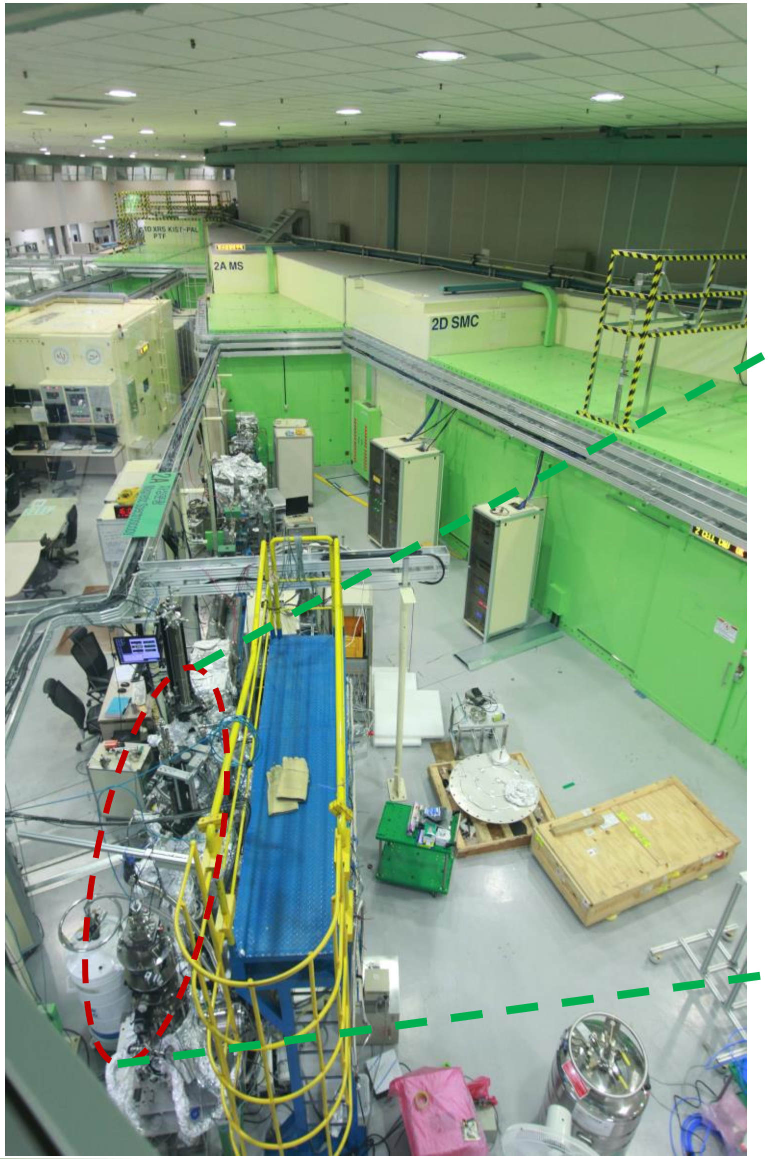
Beamline Map

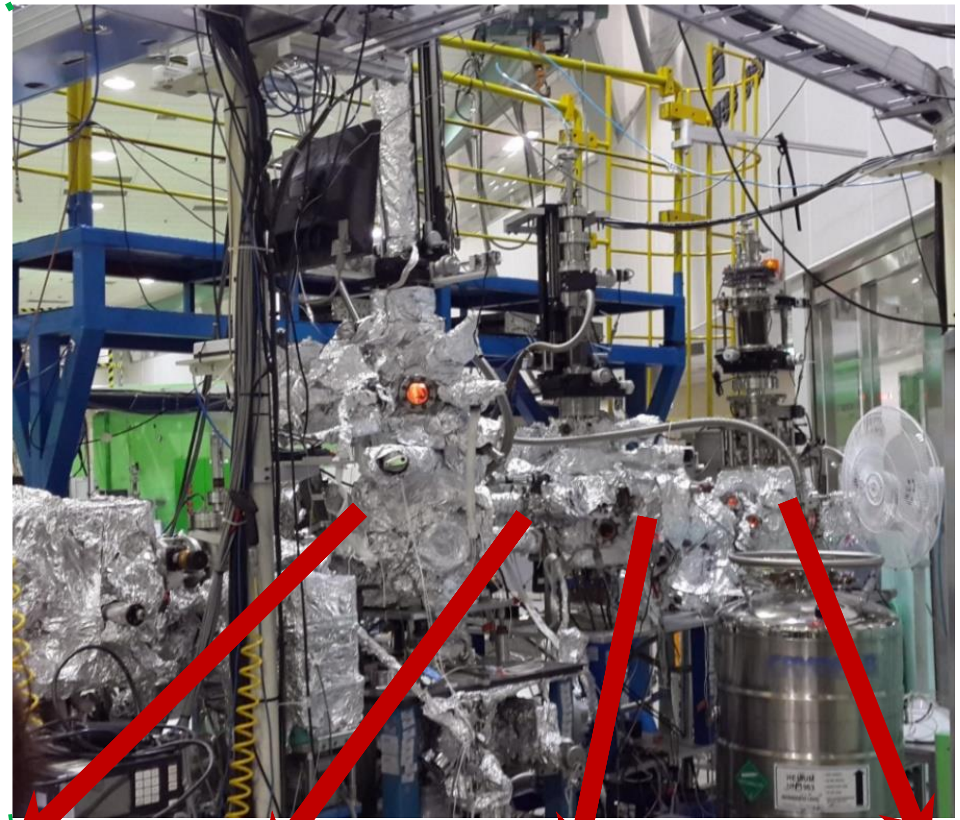
미래기반 가속기 전문인력양성 사업단



POHANG ACCELERATOR LABORATORY
Ministry of Science and ICT

2A Beamline Instruments





1st end-station
Photoemission

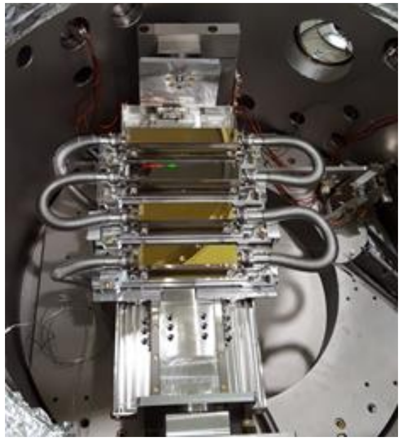
2nd end-station
XMCD

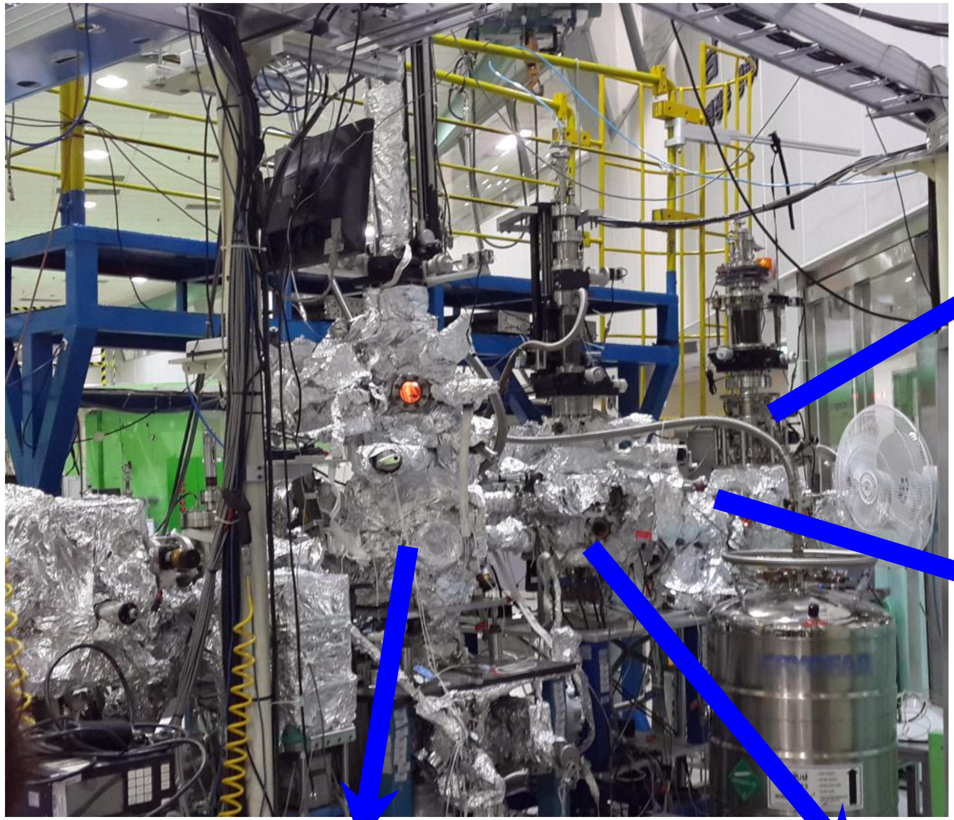
3rd end-station
XAS & XFMR

4th end-station
Soft x-ray Scattering

Four Experimental Stations of Magnetic Spectroscopy

Optics : SGM with four gratings





Fourth end-station

Soft x-ray Scattering

P ~ 1 x 10^{-10} torr (UHV)

Temperature : 5K ~ 370K

2 circle diffractometer

Electromagnet ~ 0.15 T

Third end-station

Polarization dependent XAS

P ~ 1 x 10^{-9} torr

Temperature : 7K ~ 370K

Quick measurement ~Vacuum recovered after 3 hours baking

First end-station

Photoemission

P ~ 3 x 10^{-11} torr

Temperature : 5K ~ 370K

Electron Analyzer (Scienta 100)

Second end-station

XMCD (total yield & fluorescence yield)

P ~ 4 x 10^{-10} torr

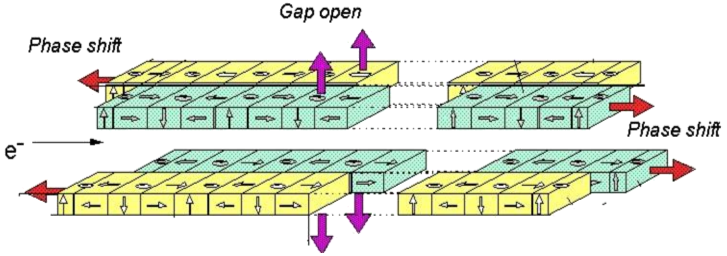
Temperature : 5K ~ 370K

Electromagnet ~ 1 T

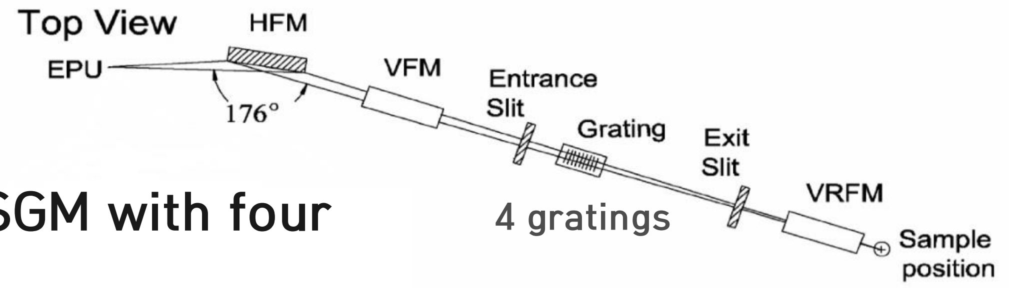
2A BL: Magnetic Spectroscopy

◆ Overview

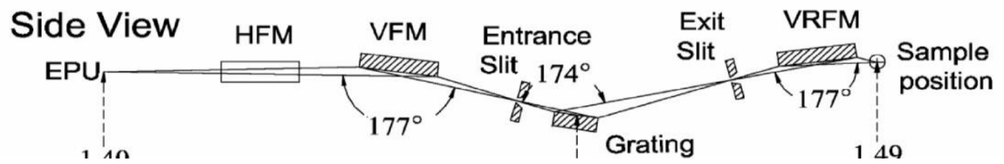
This beamline is designed for magnetic spectroscopy such as X-ray Absorption Spectroscopy (XAS), X-ray Magnetic Circular Dichroism (XMCD), X-ray Linear Dichroism(XLD) provided by elliptically or linearly polarized light



Elliptically Polarized Undulator



Optics : SGM with four gratings



◆ Research Area (Past)

→ **Spin-dependent electronic structure**

→ **Magnetic origin of transition metal oxide and rare-earth materials**

→ **Magnetic property of diluted magnetic semiconductor**

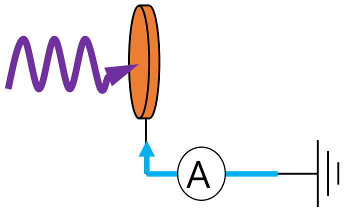
→ **Electronic orbital anisotropy**

→ **Magnetic thin film and multi-layer**

Photon energy resolving power : 5000 ~ 10000
Photon flux : 10^{10} ~ 10^{12} photons/sec
Beam size : 1 mm X 0.2 mm
Energy: 120 ~ 1500 eV (Typically 400 ~ 1400 eV)
Polarization switching : phase shift

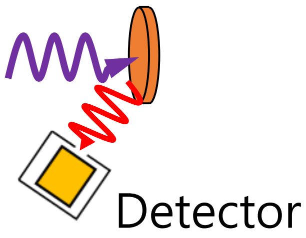
Total electron yield

measuring all excited electrons
→ proving depth ~ 150 Å



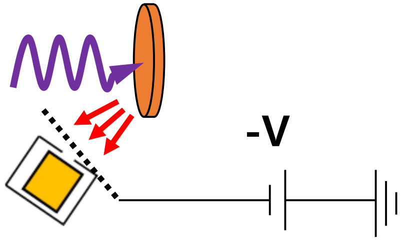
Fluorescence yield

measuring fluorescent photons
→ proving depth ~ 1000 Å



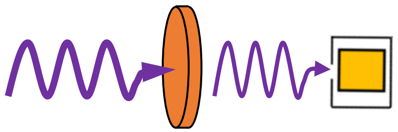
Partial electron yield

measuring electrons with higher kinetic energy than the retarding potential
→ proving depth ~ variable within ~ 50 Å



Transmission yield

measuring the intensity of transmitted light
→ proving depth ~ 1000 Å

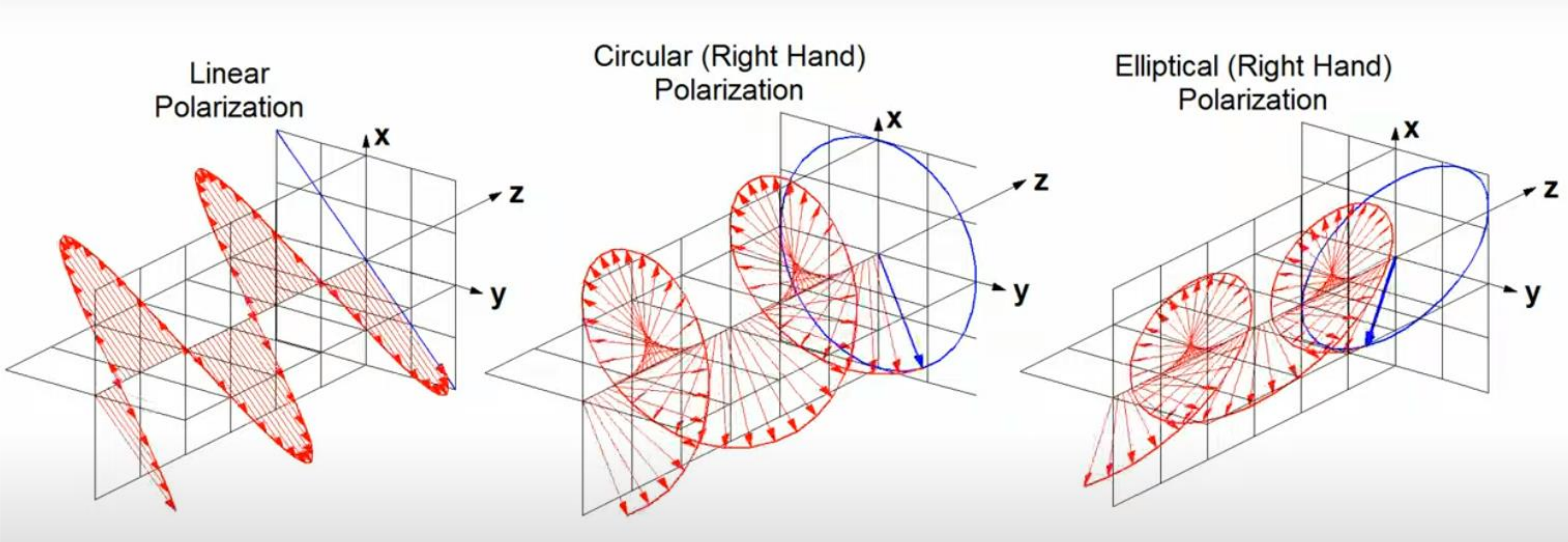


Polarization

미래기반 가속기
전문인력양성 사업단

Light: electromagnetic waves with wavelength, energy, frequency, polarization

Polarization : Describes the direction in which the electric field vector of an electromagnetic wave oscillates



출처: https://youtu.be/Q0qrU4nprB0

Magnetic Moments by XMCD

미래기반 가속기 전문인력양성 사업단

Sum rule:

$$m_{\text{orb}} = -\frac{4\int_{L_3+L_2}(\mu_+ - \mu_-)\,d\omega}{3\int_{L_3+L_2}(\mu_+ + \mu_-)\,d\omega}(10 - n_{3d}), \quad (1)$$

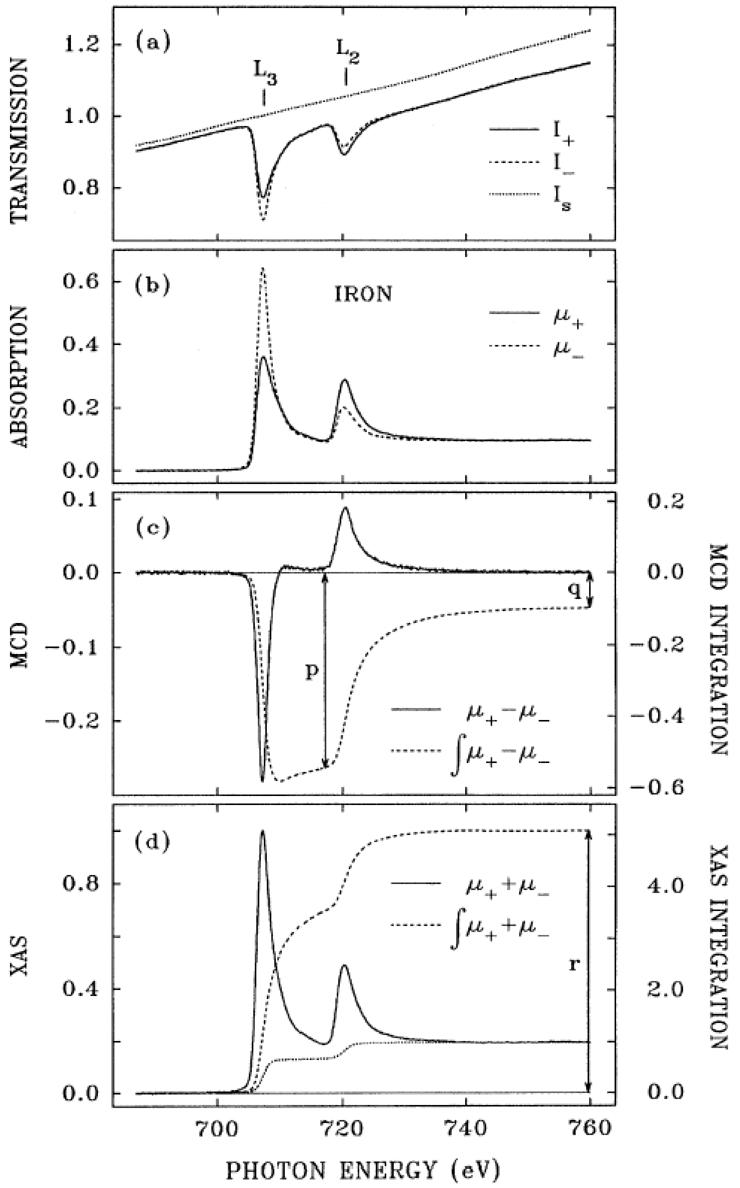
$$m_{\text{spin}} = -\frac{6\int_{L_3}(\mu_+ - \mu_-)\,d\omega - 4\int_{L_3+L_2}(\mu_+ - \mu_-)\,d\omega}{\int_{L_3+L_2}(\mu_+ + \mu_-)\,d\omega} \times (10 - n_{3d})\left(1 + \frac{7\langle T_z\rangle}{2\langle S_z\rangle}\right)^{-1}, \quad (2)$$

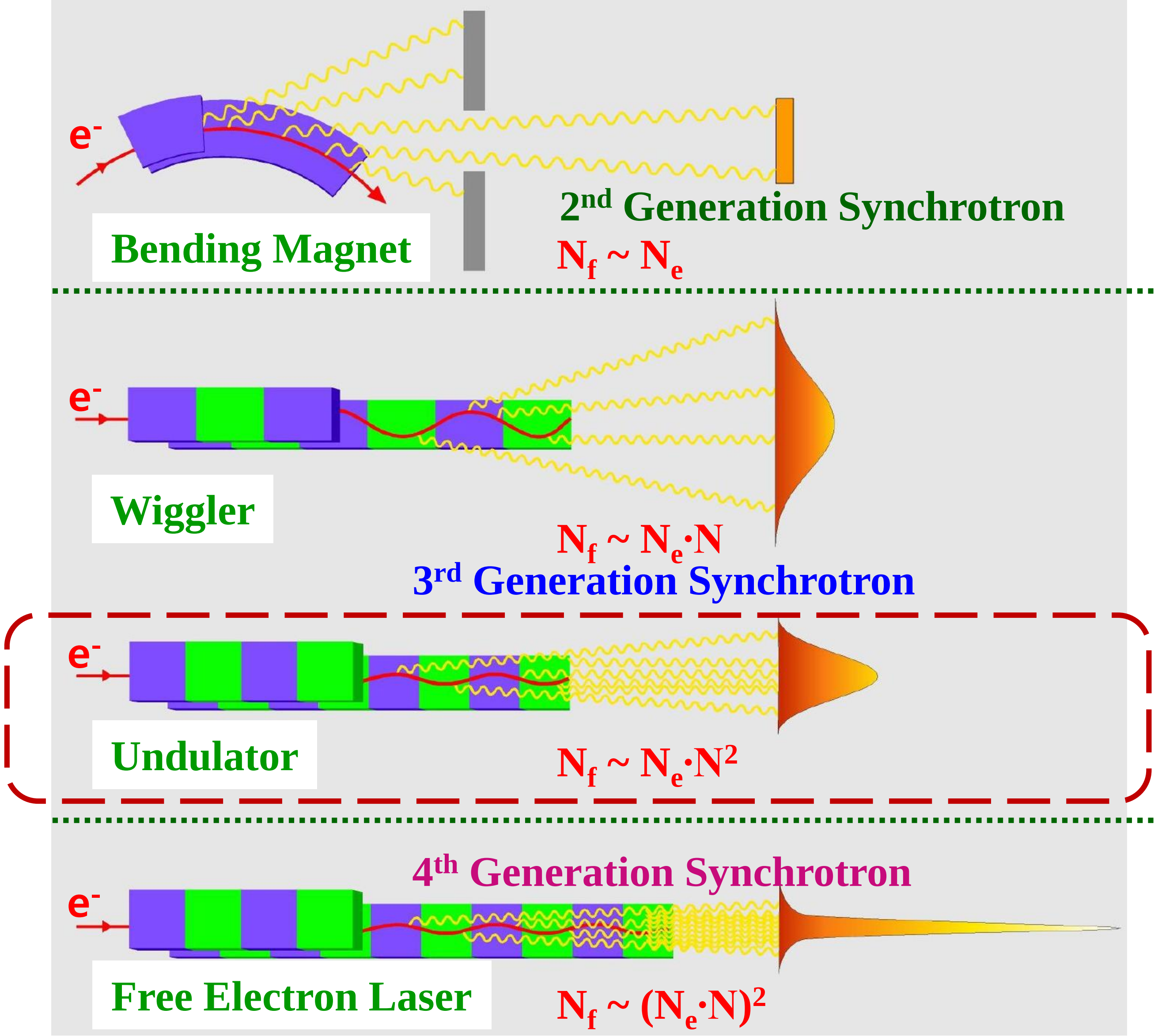
Orbital and Spin Moments
Element Specific Moments

TABLE I. Orbital and spin magnetic moments of bcc Fe and hcp Co in units of μ_B/atom.

	Fe (bcc)			Co (hcp)		
	$m_{\text{orb}}/m_{\text{spin}}$	m_{orb}	m_{spin}	$m_{\text{orb}}/m_{\text{spin}}$	m_{orb}	m_{spin}
MCD and sum rules	0.043	0.085	1.98	0.095	0.154	1.62
Gyromagnetic ratio [16]	0.044	0.092	2.08	0.097	0.147	1.52
OP-LSDA [17]	0.042	0.091	2.19	0.089	0.140	1.57
OP-LSDA (with OP off) [17]	0.027	0.059	2.19	0.057	0.090	1.57
SPR-LMTO [10]	0.020	0.043	2.20	0.054	0.087	1.60
FLAPW [11]	0.023	0.050	2.16	0.045	0.071	1.58
MCD and sum rules (corrected)	0.043	0.086	1.98	0.099	0.153	1.55

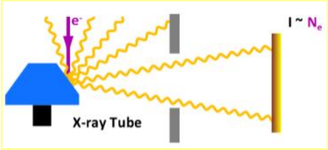
Chen et al. PRL (1995)





Synchrotron radiation is produced when particles are accelerated in a storage ring.

N_f : the number of radiated photons
N_e : the number of electrons in the beam
N : the number of undulator periods



Wiggler and undulator - linear magnetic structures - are used to improve the properties of the radiation produced.

Direct descendants of the bending magnets in a storage ring, wiggler and undulator, force the particles to travel along a zig-zag path so that the emitted light waves are superimposed.

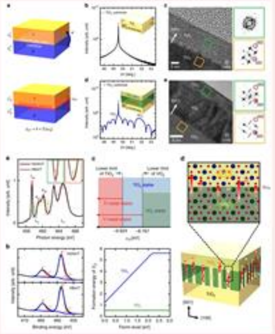
These magnetic structures are now used to produce synchrotron radiation in laboratories worldwide. Extremely intense X-ray radiation with laser-like properties is generated by free-electron lasers.

Research Highlights of BL2A

Journal paper

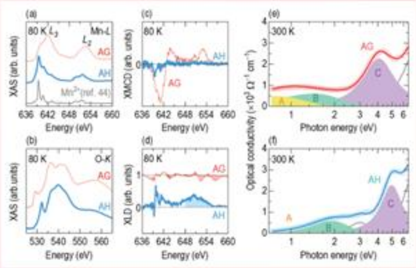
PLS-II, 2A

Directional ionic transport across the oxide interface enables low-temperature epitaxy of rutile TiO2



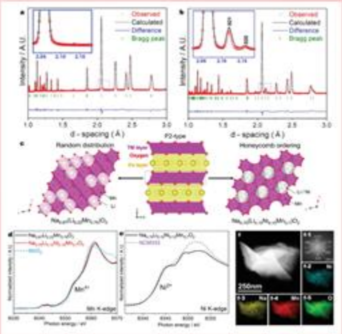
Nature communications 2021
(IF = 16.6)

Hydrogen control of double exchange interaction in La0.67Sr0.33MnO3 for ionic-electric-magnetic coupled applications



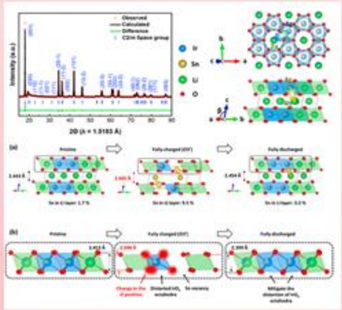
Advanced Materials 2021
(IF = 29.4)

Hysteresis-Suppressed Reversible Oxygen-Redox Cathodes for Sodium-Ion Batteries



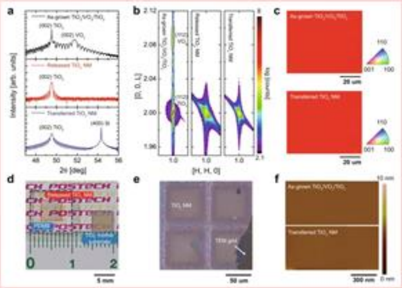
Advanced Energy Materials 2022
(IF = 27.8)

Unraveling Reversible Redox Chemistry and Structural Stability in Sn-Doped Li-Rich Layered Oxide Cathodes



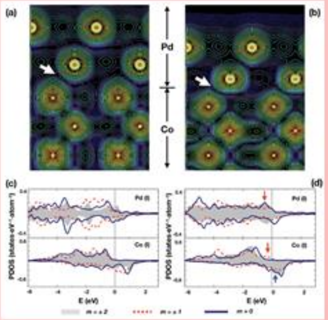
ACS Energy Letters 2022
(IF = 22.0)

Heterogeneous integration of single-crystalline rutile nanomembranes with steep phase transition on silicon substrates



Nature communications 2021
(IF = 16.6)

Giant Orbital Anisotropy with Strong Spin–Orbit Coupling Established at the Pseudomorphic Interface of the Co/Pd Superlattice

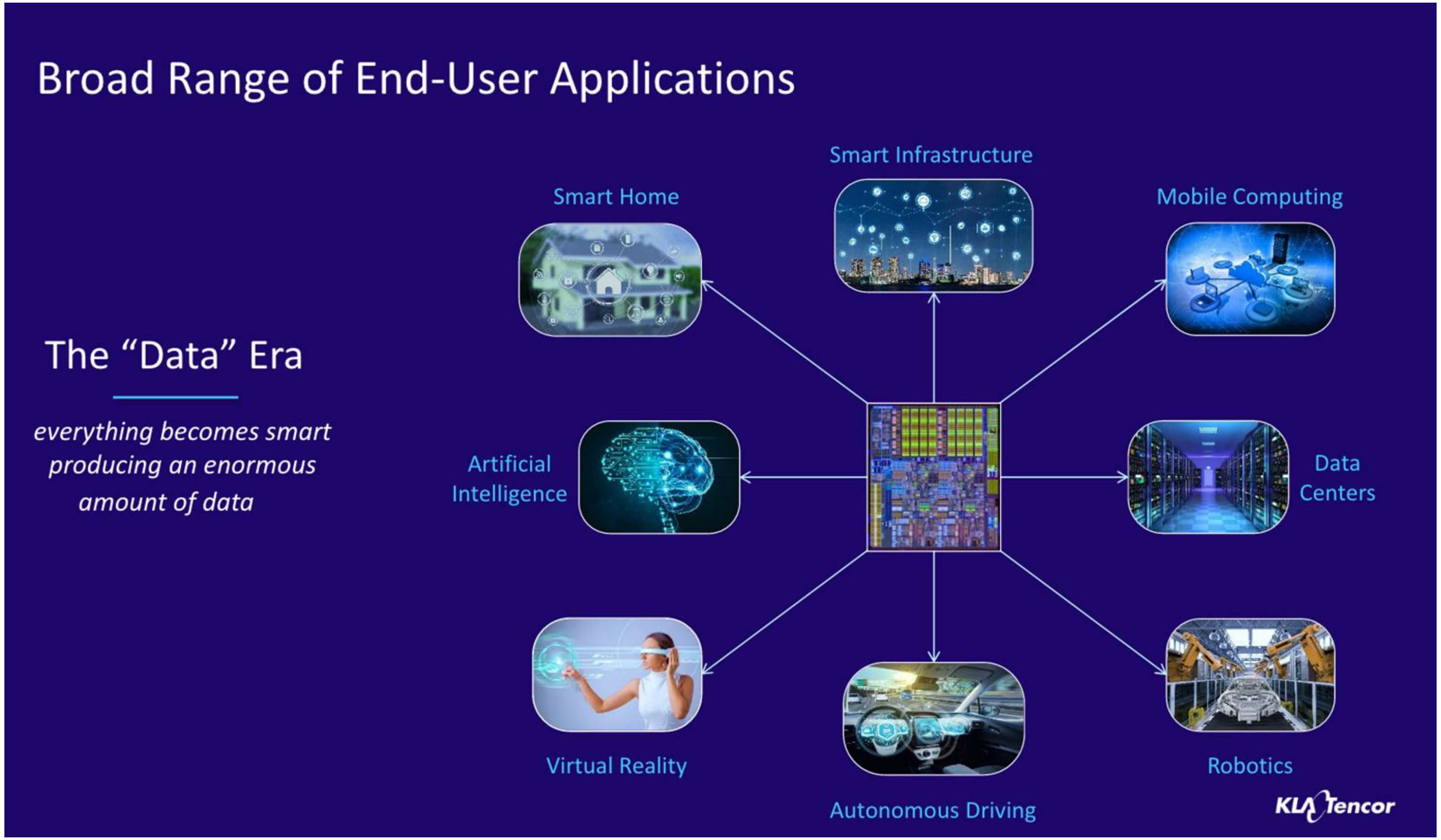


Advanced Science 2022
(IF = 15.1)

Why AI Semiconductor?

미래기반 가속기
전문인력양성 사업단

More Applications ➔ More Data and Process
➔ More Efficiency and Lower Energy
➔ AI Semiconductor (AIoT: Artificial Intelligence of Things)



Space Business

SpaceX Falcon 9

Military Unmanned Combat Aerial Vehicle (UCAV)

XQ-58 Valkyrie of the United States Air Force

Soft X-ray Spectroscopy

- Soft X-ray energy: 20 – 3000 eV
- Soft X-ray interacts strongly with materials and has weak transmission.
  → It requires Ultra-high Vacuum (UHV; below ~10^{-9} Torr).
- **X-ray absorption spectroscopy** (XAS) is a invaluable mean of determining the local geometric and electronic structure of matters.
- **X-ray Magnetic Circular Dichroism** (XMCD) is an important mean of studying the magnetic states of materials.

Soft X-ray Absorption Spectroscopy (XAS)

미래기반 가속기 전문인력양성 사업단

❖ XAS (X-ray Absorption Spectroscopy)

: To investigate electronic and local geometric structures of various materials

→ Doping and ion implantation materials for electronics

→ Materials for semiconductor devices & energy

→ Analysis of electrical and magnetic properties

→ Analysis of the composition and chemical state

❖ Probing depth

→ Total electron yield: ~15 nm

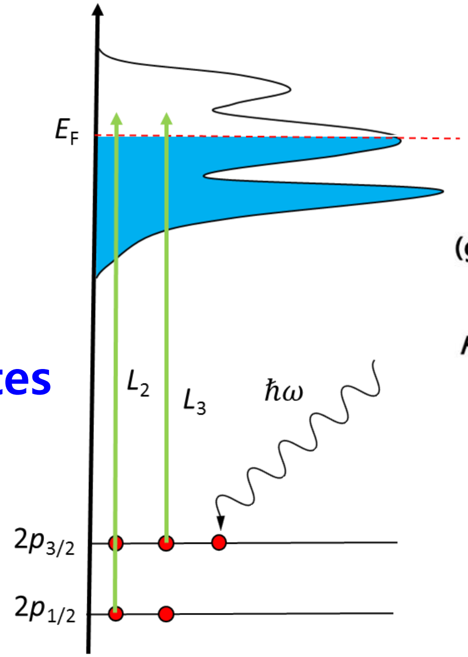
→ Fluorescence yield: ~ 100 nm

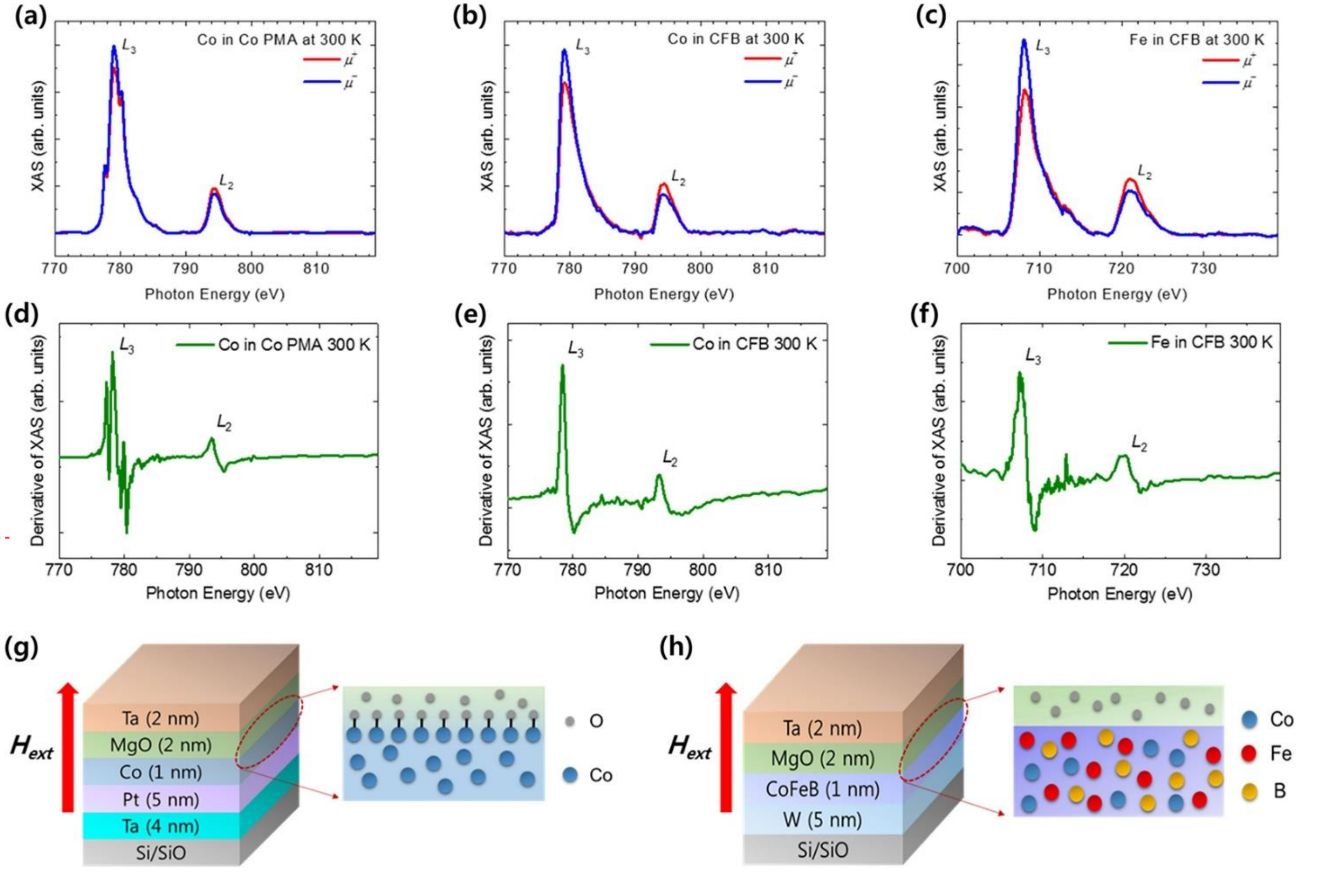
Finger Print of the Ground States

Ground State of Valence

Spin State

Ground State of Symmetry





Younghak Kim et al., Spin and orbital properties of perpendicular magnetic anisotropy for spin-orbit torque material devices, Applied Surface Science, Vol. 544 (2021)

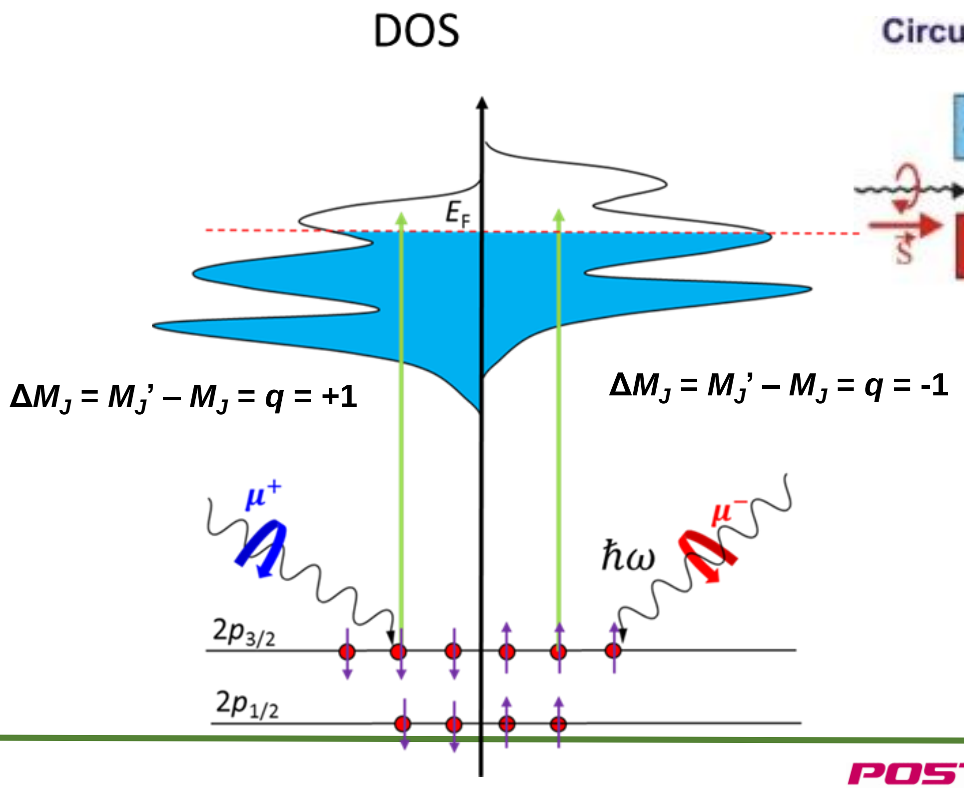
X-ray Magnetic Circular Dichroism (XMCD)

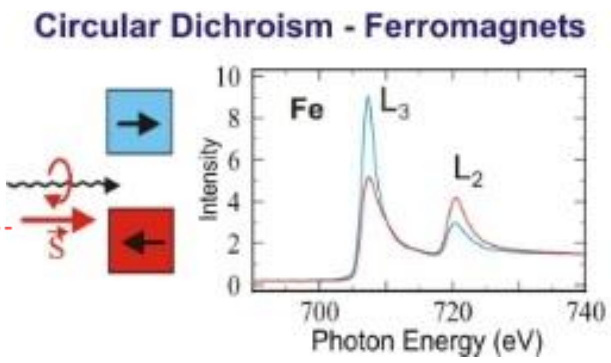
❖ XMCD (X-ray Circular Magnetic Dichroism)

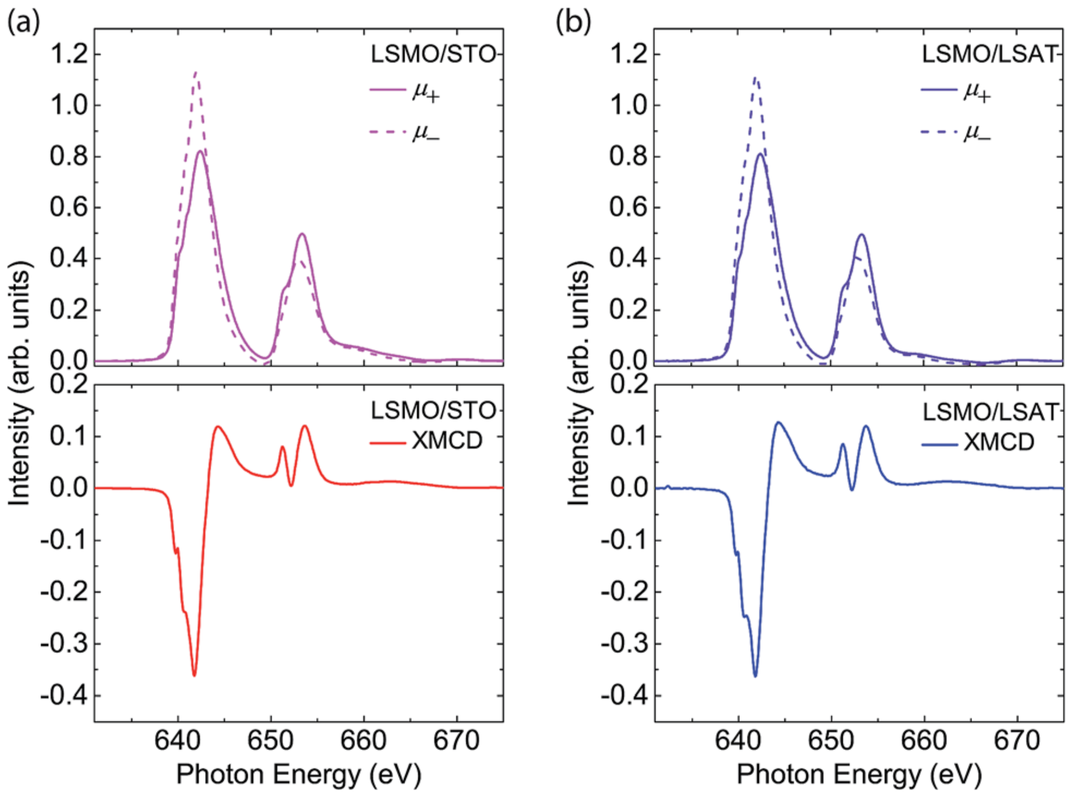
: The difference between circularly polarized x-rays with opposite helicities, which is induced by ferromagnetic ordering

→ Element and chemical specific measurement

→ Separation of spin and orbital magnetic moment (sum rule)



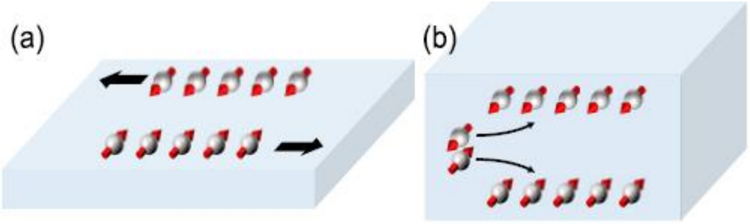




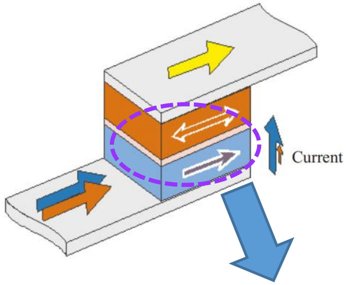
Younghak Kim et al., Strain-effected physical properties of ferromagnetic insulating $La_{0.88}Sr_{0.12}MnO_3$ thin films, RSC Adv. 9, 2645-2649 (2019)

Spin-Orbit Torque Materials for AI Memory Devices

❖ Spin-Orbit Torque:

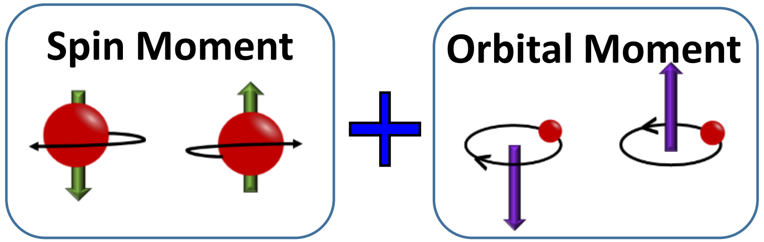


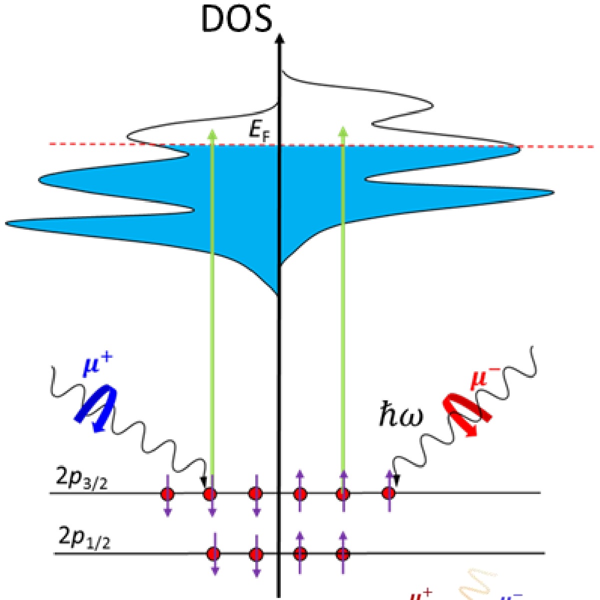
Rashba–Edelstein Effect **Spin Hall Effect**

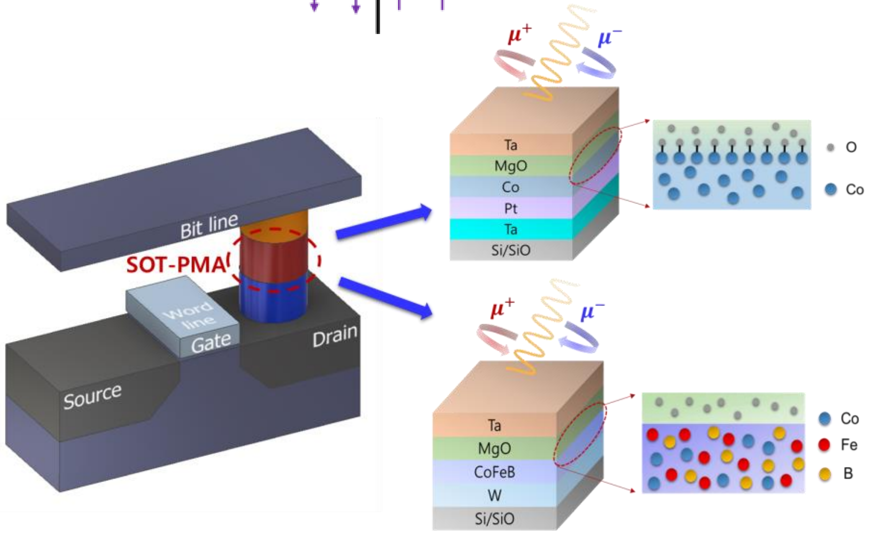


Magnetic Tunnel Junction (MTJ)
IBM. J. Res. Develop. 42, 73 (1998)

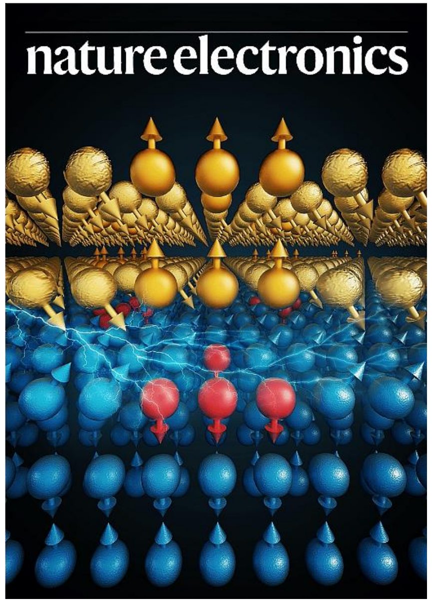
Spin-Orbit Coupling







https://doi.org/10.1038/s41928-020-00504-6



PAL 포항가속기연구소 POHANG ACCELERATOR LABORATORY

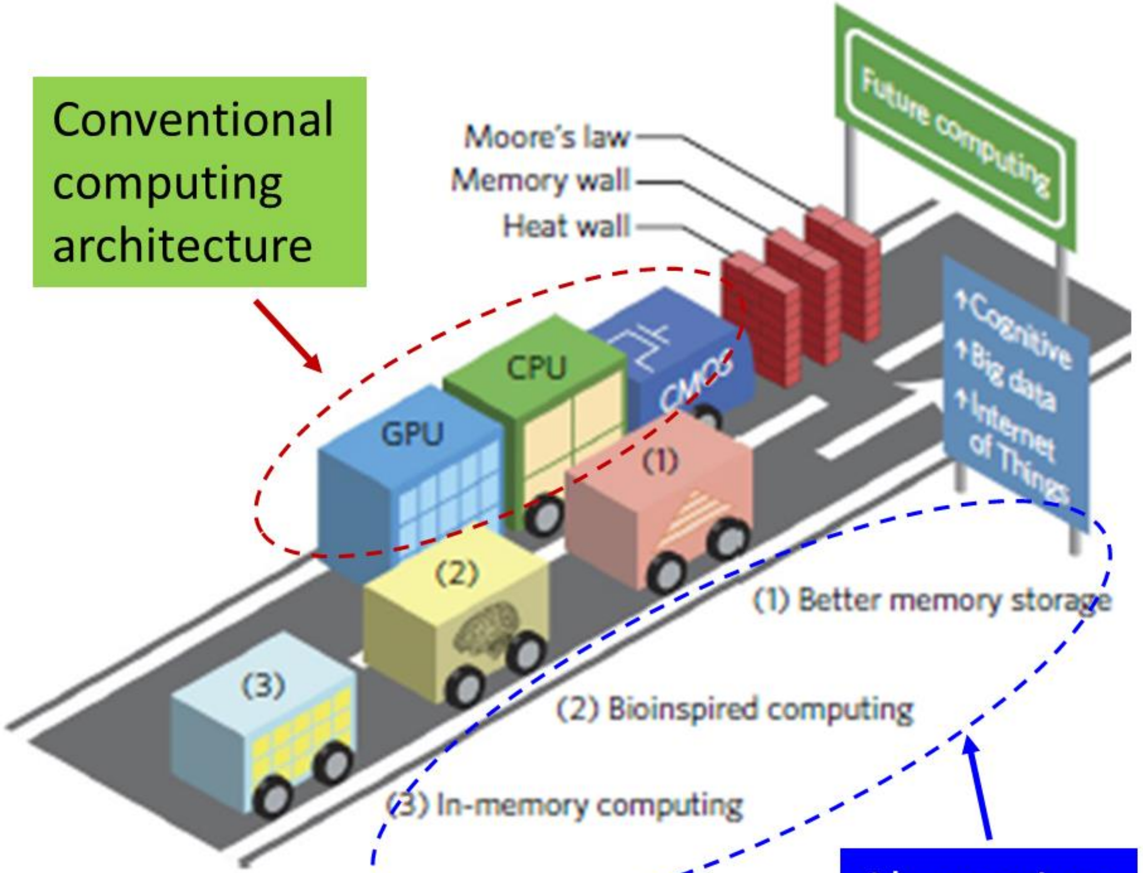
KIST 한국과학기술연구원 Korea Institute of Science and Technology

Younghak Kim et al., Spin and orbital properties of perpendicular magnetic anisotropy for spin-orbit torque material devices, Applied Surface Science, Vol. 544 (2021)

Evolution of the Computing System

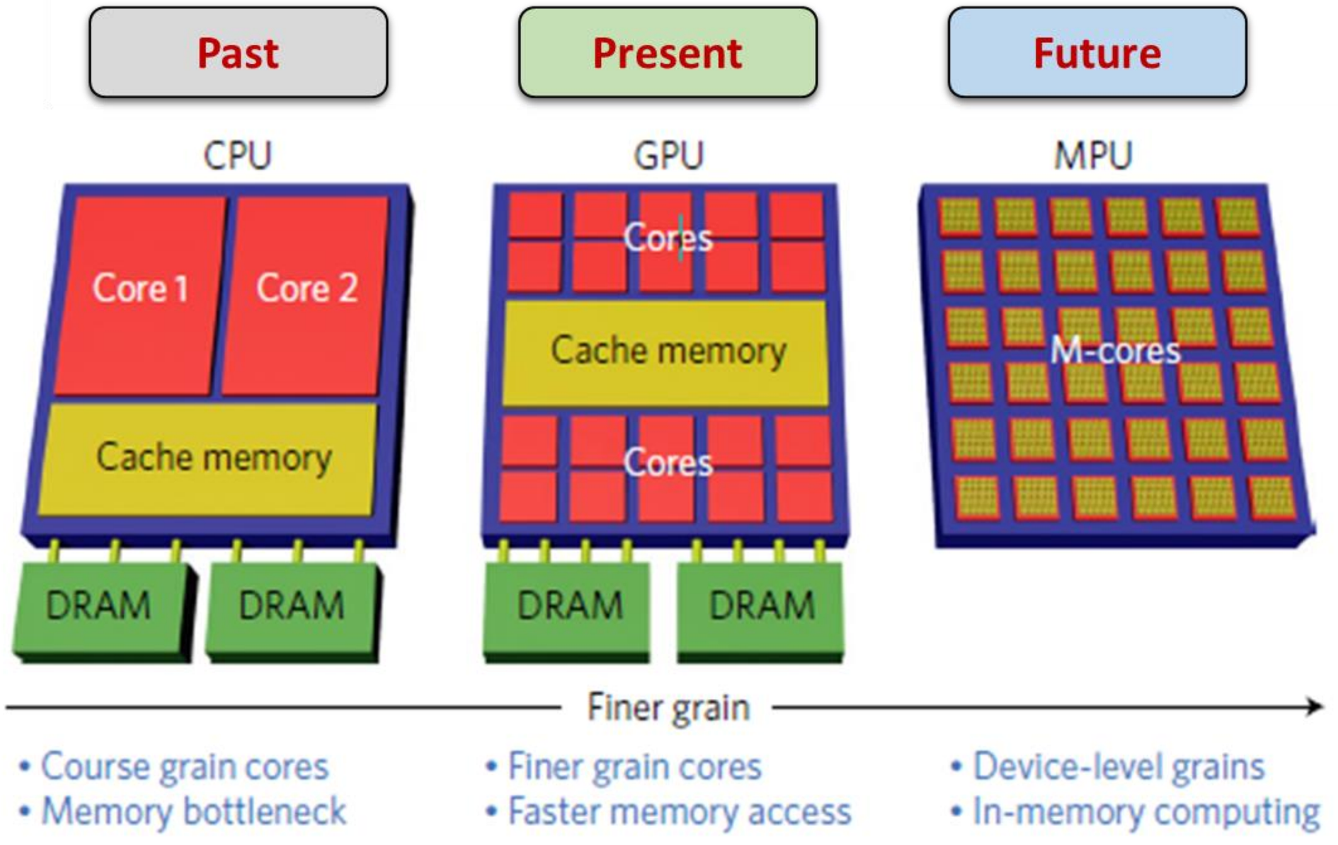
Challenges and Alternatives

Conventional computing architecture



Alternatives

Evolution of Computing Architecture



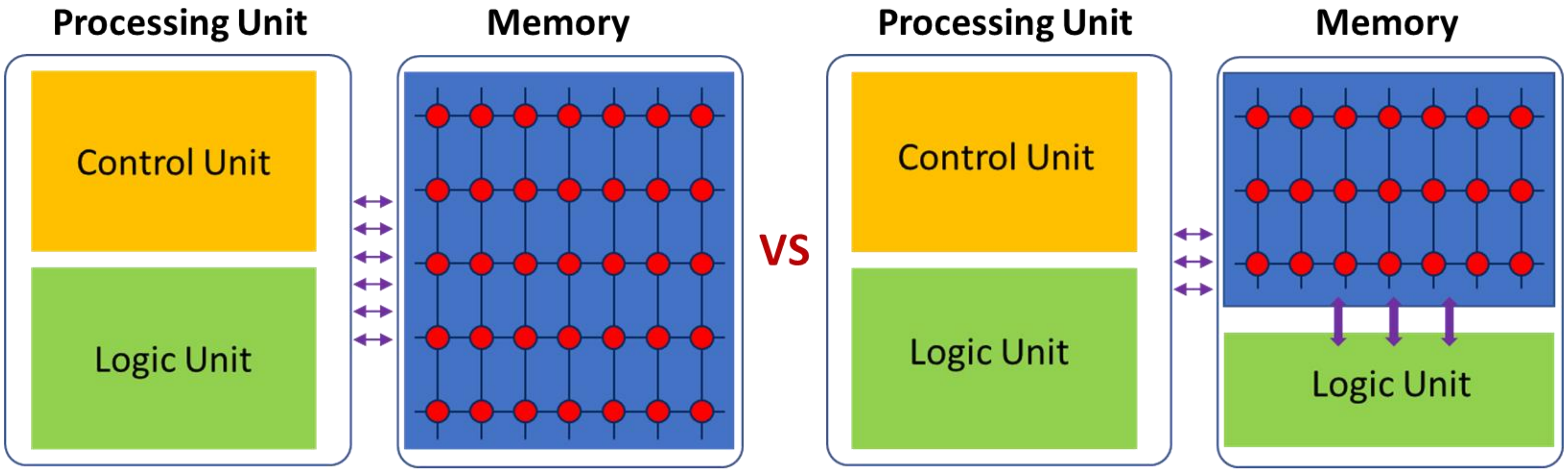
The future of electronics based on memristive systems, Nature Electronics. by M. A. Zidan et al. 2018

Processing-in Memory (In-Memory Computing)

-. Not only stores data in memory but also performs data operation

-. Efficient power utilization due to the calculations in memory

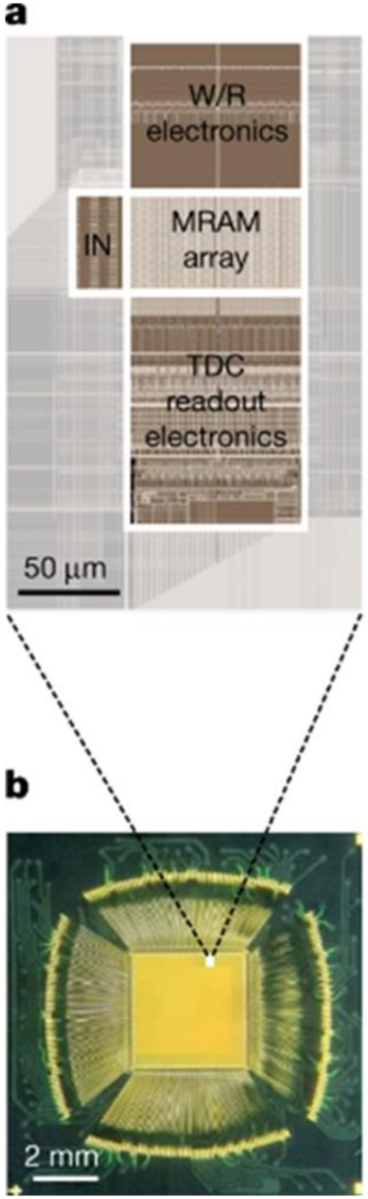
-. Enhancing overall performance of AI accelerator systems



von-Neumann architecture (Conventional computing)

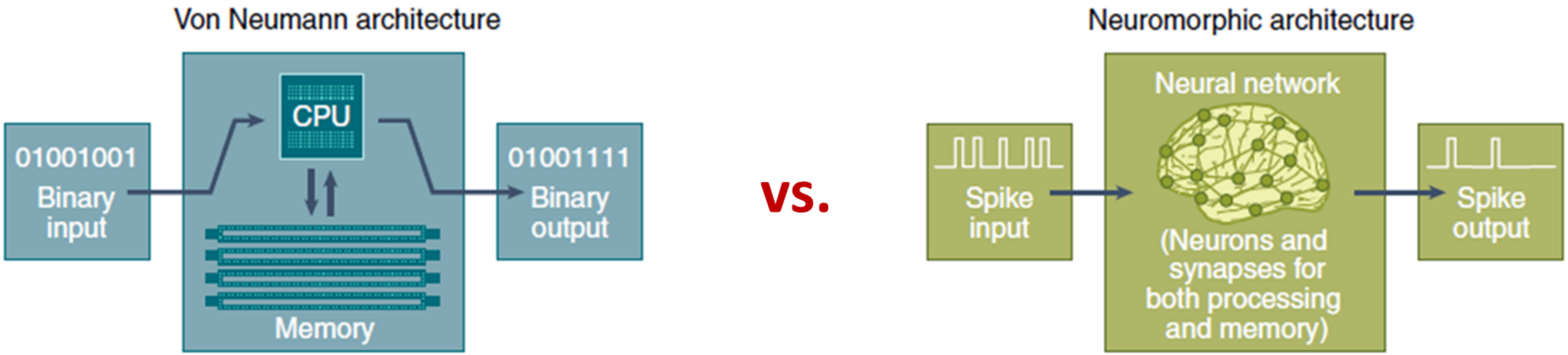
non-von Neumann (Processing-in Memory)

Samsung's Processing-in Memory based on STT-MRAM →



S. Jung et al., "A crossbar array of magnetoresistive memory devices for in-memory computing," Nature, vol. 601, pp. 211-216, (2022)

Neuromorphic Computing

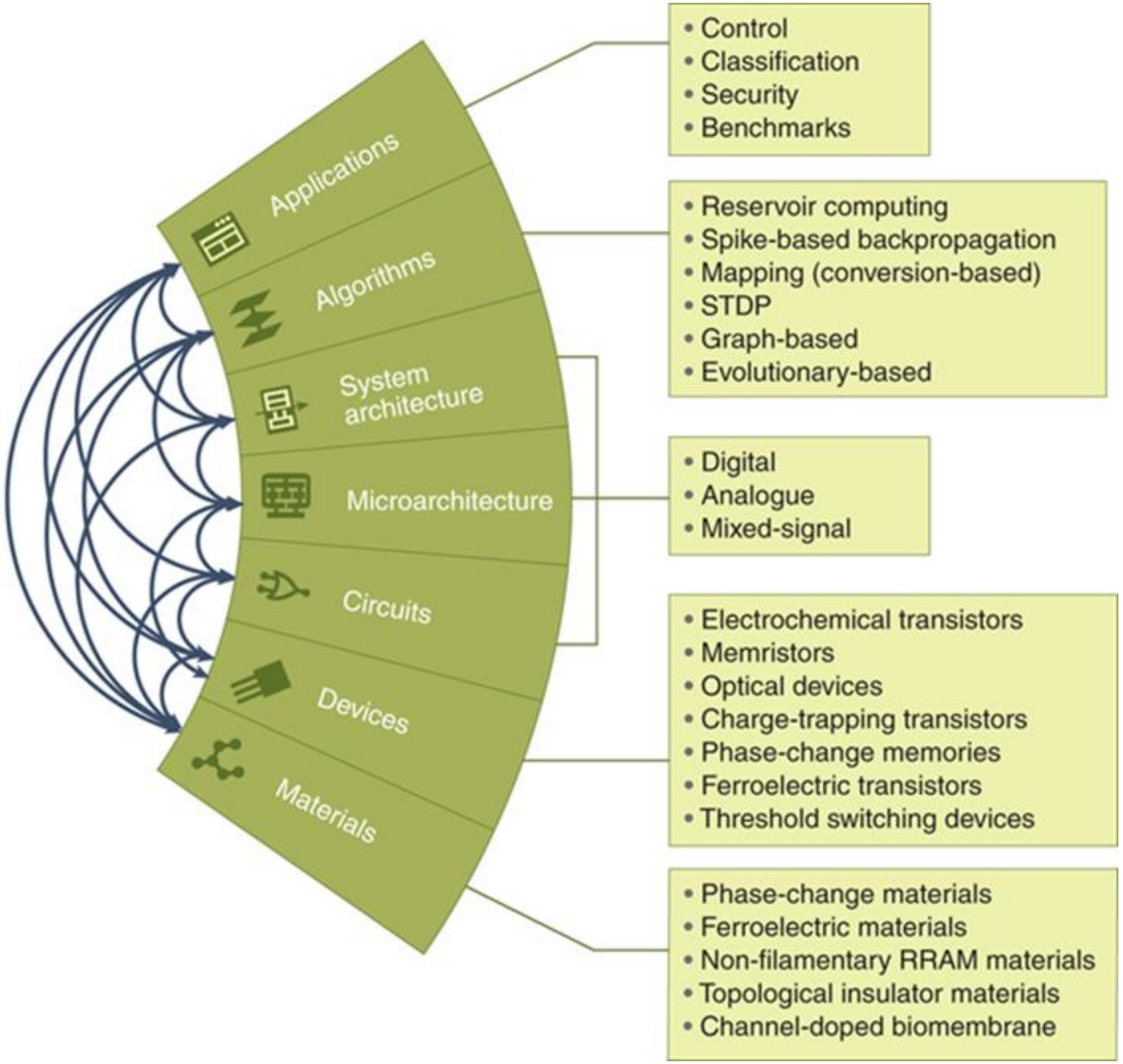


	von Neumann architecture	Neuromorphic Architecture
Operation	Sequential Processing	Massively Parallel Processing
Organization	Separated Computation and Memory	Collocated Processing and Memory
Programming	Code as Binary Instructions	Spiking Neural Network
Communication	Binary Data	Spikes
Timing	Synchronous (clock-driven)	Asynchronous (event-driven)

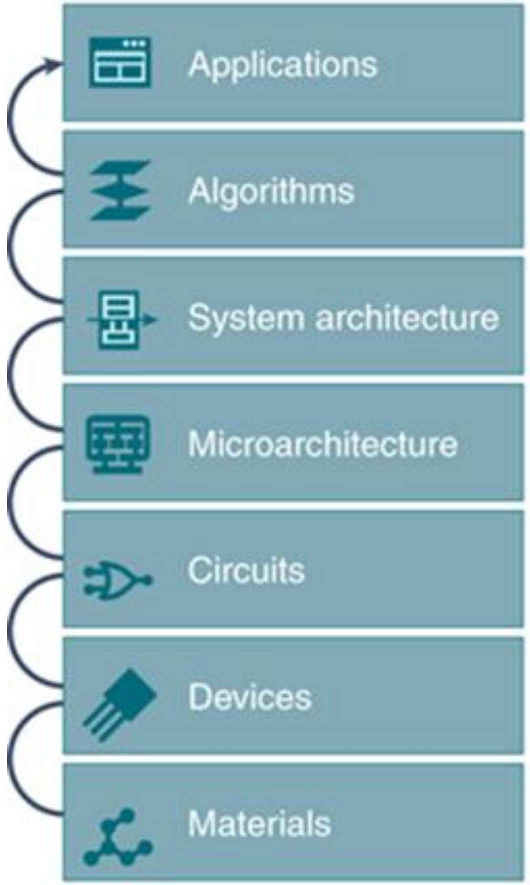
Schuman, C.D., Kulkarni, S.R., Parsa, M. *et al.* Opportunities for neuromorphic computing algorithms and applications. *Nat Comput Sci* **2**, 10–19 (2022)

New approach vs. Conventional Way

New Way: omnidirectional approach

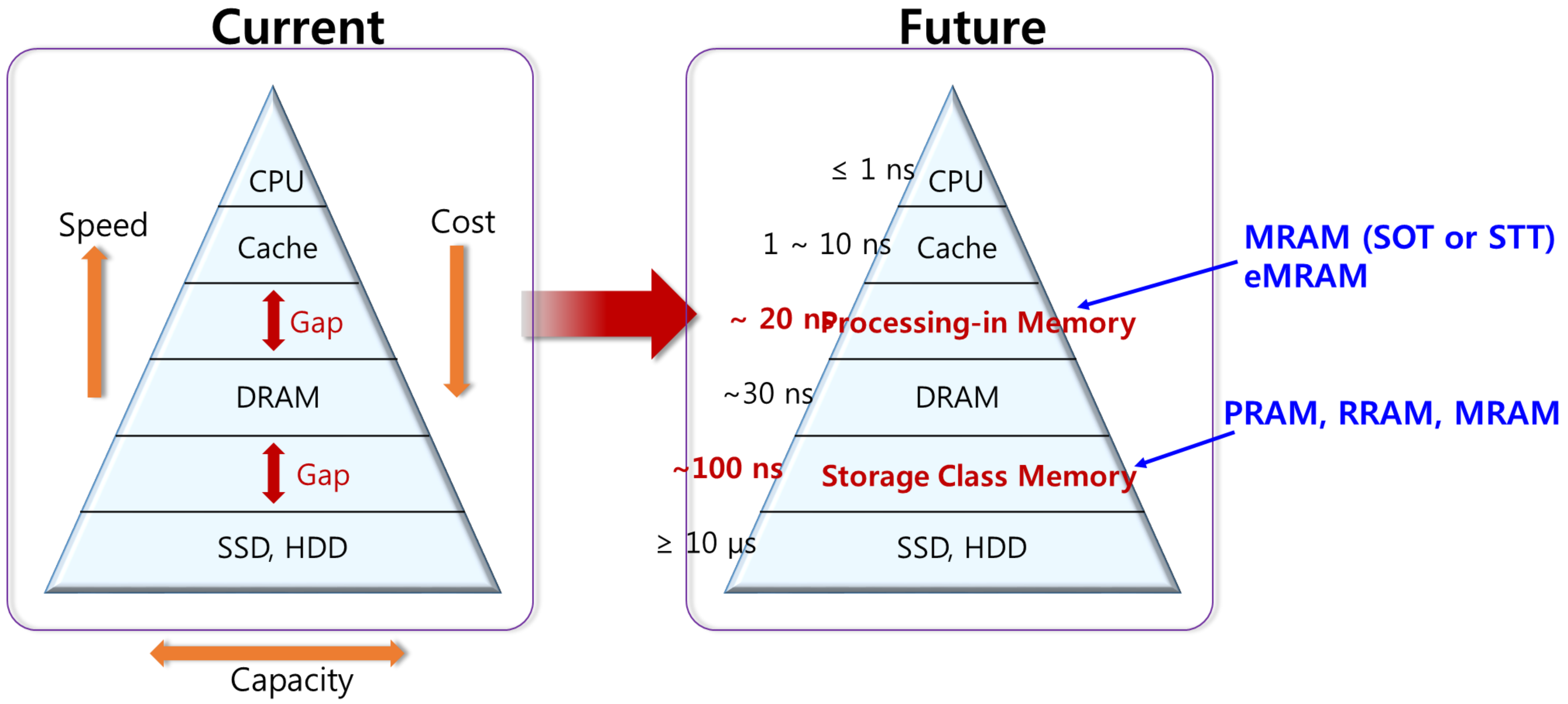


Conventional Way: bottom-up approach

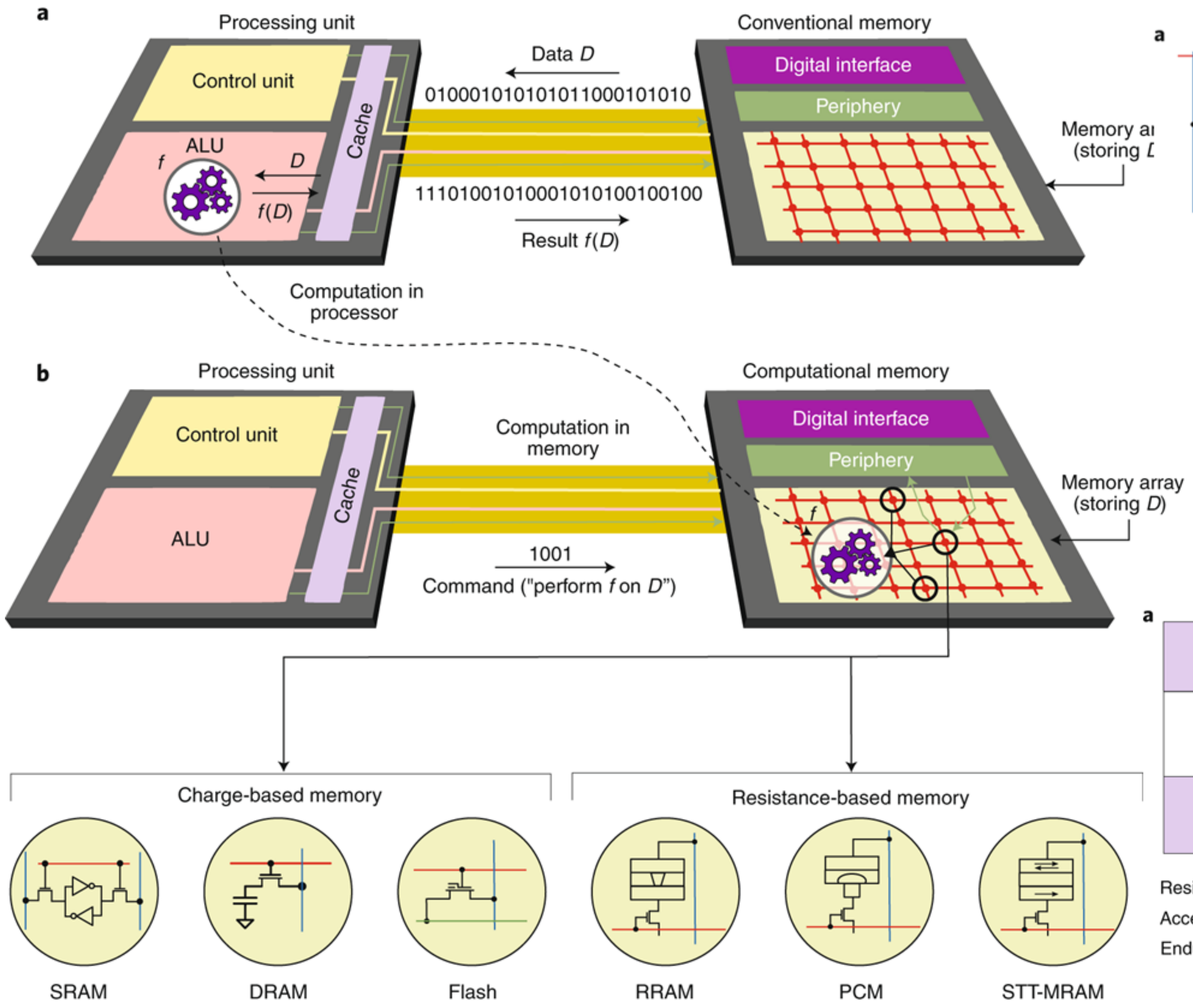


Schuman, C.D., Kulkarni, S.R., Parsa, M. *et al.* Opportunities for neuromorphic computing algorithms and applications. *Nat Comput Sci* **2**, 10–19 (2022)

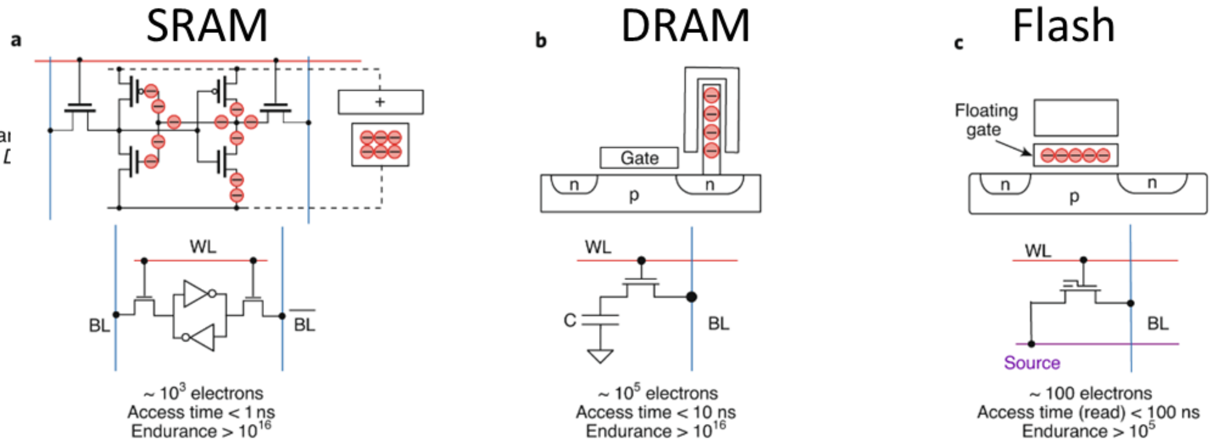
MRAM, PRAM, and ReRAM in Memory Hierarchy



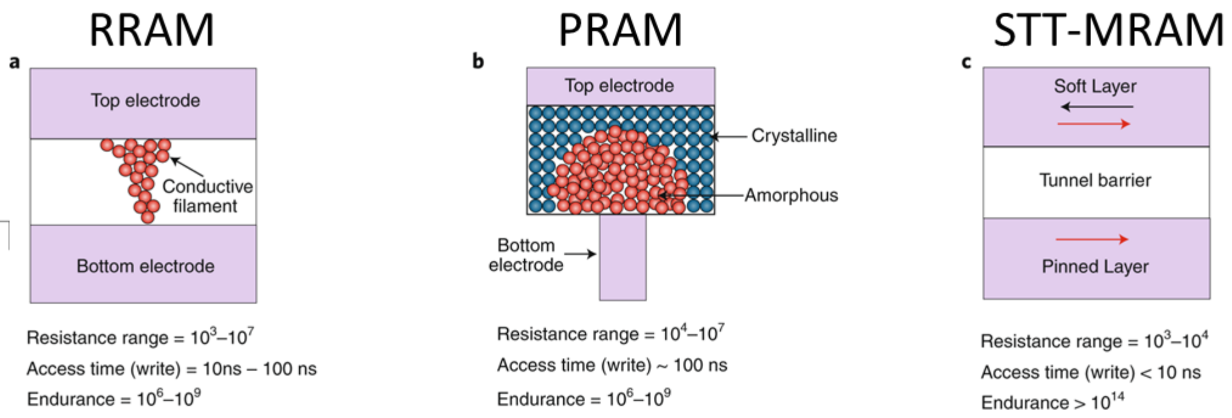
Candidates of Processing-in Memory



Charge-based memory devices

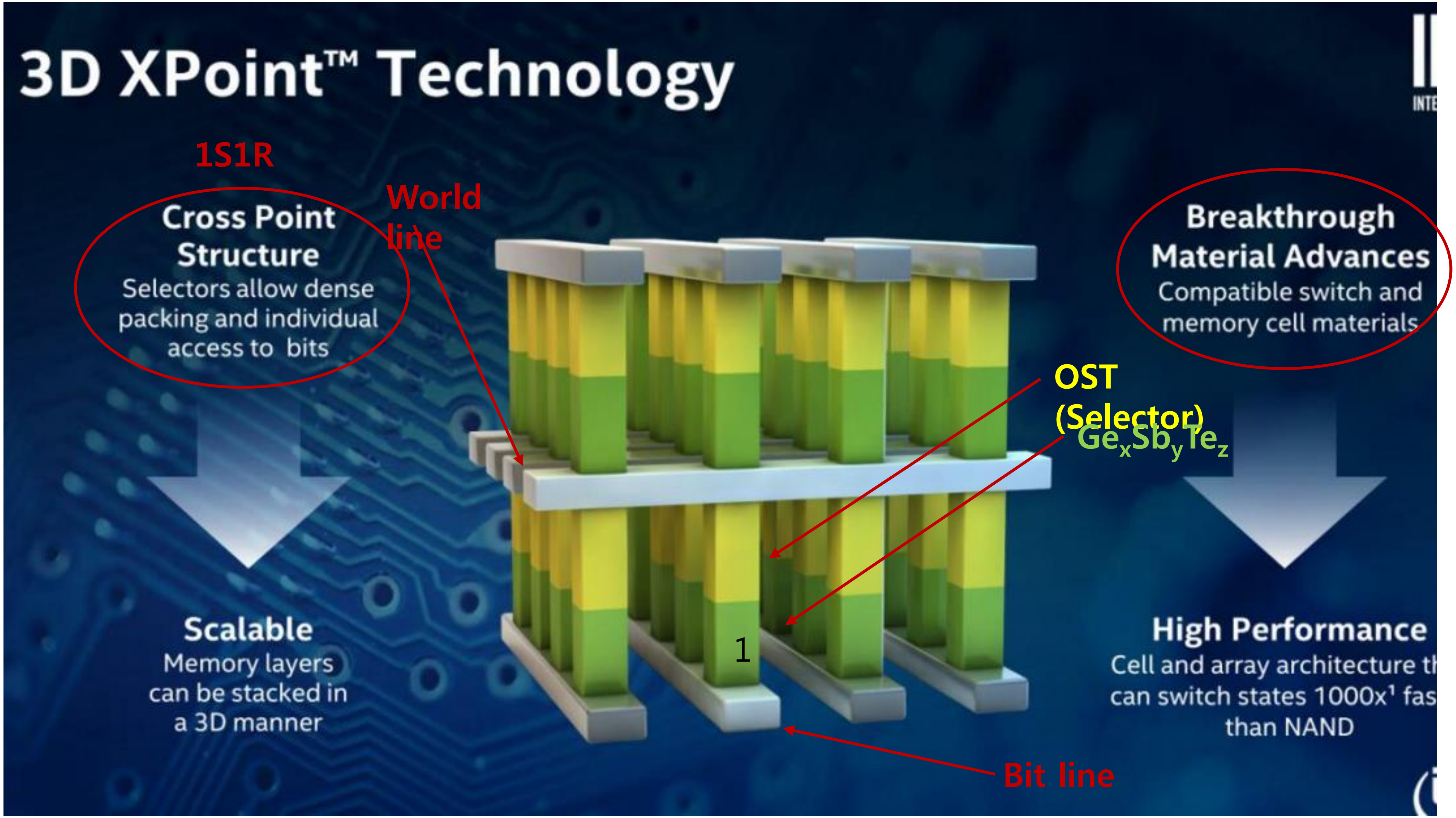


Resistance-based memory devices (Memristor)



Nat. Nanotechnol. **15,** 529–544 (2020)

R&D in Industry: PRAM

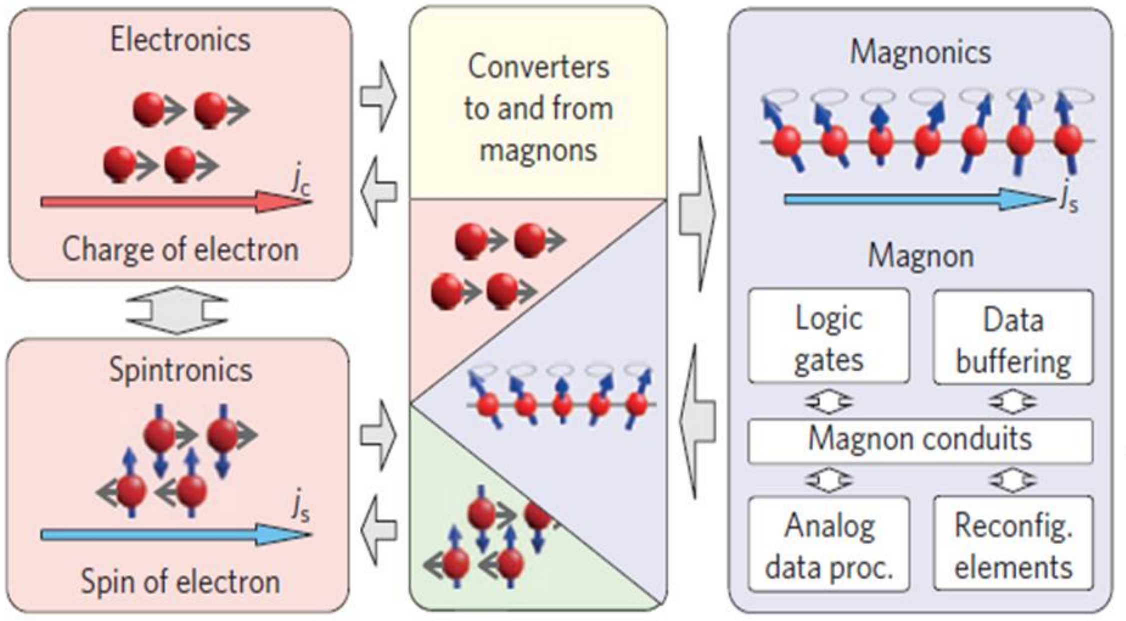


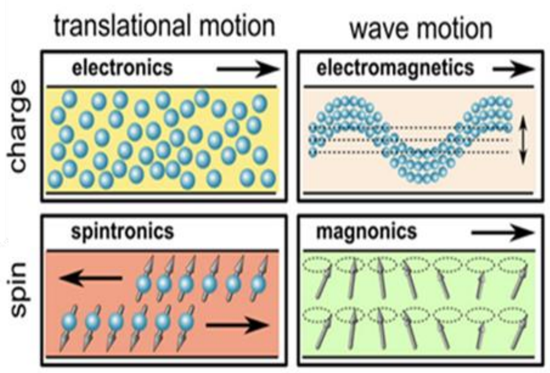
출처: Intel

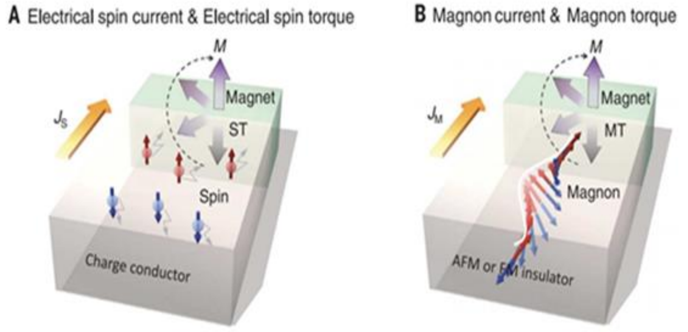
Quantum Technology: Magnonics

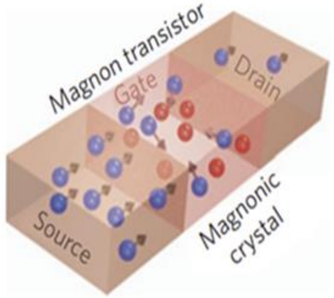
Magnonics: Use **spin waves (magnon)** for **logic devices** and **data storage**.
Spin waves are a propagating re-ordering of the magnetization in a material and arise from the precession of magnetic moments.

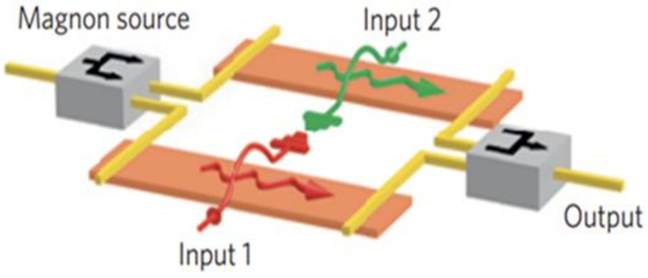
❖ **The concept of magnon spintronics**
Information is stored into charge or spin currents
→ Changed into magnon currents









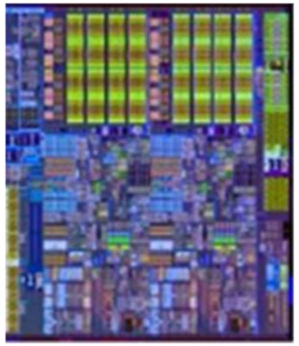


-. A. Chumak et al., Magnon spintronics, Nature Physics 11, 453 (2015)
-. C. S. Davies et al.,"Prototype magnonic device development", p. 54 in "Magnetics Technology International"(UKIP Media, Surrey, 2015)
-. Y. Wang et al., Magnetization switching by magnon-mediated spin torque through an antiferromagnetic insulator, Science 366, 1125 (2019)

Research of 2A BL at Pohang Accelerator Laboratory

미래기반 가속기
전문인력양성 사업단

Next Semiconductor
Quantum Technology

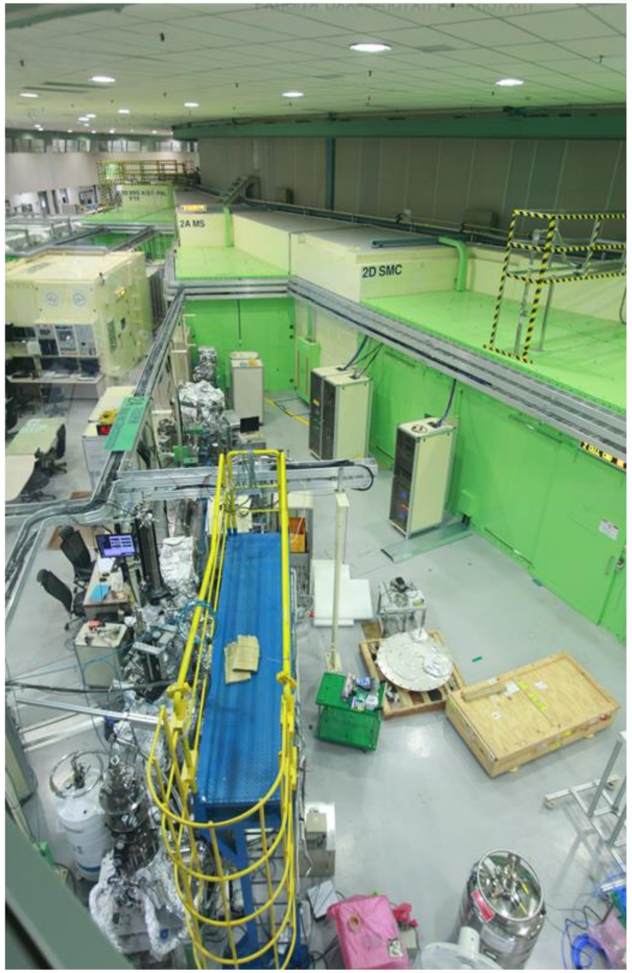


Display

Energy

Area	Materials
Next Semiconductor	Spin-orbit Torque materials, Magnetic Materials(Ferro, Antiferro, Ferri), high -k oxide (HfO2, Al2O3, SiO2), 2D Materials, Perovskite: Fe3GeTe2, Pd/Ni/Cu, Fe/Cu, Fe/Ni/W, CoFeB/Ti/NiFe, Pt/CoFeB/MgO, TiO2, V2O
Display	InGaZnO, high -k oxide (HfO2, Al2O3, SiO2)
Energy	2^{nd} Battery Cathode Material, Catalyst, Hydrogen Storage(VO2), NiCoMnAl, NiCoMn

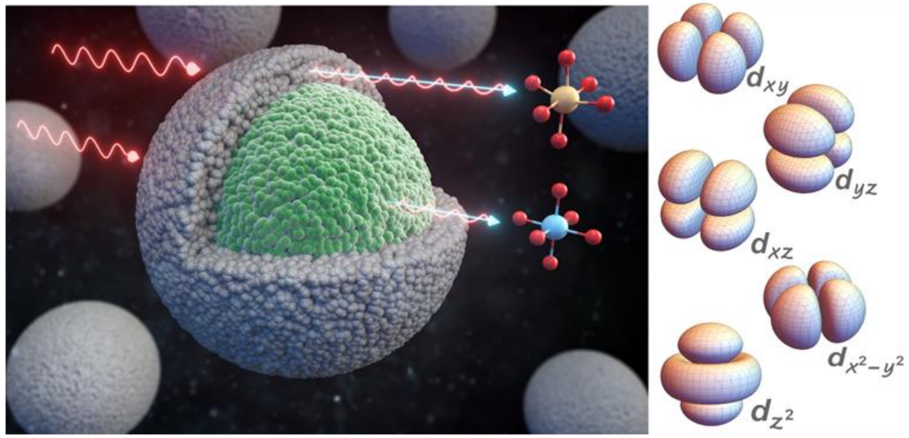
PAL 포항가속기연구소
POHANG ACCELERATOR LABORATORY

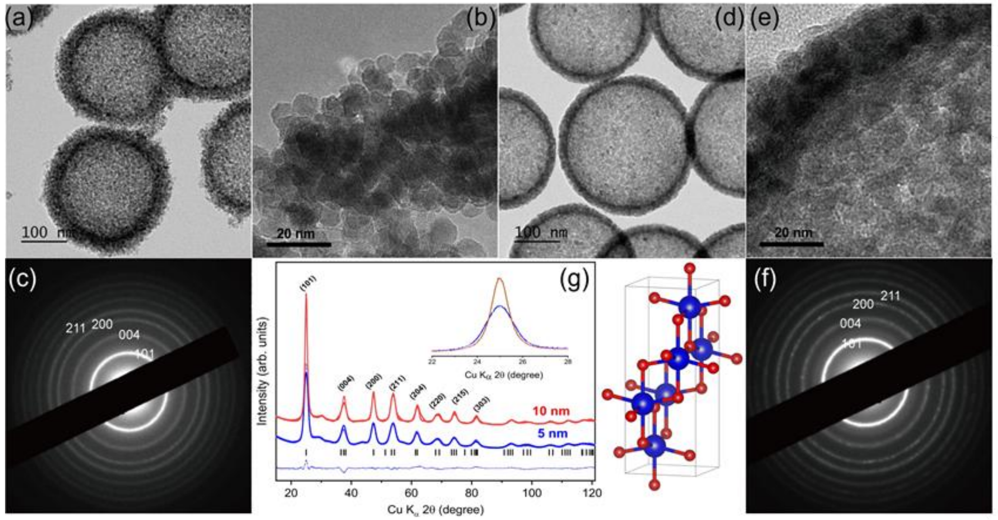


Switching Material of ReRAM: TiO2

미래기반 가속기
전문인력양성 사업단

❖ TiO_2 Nano Particles



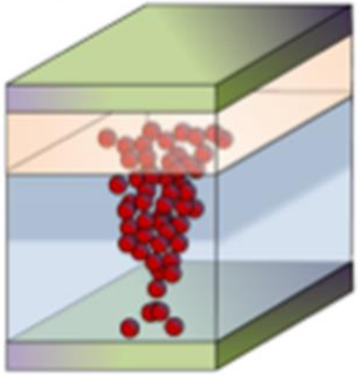


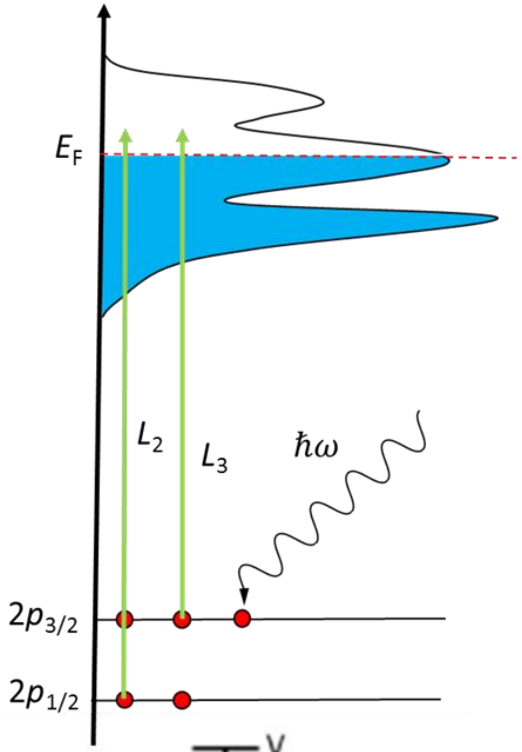
Published in Nano Letters, 2021

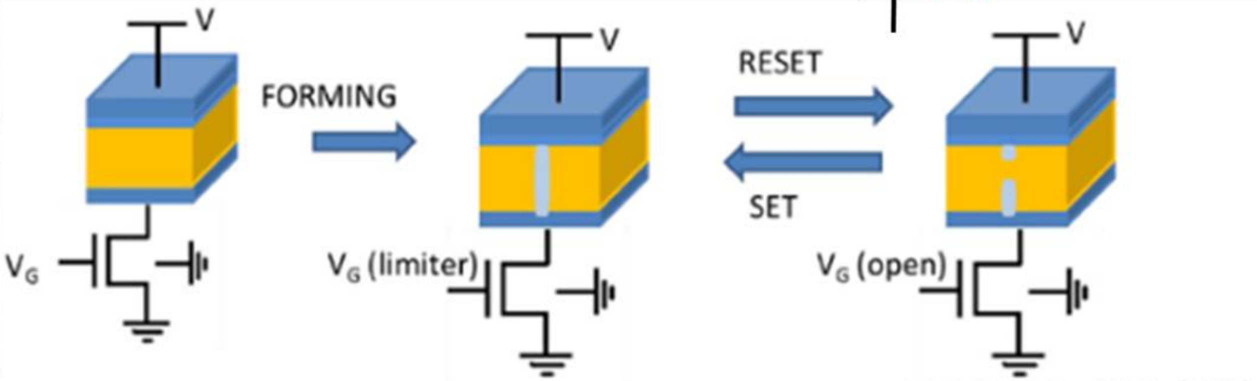
Switching Materials
→ TiO2, HfO, TaO, ...

ReRAM memory

Oxygen Vacancy Filament







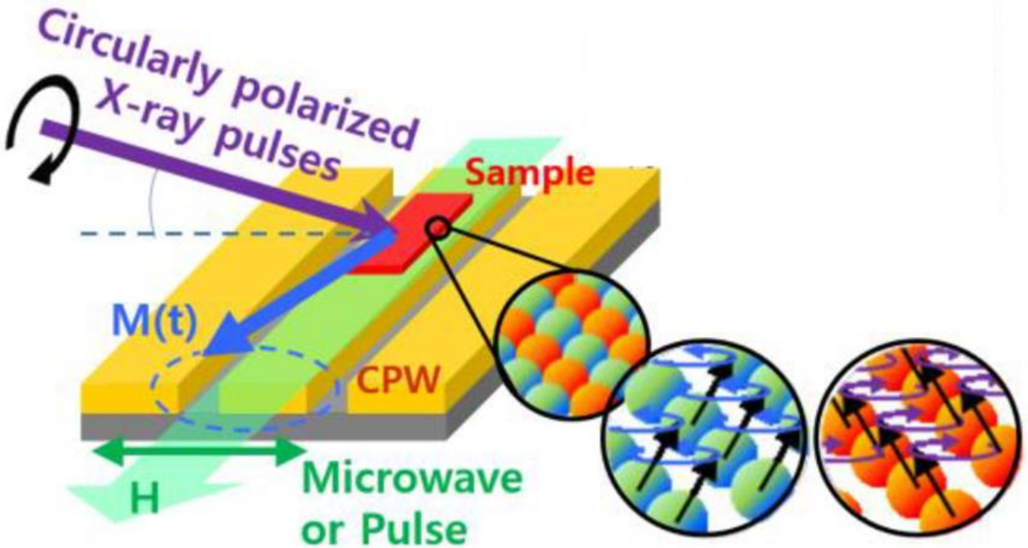
Low Resistive State (LRS)
High Resistive State (HRS)

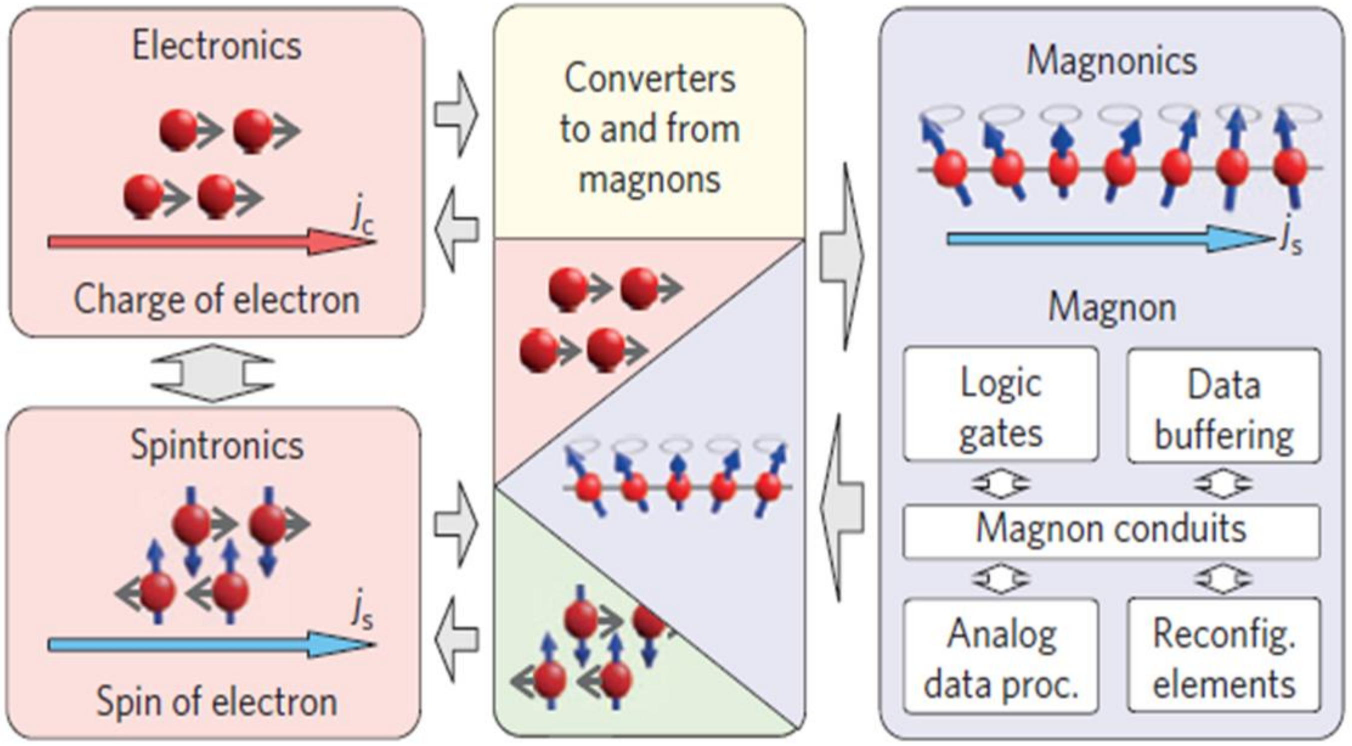
출처: Weebit Nano 제공

❖ XFMR: XMCD + FMR → Spin dynamics !
Spin wave, spin-orbit torque, other phenomena related to “Spin” → Spintronics and magnonics

PAL 포항가속기연구소 POHANG ACCELERATOR LABORATORY — KRISS 한국표준과학연구원 Korea Research Institute of Standards and Science

Schematic figure of XFMR experimental technique





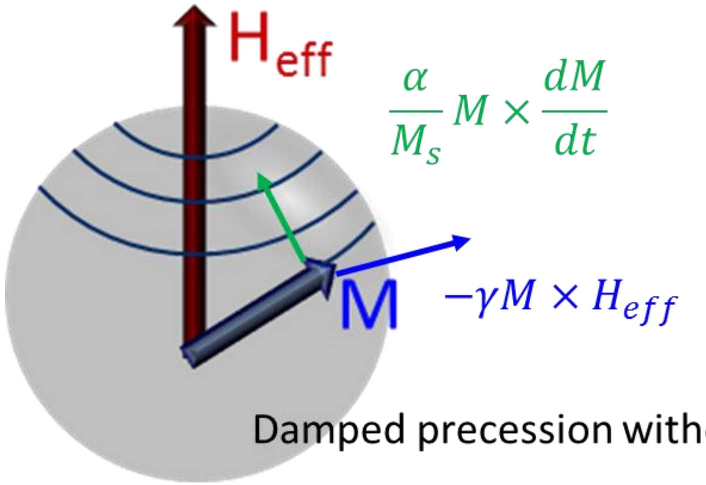
Various fundamental interaction mechanisms of spin interactions to form magnetism in time and length scales. Revised from K. L. Wang *et al*., "Electric-Field Control of Spin-Orbit Interaction for Low-Power Spintronics," in *Proceedings of the IEEE*, vol. 104, no. 10, pp. 1974-2008, Oct. 2016, doi: 10.1109/JPROC.2016.2573836.

Require research !

X-ray Ferromagnetic Resonance (XFMR)

Yi Cao, et al., Prospect of Spin-Orbitronic Devices and Their Applications, iScience, Volume 23, Issue 10 (2020)
Qiming Shao et al., Roadmap of Spin–Orbit Torques, IEEE Transactions on Magnetics (Vol. 57, Issue 7, July 2021)

FMR: **Spin Dynamics!** (Damping Constant)
no element resolution



Damped precession without driving torque

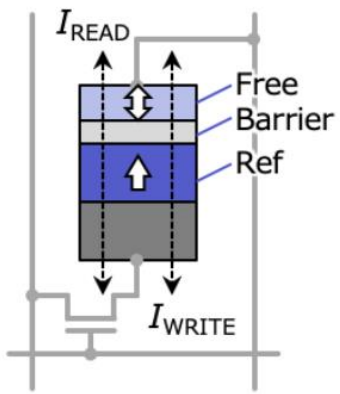
Landau Lifshits Gilbert equation

$$\frac{dM}{dt} = -\gamma M \times \left(H_{eff} - \frac{\alpha}{\gamma M_s}\frac{dM}{dt}\right) = -\gamma M \times H_{eff} + \frac{\alpha}{M_s} M \times \frac{dM}{dt}$$

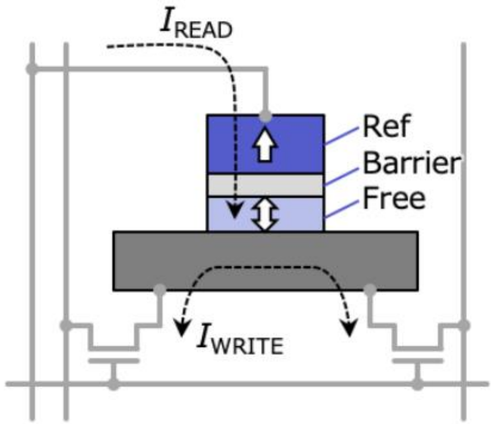
Magnetic field — Magnetic damping

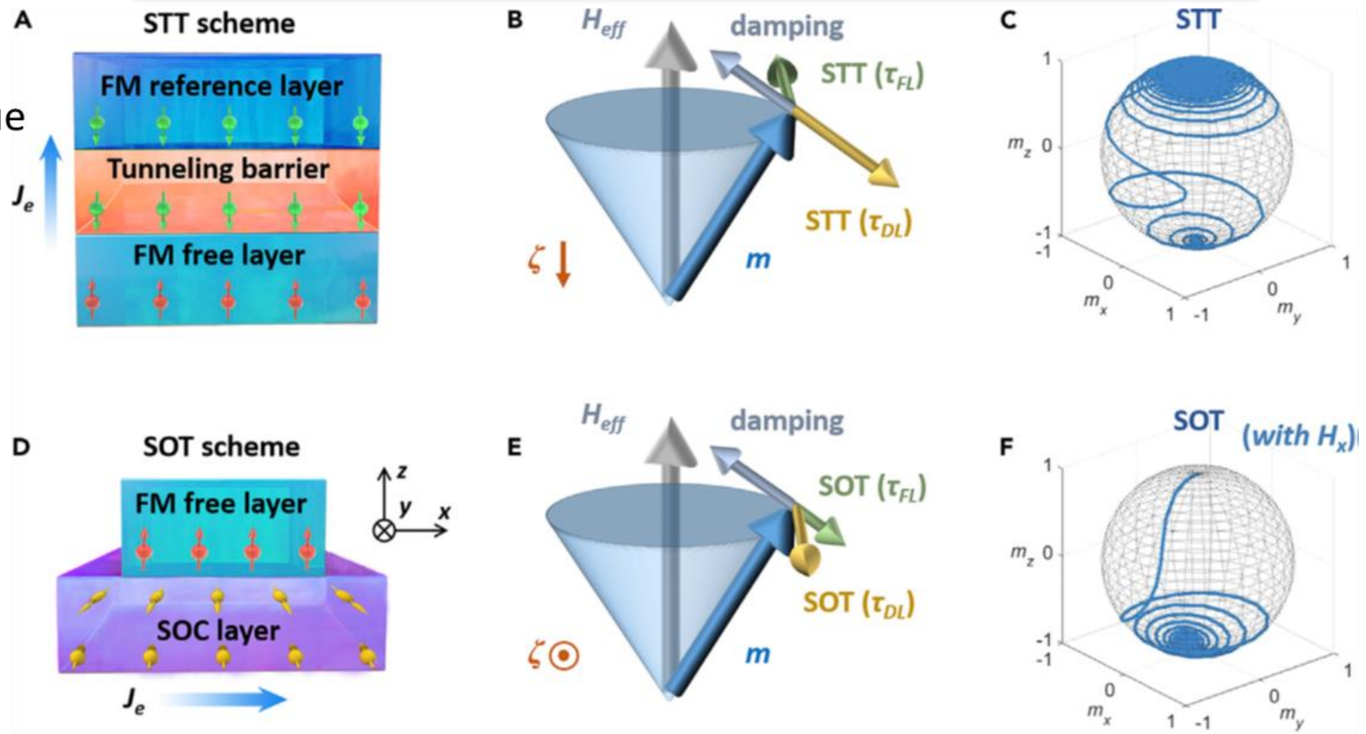
Spin-Transfer Torque (STT) vs. Spin-Orbit Torque (SOT)

STT-MRAM



SOT-MRAM



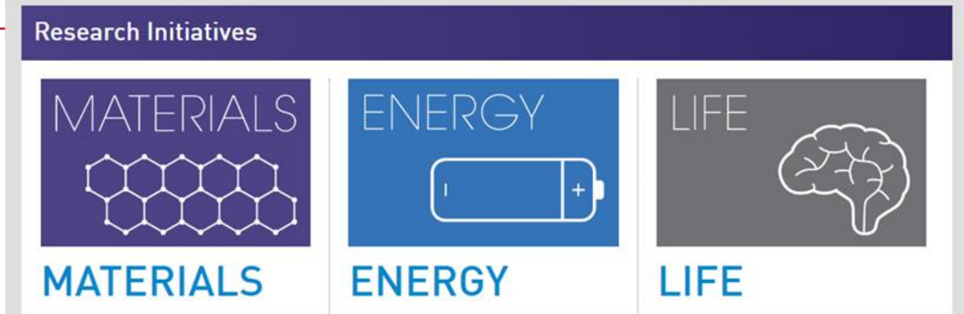


National High Magnetic Field Lab

NATIONAL MAGLAB

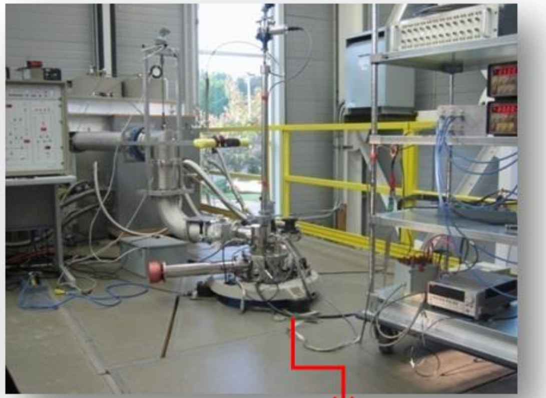
Measuring Capabilities:

-. Magneto-optics (ultra-vi
-. Magnetization
-. Specific heat and magnetocaloric effect
-. Transport
-. NMR
-. Magnetic torque
-. Dependence of optical and transport properties on field orientation

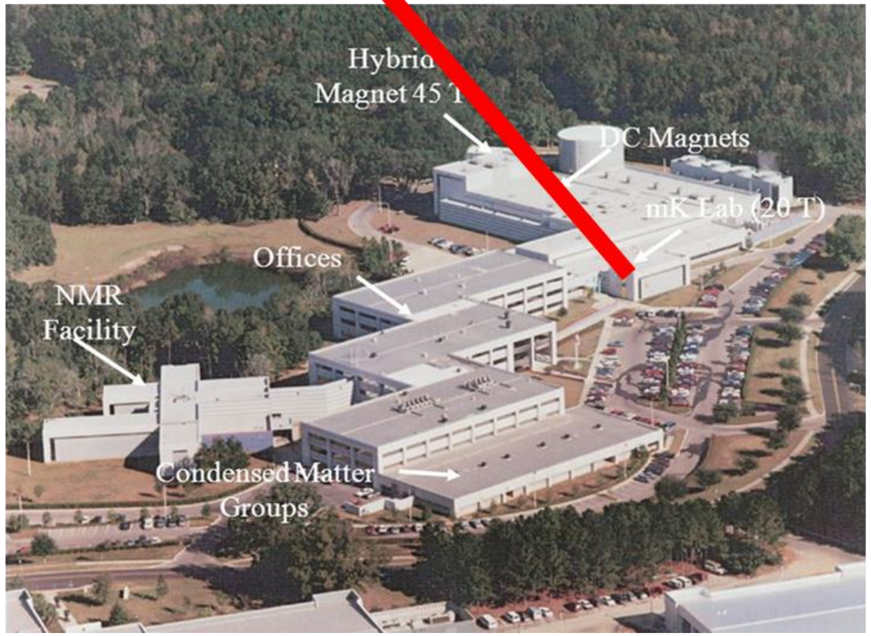


Source: NHMFL

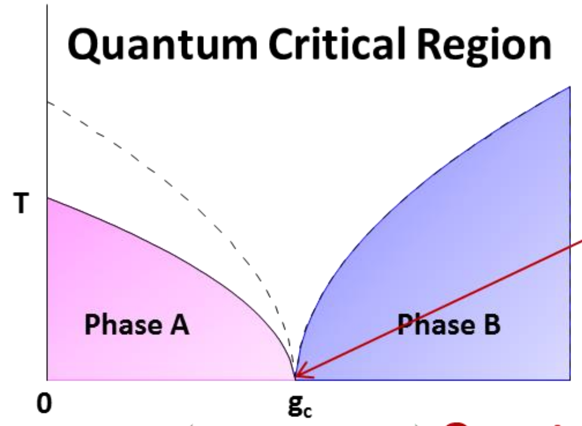
Millikelvin Facility



Superconducting magnet with a dilution fridge



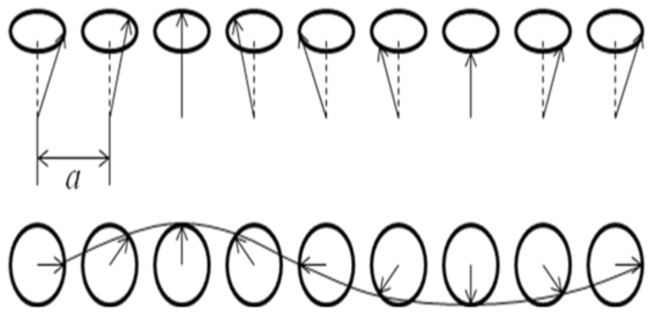
Quantum Critical Region

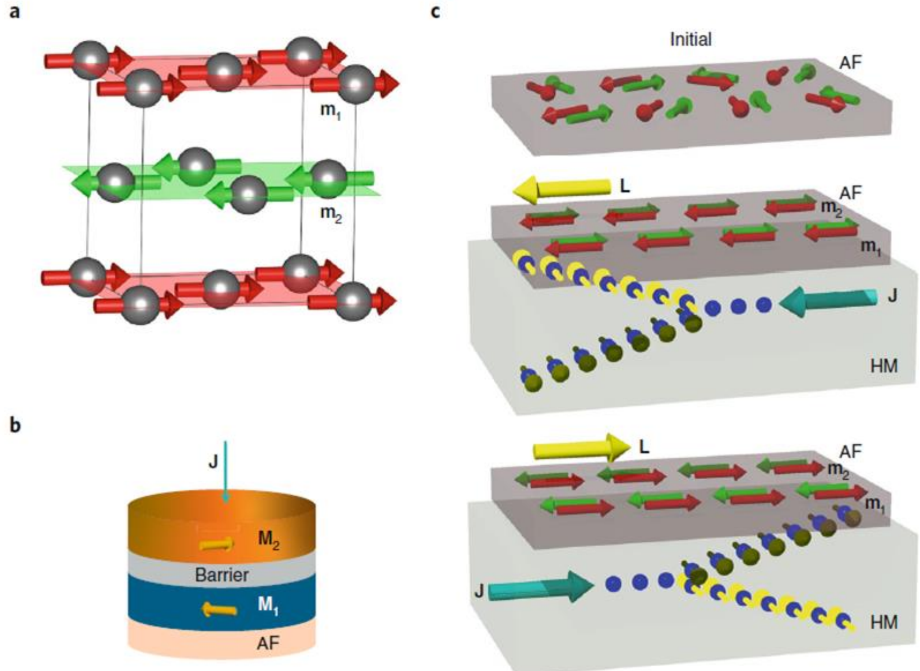


- Quantum Critical Point (g_c)
  :Tuning parameter (g)
  → Pressure
  → Chemical composition
  → Magnetic field

Quantum Phase Transition

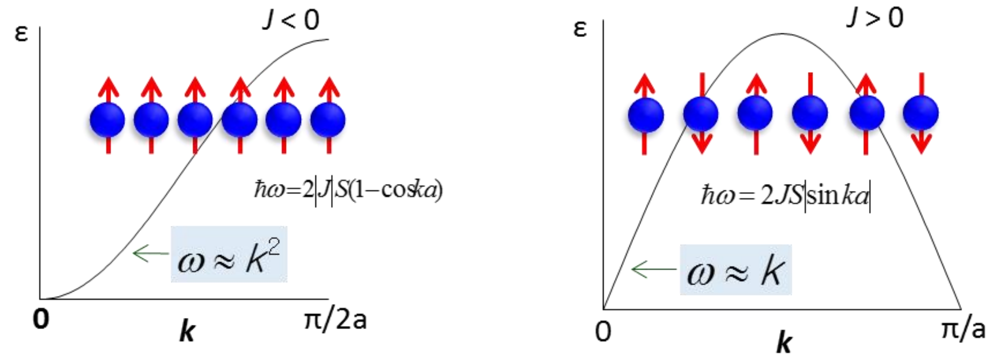
Quantum Criticality and Magnons (Spin Wave)



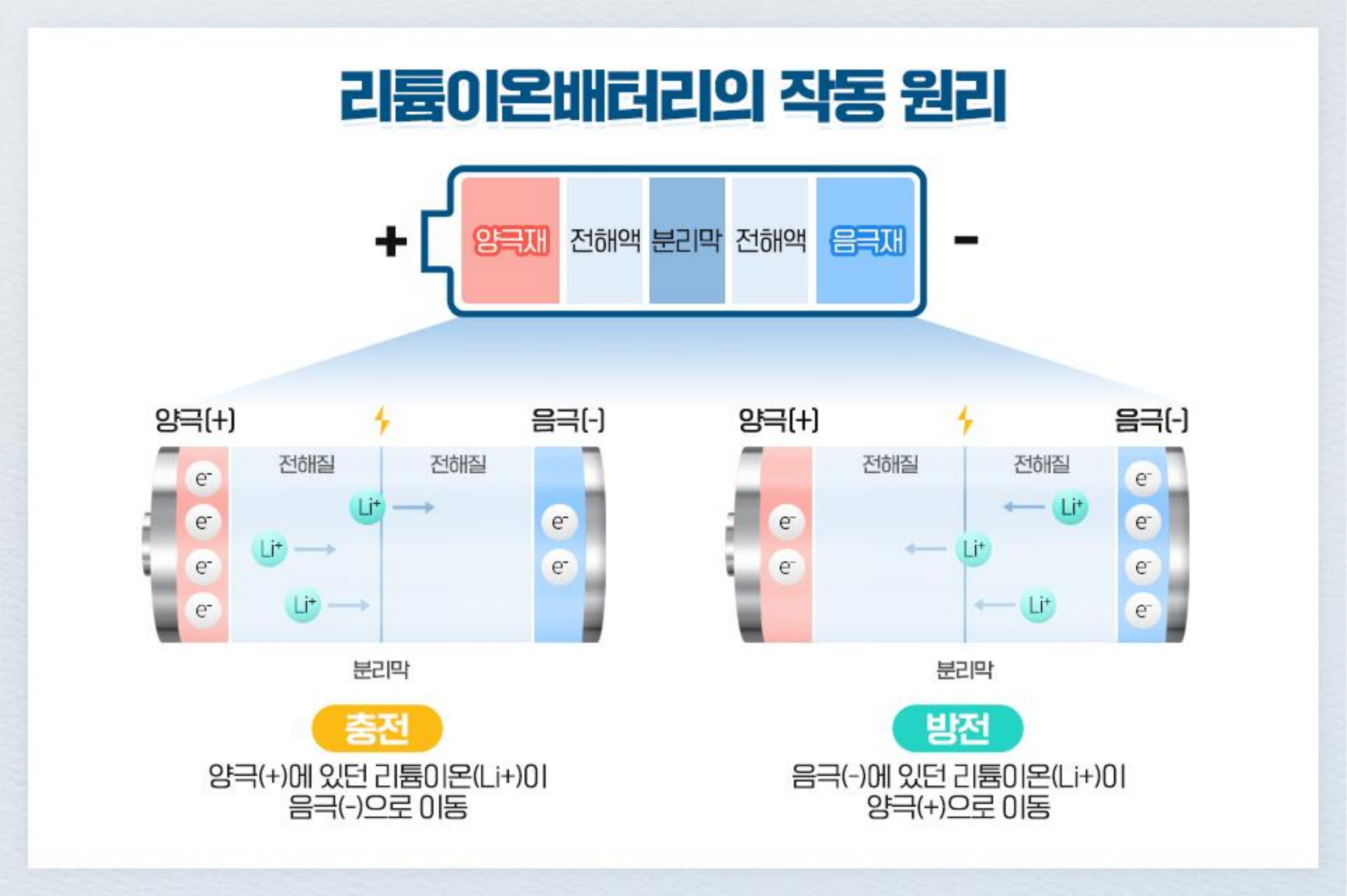


$$H = J\sum_i \vec{S}_i \cdot \vec{S}_{i+1}$$ (1D Heisenberg Hamiltonian)

Dispersions of Ferromagnet and Antiferromagnet



Reiss, G., Meinert, M. Silicon-compatible antiferromagnets switch on. *Nat Electron* **3**, 75–76 (2020)

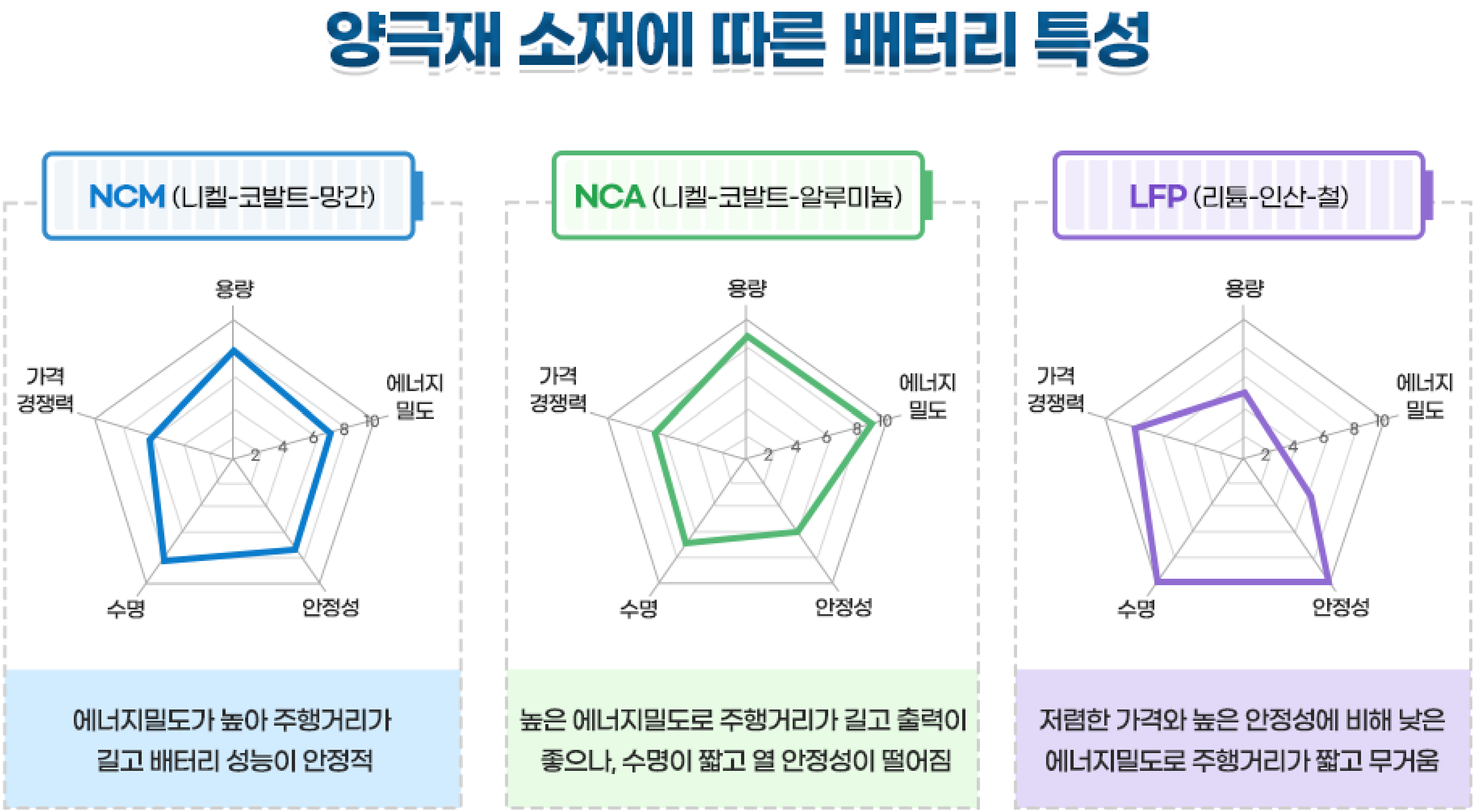

리튬이온배터리의 작동 원리
+
양극재
전해액
분리막
전해액
음극재
-
양극(+)
음극(-)
전해질
전해질
Li+
e-
분리막
충전
양극(+)에 있던 리튬이온(Li+)이
음극(-)으로 이동
양극(+)
음극(-)
전해질
전해질
분리막
방전
음극(-)에 있던 리튬이온(Li+)이
양극(+)으로 이동

출처: 포스코그룹 뉴스룸

양극재에 쓰이는 금속 원소의 특성
28 Ni 니켈 58.693
에너지 밀도
27 Co 코발트 58.933
안정성&수명
25 Mn 망간 54.938
안정성
13 Al 알루미늄 26.982
출력

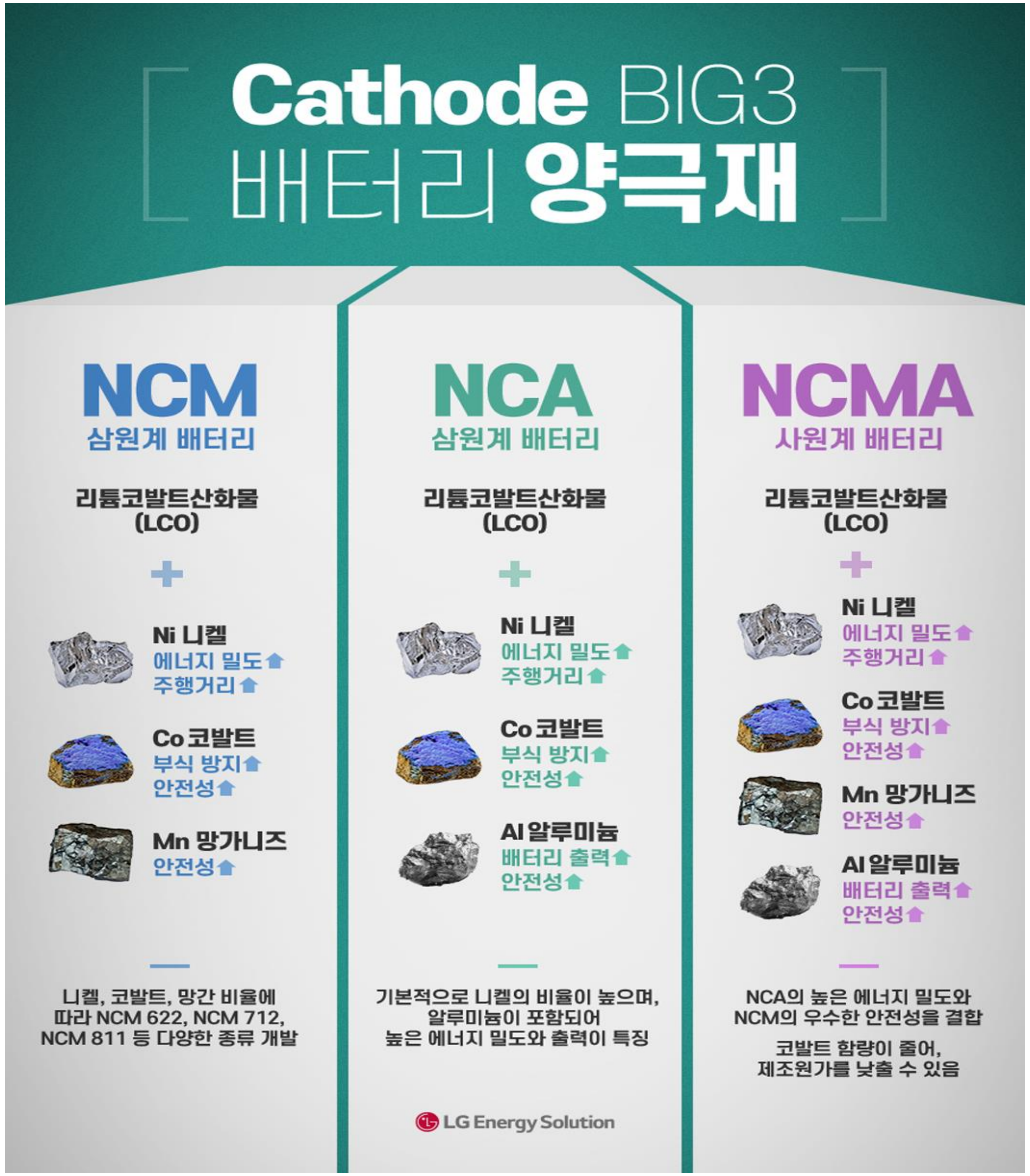
출처: 포스코그룹 뉴스룸

Battery Properties of Cathode Material Composition



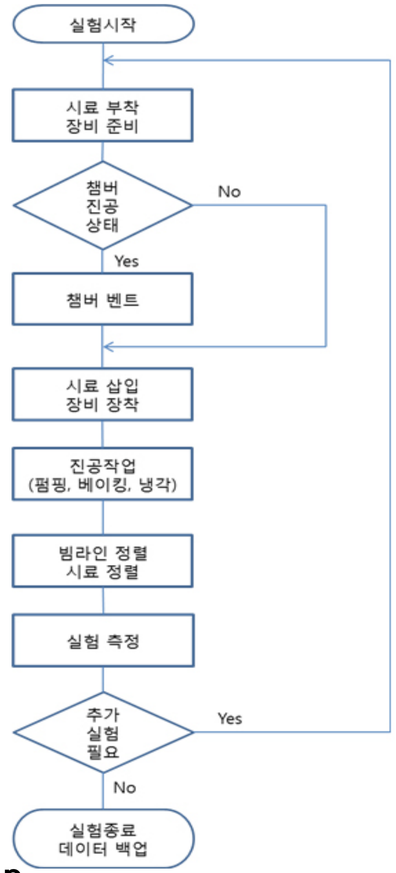
출처: 포스코그룹 뉴스룸

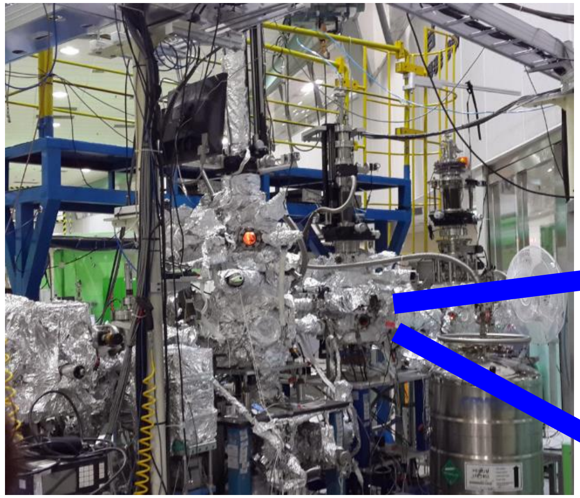
Cathode Material Big 3

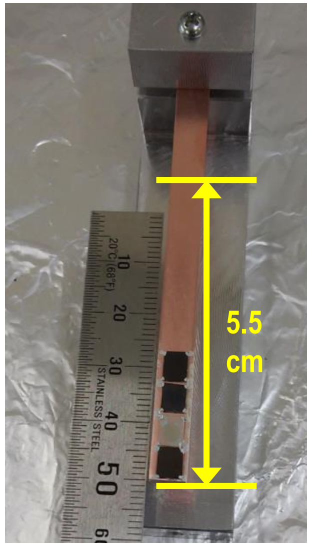


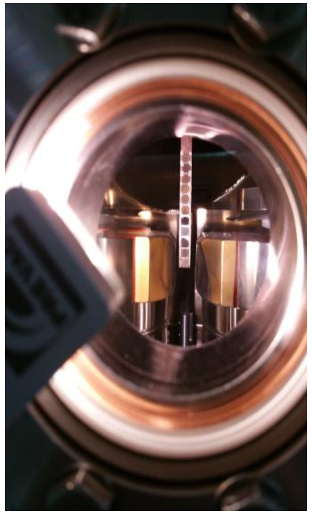
양극재구조
층상 구조 | 스피넬 구조 | 올리빈 구조
리튬이온
층상 구조
에너지 용량이 높음
(많은 양의 리튬이온 보관 가능)
대표적인 양극재
LCO, NCM, NCA
스피넬 구조
이온전도도가 높아 고출력 구현
대표적인 양극재
LMO
올리빈 구조
안정성을 높일 수 있음
대표적인 양극재
LFP
LG Energy Solution

Experiment Procedures









Sample preparation

-. 홀더에 샘플을 붙일 수 있는 길이는 5.5cm 임.

홀더의 폭은 7mm, 빔 크기는 1mm x 0.2mm

→ 샘플 크기 3mm x 3mm ~ 5mm x 3mm 정도가 적합

-. UHV(초고진공, ~ 1 x 10^{-9} torr)를 만들기 위해 Baking과 냉각 작업이 ~18시간 정도 소요됨.

Thank you for your attention