

# Advanced Light Source: The Science and Technology of Soft X-rays



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**IEEE SER Workshop 2014**  
**October 16, 2014**

Overview of the ALS

Industrial R&D at the ALS: Memristors

Basic R&D at the ALS: Skyrmions

A look to the future: ALS Upgrade

# Overview of the Advanced Light Source

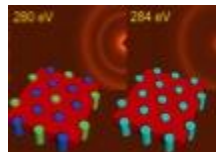
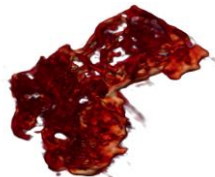
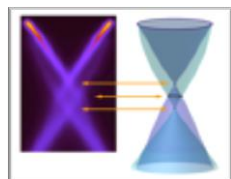
ALS mission is to *support users in doing outstanding science in a safe environment*

- Funded by DOE Office of Basic Energy Sciences, ~\$60M/year
- 21 years of operation
- 40 beamlines with unique capabilities, from IR to hard x-ray, niche area is soft x-rays
- Presently supports ~2400 users who produce >800 publications annually
- User base in chemical, physical, material, biological, environmental, and geological sciences
- X-ray spectroscopy, scattering, microscopy, and diverse combinations of all three of these

# ALS Users Are Productive Across a Broad Range of High Impact Science and Critical Technologies

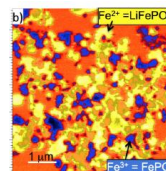
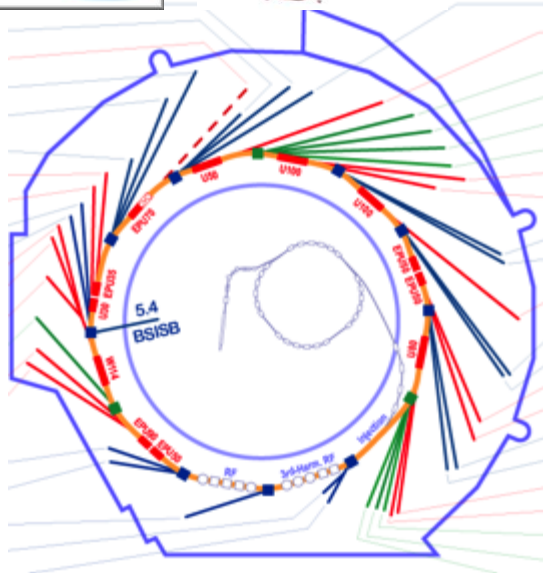
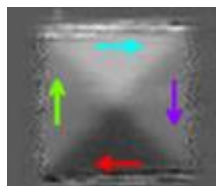
Hard materials: 5.3.2, 8.3.2, 10.3.2, 12.2.2, 12.3.2

Electronic textures: 4.0.3,  
7.0.1, 10.0.1, 12.0.1



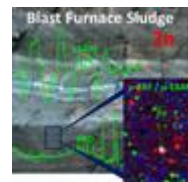
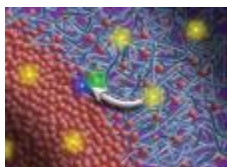
Polymers and resist materials: 5.3.2, 7.3.2,  
11.0.1, 12.0.1

Magnetism: 4.0.2,  
6.1.2, 11.0.1, 12.0.2



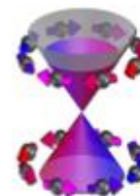
Electrochemical devices: 5.3.2, 9.3.1, 11.0.2

Organic PVs: 5.3.2, 7.3.3



Environmental textures: 5.3.2, 10.3.2

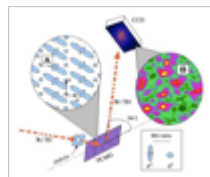
Cellular nanotomography: 2.1



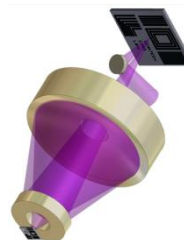
Spin textures: 10.0.1, 12.0.1



Living tissues: 1.4, 5.4, 8.3.2



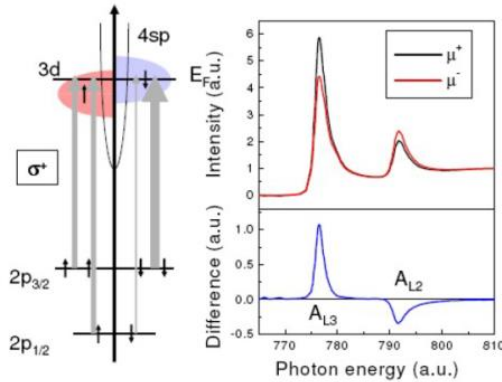
Orbital textures: 5.3.2, 12.0.2



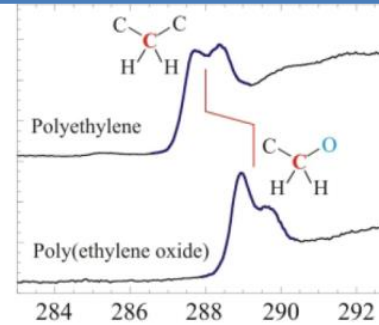
EUV technology: 6.3.1,  
11.3.2, 12.0.1

# Soft X-rays Enable Chemical, Electronic and Magnetic Imaging

## 3d transition metals



## Light elements



1	
<b>H</b>	
1.0079	
3	4
<b>Li</b>	<b>Be</b>
6.941	9.012
11	12
<b>Na</b>	<b>Mg</b>
22.99	24.30

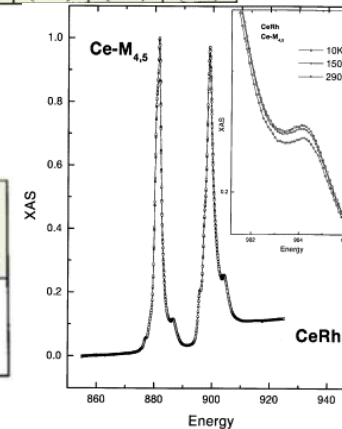
2
<b>He</b>
4.0026

5	6	7	8	9	10
<b>B</b>	<b>C</b>	<b>N</b>	<b>O</b>	<b>F</b>	<b>Ne</b>
10.811	12.011	14.007	16.00	19.00	20.179
13	14	15	16	17	18
<b>Al</b>	<b>Si</b>	<b>P</b>	<b>S</b>	<b>Cl</b>	<b>Ar</b>
26.98	28.09	30.974	32.06	35.453	39.948

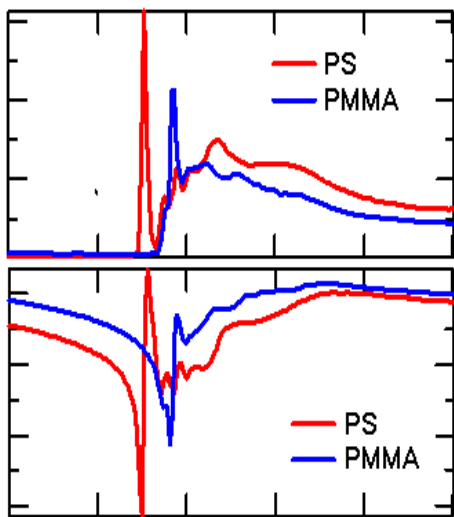
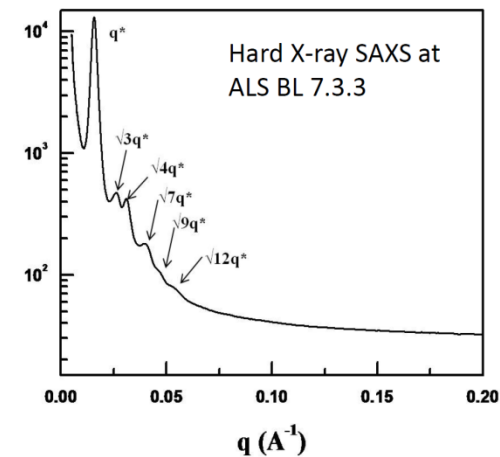
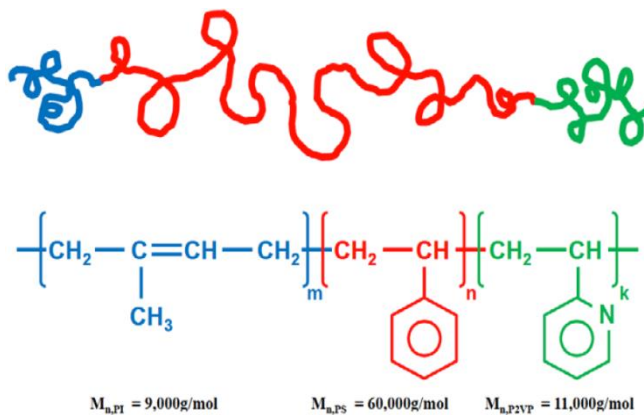
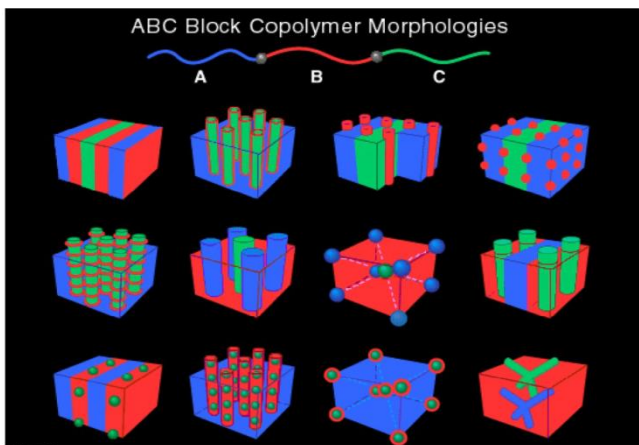
19	20	21	22	23	24	25	26	27	28	29	30	31	32	33	34	35	36
<b>K</b>	<b>Ca</b>	<b>Sc</b>	<b>Ti</b>	<b>V</b>	<b>Cr</b>	<b>Mn</b>	<b>Fe</b>	<b>Co</b>	<b>Ni</b>	<b>Cu</b>	<b>Zn</b>	<b>Ga</b>	<b>Ge</b>	<b>As</b>	<b>Se</b>	<b>Br</b>	<b>Kr</b>
39.10	40.08	44.96	47.90	50.94	52.00	54.938	55.85	58.93	58.69	63.55	65.39	69.72	72.59	74.92	78.96	79.90	83.80
37	38	39	40	41	42	43	44	45	46	47	48	49	50	51	52	53	54
<b>Rb</b>	<b>Sr</b>	<b>Y</b>	<b>Zr</b>	<b>Nb</b>	<b>Mo</b>	<b>Tc</b>	<b>Ru</b>	<b>Rh</b>	<b>Pd</b>	<b>Ag</b>	<b>Cd</b>	<b>In</b>	<b>Sn</b>	<b>Sb</b>	<b>Te</b>	<b>I</b>	<b>Xe</b>
85.47	87.62	88.91	91.22	92.91	95.94	(98)	101.1	102.91	106.42	107.87	112.41	114.82	118.71	121.75	127.60	126.91	131.29
55	56	57	72	73	74	75	76	77	78	79	80	81	82	83	84	85	86
<b>Cs</b>	<b>Ba</b>	<b>*La</b>	<b>Hf</b>	<b>Ta</b>	<b>W</b>	<b>Re</b>	<b>Os</b>	<b>Ir</b>	<b>Pt</b>	<b>Au</b>	<b>Hg</b>	<b>Tl</b>	<b>Pb</b>	<b>Bi</b>	<b>Po</b>	<b>At</b>	<b>Rn</b>
132.91	137.33	138.91	178.49	180.95	183.85	186.21	190.2	192.2	195.08	196.97	200.59	204.38	207.2	208.98	(209)	(210)	(222)
87	88	89	104	105	106	107	108	109	110	111	112						
<b>Fr</b>	<b>Ra</b>	<b>†Ac</b>	<b>Rf</b>	<b>Db</b>	<b>Sg</b>	<b>Bh</b>	<b>Hs</b>	<b>Mt</b>	<b>§</b>	<b>§</b>	<b>§</b>						
(223)	226.02	227.03	(261)	(262)	(263)	(262)	(265)	(266)	(269)	(272)	(277)						

## Rare earths

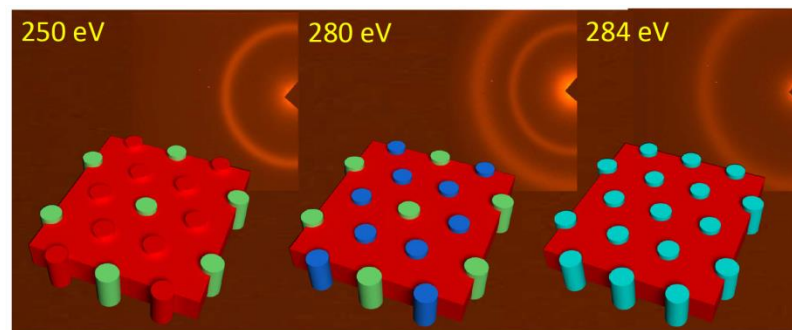
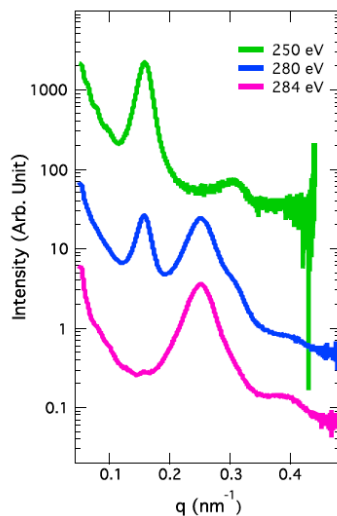
58	59	60	61	62	63	64	65	66	67	68	69	70	71
<b>Ce</b>	<b>Pr</b>	<b>Nd</b>	<b>Pm</b>	<b>Sm</b>	<b>Eu</b>	<b>Gd</b>	<b>Tb</b>	<b>Dy</b>	<b>Ho</b>	<b>Er</b>	<b>Tm</b>	<b>Yb</b>	<b>Lu</b>
140.12	140.91	144.24	(145)	150.4	151.97	157.25	158.93	162.50	164.93	167.26	168.93	173.04	174.97
90	91	92	93	94	95	96	97	98	99	100	101	102	103
<b>Th</b>	<b>Pa</b>	<b>U</b>	<b>Np</b>	<b>Pu</b>	<b>Am</b>	<b>Cm</b>	<b>Bk</b>	<b>Cf</b>	<b>Es</b>	<b>Fm</b>	<b>Md</b>	<b>No</b>	<b>Lr</b>
232.04	231.04	238.03	237.05	(244)	(243)	(247)	(247)	(251)	(252)	(257)	(258)	(259)	(260)



# Chemically Sensitive Soft X-ray Scattering



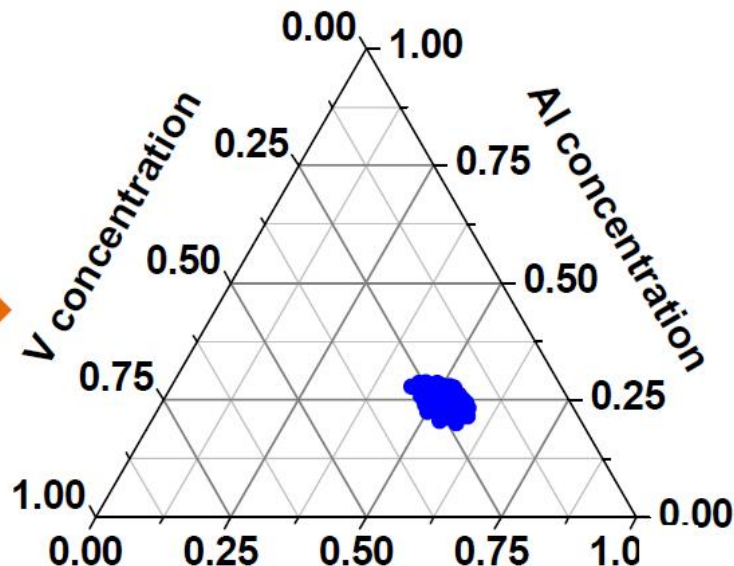
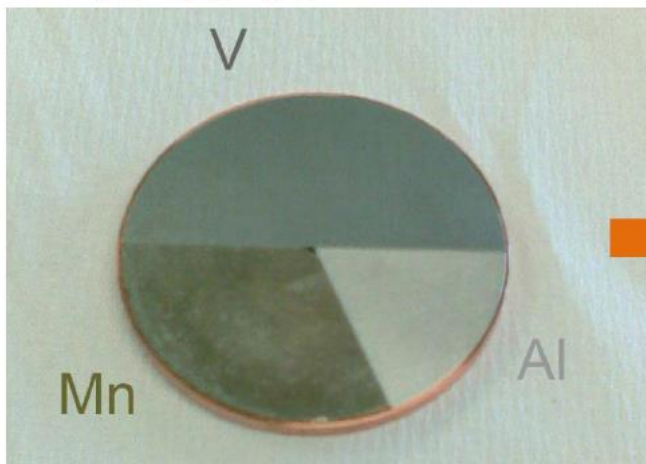
Photon Energy (eV)



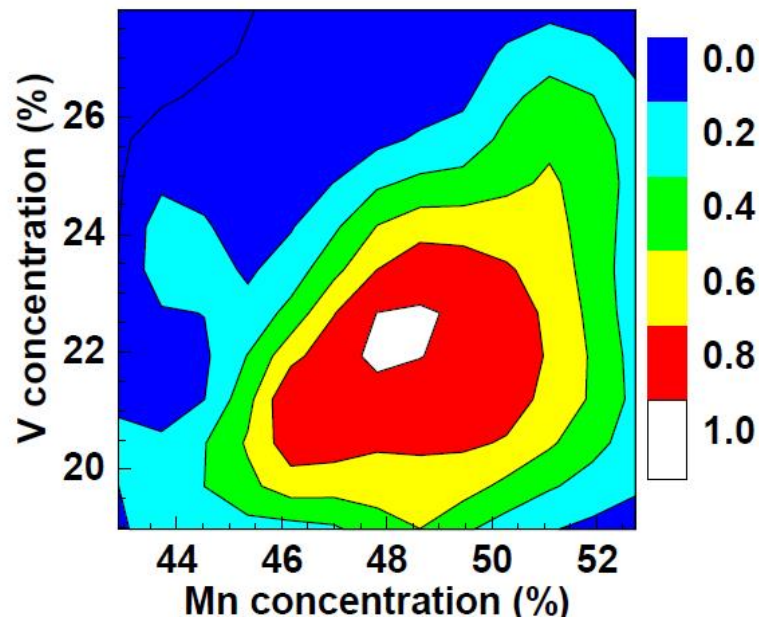
C. Wang et al., Nano Letters 2011

# Materials Discovery: Magnetism in Heusler Alloys

$\text{Mn}_{2-\delta}\text{V}_{1+\delta}\text{Al}_{1+\delta}$  – Sputter Target



- + Mn moment as function of composition (~200 XMCD spectra)
- + Maximum moment for  $\text{Mn}_{48.5}\text{V}_{22.5}\text{Al}_{29}$



Jan Schmalhorst, Daniel Ebke  
Univ. Bielefeld, Germany

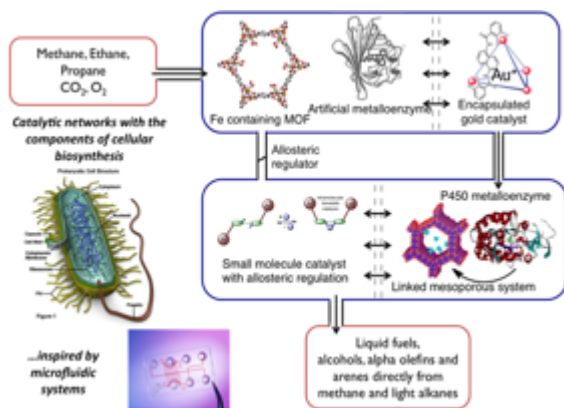


# ALS and ALS-U Research Trends

## Enabling directed chemistry

*Designed nanostructure to control kinetics and optimize efficiency of diverse chemical systems*

- Optimize electrodes of nano- and mesoscale structures for efficient energy conversion and storage
- Connect nanostructures in mesoscale functional networks for efficient and selective catalysis, electrocatalysis, and photocatalysis

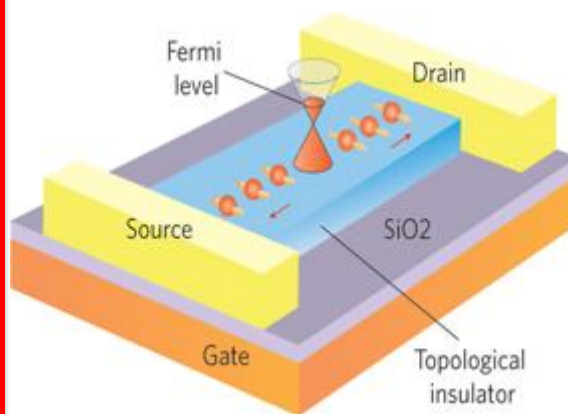


Toste/Hartwig, UC/LBNL

## Materials to enabling human scale computing

*New materials for controlling energy flow, and neural, quantum, and spintronic processing*

- Map and optimize nanoscale spin currents and spin textures in operating spintronic structures
- Control emergent, strongly coupled excitations for low power applications
- Develop candidate materials and structures for neural processing

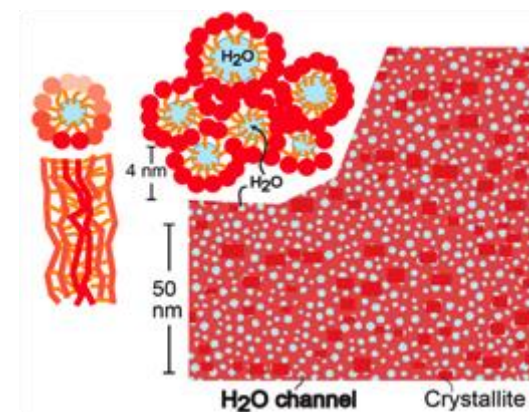


Xue, Nature Nano. **6**, 197–198 (2011)

## Materials and chemistry to address global challenges

*Material structures that rival the function of bio/enviro-systems*

- Design nanoporous membranes with extreme chemical selectivity of a biological membrane
- Optimize porous materials for carbon capture and sequestration, environmental remediation, water purification

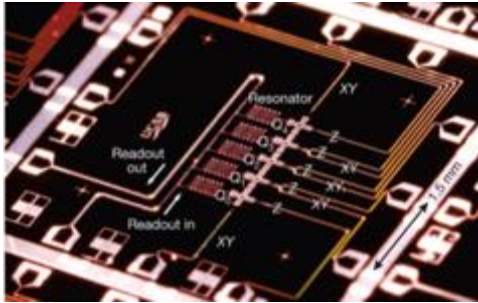


Schmitt-Rohr, Nature Mat. **7**, 75 (2008)

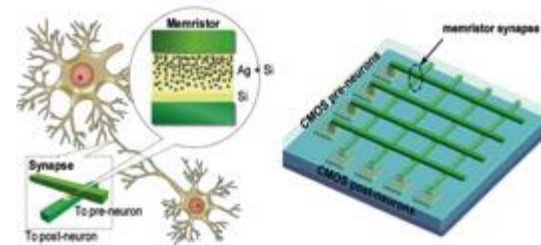


# Materials for Computing at the Human Scale

*Transformative material and device concepts for ultralow power future generation processing*



Superconducting Q-bits

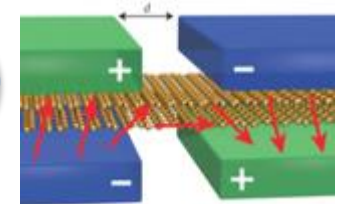


Neuromorphological processing

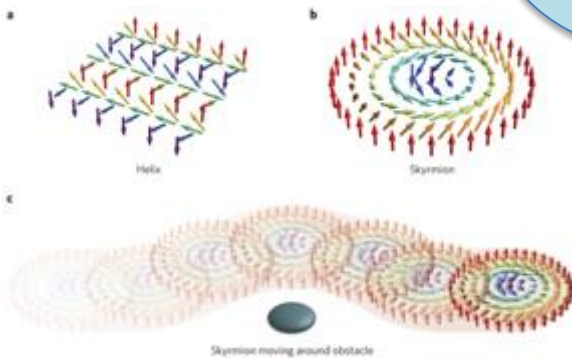
Composite Particles

Charges and orbitals

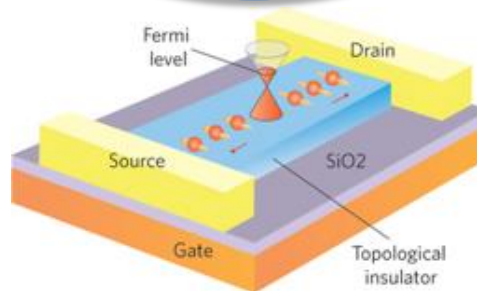
Spin currents and structures



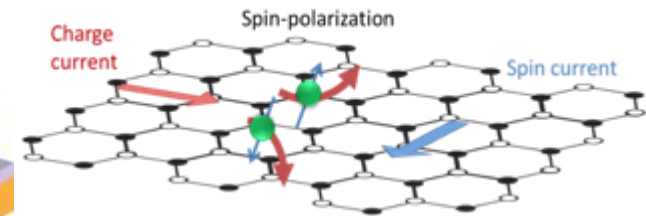
Graphene pseudo-spintronics



Classical and quantum processing with skyrmions

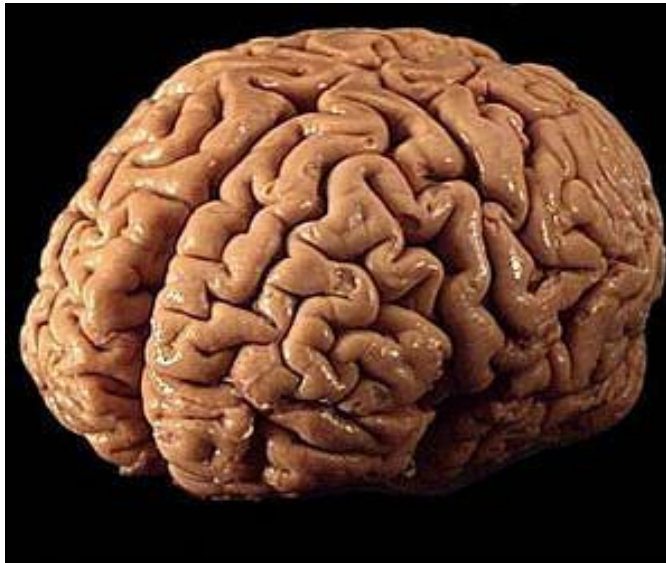


Spin currents and devices using topological insulators



Spin switching with currents

# A Model in Power Efficient Computing

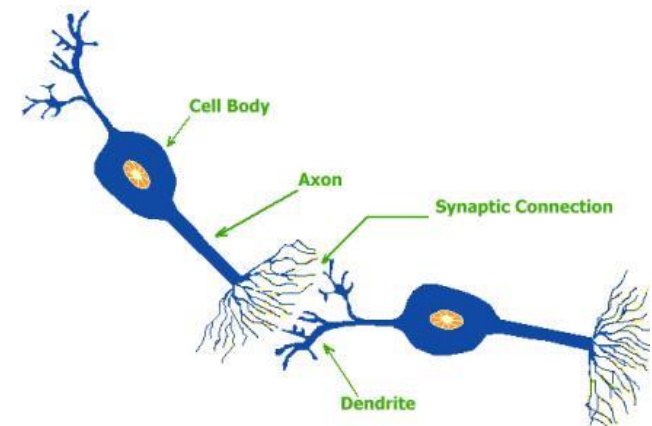


- ~20 W
- ~40 pFLOPS/sec
- ~100 Hz
  
- ~ $10^{11}$  neurons
- ~ $10^{14}$  synapses
  
- Massively parallel
- Usually off – neurons spike
- 3D architecture with distributed memory

Many biological systems switch with energy near the Landauer limit  $kT \ln(2) \sim 10^{-20}$  J

- - *even though they are immersed in a thermal bath which will drive fluctuations at the same energy scale*
- - *ingredients of the model are low frequency, low power, massive parallelism*

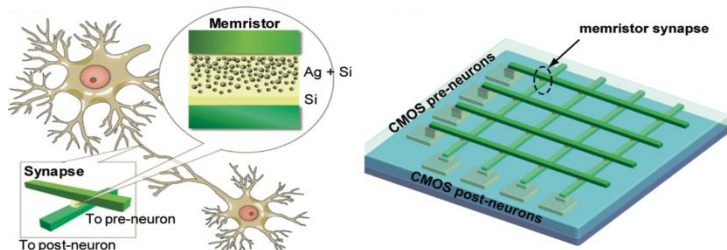
CMOS gates require  $\sim 10^5$   $kT$  to switch at present



Biological Processing Element(s)

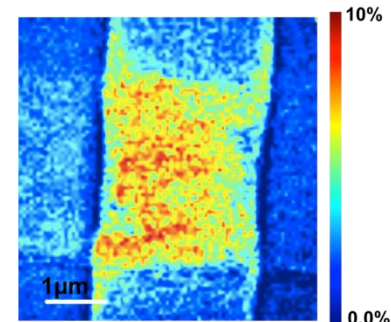
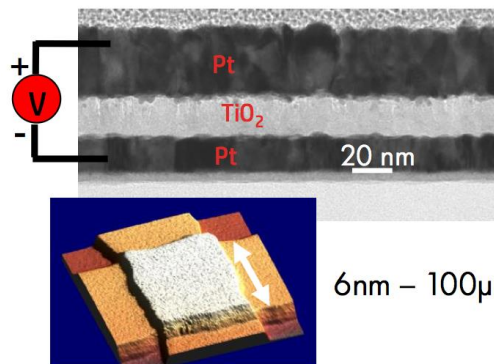
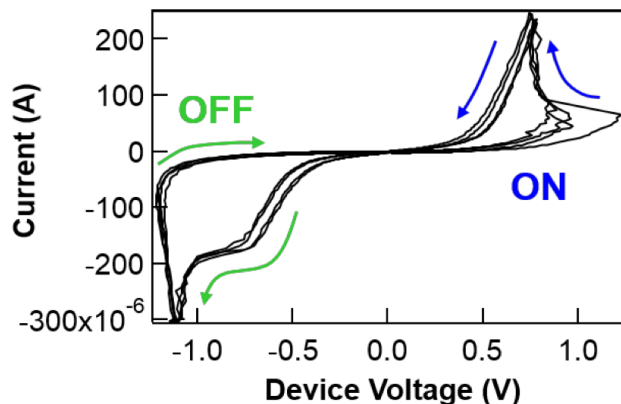
# Low-hanging Fruit: Memristor Memory

(Stan Williams, et. al, HP Labs)



## Memristor

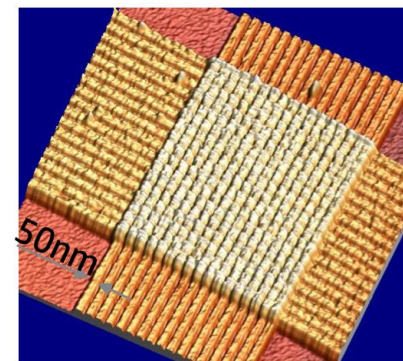
- Resistance depends on current history
- Resistance  $\sim$  strength of a synapse



2D map of oxidation states (ALS STXM)

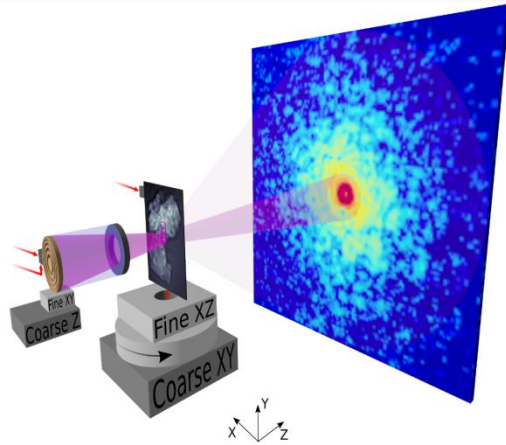
## Memristor Crossbar Array

- Nonvolatile memory
- High density (30 nm  $\rightarrow$  10 nm)
- Low switching energy (1 pJ)
- Fast ( $< 100$  ps)
- Scalable in 3D (Tb cache?)

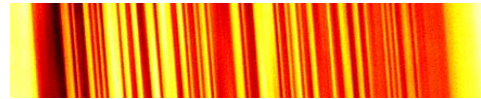


***Chemical heterogeneity of the junction determines nonlinear behavior, switching energy, cycle life, etc.***

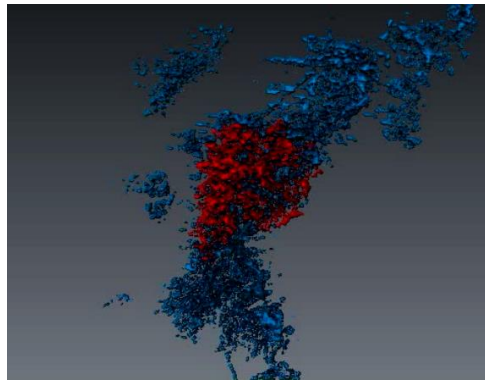
# Chemical X-ray Microscopy to the Nanoscale



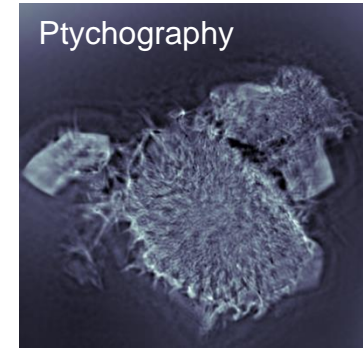
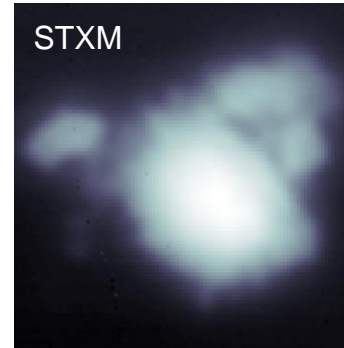
Diffractive imaging with ptychography



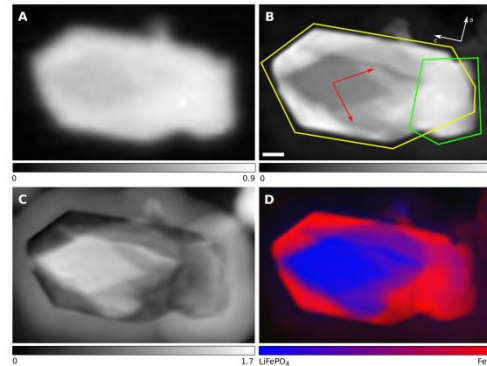
3 nm resolution on test objects



Sintered Zirconia in 3D



Resolving concrete chemistry



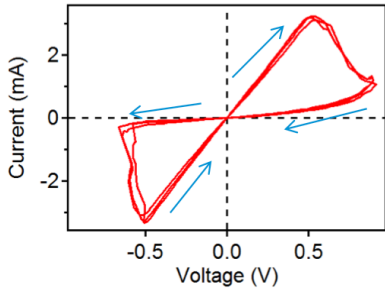
- A. STXM @ 30 nm
- B. Ptychography @ 10 nm: absorption
- C. Ptychography @ 10 nm: phase
- D. Chemical map @ 10 nm

Mapping battery oxidation states

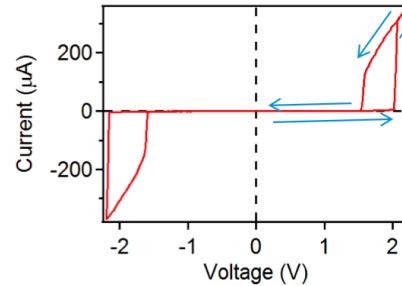
# Memristor Networks: Neural-morphological processing

[Pickett, *et. al.*, Nature Materials 12, 114–117 (2013)]

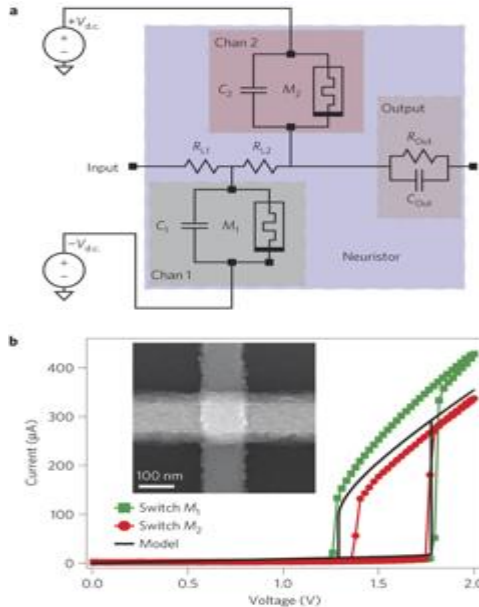
Nonvolatile memristor:  
*Emerging memory element*



Volatile memristor:  
*Emerging processing element*

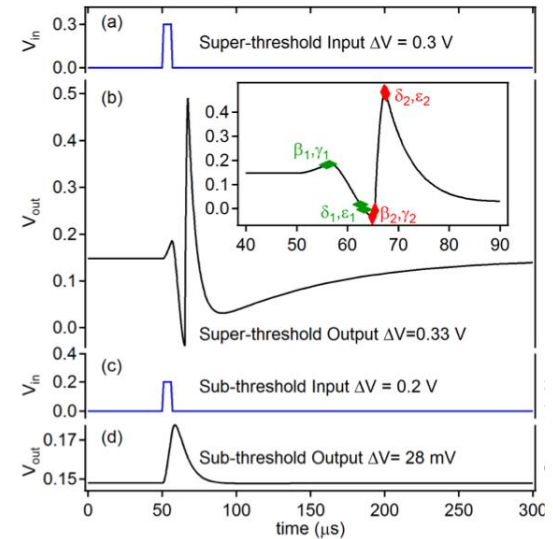


**Chemical heterogeneity of the junction determines nonlinear behavior, switching energy, cycle life, etc.**

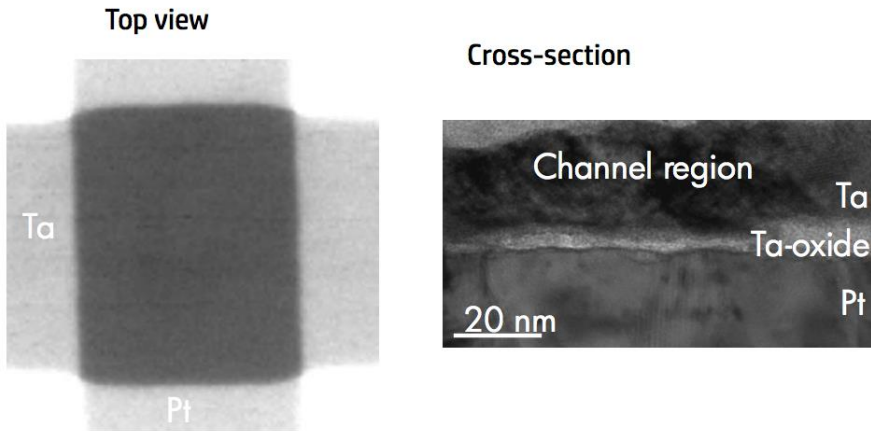


Memristive Neuristor:

- Two volatile memristors
- Two parallel capacitors
- All-or-nothing spiking

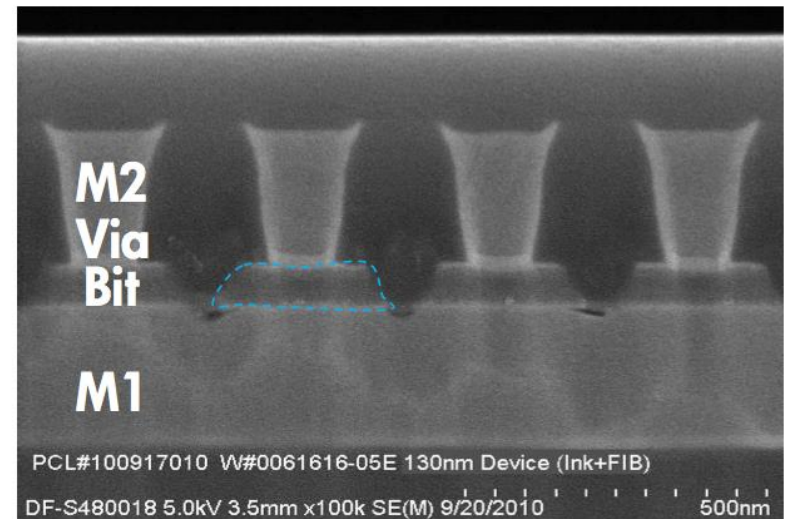


# Adding Color to Optimize Memristor Junctions

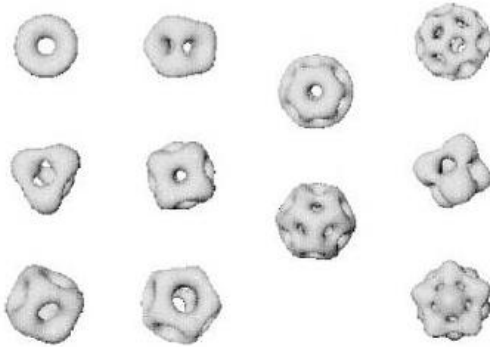


Full 3D mapping of chemical/structural/electronic properties with few nm resolution

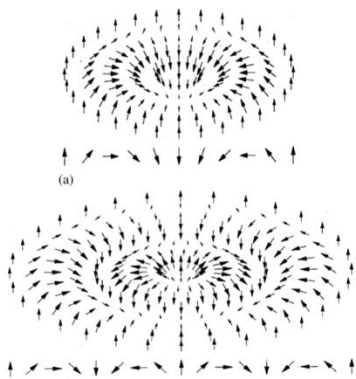
3D mapping of functioning device – junction, sidewalls, cycling lifetimes, etc.,



# Changing Gears: Magnetic Skyrmions

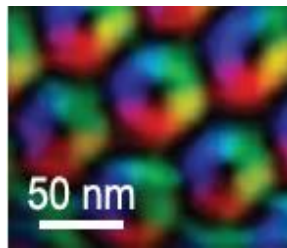


Tony Skyrme, 1962: *Topological Model of Baryons*



Skyrmions

Bogdanov, J. Mag. Magn. Materials 195, 182 (1999)



Skyrmion lattice

Seki et. al. Science 336, 198 (2012).

*What magnetic interactions conspire to cause magnetic skyrmions?*

Micromagnetic Ginsberg-Landau free energy density

$$w = A \left( \nabla \vec{M} \right)^2 - \vec{M} \cdot \vec{H} - K \left( \vec{M} \cdot \hat{n} \right)^2 - \frac{1}{2} \vec{M} \cdot \vec{H}_m + w_D$$

Symmetric exchange: ferro- or ferri-magnetic

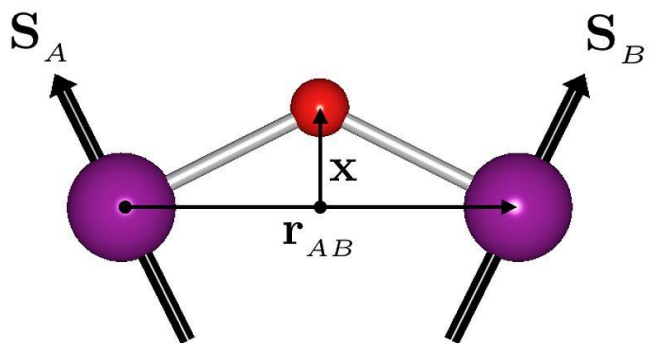
Zeeman energy: non-zero field

Perpendicular magnetic anisotropy (in films)

Demagnetizing (self) energy

Asymmetric exchange: Dzyaloshinskii–Moriya interaction

# Dzyaloshinskii–Moriya Interaction



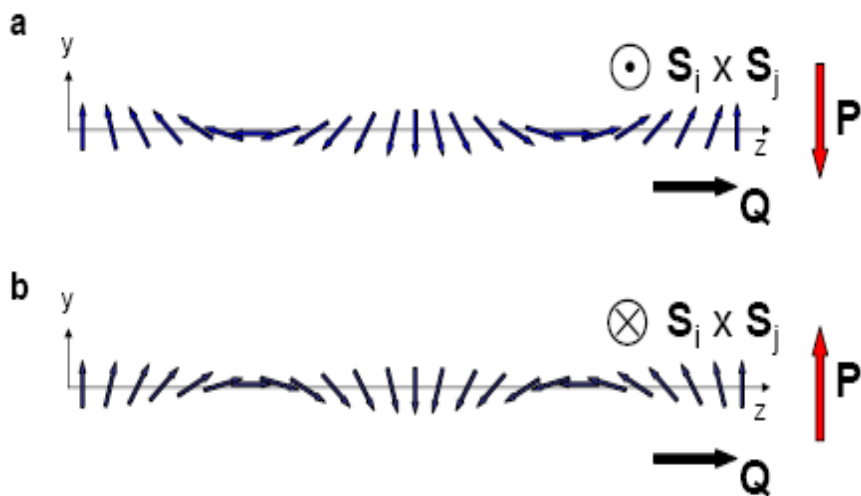
Symmetric (super)exchange (e.g., Heisenberg):

$$H_H = J \vec{S}_A \times \vec{S}_B$$

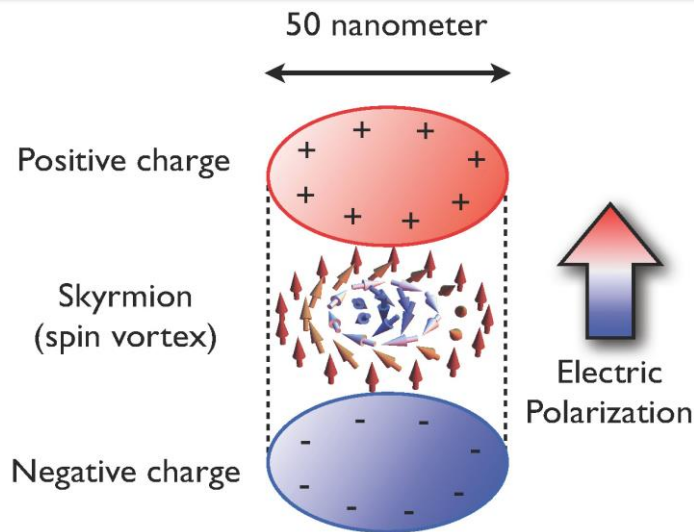
Asymmetric (super)exchange (e.g., DM via spin-orbit):

$$H_{DM} = \vec{D}_{DM} \times \vec{S}_A \cdot \vec{S}_B \quad \vec{D}_{DM} \propto \vec{x} \times \vec{r}_{AB}$$

Exchange between neighboring spins



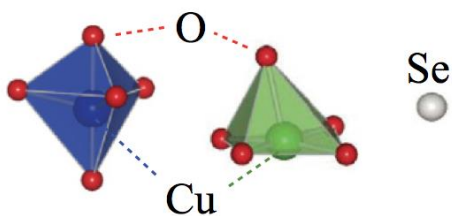
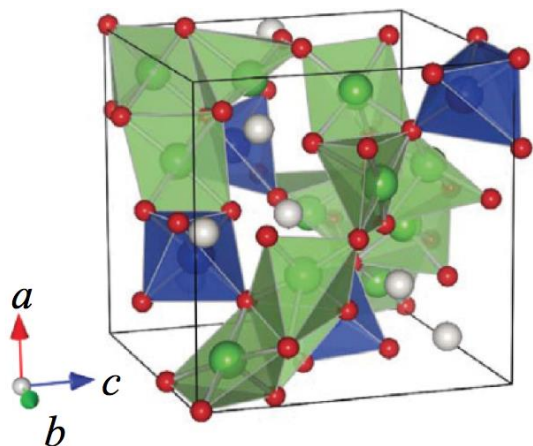
Spin-spirals in many materials with helicity set by the sign of  $D_{DM}$



Skyrmions in non-centrosymmetric materials – also with magnetoelectric effects



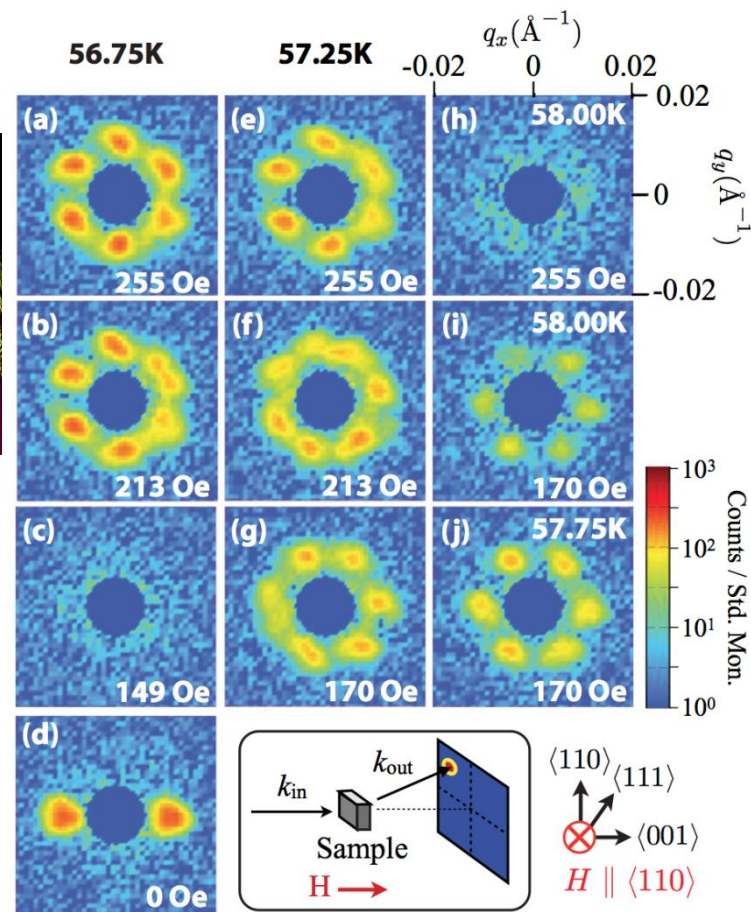
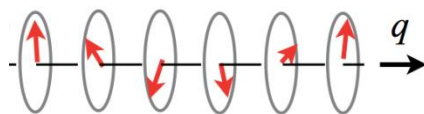
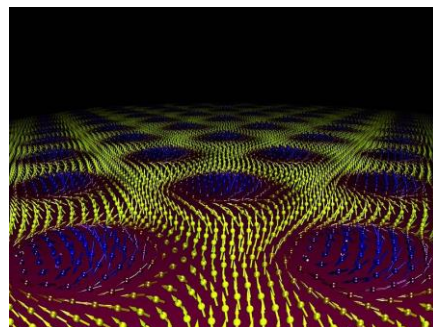
# Chiral Skyrmions in $\text{Cu}_2\text{OSeO}_3$



Cubic lattice

Ferrimagnetic

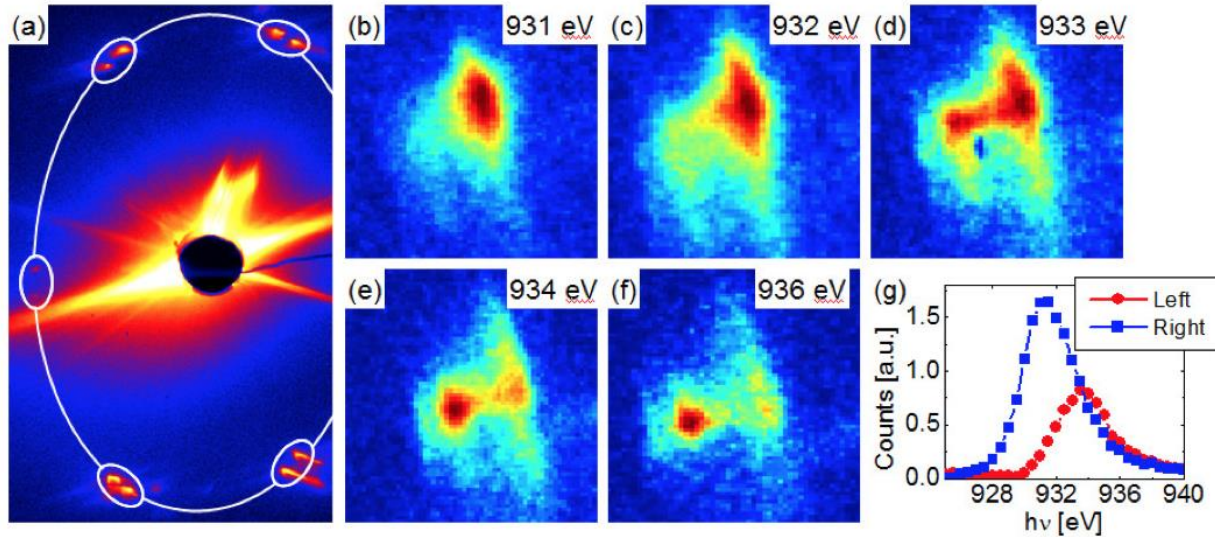
Non-centrosymmetric



Neutron diffraction from spiral and skyrmion lattices

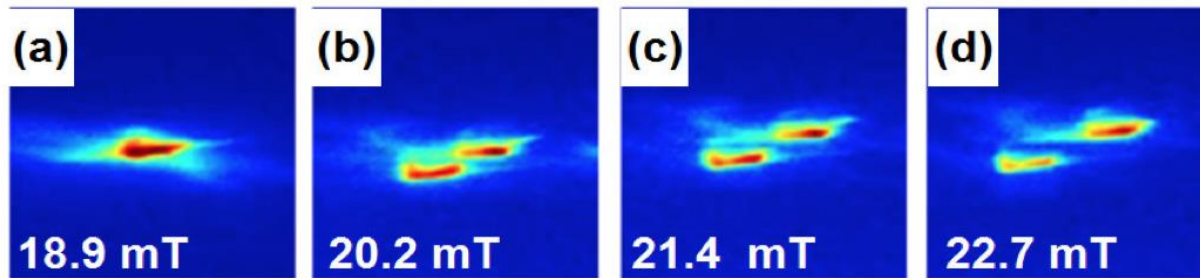
Seki, et. al., Phys. Rev. B **85**, 220406(R) (2012)

# Split Skyrmion Phases in $\text{Cu}_2\text{OSeO}_3$



Skyrmion satellites split suggesting two phases

Different phases resonate at different energies and have different chemical character.



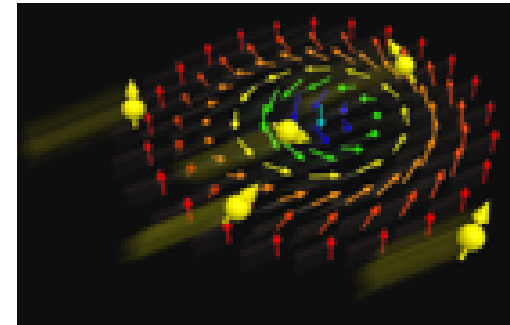
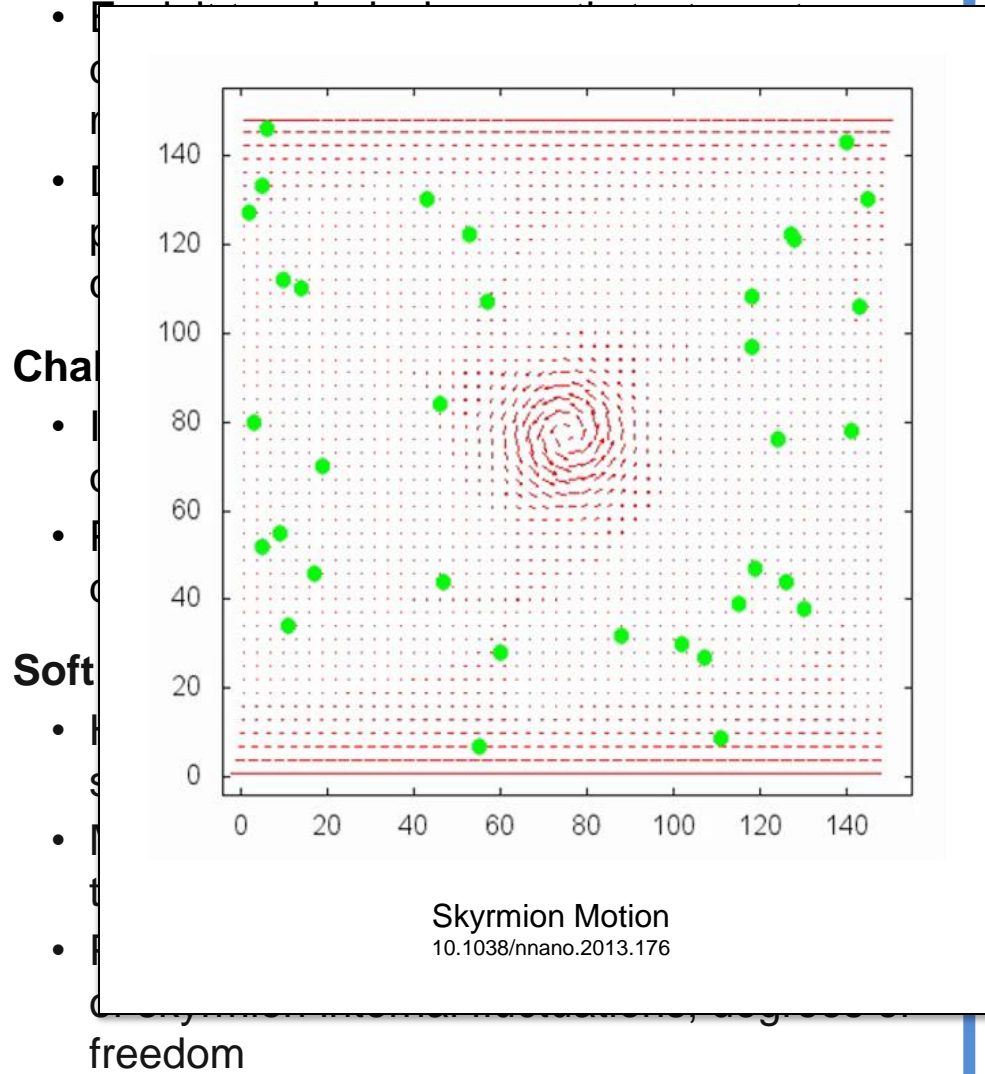
... and the splitting varies ~continuously with T and H

Rotational splitting is more robust than coupling to the lattice

We must be missing a term in the Hamiltonian. Ferrimagnetic?

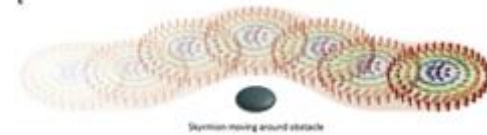
# Skyrmionic Information Processing

## Opportunity

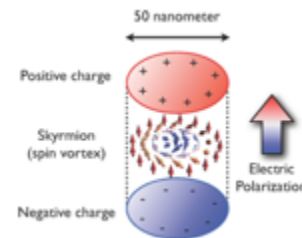


Skyrmions: Stable Topological Vortices

- - Two quantum numbers – store information
- - Weak lattice coupling - low dissipation

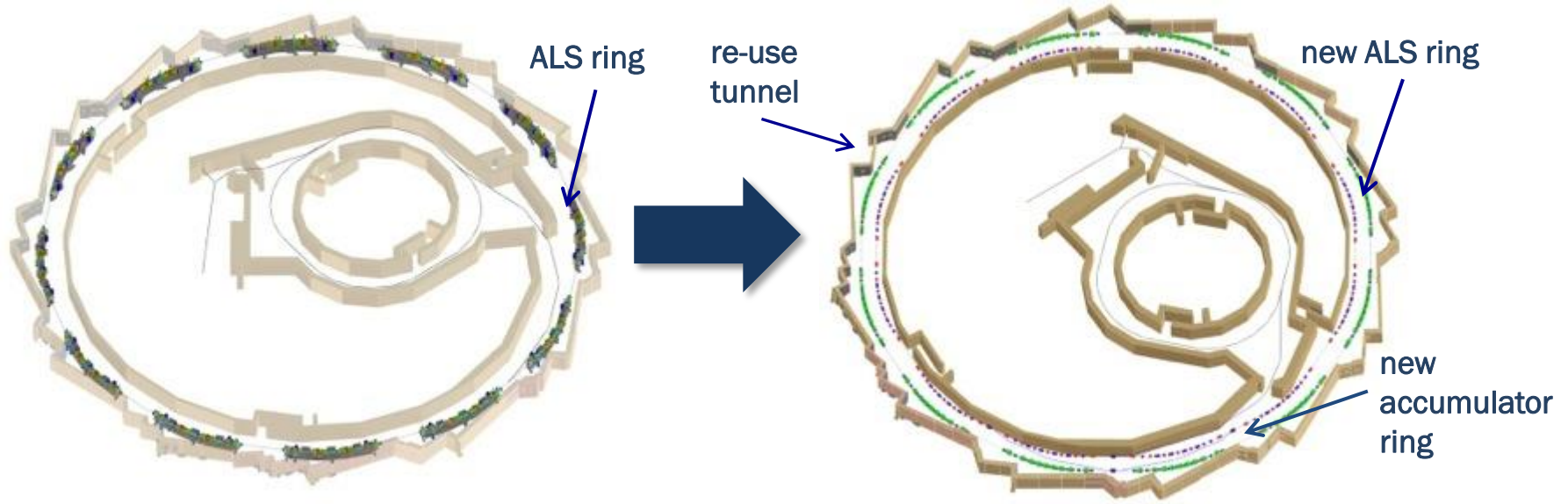


- - Magnetoelectric – external field control



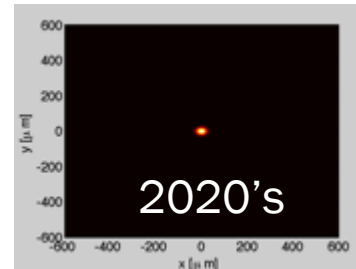
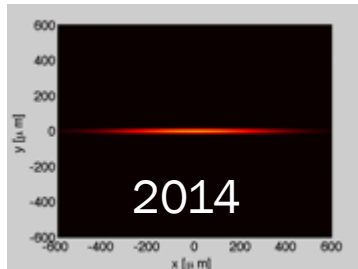
Candidate for low power classical and quantum information storage and processing

# Scope of ALS-U



ALS today

ALS-U

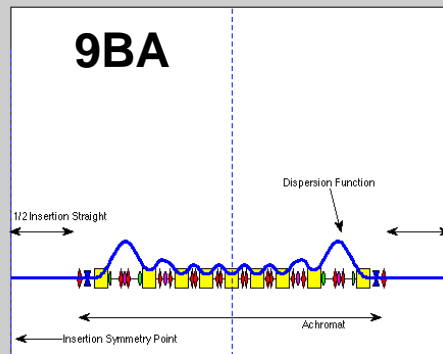
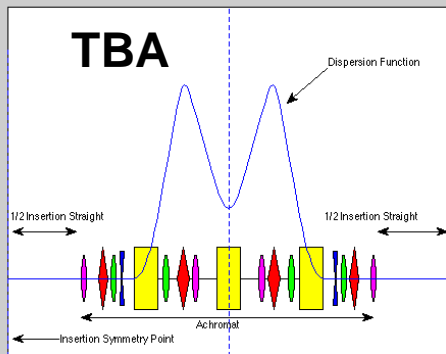
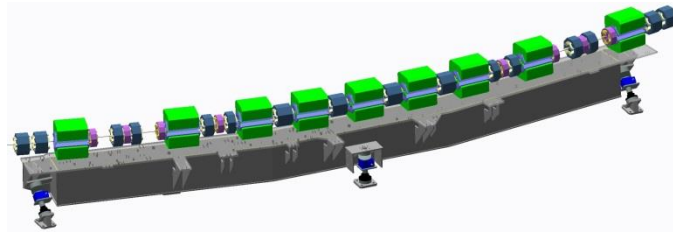
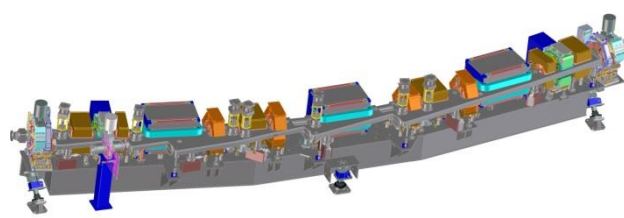


- New accumulator and storage ring based on multi-bend achromat lattice
- Re-use tunnel and beamlines

***ALS-U will provide highest-power coherent soft x-rays from a synchrotron***

# Multi-bend achromats pave way to the diffraction limit

Lattice design of ALS would evolve from a triple-bend achromats (TBA) to a multi-bend (9BA) achromat for ALS-II. Result is a large reduction in emittance



**MBA: Strong Focusing and Low Dispersion**  
**First used for MAX-IV.**

$$e_x = 2000 \text{ pm} @ 1.9 \text{ GeV}$$

$$e_x = 52 \text{ pm} @ 2.0 \text{ GeV}$$

$$e_x = C \frac{E^2}{N_D^3}, \quad e_x \propto \frac{E^2}{C^3}$$

$C_L$  = lattice constant  
 $N_D$  = # dipoles  
 $C$  = Circumference

# Some Parting Thoughts

DOE x-ray facilities have a large impact on diverse areas of science and technology, serving over 10,000 users/year

ALS fills an important soft x-ray niche that offers useful electronic, chemical, and magnetic contrast

Increasing source brightness combine spectroscopic contrast with imaging and time resolution and broadens the application areas

A planned upgrade of the ALS will provide intense, diffraction-limited soft x-ray beams that will continue these trends

