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

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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:
\[ UD(E_F) > 1 \quad \text{--- too crude} \]

**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)

(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 $\neq$ atomic limit

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

*** NO! ***

(Kusakabe & Aoki, PRL 1994)

Spin stiffness = finite for both of

Weak coupling ($U \ll t$)

Strong coupling ($U \gg t$)

Ferromag spin stiffness ~

Connectivity condition (unusual "Wannier" orbits)

spin stiffness ~ t

Correlation effect in the multi-band system
Reasonable, since in both cases Perron–Frobenius theorem $\rightarrow$ F ground state
Realisations: 1D polymers

Atomic quantum wire
Arita et al, PRB (1998)

As adatoms on Si
(Hashizume et al, Jpn J Appl Phys 1996)

(See Gulacsi, ..., Vollhardt, PRL 2007 for diamond chain)
Design of a ferromagnetic polymer

A new material: polyaminotriazole

(Arita et al, PRL 2002; PRB 2003)

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 $\psi_f \rightarrow$ Bloch $\psi$ strongly dep on $k$

(Suwa et al, PRB 2010)
Spin density = 0.42/molecule
Q-1D ferromagnet CeRh$_3$B$_2$

(Yamada et al, JPSJ 2004)

(Kono & Kuramoto, JPSJ 2006)
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Realisations in 2D?
Ferromagnetism in carbon structures — long-period graphene or graphene antidot array

Electron correlation → Flat-band ferromagnetism

(Shima & Aoki, PRL 1993)
Long-period graphene --- $\Gamma$–$K$ manipulation

(Shima & Aoki, PRL 1993)

<table>
<thead>
<tr>
<th>Type</th>
<th>Formula unit</th>
<th>$\Gamma$</th>
<th>$K$</th>
<th>Bipartite</th>
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<td>$A_0$</td>
<td>$(C_{3m})_2$</td>
<td></td>
<td></td>
<td>$sc + n(\geq 0)$ flat band(s)</td>
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<tr>
<td>$A_C$</td>
<td>$(C_{3m+1})_2$</td>
<td></td>
<td>$E$</td>
<td>$sm + n(\geq 0)$ flat band(s)</td>
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<td>$(C_{3m+3/2})_2$</td>
<td>$A, E$</td>
<td>$A, E$</td>
<td>$sc + n(\geq 3)$ flat bands</td>
</tr>
<tr>
<td>$B_C$</td>
<td>$(C_{3m+5/2})_2$</td>
<td>$A, E$</td>
<td>$A$</td>
<td>$sm + n(\geq 1)$ flat band(s)</td>
</tr>
</tbody>
</table>
Graphene nanomesh

Graphene nanomesh → diodes
(Bai et al, Nature Nanotech 2010)
"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?
Figure 1. Schematic illustration and chemical structure of monolayer nickel bis(dithiolene) complex nanosheet. Countercations have been omitted for clarity. Gray, C; yellow, S; green, Ni.
High Electrical Conductivity in Ni₃(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 Dinca*†
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)
phenalenyl-based ligands

\[ Z = C^\bullet \] neutral radical

\[ X', Y' = \text{OH, SH, NH}_2 \]
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)
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Can we even make the ferromagnetic flat-band topological?
   * Spin-orbit coupling → can the ferromagnetic flat band become topological?
usual Kane–Mele

present
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 $\rightarrow$ tight-binding model

$$H = E_0 + H_0 + H_{SO} + H_Z,$$

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

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

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

Chern $\#$ calculated by a numerical integration of the Berry's curvature with the Fukui-Hatsugai-Suzuki method

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

(Yamada et al, arXiv:1510.00164)
Topological edge modes

1st 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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Tomohiro Soejima, Mircea Dinca  Dept Chemistry, MIT