THE UNIVERSITY OF TOKYO

Quantum Magnets 2015 WORKSHOP

Crete, 15 Sept 2015

Designing flat-band ferromagnets — a path to make organic-based systems magnetic and topological

Hideo Aoki
Dept of Physics, Univ Tokyo

Thesis of today's talk

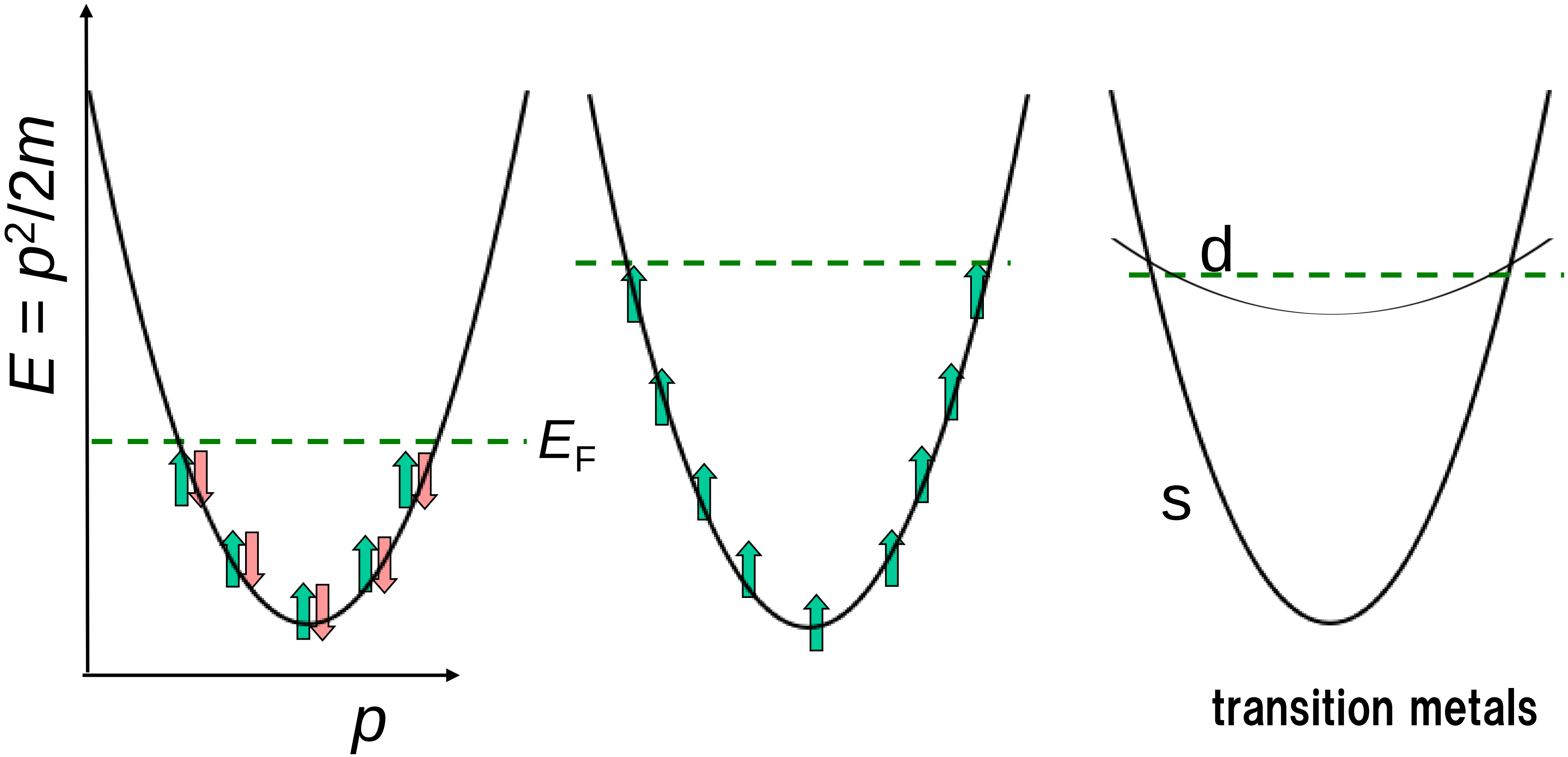
(a) Can we realise the "flat-band ferromagnetism"?

* What is the flat-band ferromagnetism
* A design for a metal organic framework (MOF) predicts we can just realise it.

(b) Can we even make the ferromagnetic flat-band topological ?

Yes, for a kagome MOF designed here, SOC can realise a ferromagnetic and topological (nearly) flat-band

Itinerent ferromagnetism ← very difficult to realise



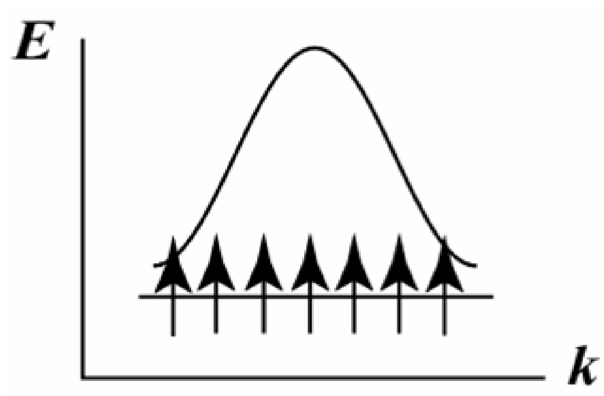
Criterion (Stoner's) for band F:

$UD(E_F) > 1$ --- too crude

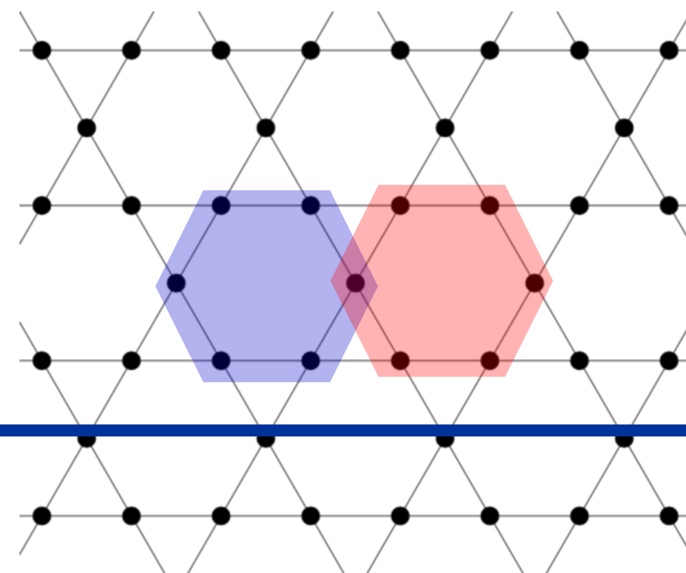
Flat-band ferromagnetism

(Lieb 1989; Mielke 1991; Tasaki 1992)

(a) Flat one-electron band

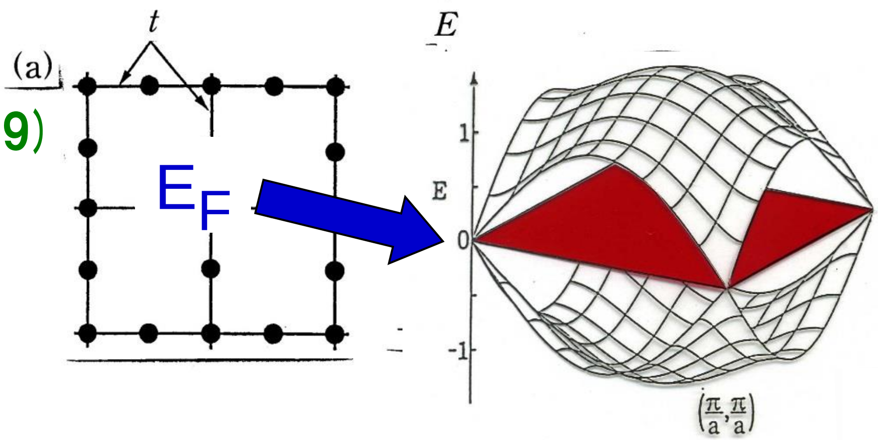


(b) Connectivity condition
(unusual ``Wannier'' orbits)
i.e., totally different from $t \to 0$



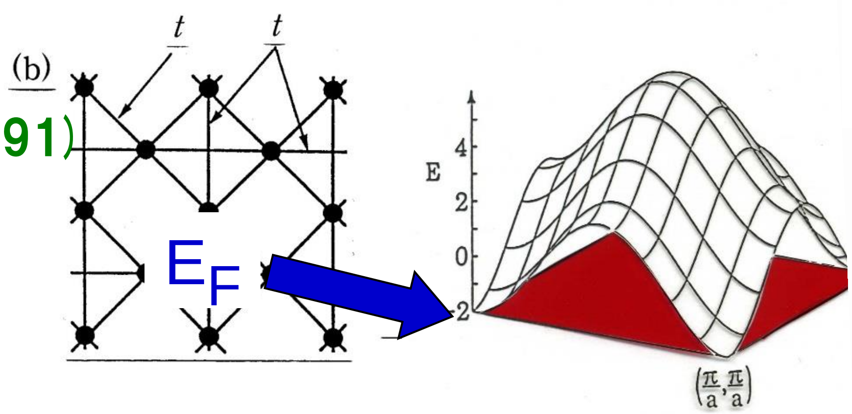
Hubbard model on flat-band systems

(Lieb, 1989)

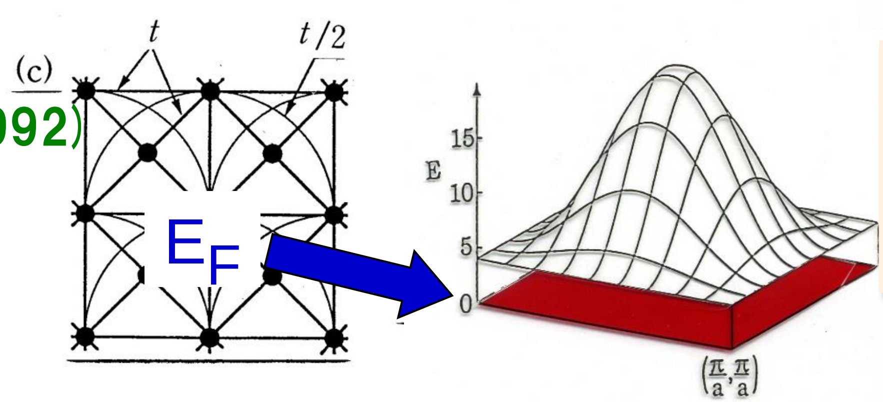


(Mielke, 1991)

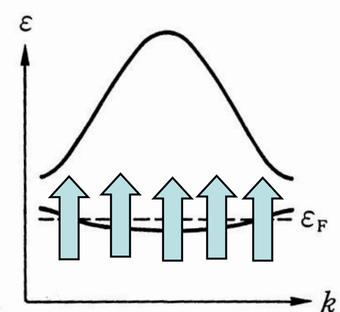
∋ Kagome



(Tasaki, 1992)



Ferromagnetism can be shown rigorously for U>0



Generalised Hund's coupling from the unusual "Wannier orbits"
(Kusakabe & Aoki, J Phys Soc Jpn 1992)

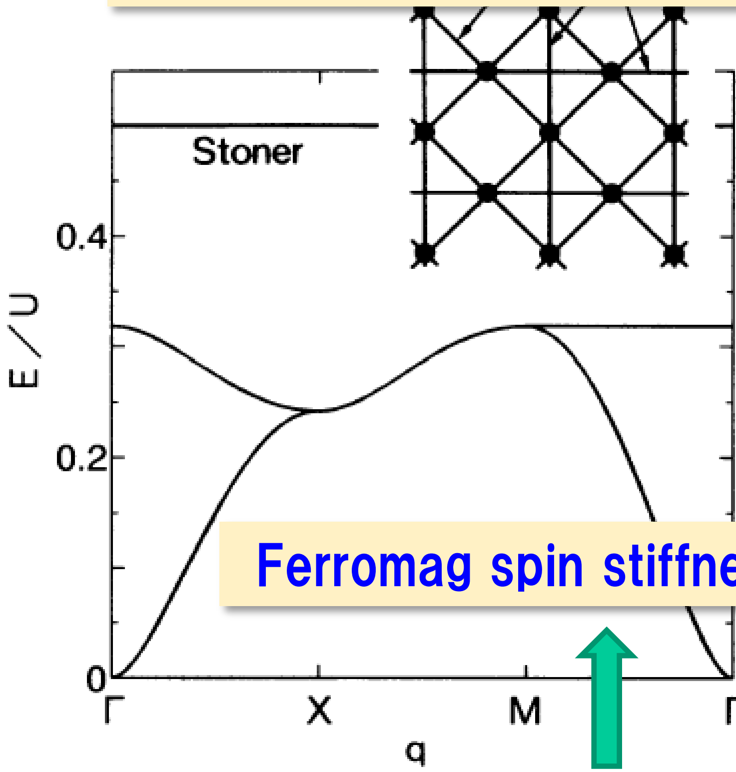
An indication that the flat band ≠ atomic lim

Ferromag on a flat band → magnon dispersion also flat? (ie spin stiffness = 0) ?

*** NO ! *** (Kusakabe & Aoki, PRL 1994)

Spin stiffness = finite for both of

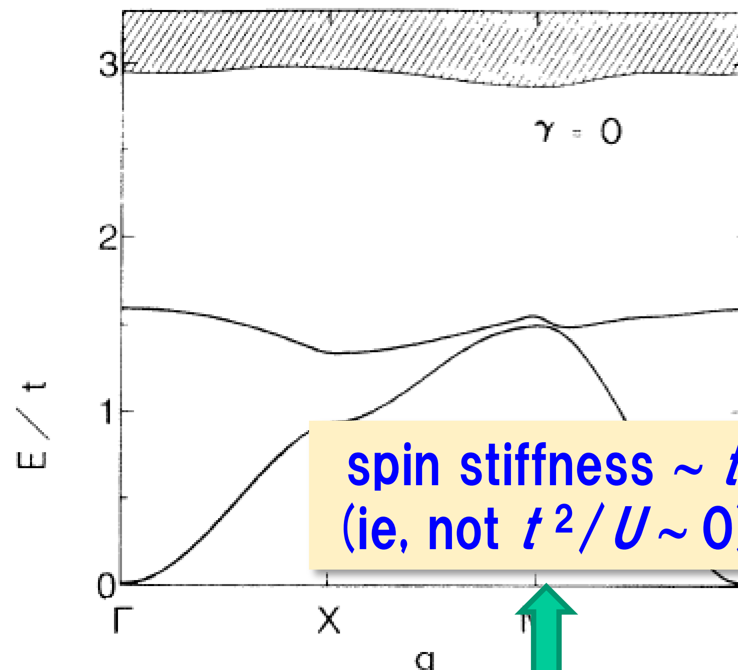
Weak coupling ($U \ll t$)



Ferromag spin stiffness ~

Connectivity condition
(unusual ``Wannier'' orbits)

Strong coupling ($U \gg t$)

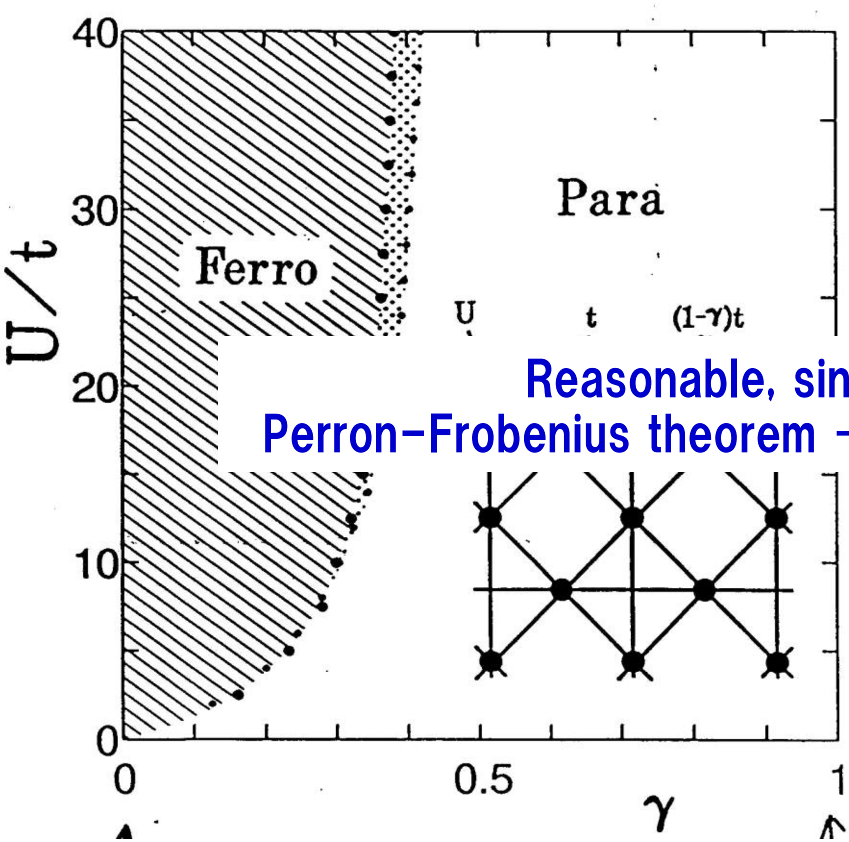


spin stiffness ~ t
(ie, not $t^2/U \sim 0$)

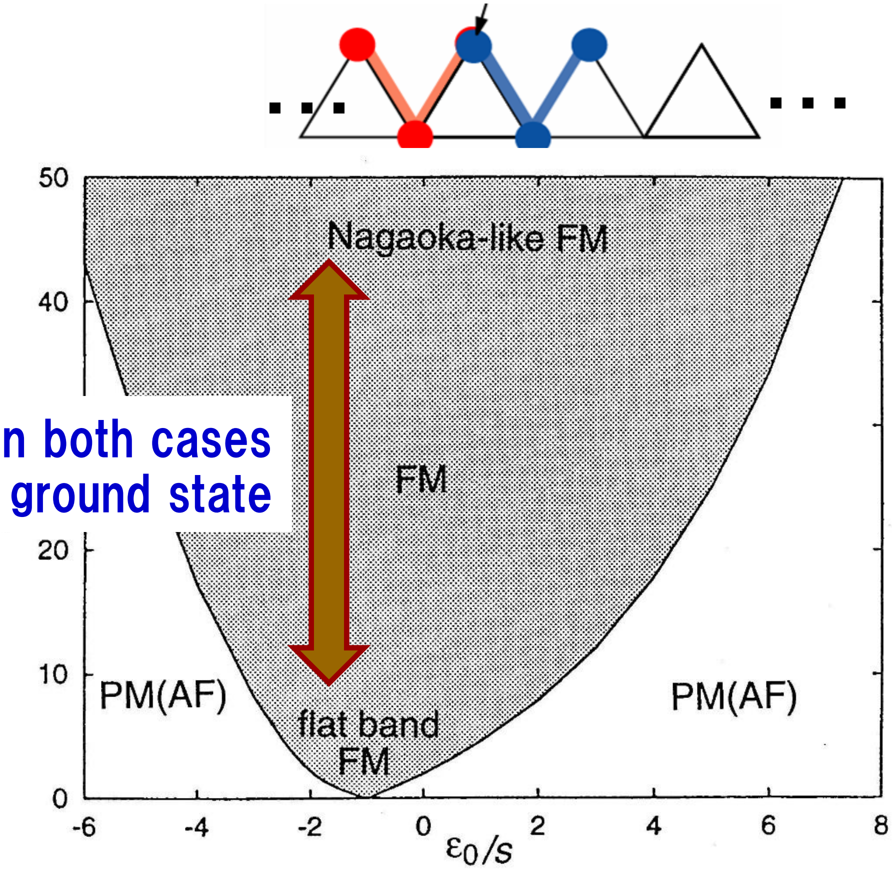
Correlation effect in the
multi-band system

Phase diagram for warped flat bands

Kusakabe & Aoki 1994

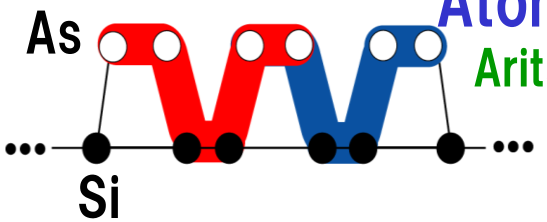


Penc, Shiba et al 1996



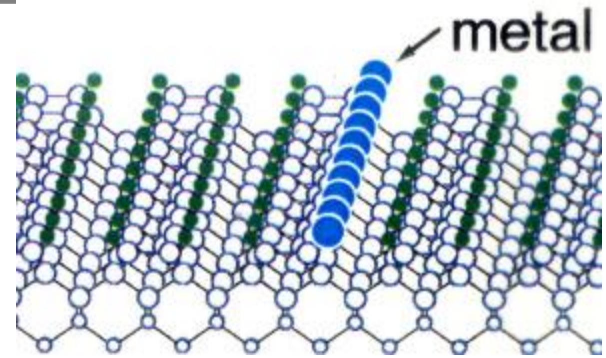
Reasonable, since in both cases
Perron-Frobenius theorem → F ground state

Realisations: 1D polymers



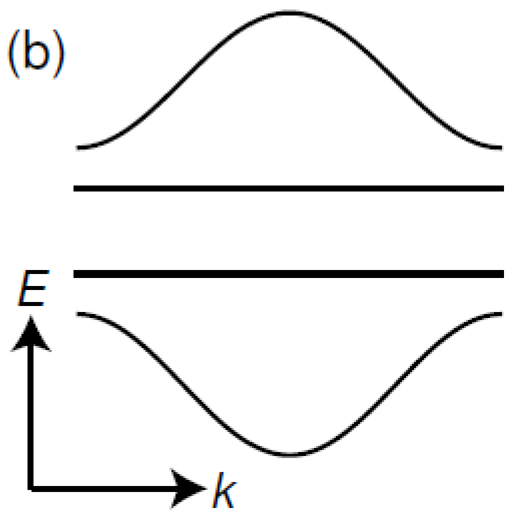
Atomic quantum wire

Arita et al, PRB (1998)



As adatoms on Si

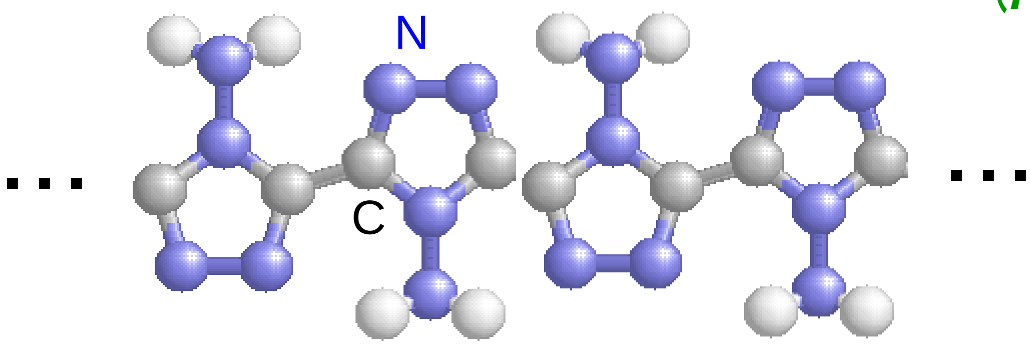
(Hashizume et al, Jpn J Appl Phys1996)



(See Gulacsi, ..., Vollhardt,
PRL 2007 for diamond chain)

Design of a ferromagnetic polymer

(Arita et al, PRL 2002; PRB 2003)

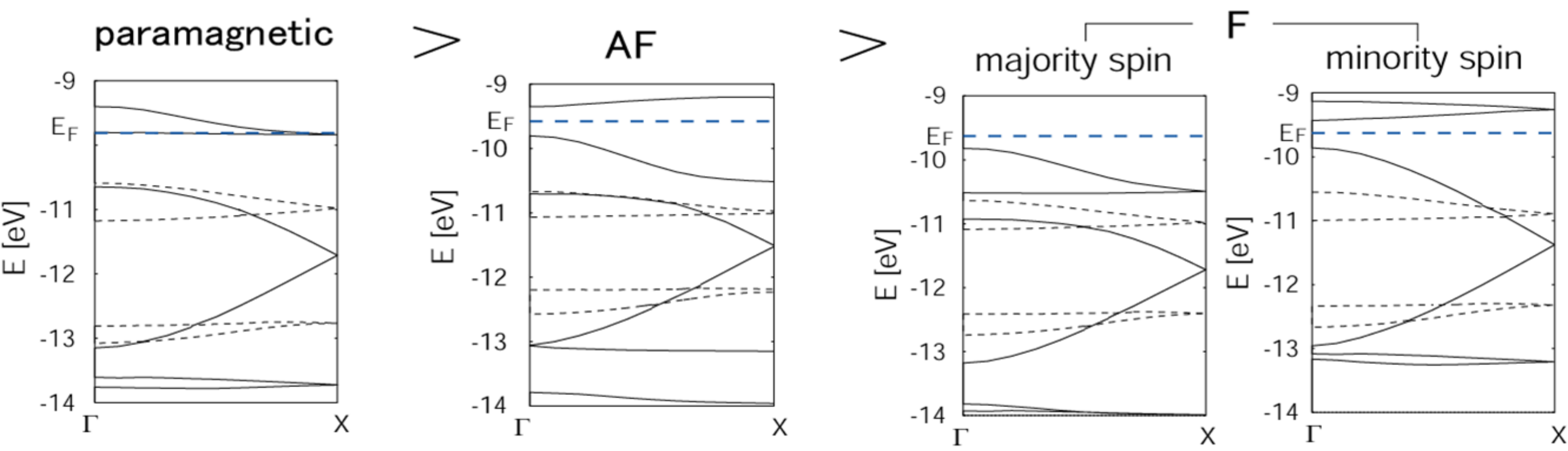


A new material: polyaminotriazole

Connectivity condition
(``Wannier" orbits overlap)

GGA-SDFT result

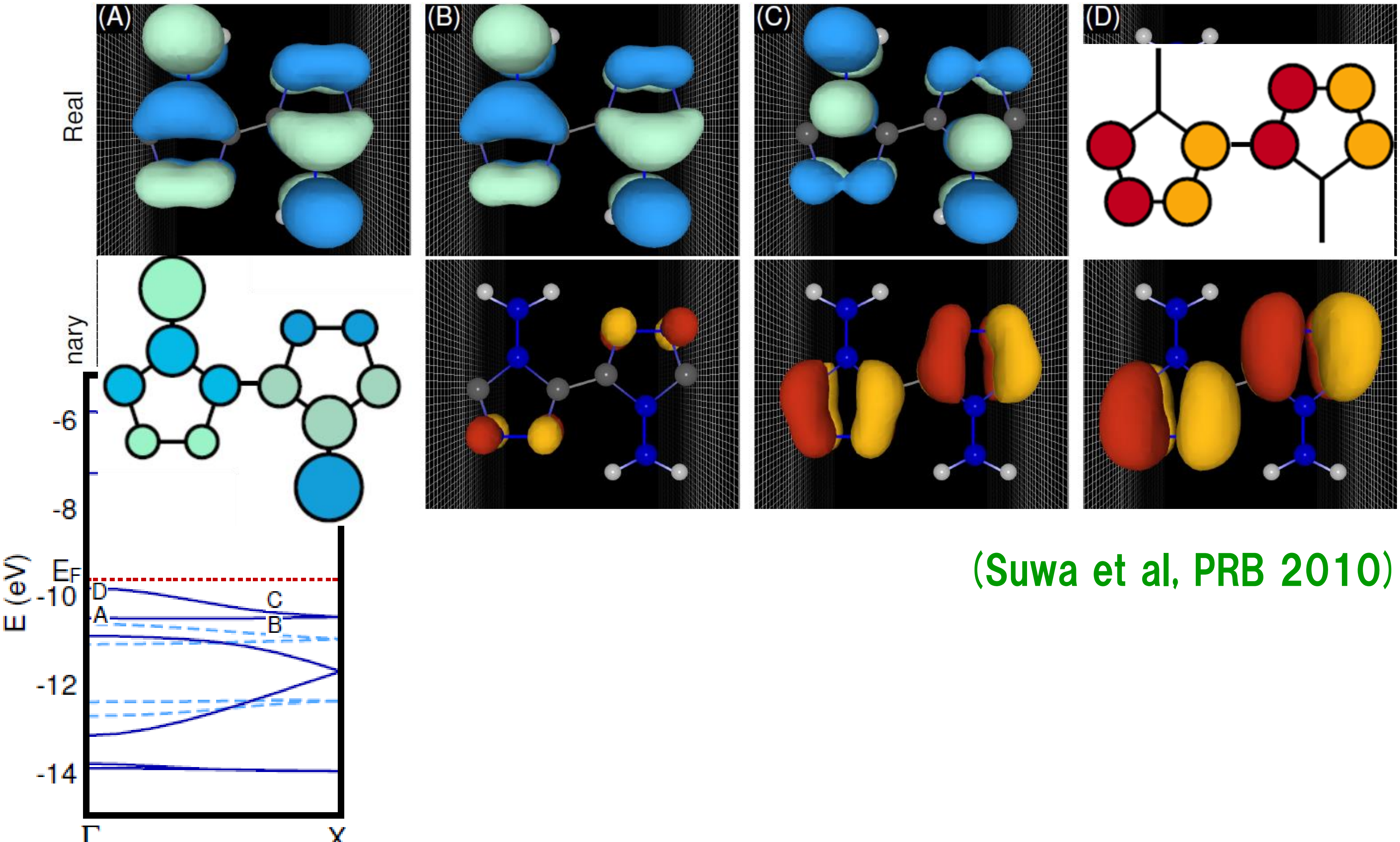
Exchange splitting = O(1) eV



A peculiarity of the flat-band system (≠ atomic lim)

Flat band arises from interference in wf → Bloch wf $\exp(i\mathbf{k}\mathbf{r})u_{\mathbf{k}}(\mathbf{r})$

strongly dep on ***k***



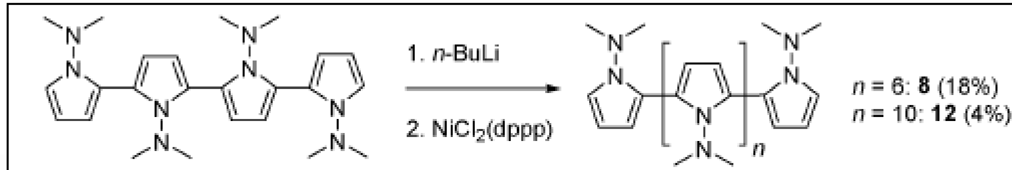
(Suwa et al, PRB 2010)

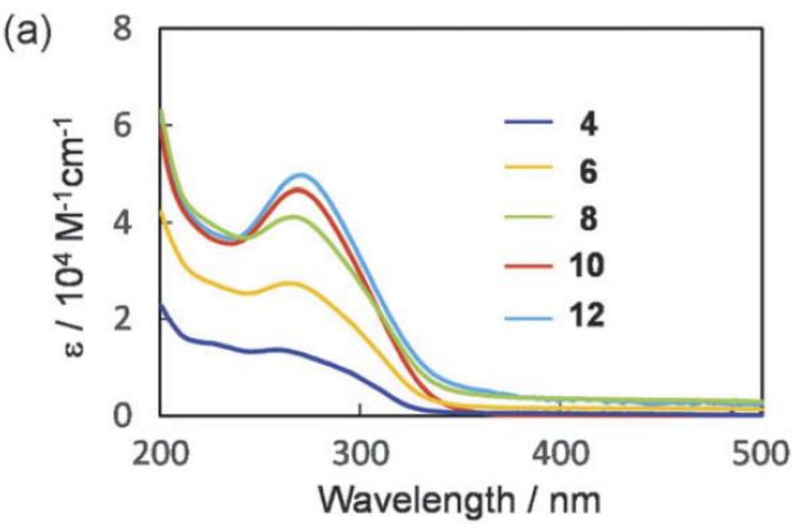
Journal of
Materials Chemistry C
2015

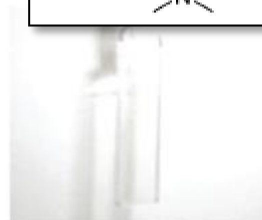
Synthesis, characterization, and physical properties of oligo(1-(*N*,*N*-dimethylamino)pyrrole)s and their doped forms, precursors of candidates for molecular flat-band ferromagnets†

(Nishihara's group, 2015)

Yoshinori Yamanoi,* Kazuhiro Takahashi, Takeshi Hamada, Norikazu Ohshima, Masashi Kurashina, Yohei Hattori, Tetsuro Kusamoto, Ryota Sakamoto, Mariko Miyachi and Hiroshi Nishihara*

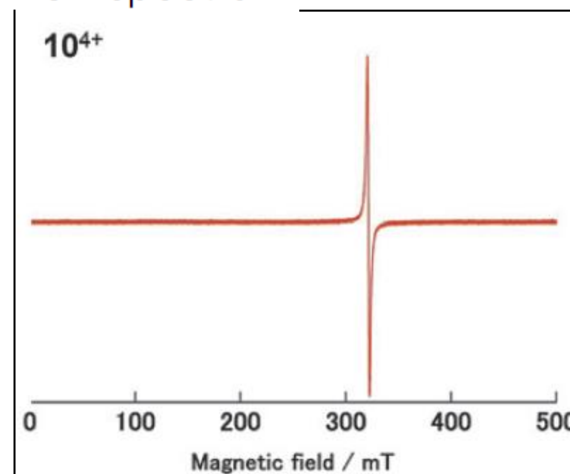




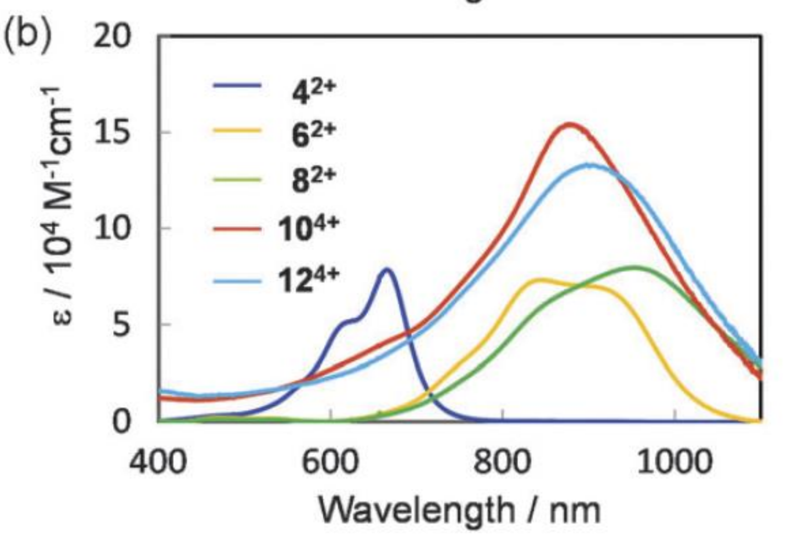


4
(λ_{max} = 280 nm)

ESR spectrum

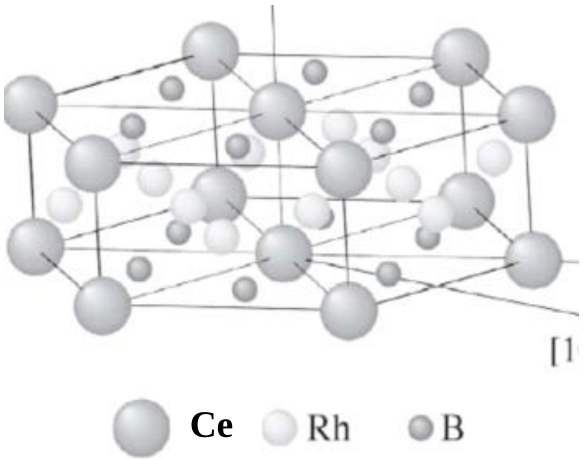


spin density = 0.42/molecule

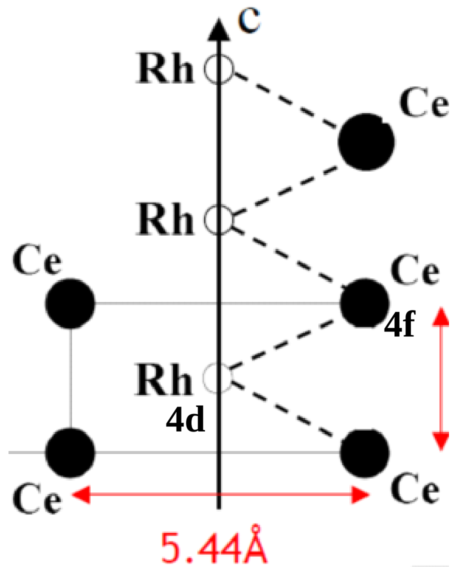


4^{2+}(SbCl$_6^-$)$_2$
(λ_{max} = 660 nm)

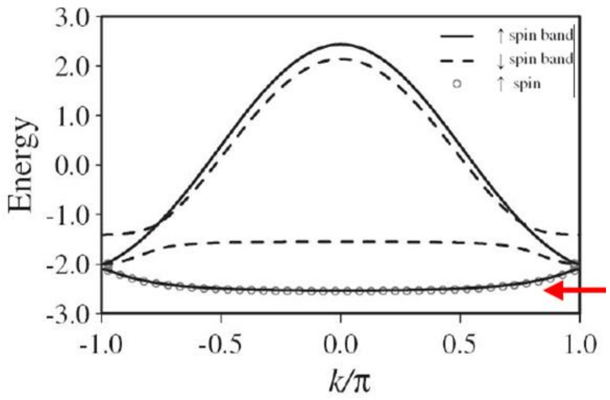
Q-1D ferromagnet $CeRh_3B_2$



(Yamada et al, JPSJ 2004)



(a,c) plane



(Kono & Kuramoto, JPSJ 2006)

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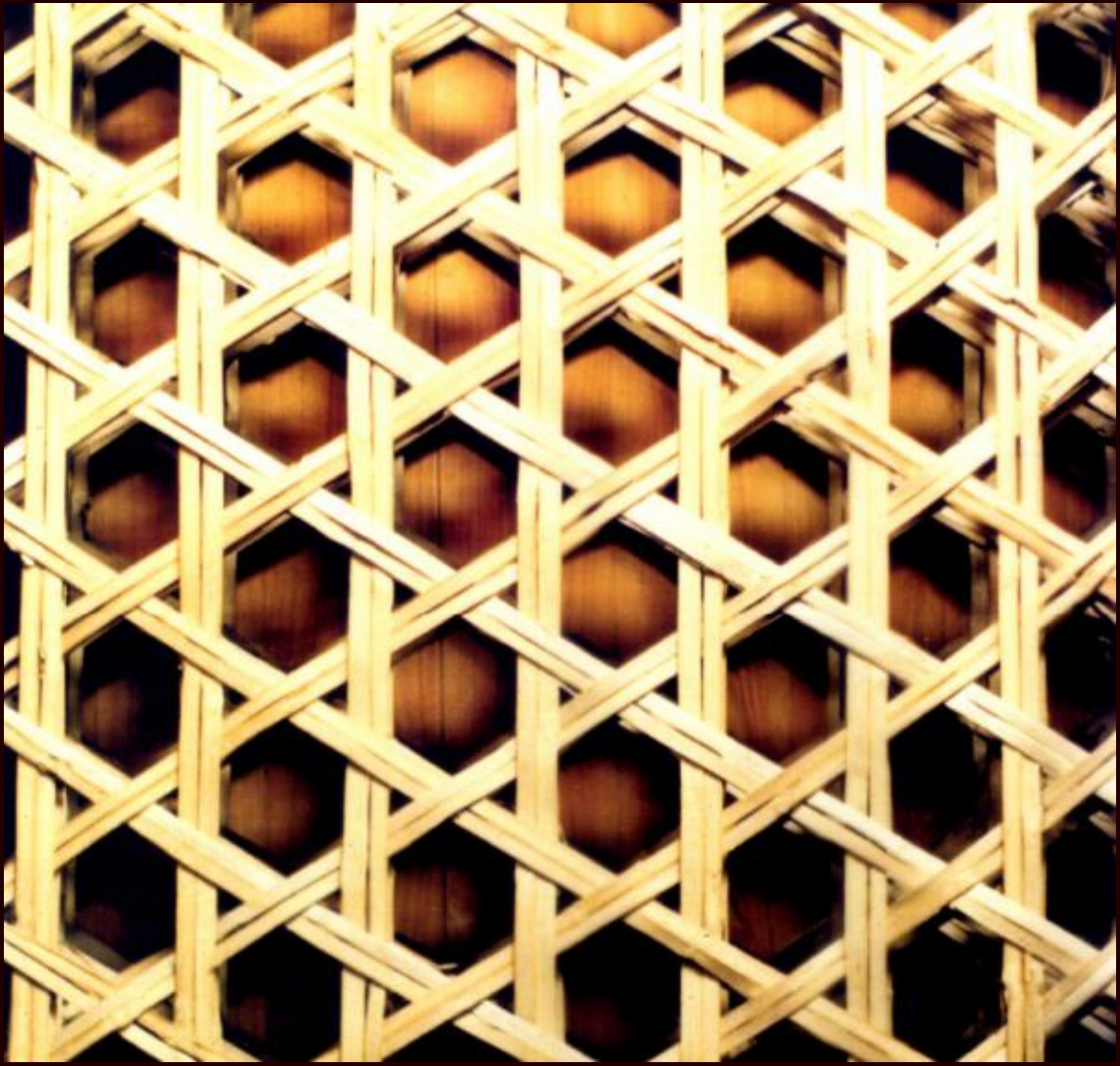
(a) Can we realise the "flat-band ferromagnetism"?

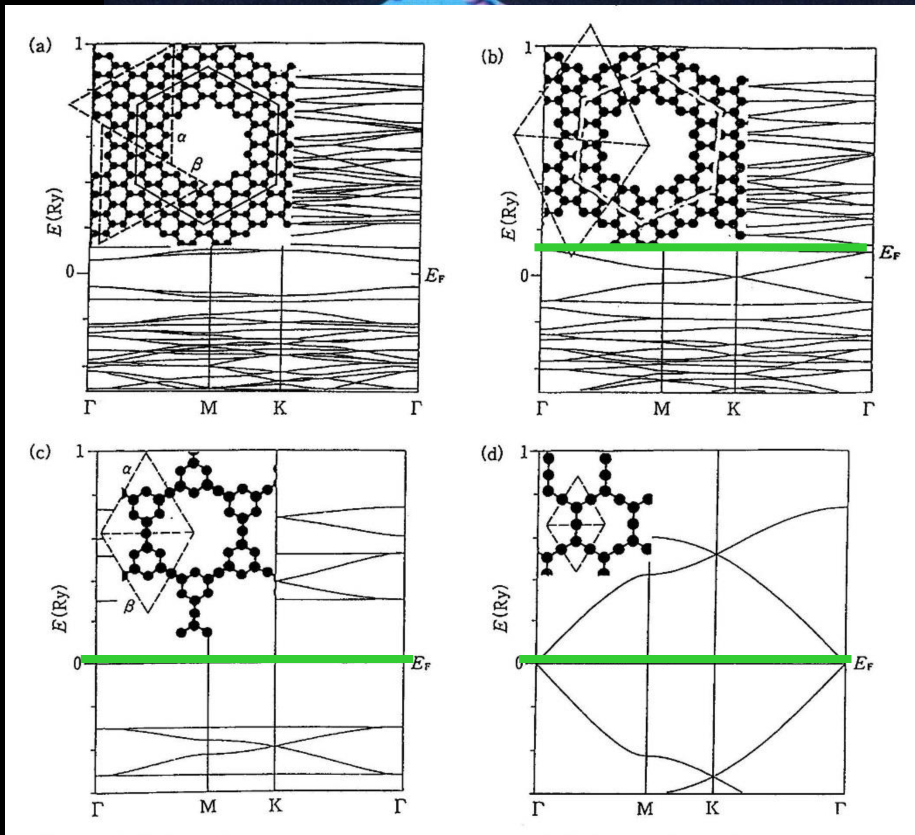
* What is the flat-band ferromagnetism
* A design for a metal organic framework (MOF) predicts we can just do that.

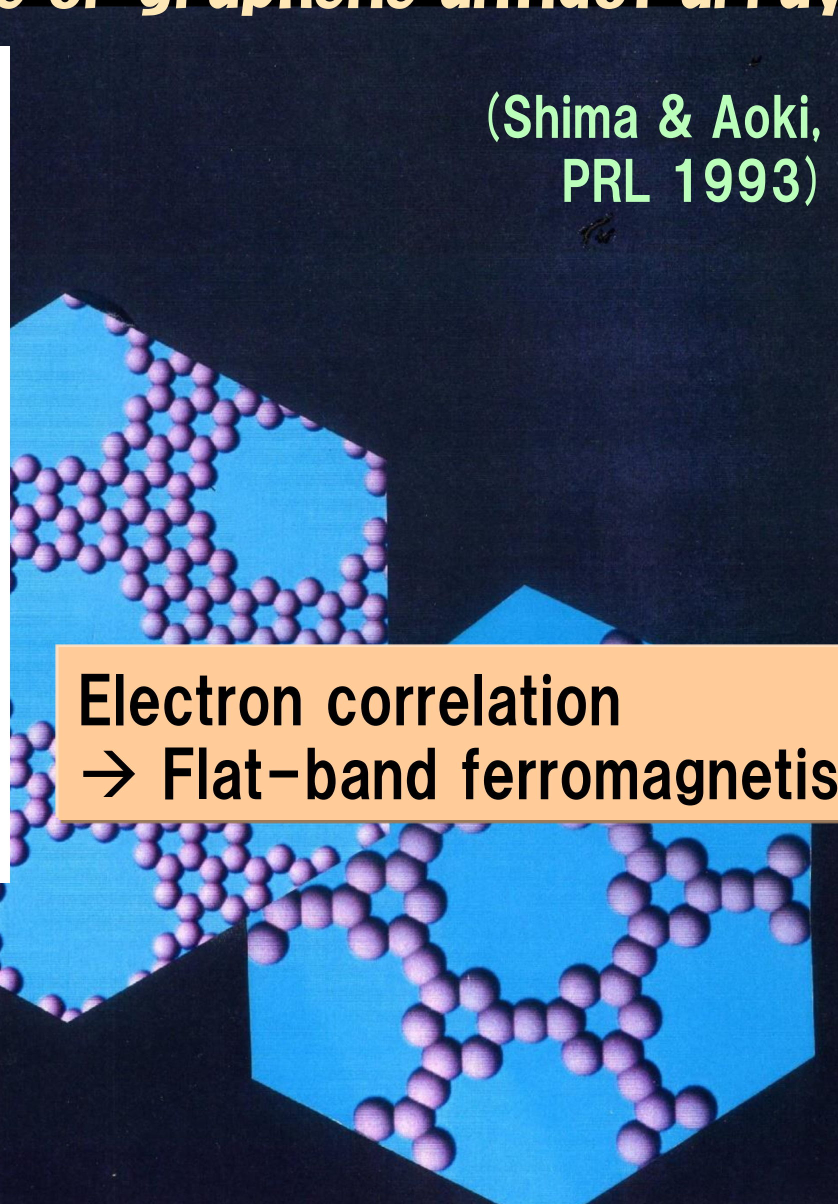
Realisations in 2D?



Ferromagnetism in carbon structures
— long-period graphene or graphene antidot array

(Shima & Aoki, PRL 1993)

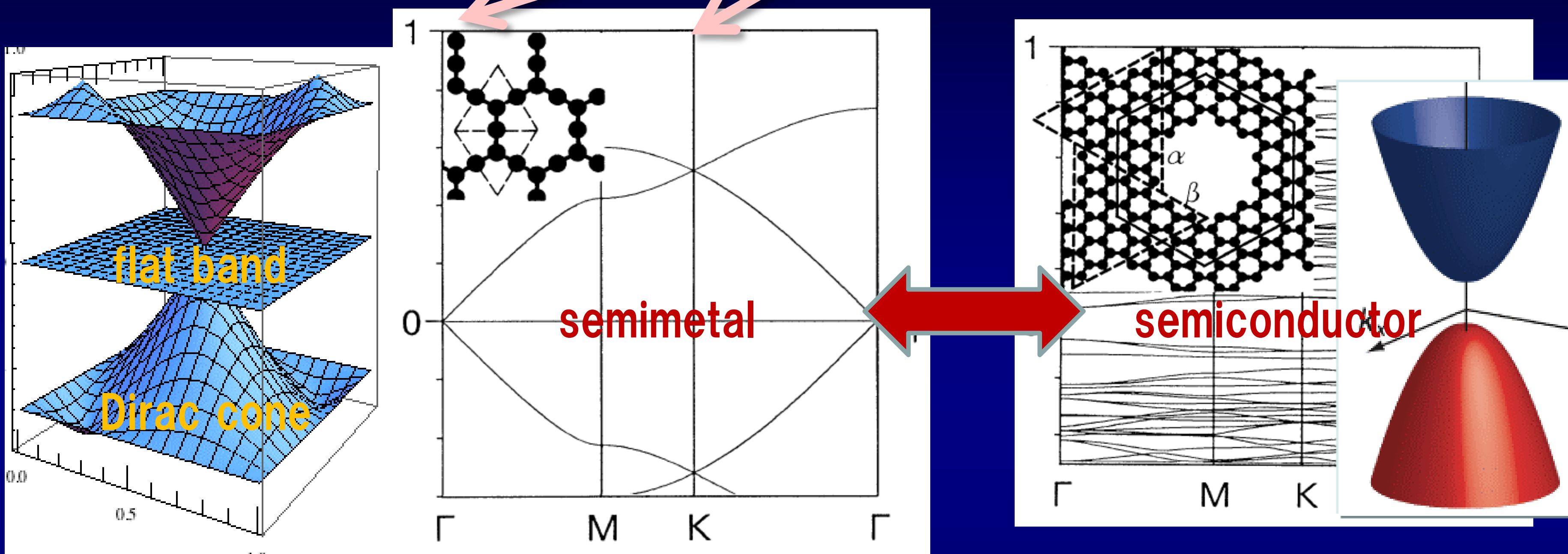




Electron correlation
→ Flat-band ferromagnetism

Long-period graphene --- Γ-K manipulation

(Shima & Aoki, PRL 1993)



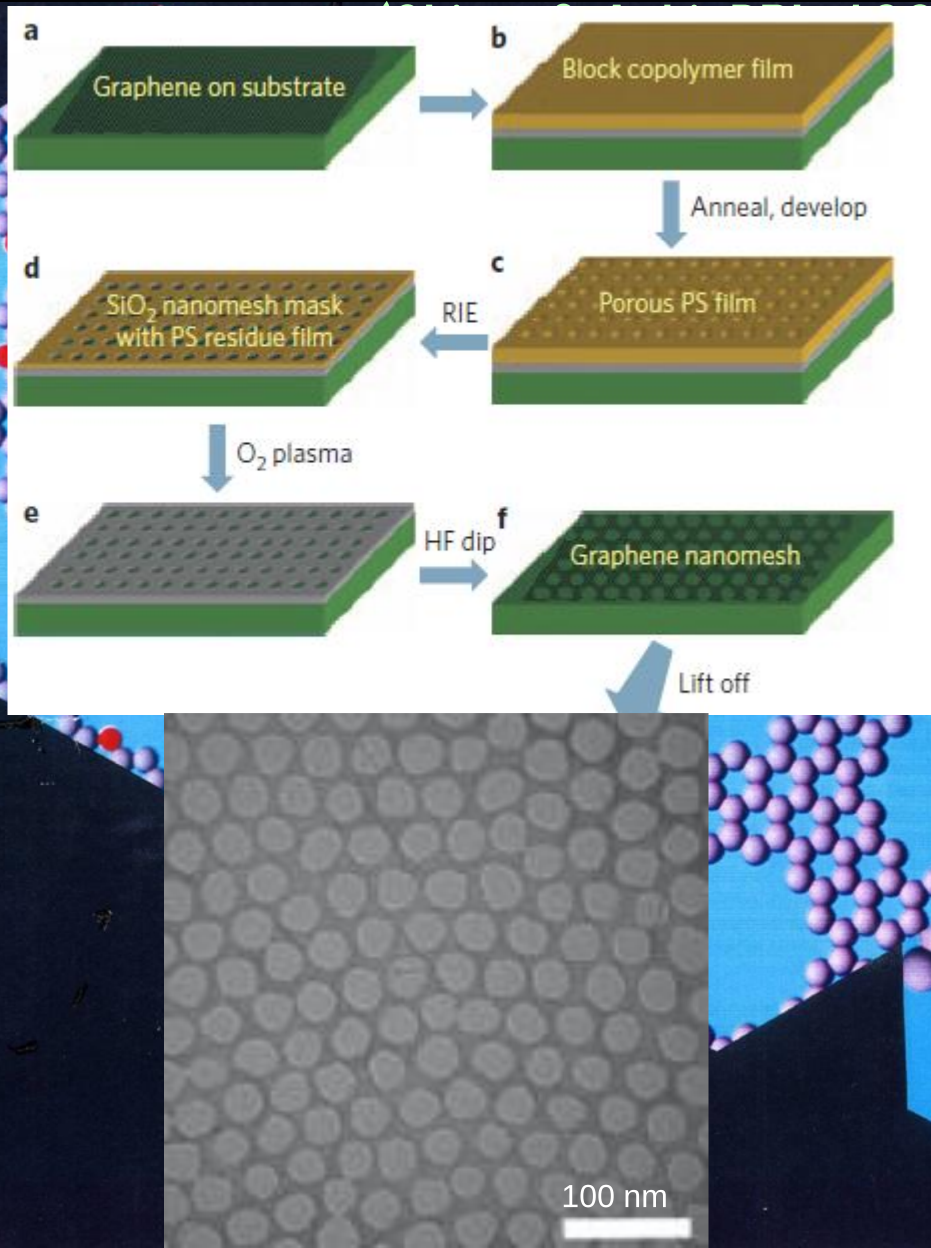
Type	Formula unit	Γ	K	Bipartite
A_0	$(C_{3m})_2$			sc + $n(\geq 0)$ flat band(s)
A_C	$(C_{3m+1})_2$		E	sm + $n(\geq 0)$ flat band(s)
B_0	$(C_{3m+3/2})_2$	A, E	A, E	sc + $n(\geq 3)$ flat bands
B_C	$(C_{3m+5/2})_2$	A, E	A	sm + $n(\geq 1)$ flat band(s)

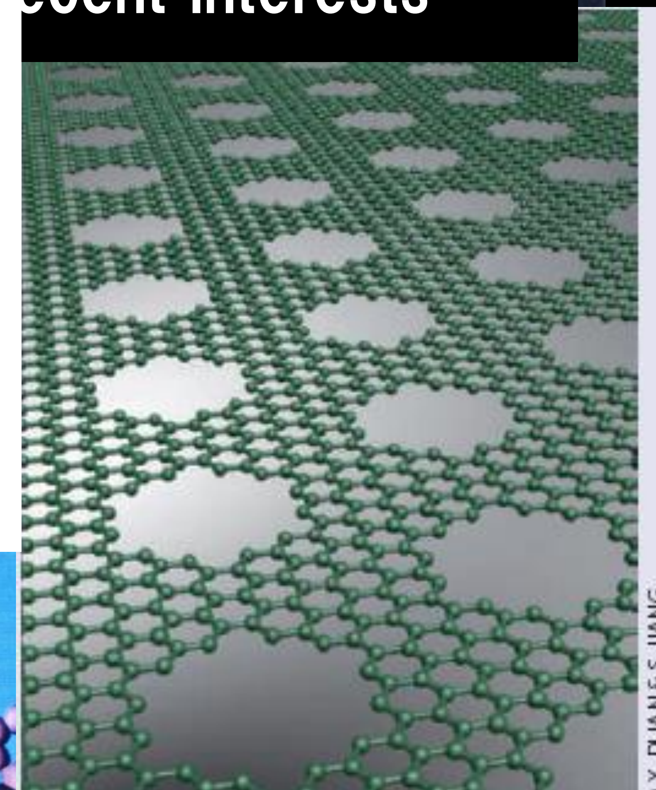
1D irred rep
2D irred rep

Graphene nanomesh

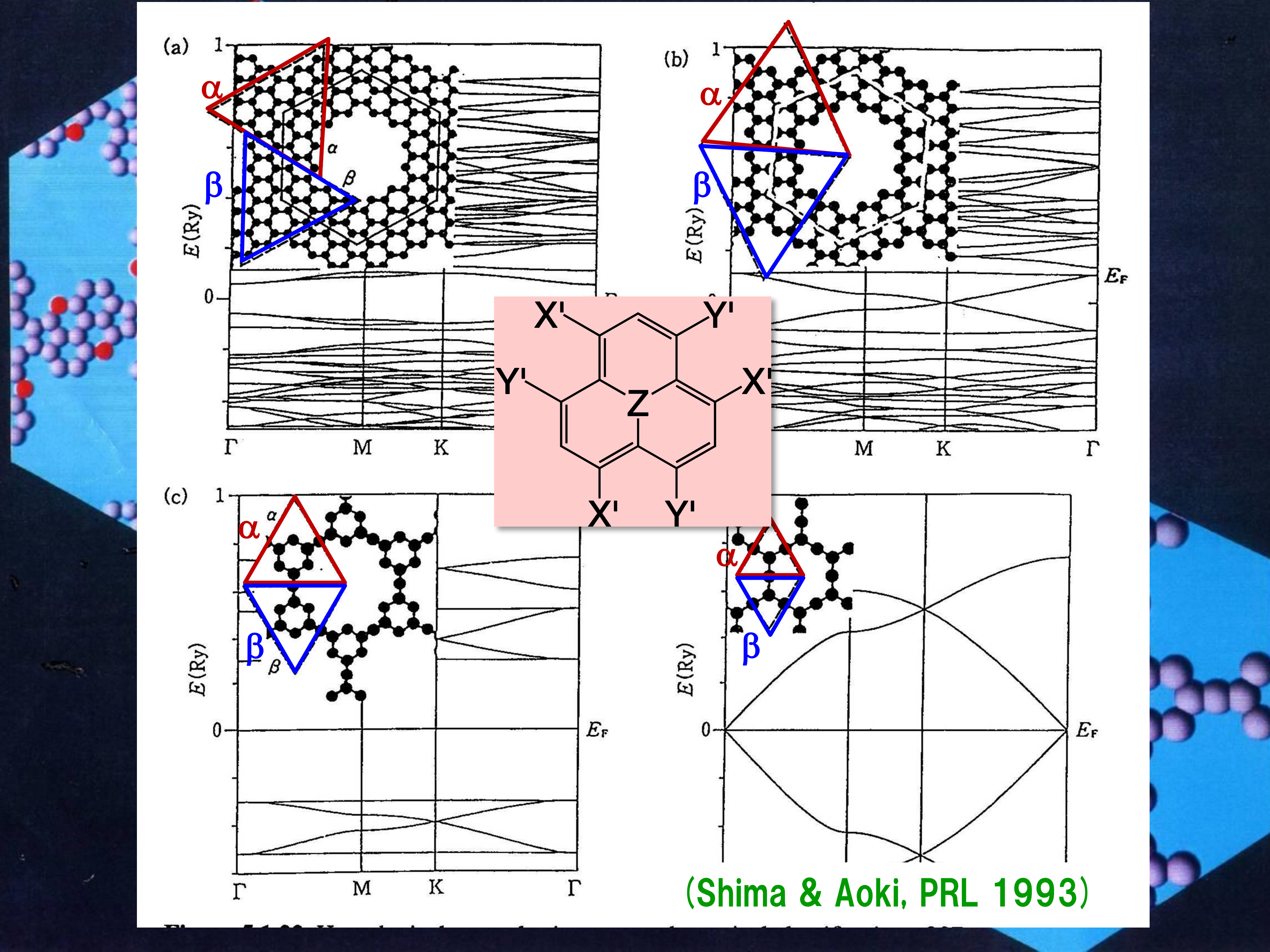
3)

ecent interests



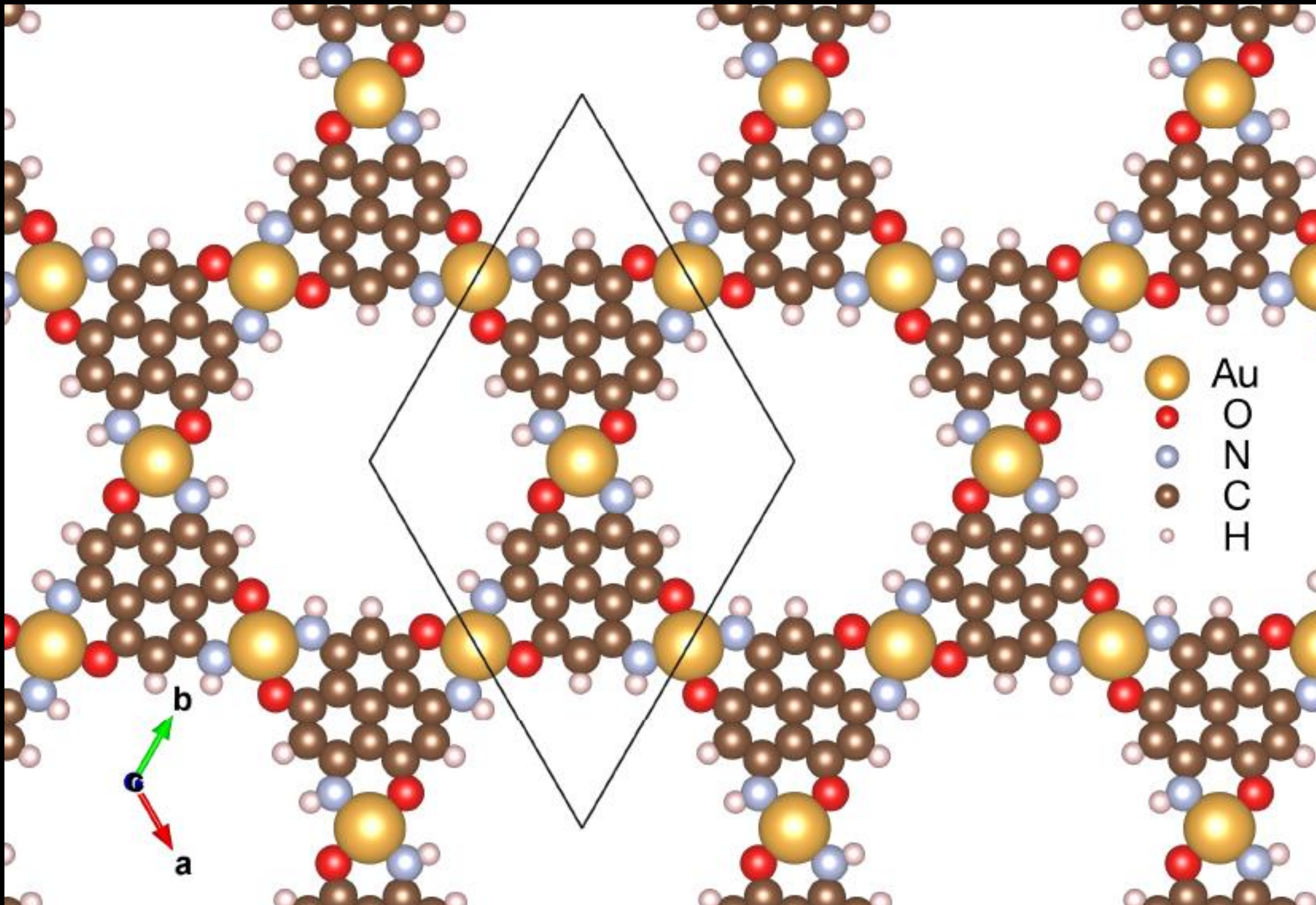


Graphene nanomesh → diodes
(Bai et al, Nature Nanotech 2010)



(a)
(b)
(c)
1
0
E(Ry)
α
β
Γ
M
K
E_F
X'
Y'
Z
(Shima & Aoki, PRL 1993)

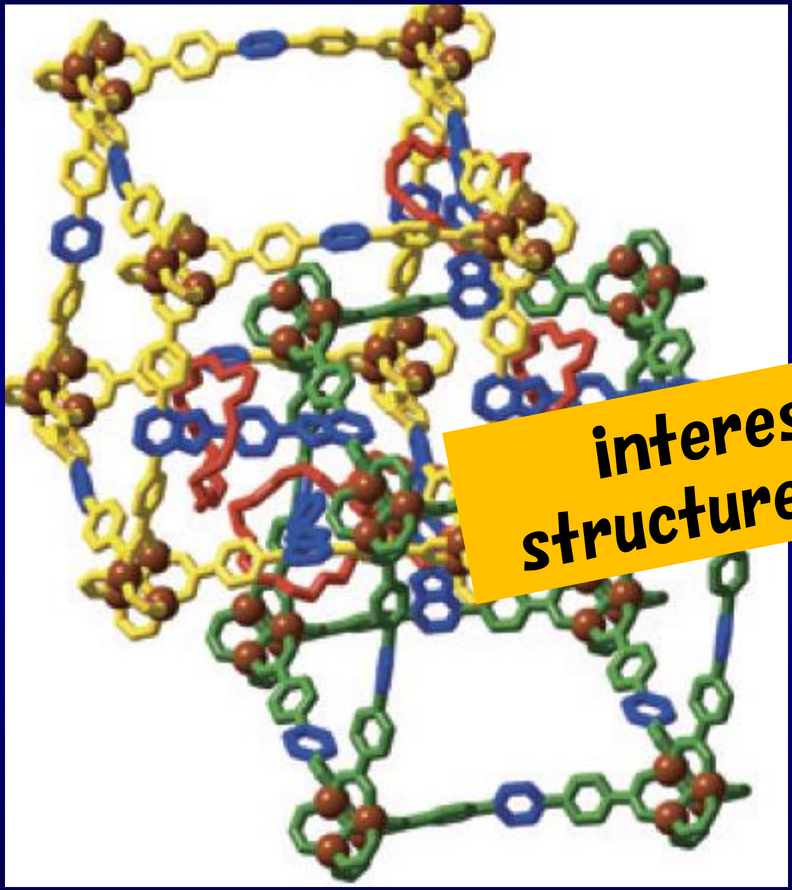
"Organic ferromagnetic topological" MOF systems
(Yamada et al, arXiv:1510.00164, a collaboration with MIT)



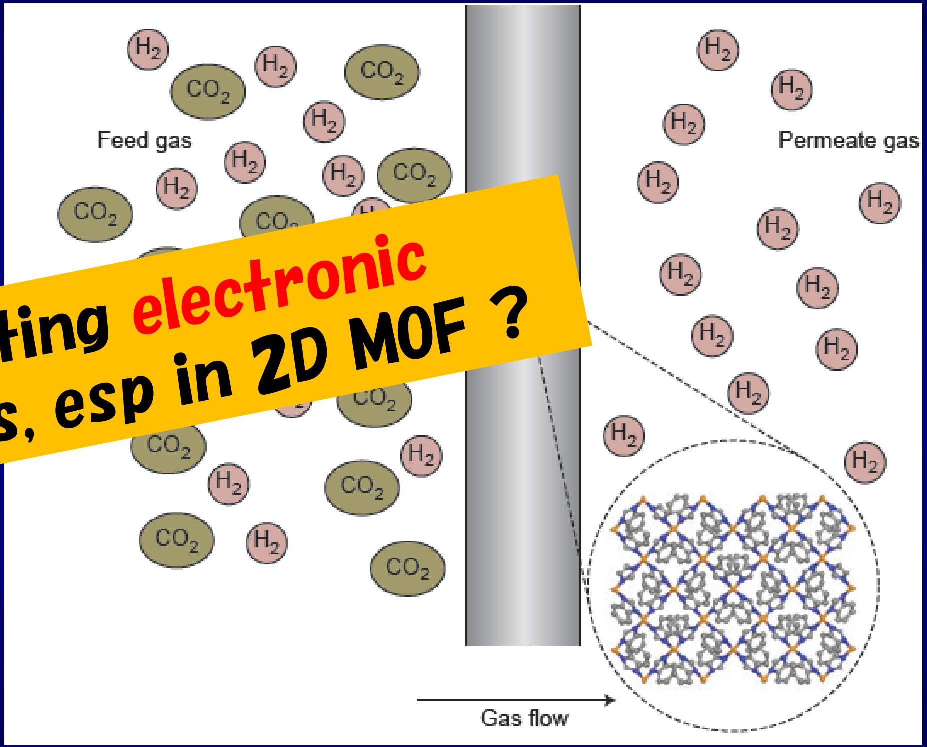
MOF
(Metal-organic frameworks)

What is MOF?

3D MOF
(Olson, nature chem 2015)



(Nenoff, nature chemistry 2015)



interesting electronic structures, esp in 2D MOF ?

JACS JOURNAL OF THE AMERICAN CHEMICAL SOCIETY
J. Am. Chem. Soc. 2013, 135, 2462–2465
Communication
pubs.acs.org/JACS

Nishihara's group (2013)

π-Conjugated Nickel Bis(dithiolene) Complex Nanosheet

Tetsuya Kambe,† Ryota Sakamoto,† Ken Hoshiko,† Kenji Takada,† Mariko Miyachi,† Ji-Heun Ryu,‡ Sono Sasaki,§,∥ Jungeun Kim,∥ Kazuo Nakazato,‡ Masaki Takata,∥,⊥ and Hiroshi Nishihara*,†

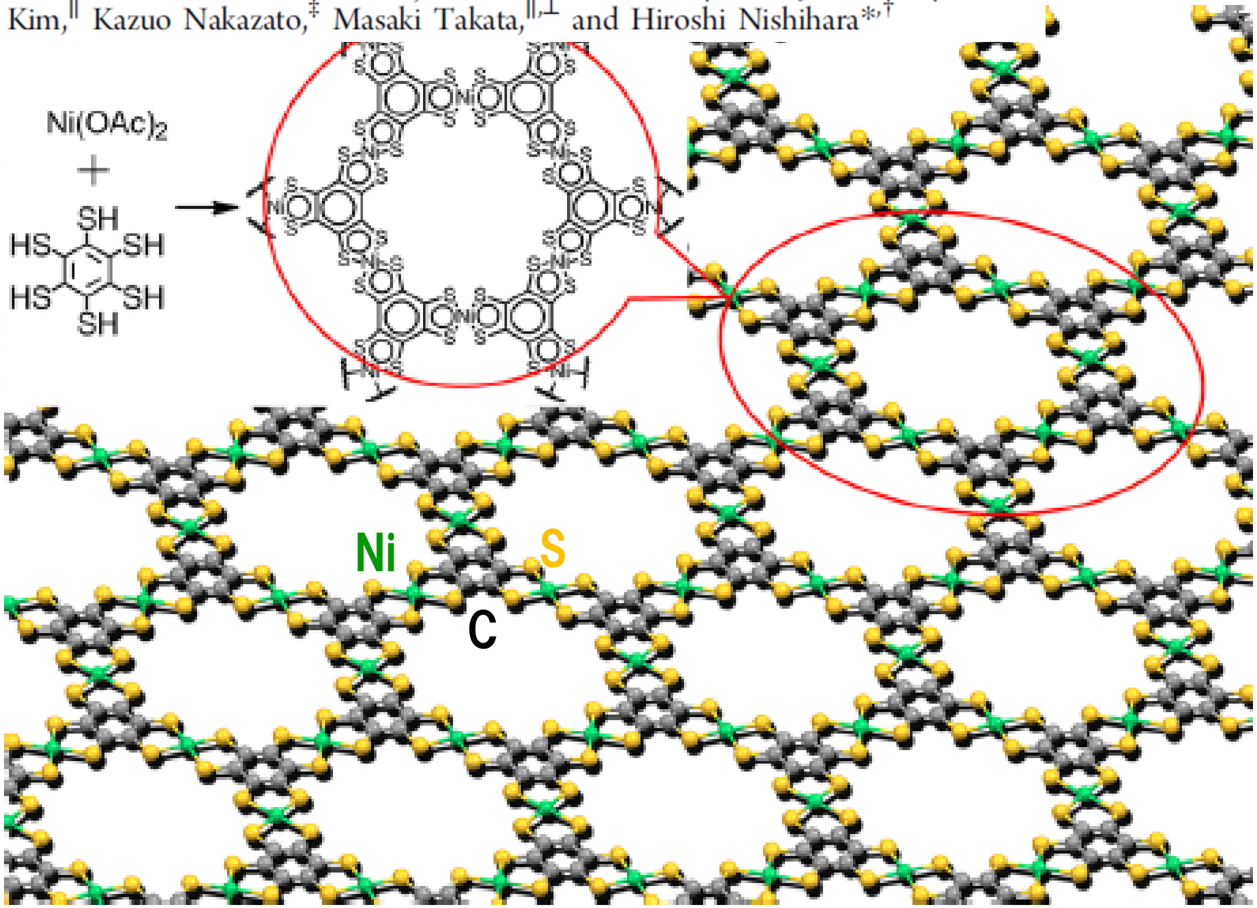


Figure 1. Schematic illustration and chemical structure of monolayer nickel bis(dithiolene) complex nanosheet 1. Countercations have been omitted for clarity. Gray, C; yellow, S; green, Ni.

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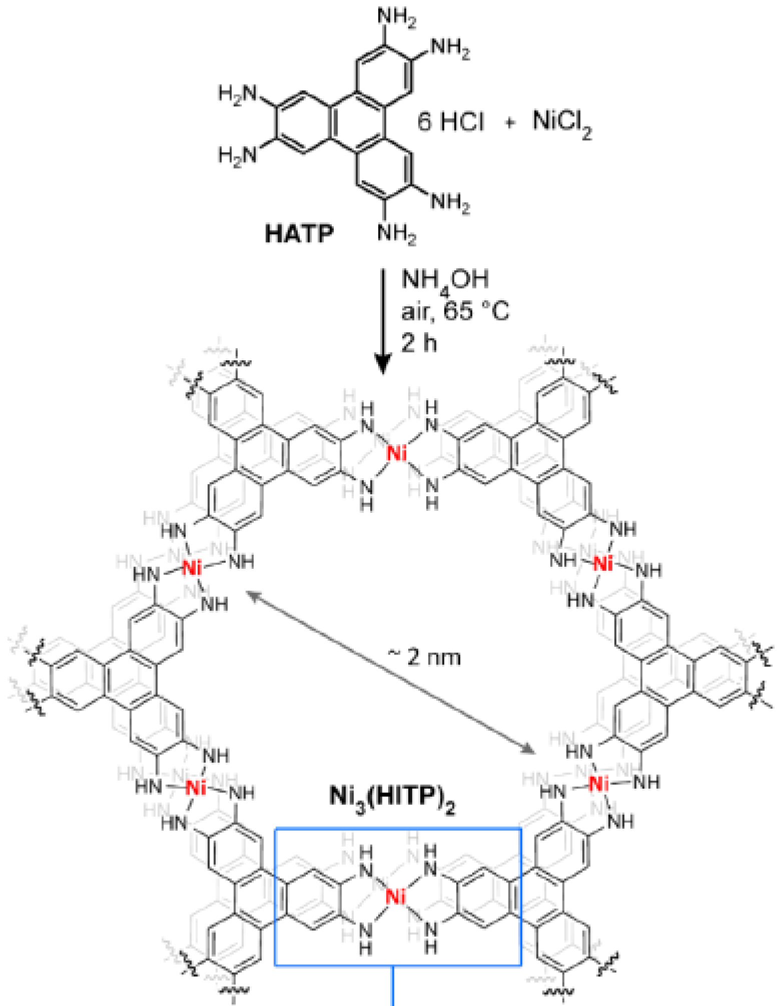
Communication
pubs.acs.org/JACS

Dinca’ s group (2014)

High Electrical Conductivity in Ni_3(2,3,6,7,10,11-hexaiminotriphenylene)$_2$, a Semiconducting Metal–Organic Graphene Analogue

Dennis Sheberla,† Lei Sun,† Martin A. Blood-Forsythe,‡ Süleyman Er,‡ Casey R. Wade,† Carl K. Brozek,† Alán Aspuru-Guzik,‡ and Mircea Dincă*,†

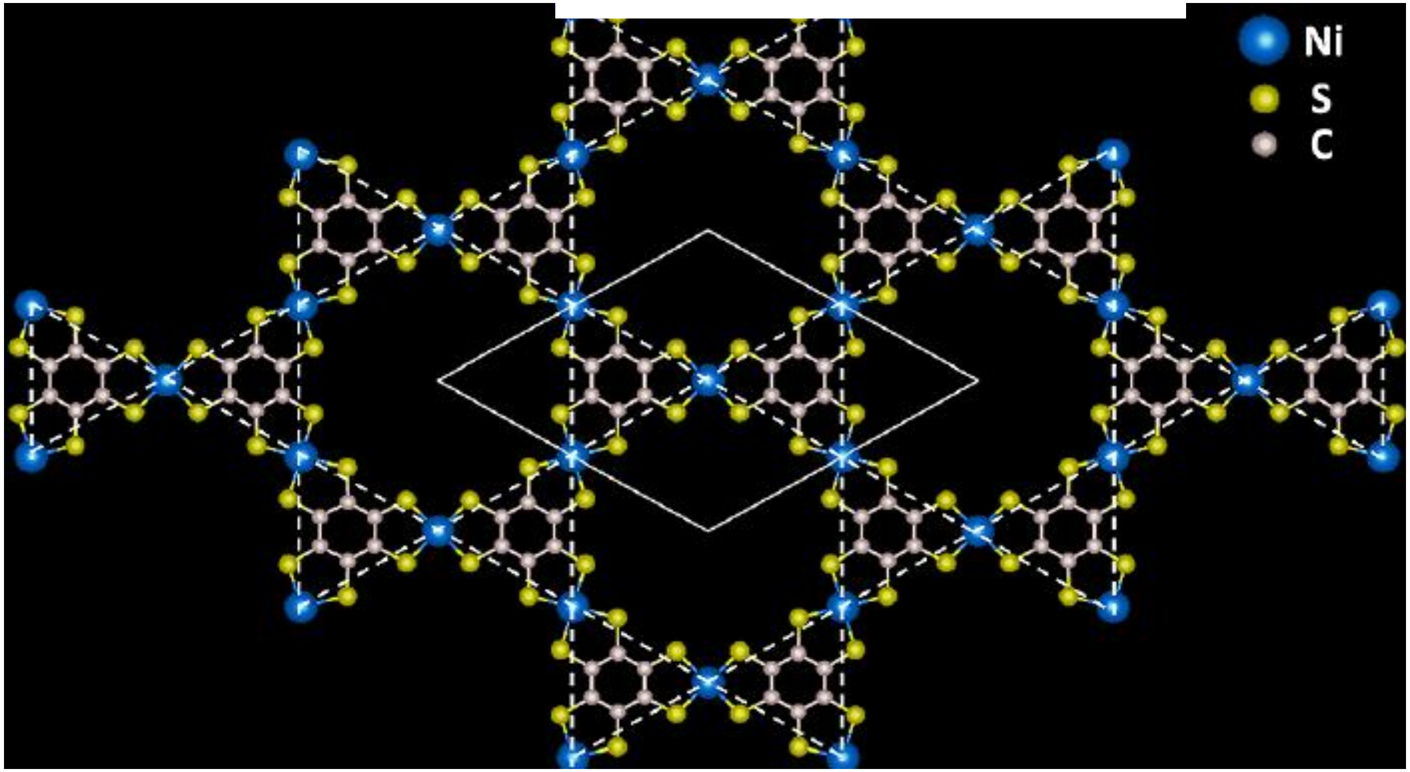


NANO LETTERS
Letter
pubs.acs.org/NanoLe

Prediction of a Two-Dimensional Organic Topological Insulator

Z. F. Wang, Ninghai Su, and Feng Liu*

Nano Lett. 2013, 13, 2842–2845

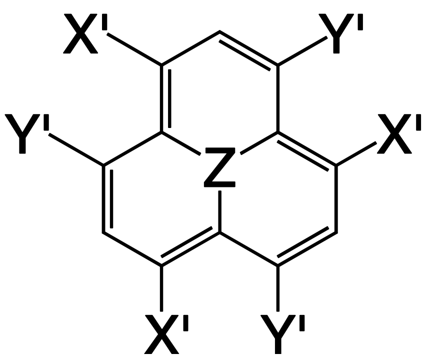


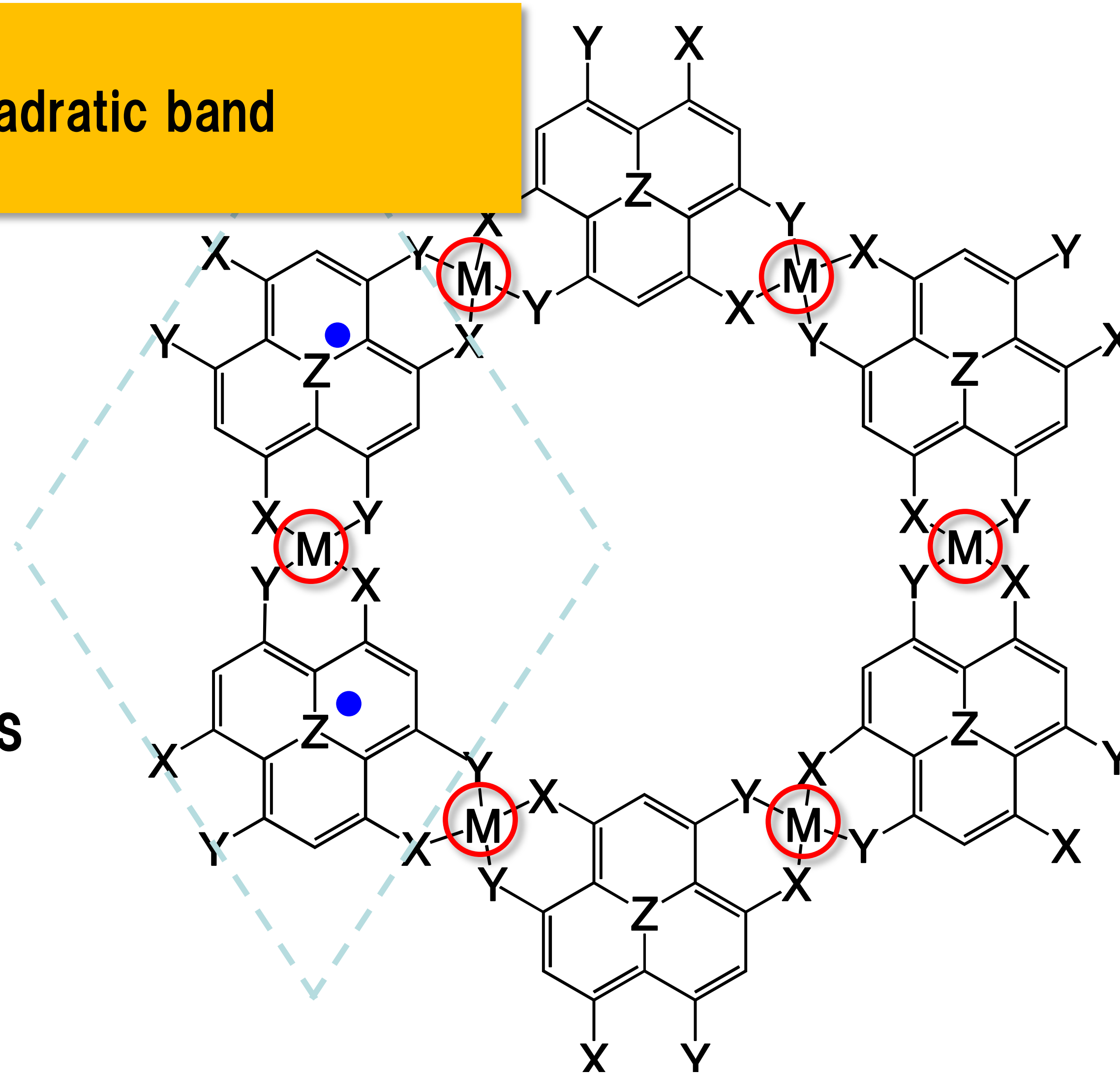
(see also Zhao et al, Nanoscale 2013;
Zhao et al, PRB 2014;
Zhou et al, Nanoscale 2015)

Our design of MOF for flat-band F

(Yamada et al, arXiv:1510.00164)

✔ E_F right at the flat band
✔ Flat band touches with a quadratic band
→ topological ?



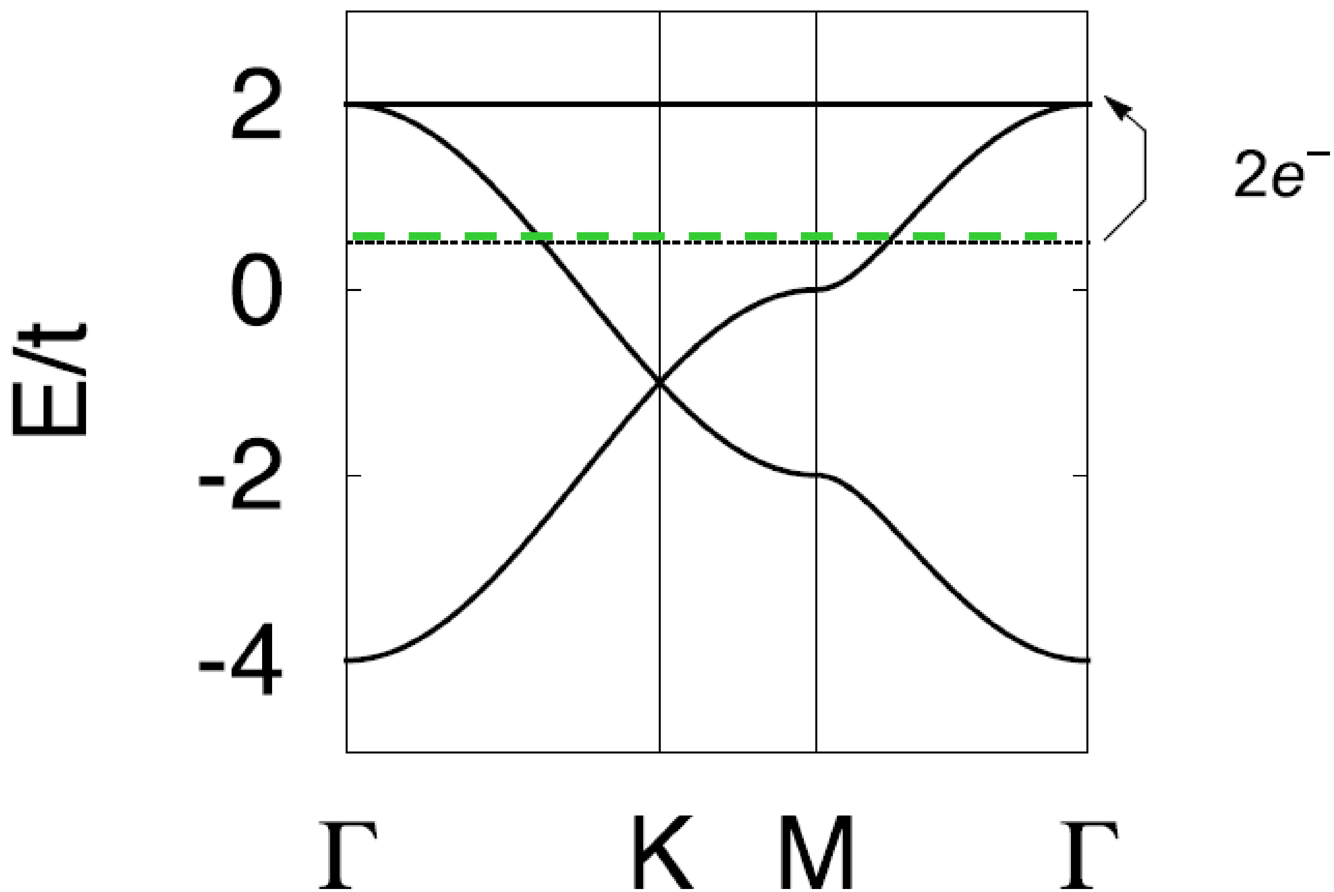


phenalenyl-based ligands

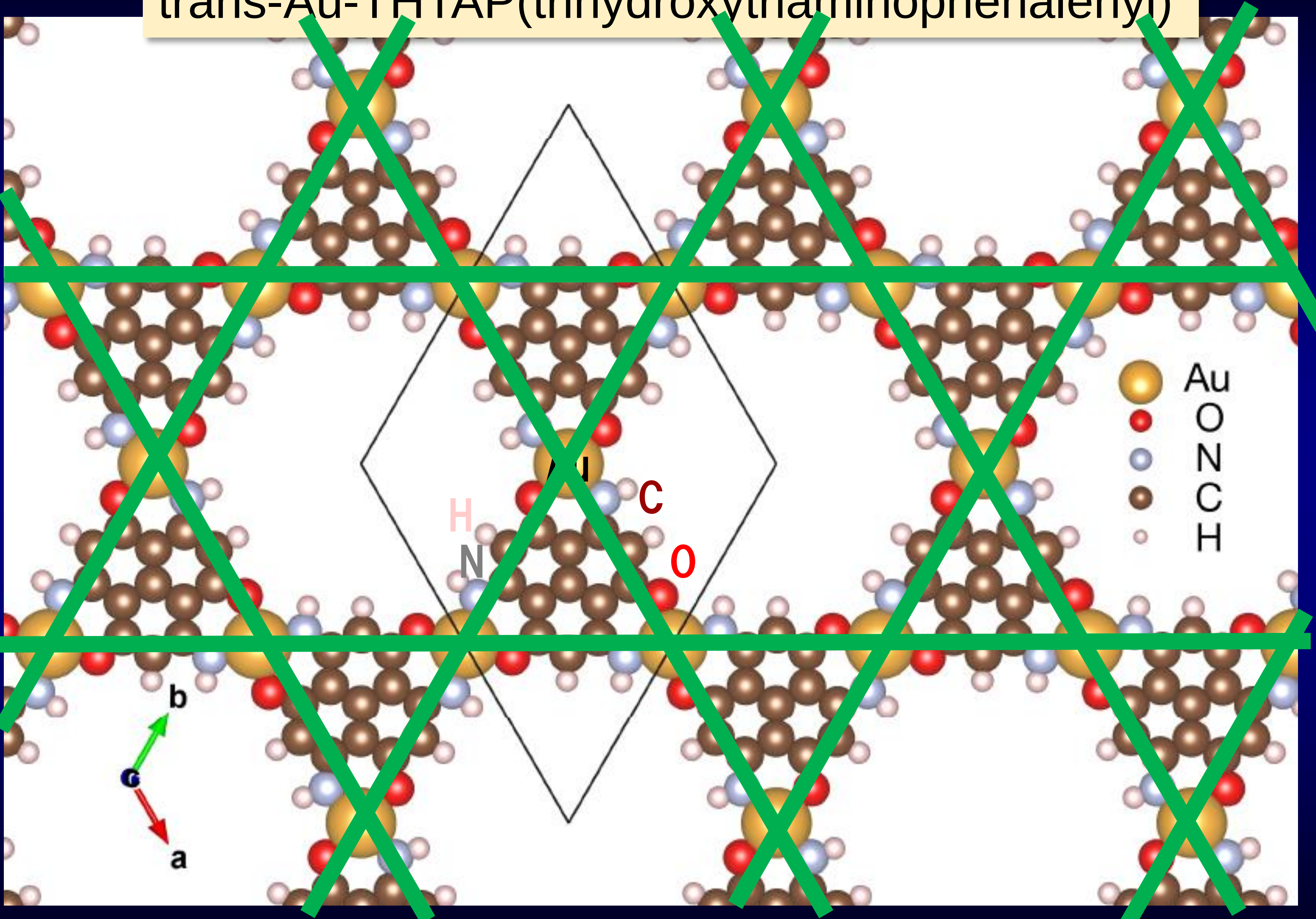
$Z = C^{\bullet}$ neutral radical

$X', Y' = OH, SH, NH_2$

We want to have E_F at the flat band

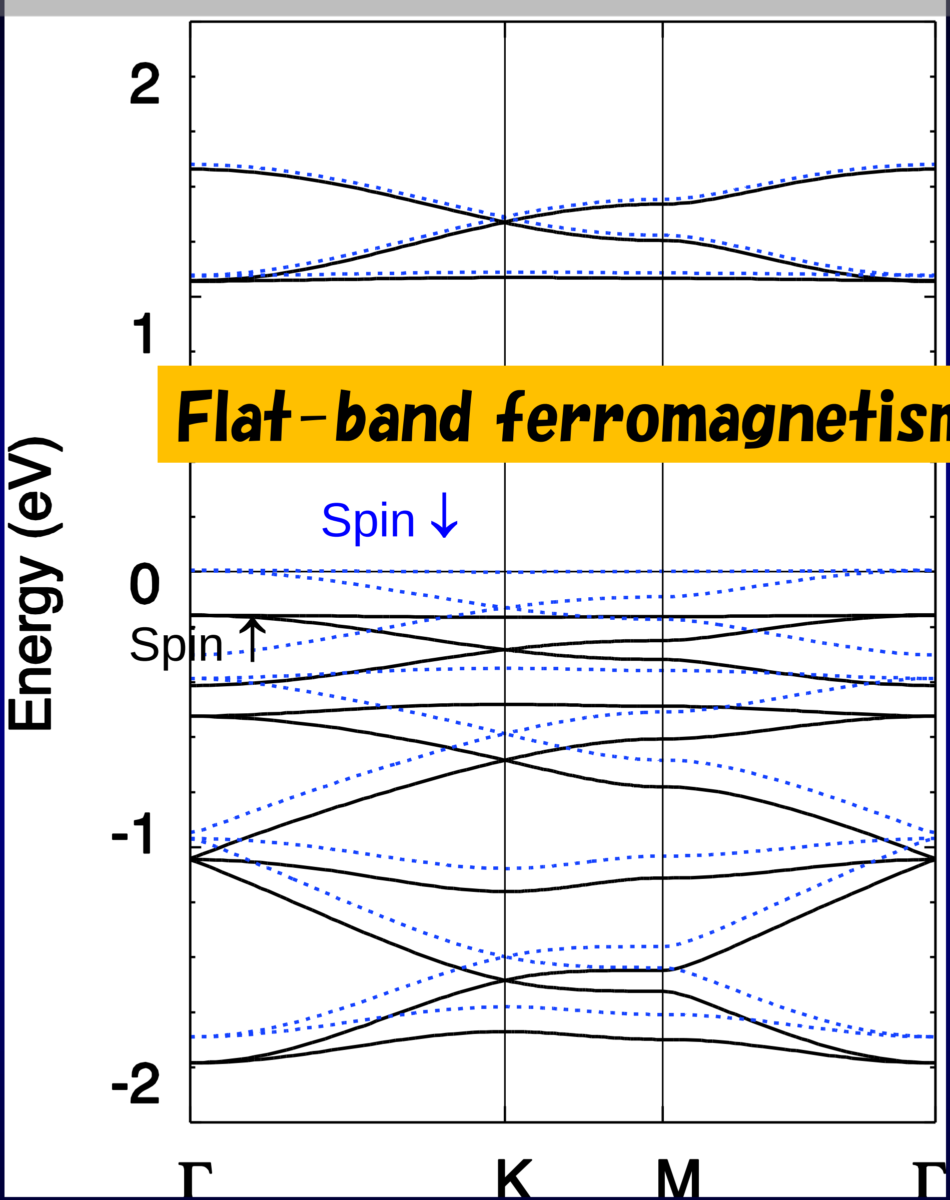


after many trials we hit upon a right material,
trans-Au-THTAP(trihydroxytriaminophenalenyl)



(Yamada et al, arXiv:1510.00164)

First-principles bands

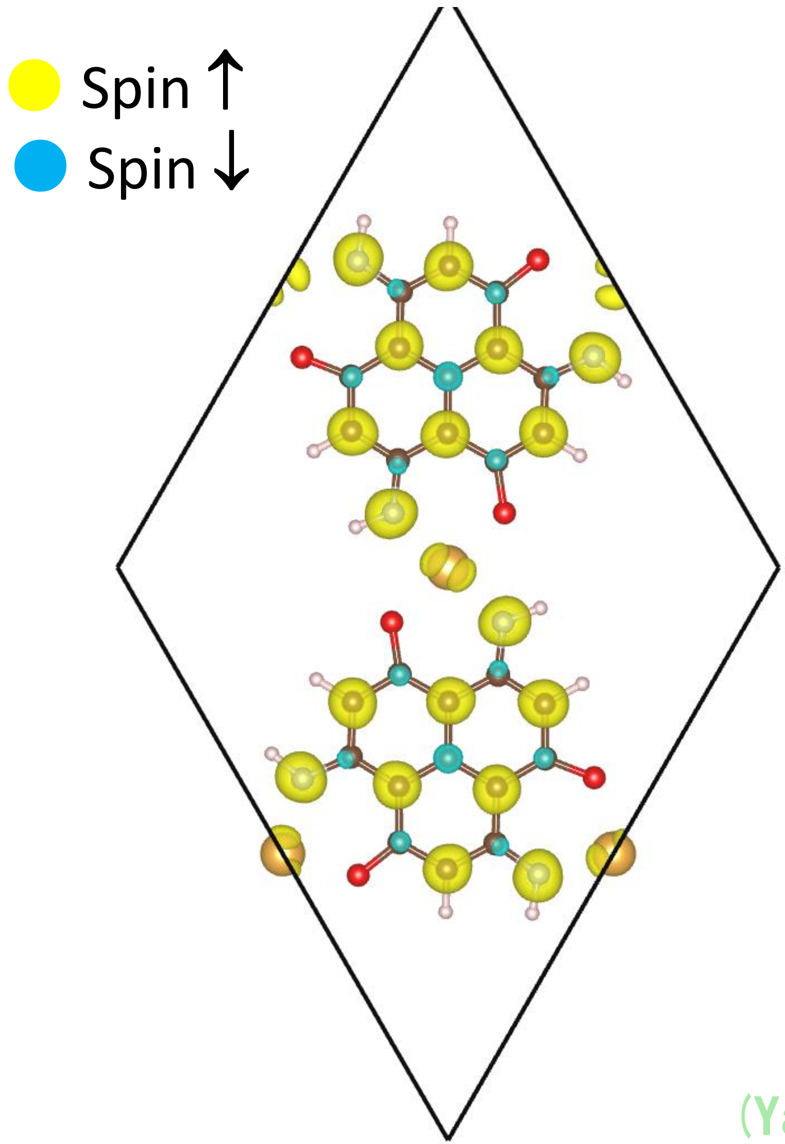


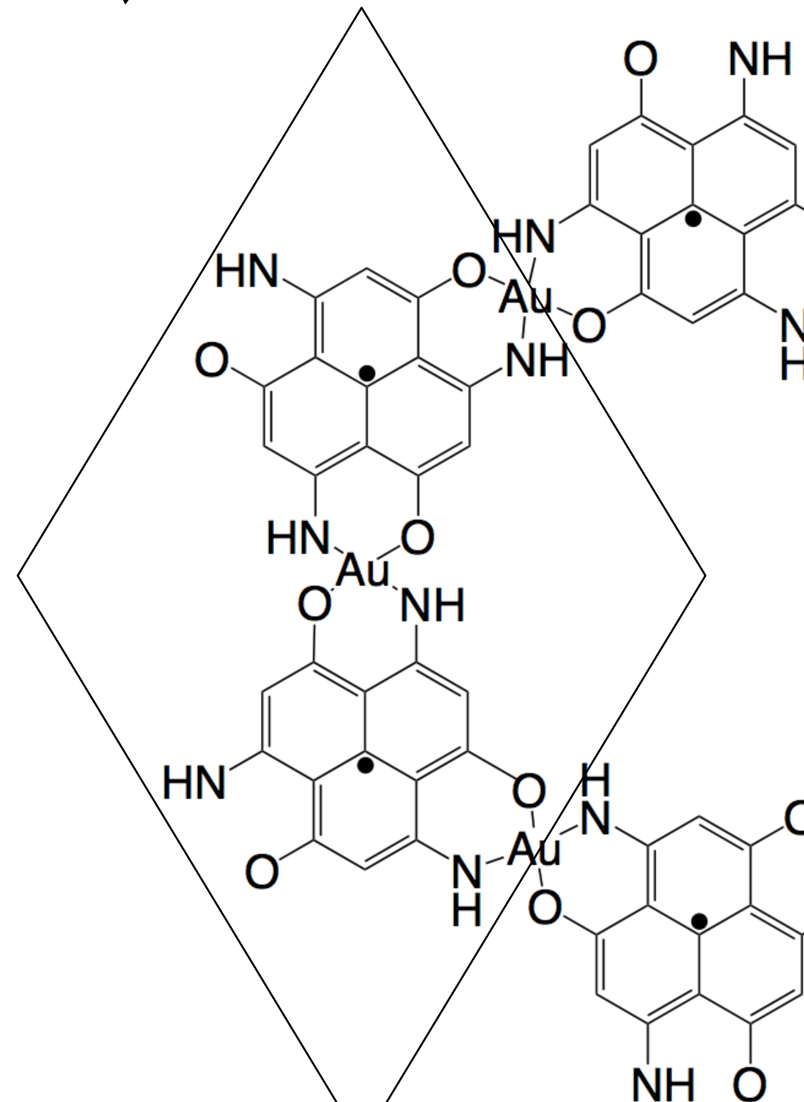
Flat-band ferromagnetism does arise

(Yamada et al, arXiv:1510.00164)

Which elements responsible for magnetism ?

Spin density: mostly on C (and N)





(Yamada et al, arXiv:1510.00164)

Thesis of today's talk

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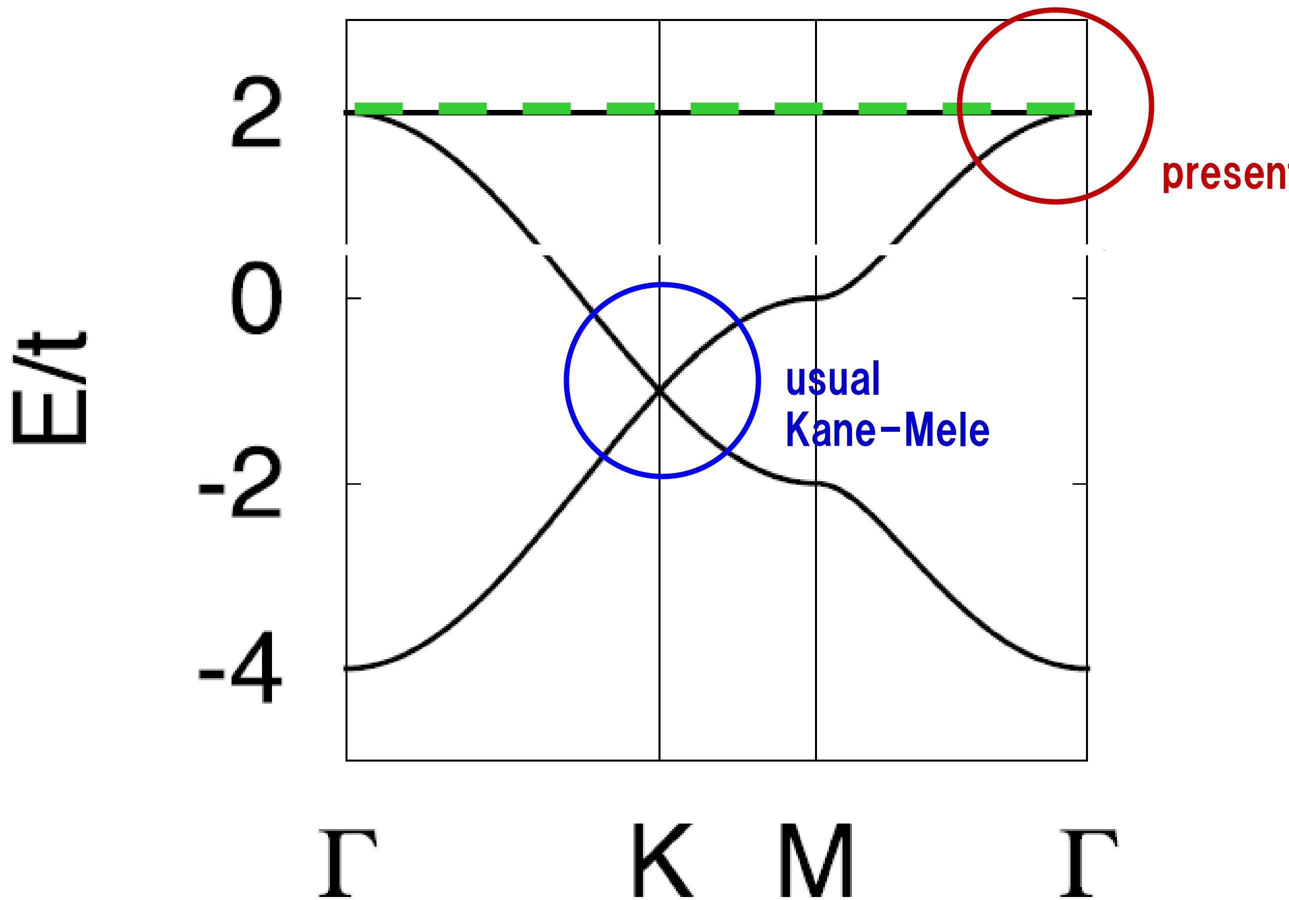
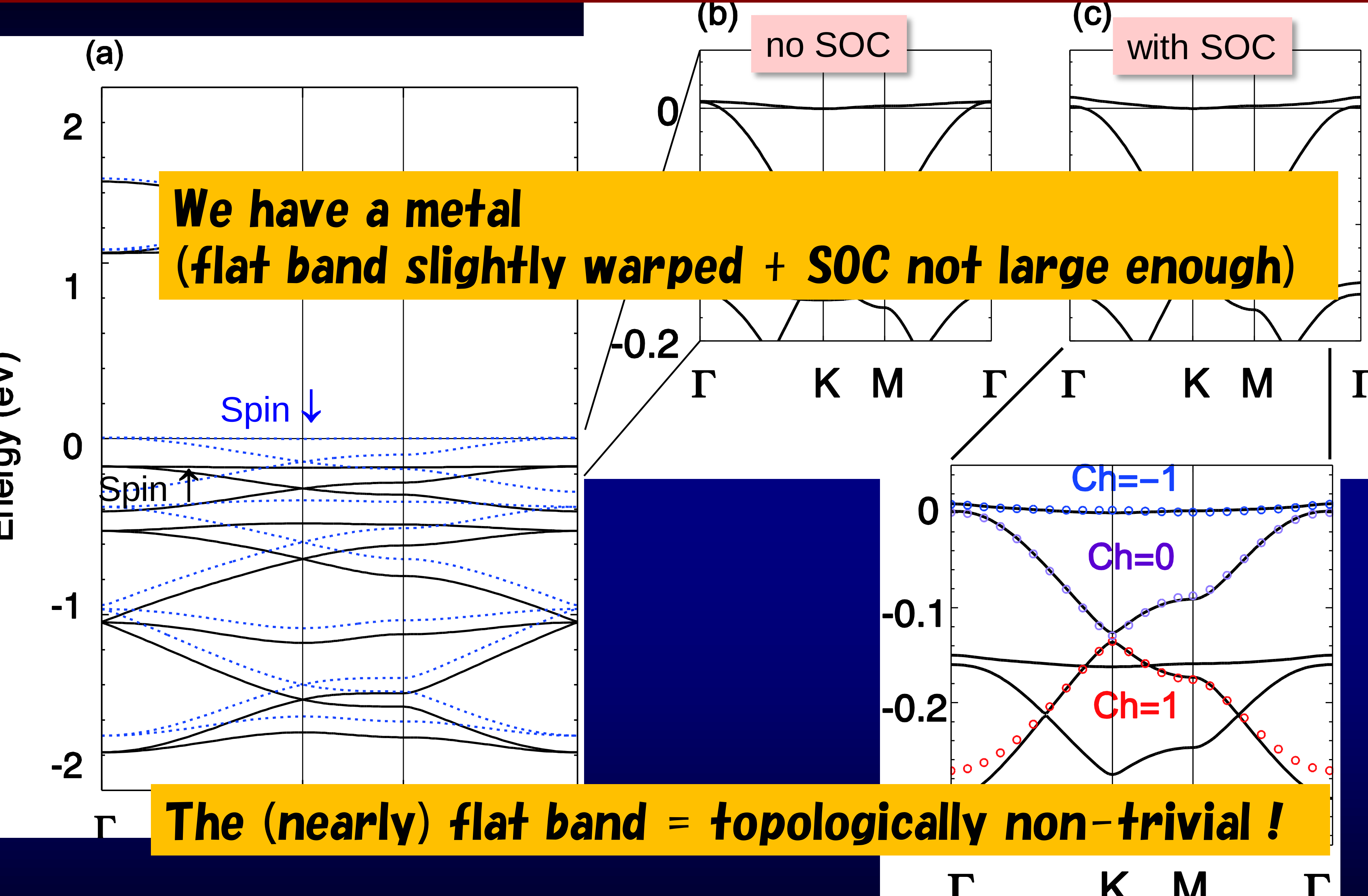
Can we even make the ferromagnetic flat-band topological ?

* Spin-orbit coupling → can the ferromagnetic flat band become topological ?



E/t
2
0
-2
-4
present
usual
Kane-Mele
Γ
K
M
Γ

SOC bands (Dirac eqn solved)



We have a metal
(flat band slightly warped + SOC not large enough)

The (nearly) flat band = topologically non-trivial !

(Yamada et al, arXiv:1510.00164)

Calculation of the Chern

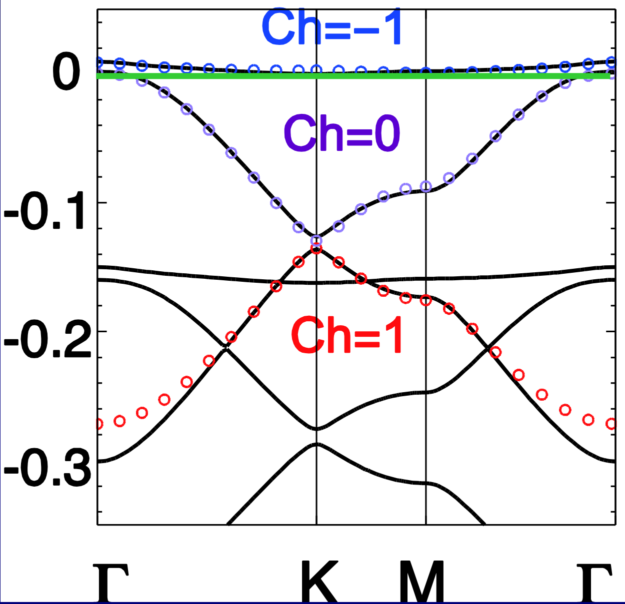
Map the system → tight-binding model

$$H = E_0 + H_0 + H_{\rm SO} + H_{\rm Z},$$

$$H_0 = -t_1 \sum_{\langle ij\rangle\sigma} c_{i\sigma}^{\dagger} c_{j\sigma} - t_2 \sum_{\langle\langle ij\rangle\rangle\sigma} c_{i\sigma}^{\dagger} c_{j\sigma},$$

$$H_{\rm SO} = i\lambda_1 \sum_{\langle ij\rangle\alpha\beta} \nu_{ij}\sigma^z_{\alpha\beta} c_{i\alpha}^{\dagger} c_{j\beta},$$

$$H_{\rm Z} = b\sum_i (-1-\sigma^z_{\alpha\beta}) c_{i\alpha}^{\dagger} c_{i\beta}.$$



$t_1 = 45.1\,\mathrm{meV},\ t_2 = 1.0\,\mathrm{meV},\ \lambda_1 = 1.2\,\mathrm{meV}.$

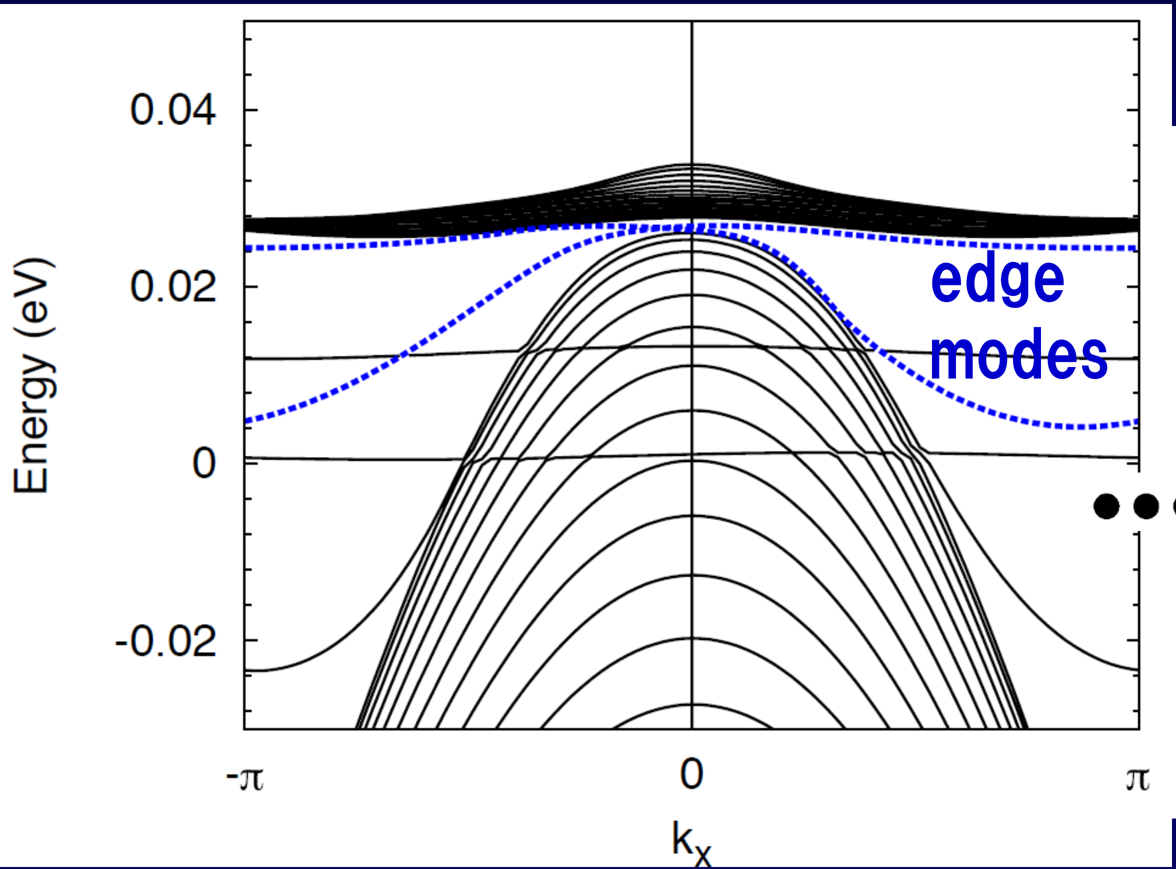
Chern # calculated by a numerical integration of the

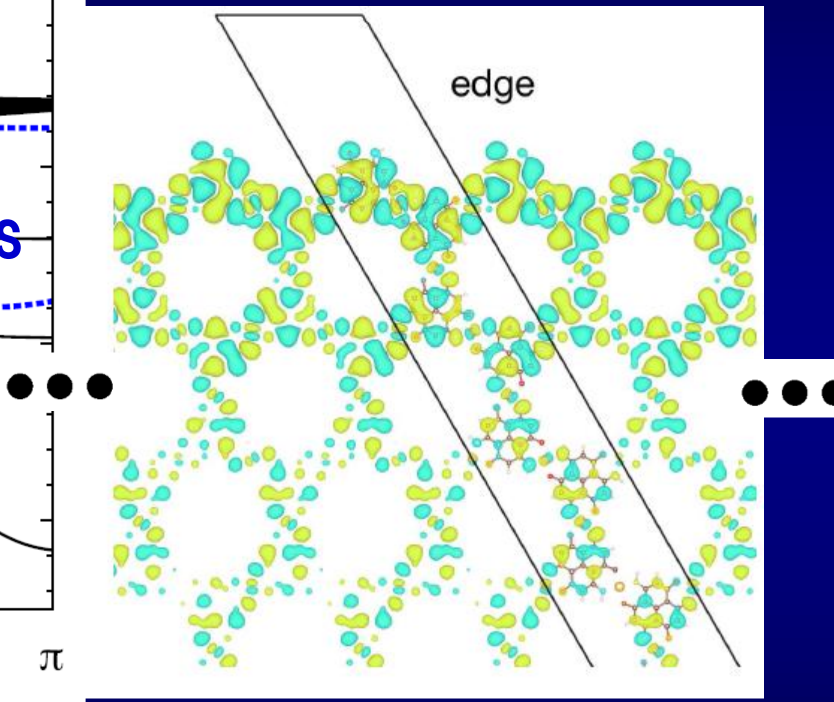
Berry's curvature with the Fukui-Hatsugai-Suzuki method

(Yamada et al, arXiv:1510.00164)

Topological edge modes

1[st] principles electronic structure
for a strip (20 uc wide)



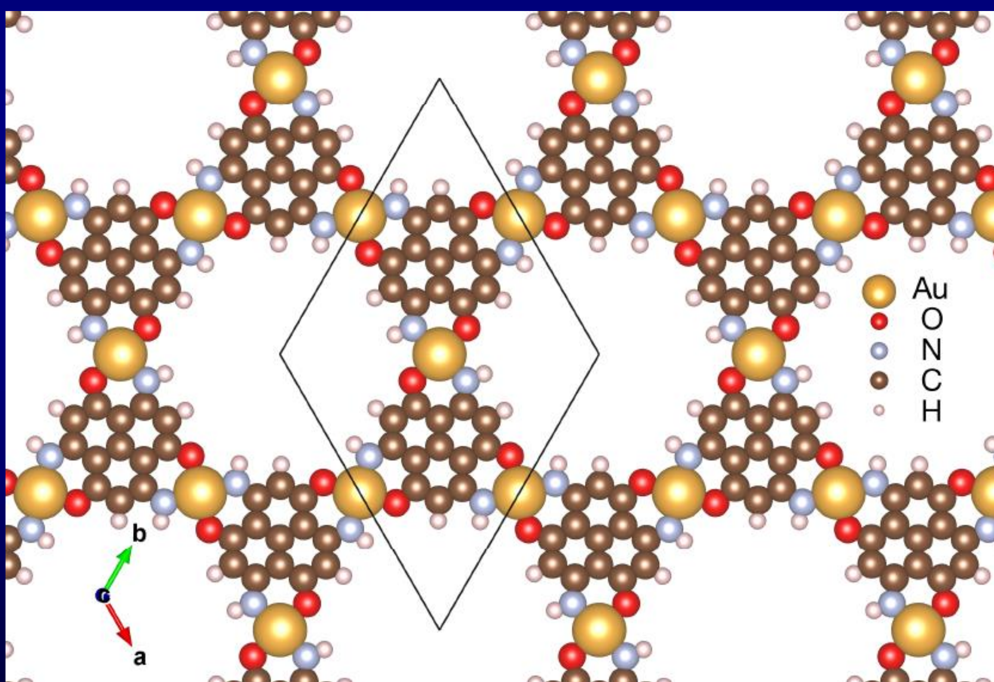


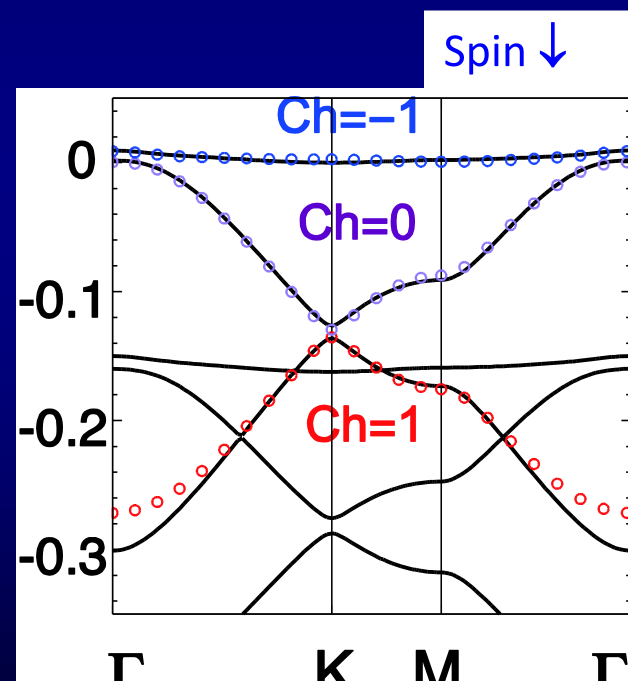
(Yamada et al, arXiv:1510.00164)

Summary

A designed MOF (Metal-organic frameworks)

- ✔ flat-band ferromagnetism realised
- ✔ + SOC → the flat band has a nonzero Chern # with edge modes, but metallic





Future works

✔ the proposed material = metallic
(due to a slight warping of the flat band
while the topological gap not large enough)

⇩

* If the system can be made an insulator,
→ quantum anomalous Hall effect or Chern insulator
(eg, Chang et al, Science 2013)

* fractional topological insulator
(Qi, PRL 2011; Maciejko & Fiete, Nat Phys 2015)
or fractional Chern insulator
(Regnault & Bernevig, PRX 1, 2011;
Bergholtz & Liu, Int J Mod Phys 2013)
[analogue of FQHE without Landau levels]
(Sheng et al, Nat Commun 2011)

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