Ising dipolar model and the structure of the spin-orbital liquid $Ba_3CuSb_2O_9$

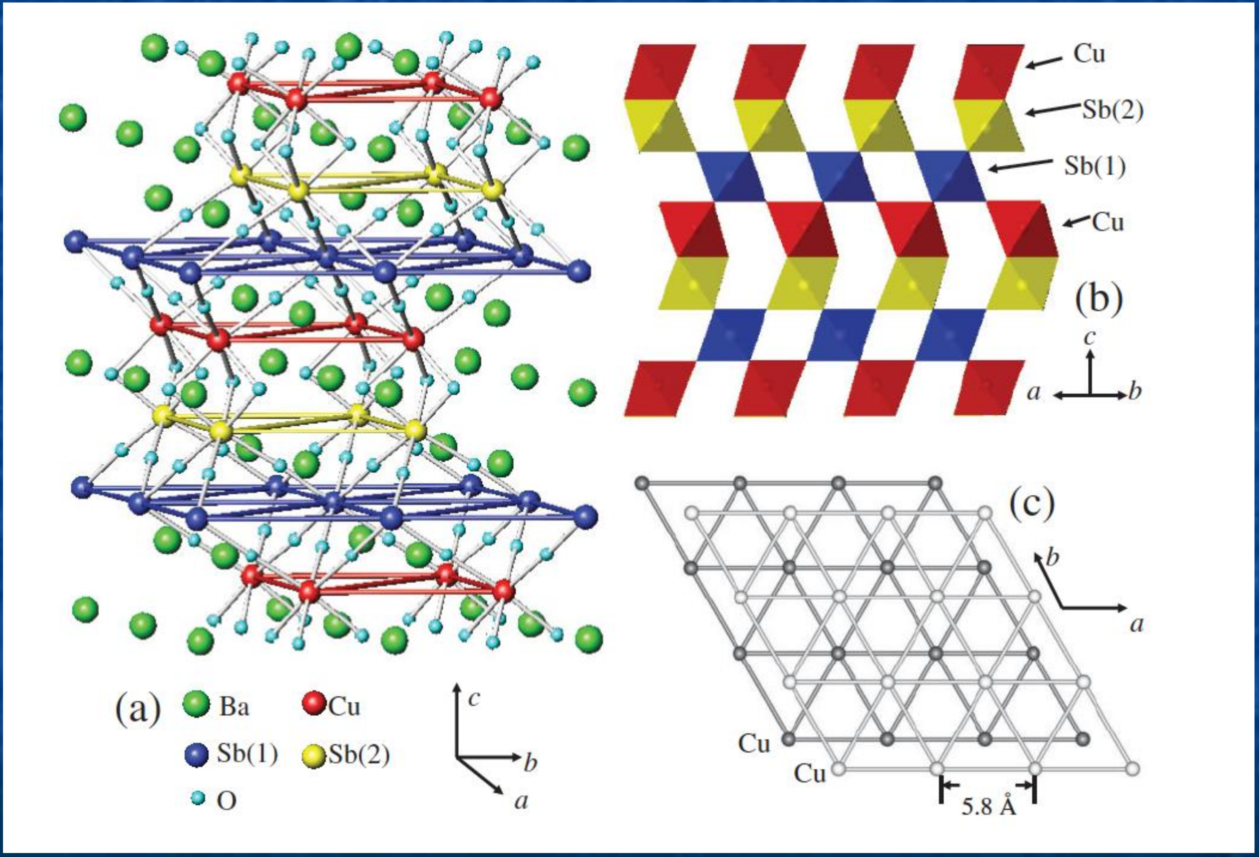
Frédéric Mila and Andy Smerald
EPFL, Lausanne, Switzerland

Scope

- The family $Ba_3AB_2O_9$
- Basic facts about $Ba_3CuSb_2O_9$
  → spin-orbital liquid
- Kugel-Khomskii model on honeycomb
  → orbital order
- Dipolar Ising model of CuSb dumbbell ordering
- Conclusions

Spin Liquid State in the $S = 1/2$ Triangular Lattice $Ba_3CuSb_2O_9$

H. D. Zhou,[1,*] E. S. Choi,[1] G. Li,[1] L. Balicas,[1] C. R. Wiebe,[1,2,3] Y. Qiu,[4,5] J. R. D. Copley,[4] and J. S. Gardner[4,6]



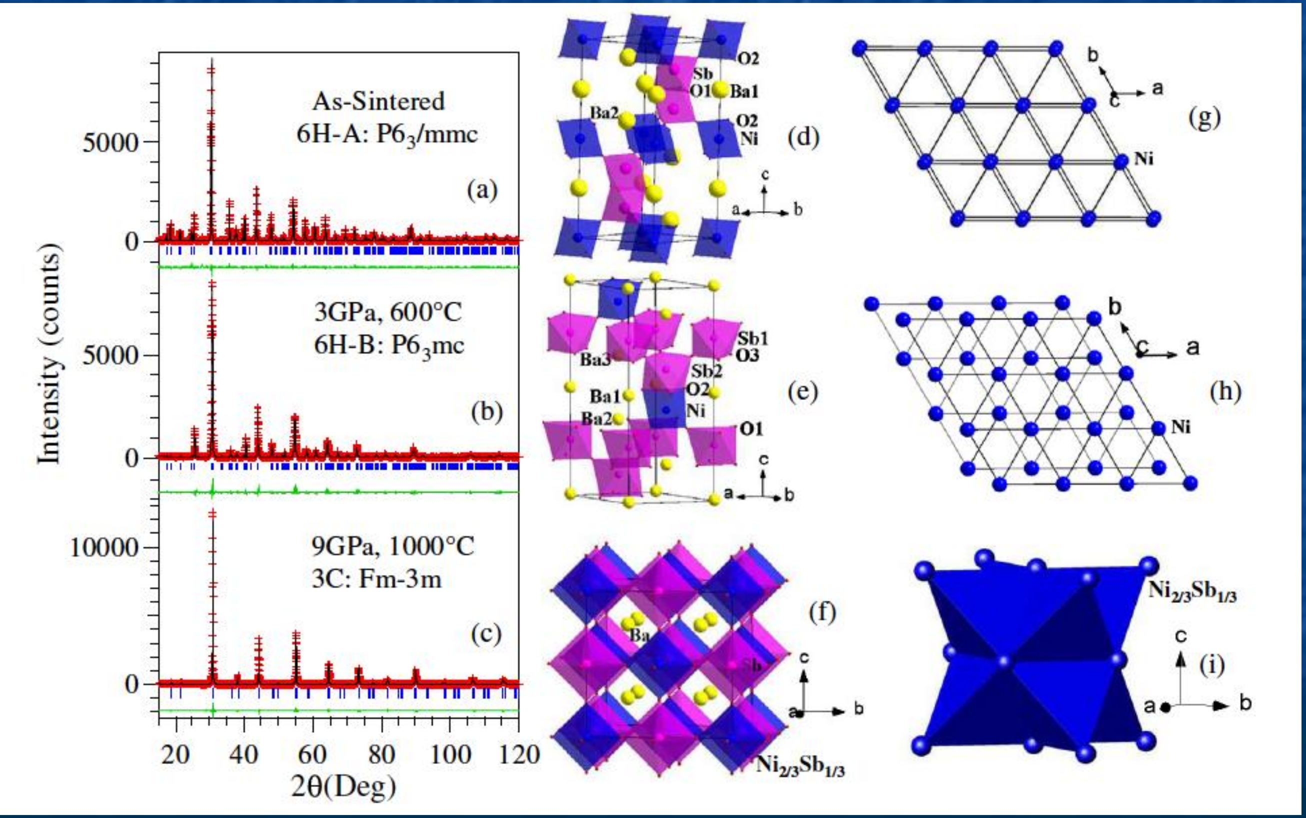
PRL 107, 197204 (2011) PHYSICAL REVIEW LETTERS week ending 4 NOVEMBER 2011

High-Pressure Sequence of $Ba_3NiSb_2O_9$ Structural Phases: New $S = 1$ Quantum Spin Liquids Based on Ni^{2+}

J. G. Cheng,[1] G. Li,[2] L. Balicas,[2] J. S. Zhou,[1] J. B. Goodenough,[1] Cenke Xu,[3] and H. D. Zhou[2,*]

Co^{2+} $S=1/2$

Mn^{2+} $S=5/2$

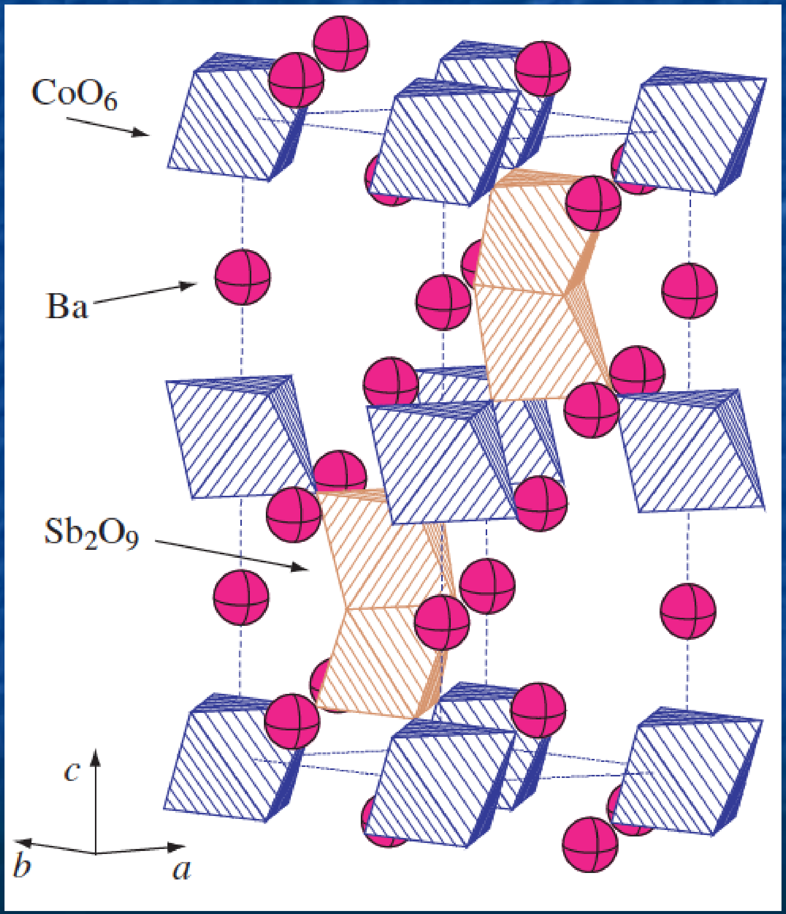


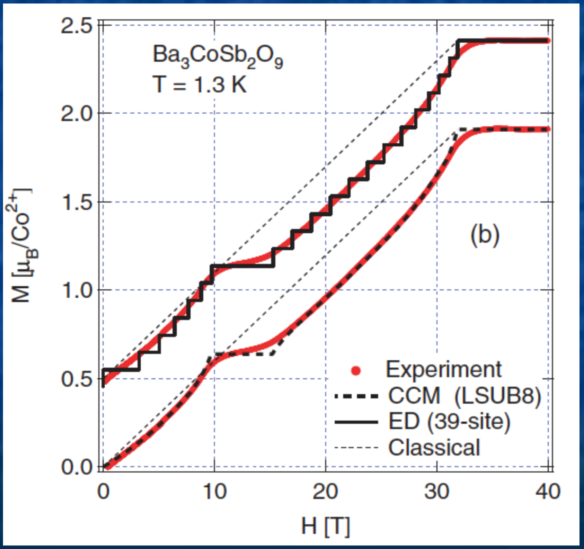
PRL **108**, 057205 (2012) PHYSICAL REVIEW LETTERS week ending 3 FEBRUARY 2012

Experimental Realization of a Spin-1/2 Triangular-Lattice Heisenberg Antiferromagnet

Yutaka Shirata,[1] Hidekazu Tanaka,[1] Akira Matsuo,[2] and Koichi Kindo[2]

Spin-1/2 AF on triangular lattice



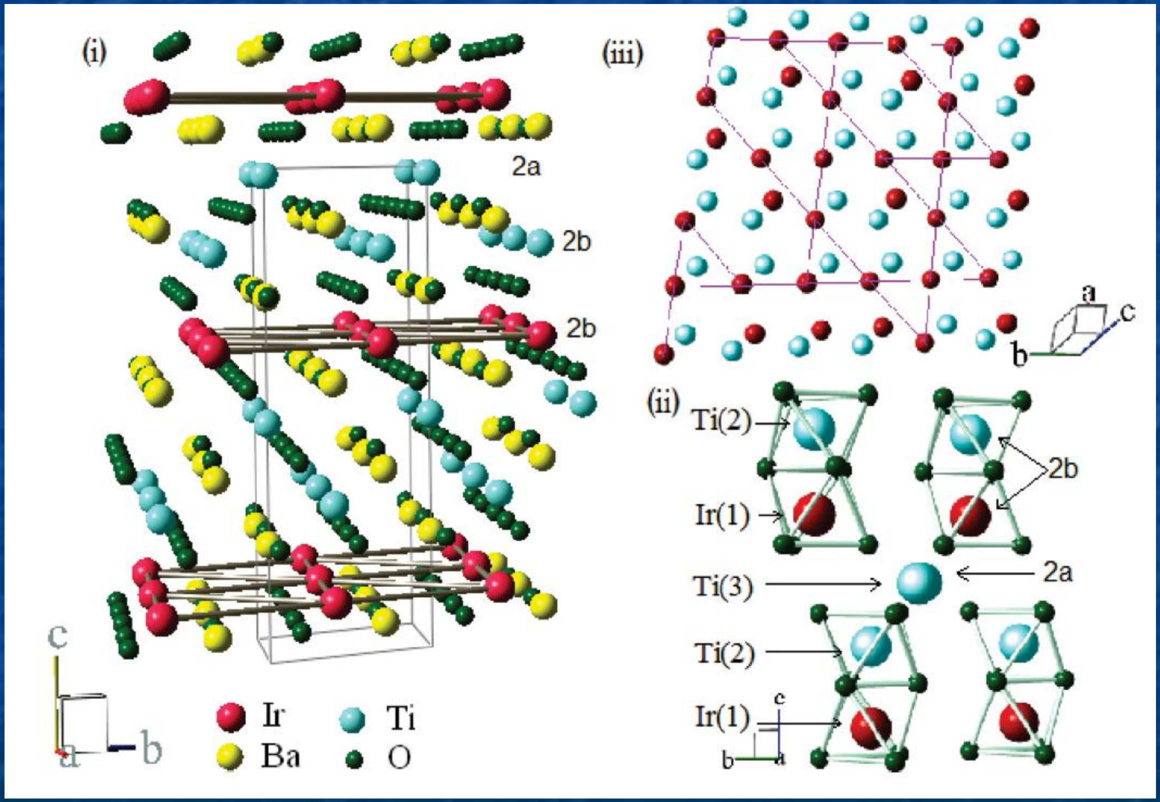


RAPID COMMUNICATIONS

PHYSICAL REVIEW B **86**, 140405(R) (2012)

Spin-liquid behavior in $J_{eff} = \frac{1}{2}$ triangular lattice compound $Ba_3IrTi_2O_9$

Tusharkanti Dey,[1] A. V. Mahajan,[1,*] P. Khuntia,[2] M. Baenitz,[2] B. Koteswararao,[3] and F. C. Chou[3]

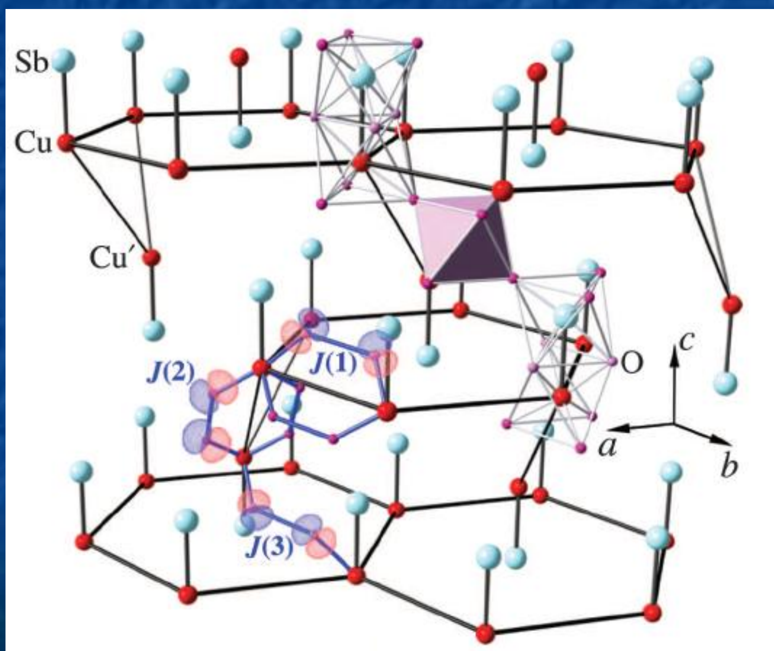


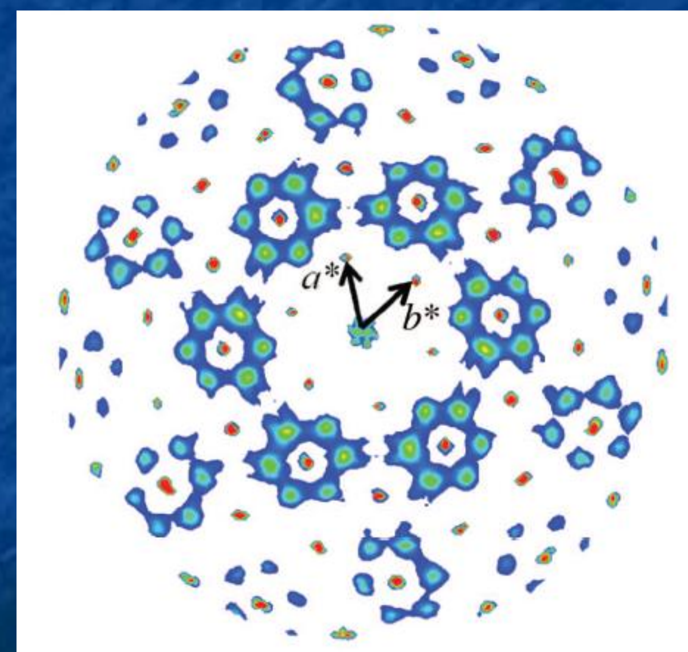
Ti^{4+}/Ir^{4+} site disorder, synthesis dependent

Spin-Orbital Short-Range Order on a Honeycomb-Based Lattice

S. Nakatsuji,[1*] K. Kuga,[1] K. Kimura,[1] R. Satake,[2] N. Katayama,[2] E. Nishibori,[2] H. Sawa,[2] R. Ishii,[3] M. Hagiwara,[3] F. Bridges,[4] T. U. Ito,[5] W. Higemoto,[5] Y. Karaki,[6] M. Halim,[7] A. A. Nugroho,[7] J. A. Rodriguez-Rivera,[8,9] M. A. Green,[8,9] C. Broholm[8,10]

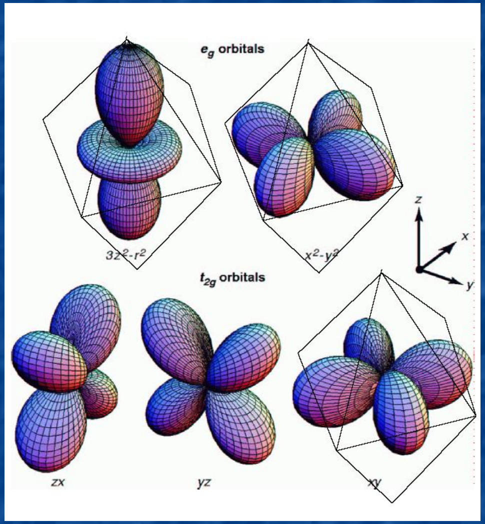
www.sciencemag.org SCIENCE VOL 336 4 MAY 2012

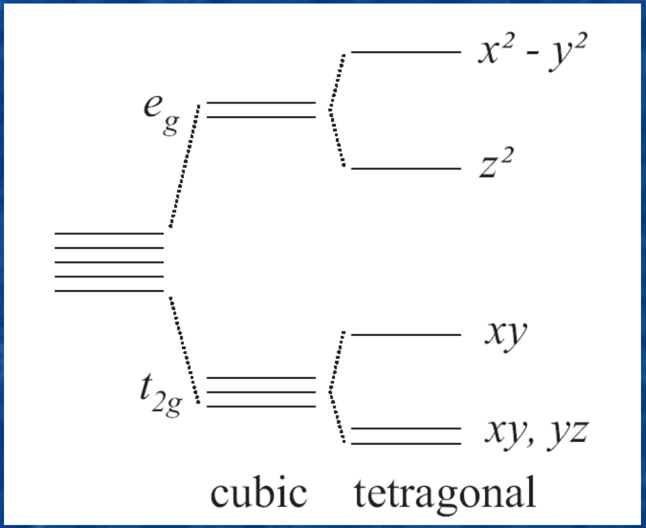


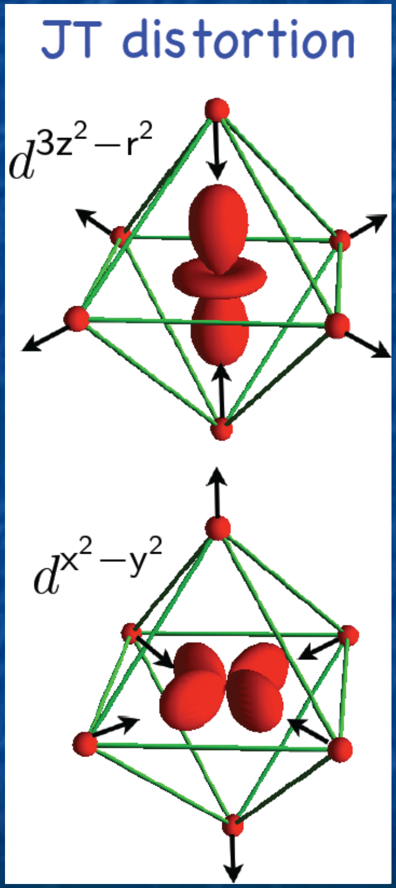


$Ba_3CuSb_2O_9$

Orbital degrees of freedom







- Cu^{2+} → 1 hole in e_g
- Orbital degeneracy

Orbital ordering in $Ba_3CuSb_2O_9$

- Off-stoichiometric sample $Ba_3Cu_{1-x}Sb_{2+x}O_9$
  → orthorhombic distortion for x≈10%
  → Cooperative Jahn-Teller distortion
  → ferro-orbital ordering
- Stoichiometric sample $Ba_3CuSb_2O_9$
  → no static JT, no orbital ordering
  → honeycomb lattice of CuSb dumbbells

Stoichiometric $Ba_3CuSb_2O_9$

- No orbital ordering
- NMR: Spin gap Quilliam et al, PRL 2012
- ESR: Orphan spins, between 5% and 16%

Can we understand this?

Kugel-Khomskii model

Two degrees of freedom per site

Spin

Pseudo-spin (orbital)

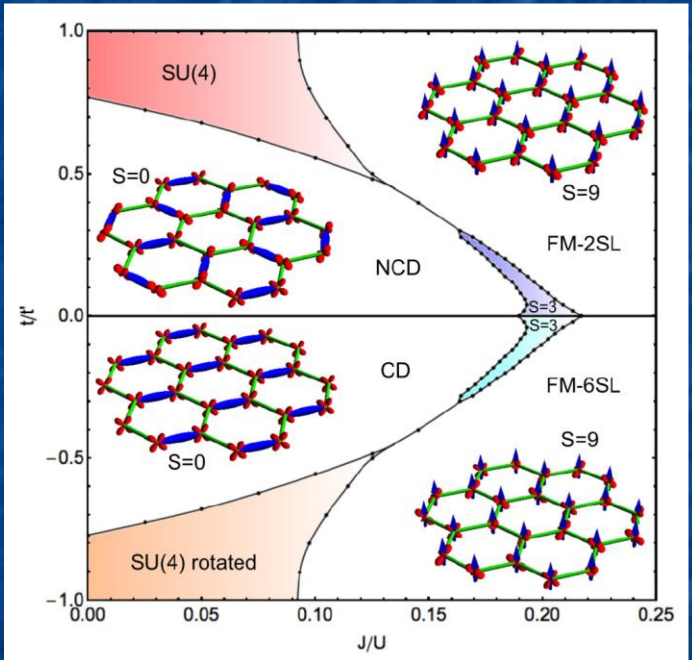
$$H = \sum_{i,j} J_{ij} \left(\vec{S}_i . \vec{S}_j \, K_1(\vec{\tau}_i, \vec{\tau}_j) + K_2(\vec{\tau}_i, \vec{\tau}_j) \right)$$

PHYSICAL REVIEW B **90**, 094422 (2014)

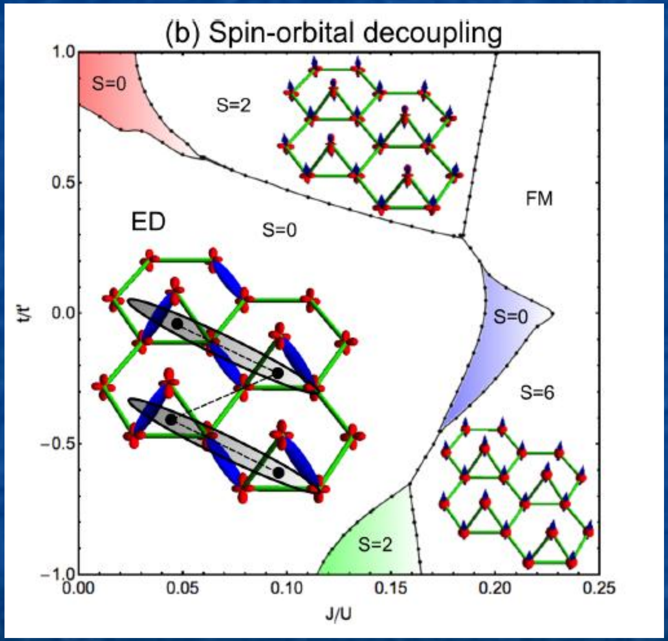
Exploring the spin-orbital ground state of $Ba_3CuSb_2O_9$

Andrew Smerald and Frédéric Mila

Honeycomb



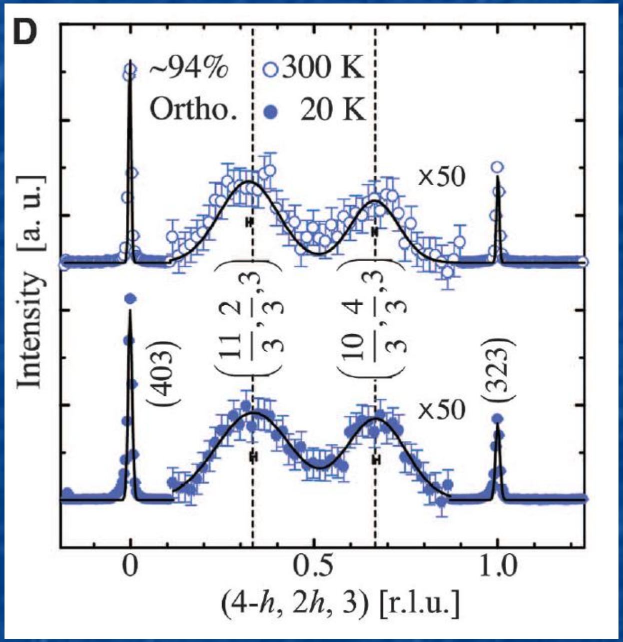
Decorated honeycomb



Strong tendency toward orbital order!

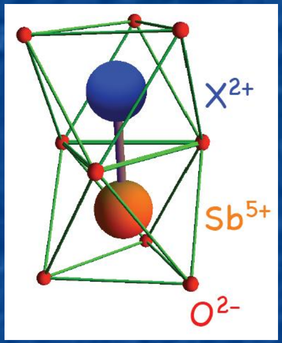
Disorder in $Ba_3CuSb_2O_9$

- Orphan spins
- Short correlation length
- No orbital order

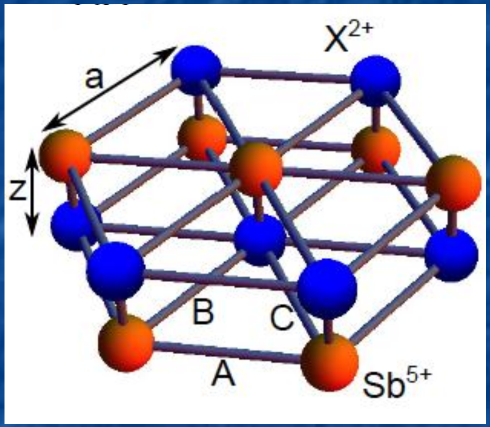


Which kind of disorder?

Dipolar Ising model



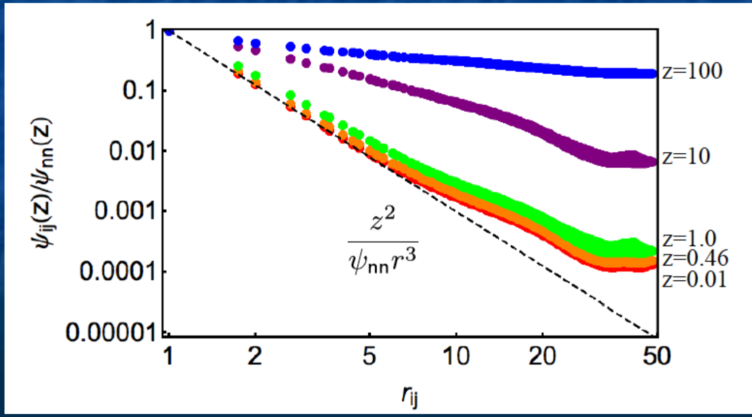
Cu^{2+}-Sb^{5+} → dipole



$$E_{\text{Coul}} = \frac{1}{2}\sum_{i\neq j}\frac{q_i q_j}{r_{ij}}$$

Ewald ↓

$$E_{\text{Coul}} = E_0 + \frac{1}{2}\sum_{i,j}\psi_{ij}(z)\sigma_i\sigma_j$$



AF Ising model on triangular lattice

- Infinite degeneracy

  → 2 up – 1 down or 2 down – 1 up / triangle

  Entropy/site: 0.3230…

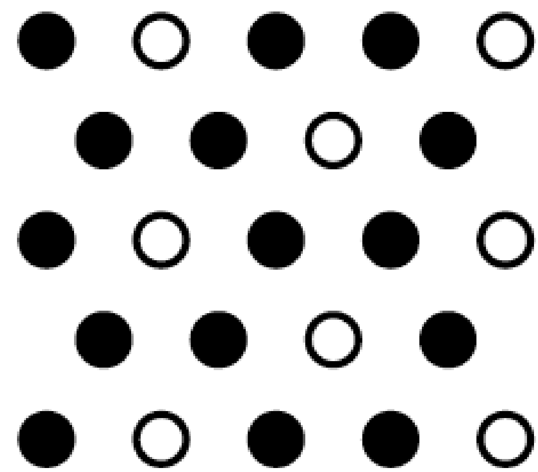
  Wannier, 1950

- Algebraic ground state correlations

  $<\sigma_i \sigma_j> \alpha\ 1/r_{ij}^{1/2}$

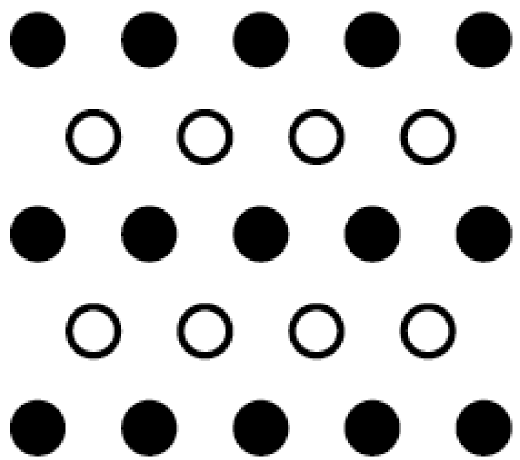
  Stephenson, 1964

J_1-J_2 Ising model on triangular lattice



$J_2<0$

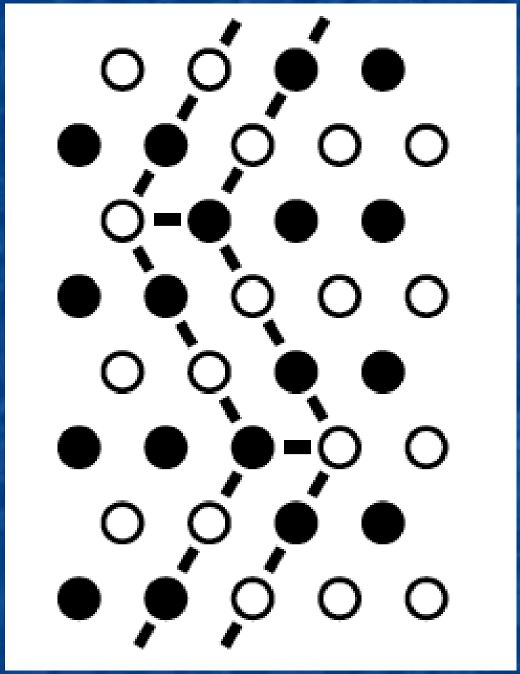
Honeycomb



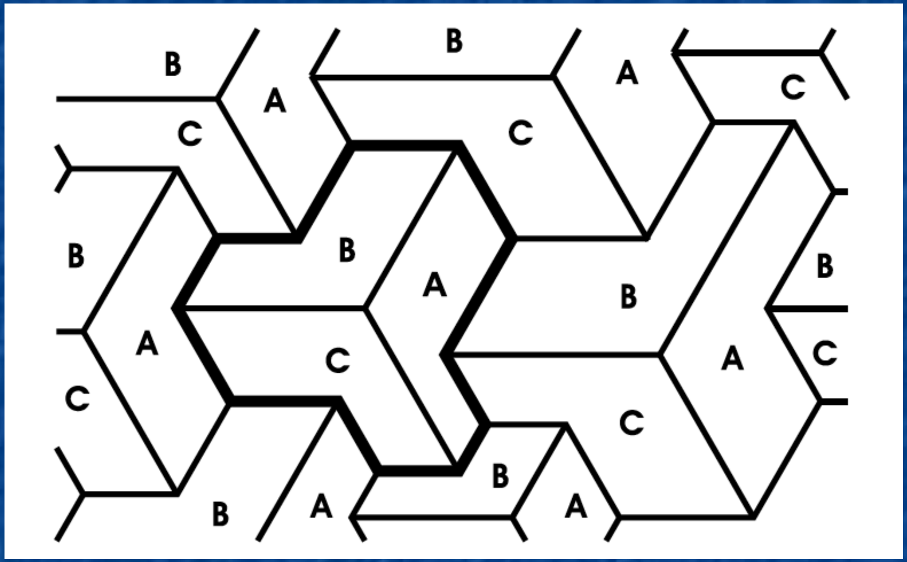
$J_2>0$

Stripe

Possible phase transitions



Ising, T_2



1st order, T_3

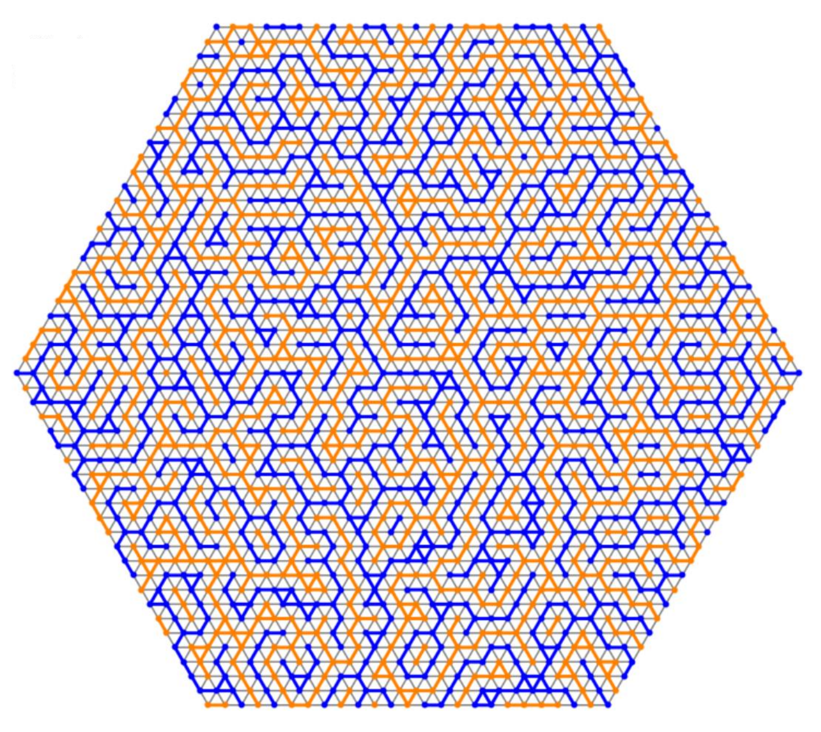
Sergey Korshunov, PRB 2005

One or two phase transitions?

- J_1-J_2: $T_3<T_2$ → only 1st order transition
- J_5 large enough, J_4 small: $T_2<T_3$
  → 2 phase transitions Korshunov 2005
- Dipolar model
  → stripe order at zero temperature
  → 1st order transition (Monte Carlo)

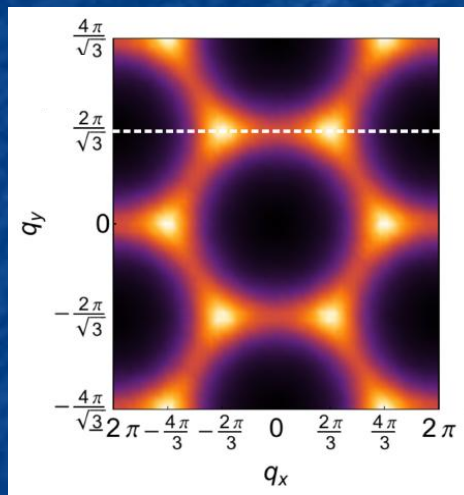
  A. Smerald, FM, unpublished

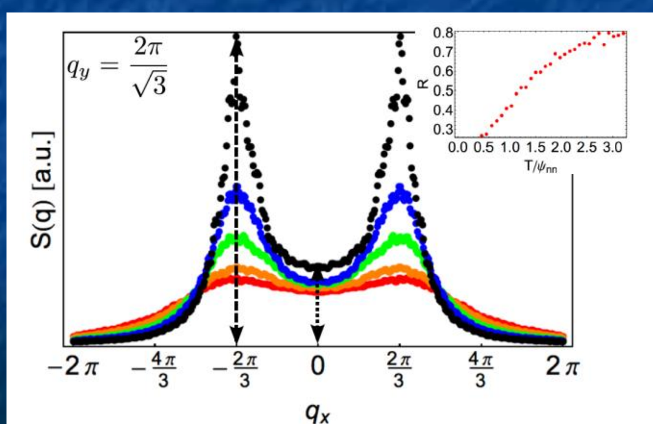
High temperature phase



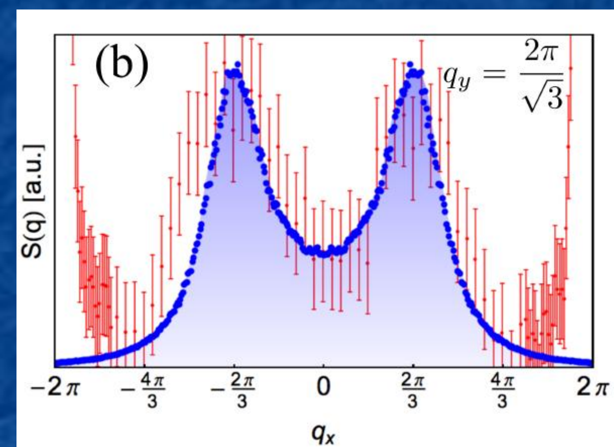
‘branch’ lattice

Structure factor





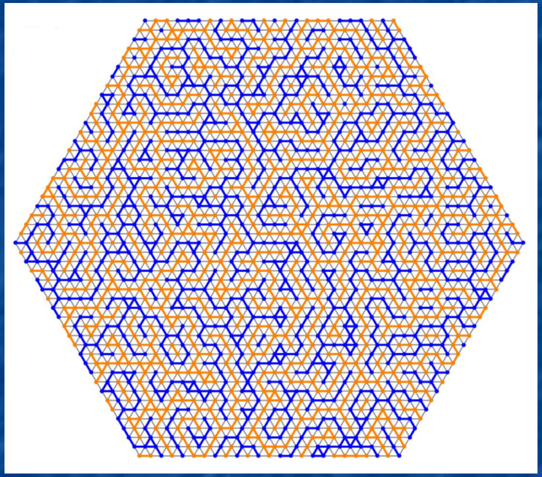
Theory/exp[t]

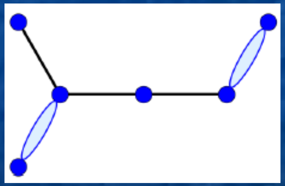


$T=0.9\ \Psi_{nn}$

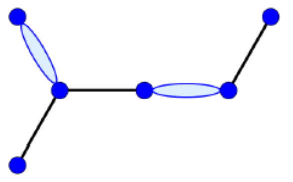
Properties of branch lattice

- Very few hexagons
- Fractal dimension: d=1.9
- Orphan spins: 6%
- Resonating dimers

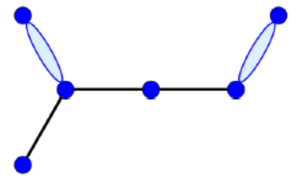




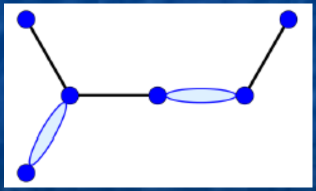
-0.415



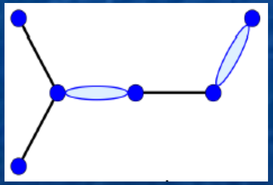
-0.243



+0.387

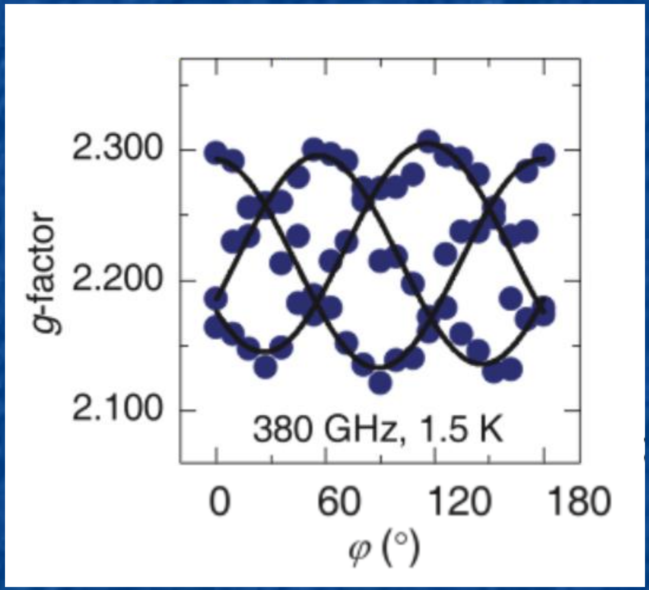


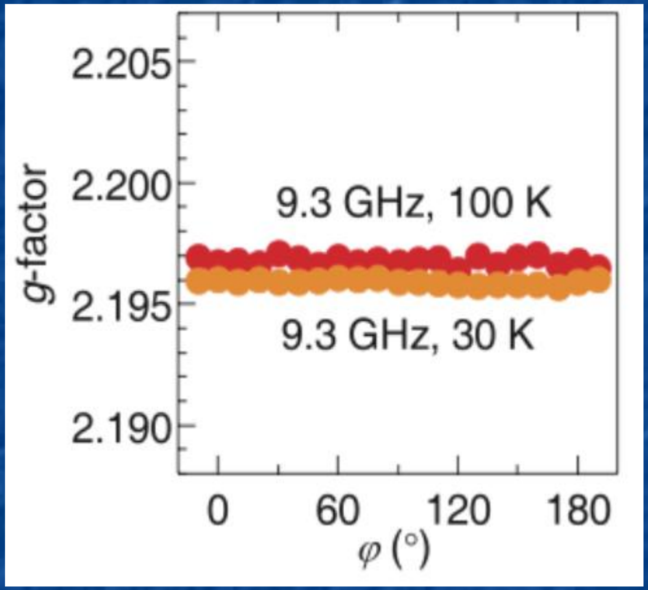
-0.235



-0.405

Dynamic Jahn-Teller effect





N. Katayama et al, unpublished

Conclusion

- Structure of $Ba_3AB_2O_9$ family: quite subtle!
- $Ba_3CuSb_2O_9$
  → CuSb dumbbells: dipolar Ising model
  → Low-temperature: stripe structure
  → High-temperature: branch lattice
- Kugel-Khomskii model on branch lattice
  → Resonating dimers + orphan spins
- Implications for other members of the family?