Anomalous quantum oscillations in $RuCl_3$/graphene heterostructures

KITP: Unconventional Magnetism and Novel Probes in Heterostructures, 1st October 2020

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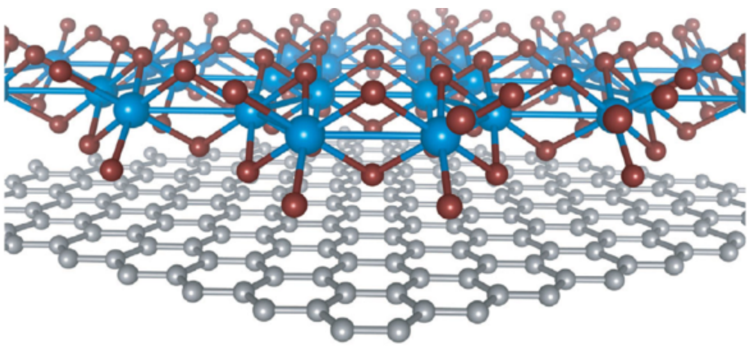
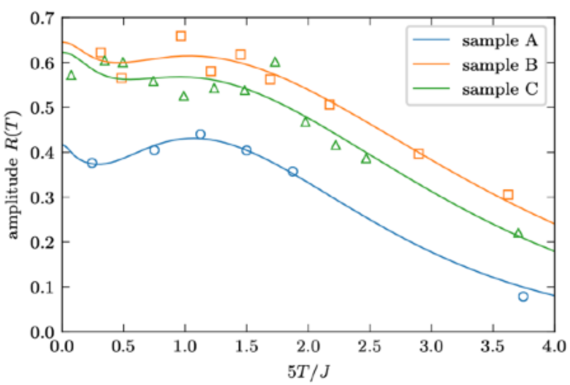
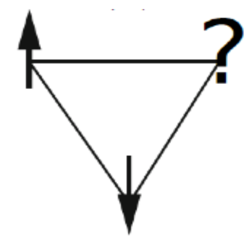




Leeb, Polyudov, Mashhadi, Biswas, Valenti, Burghard, JK, *arXiv:2010.01649*

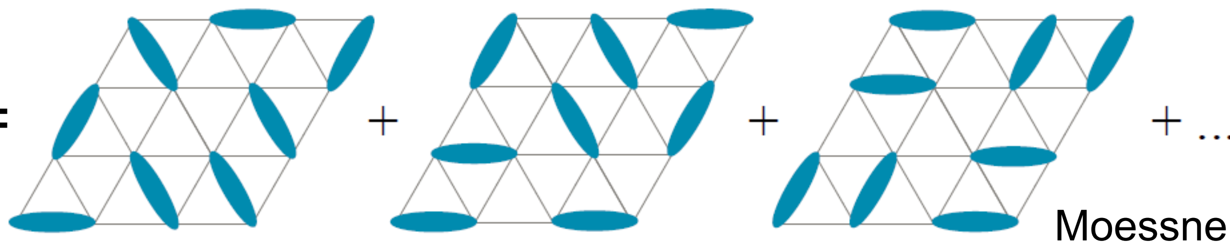
Quantum Spin Liquids



- QSL predicted in **frustrated magnets** which can evade ordering down to T=0. Anderson 1973

- **QSL:** State of interacting spins that breaks no rotational or translational symmetry and has only short range spin correlations.

RVB: $\Psi=$

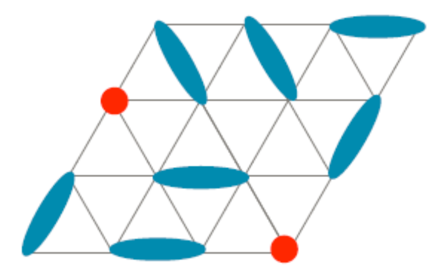


Moessner, Sondhi 2000

 $= \frac{1}{\sqrt{2}}\left(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle\right)$

- **Long-range entangled & topologically ordered**

- **Fractional excitations with Anyonic statistics**



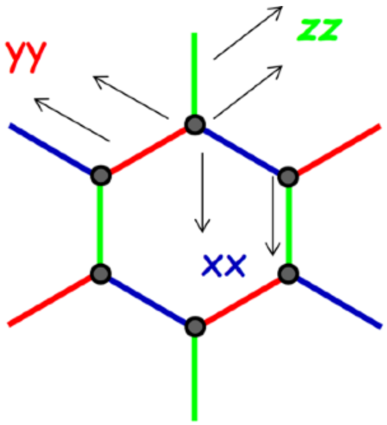
Wen 1990

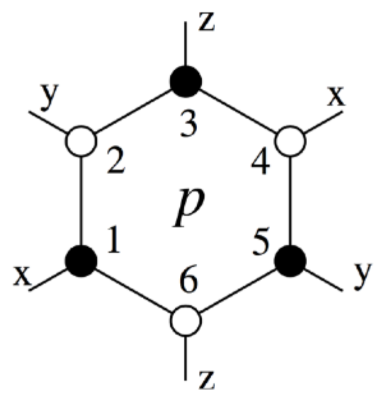
Kitaev's QSL

- Spin ½ on the honeycomb lattice with strong spin orbit coupling

Kitaev 2006

$$H = -J_x \sum_{x \text{ links}} \sigma_j^x \sigma_k^x - J_y \sum_{y \text{ links}} \sigma_j^y \sigma_k^y - J_z \sum_{z \text{ links}} \sigma_j^z \sigma_k^z$$

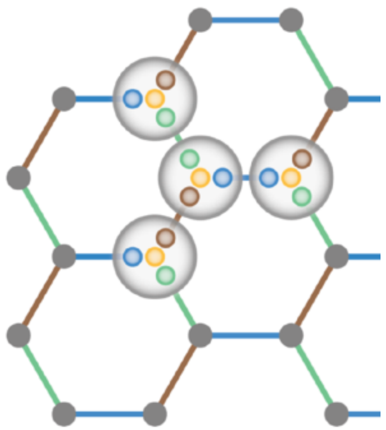




$$W_p = \sigma_1^x \sigma_2^y \sigma_3^z \sigma_4^x \sigma_5^y \sigma_6^z$$

- **Exactly solvable interacting 2D model!**
- Large number of conserved quantities, local plaquette operators.

Kitaev‘s QSL: Exact Solution



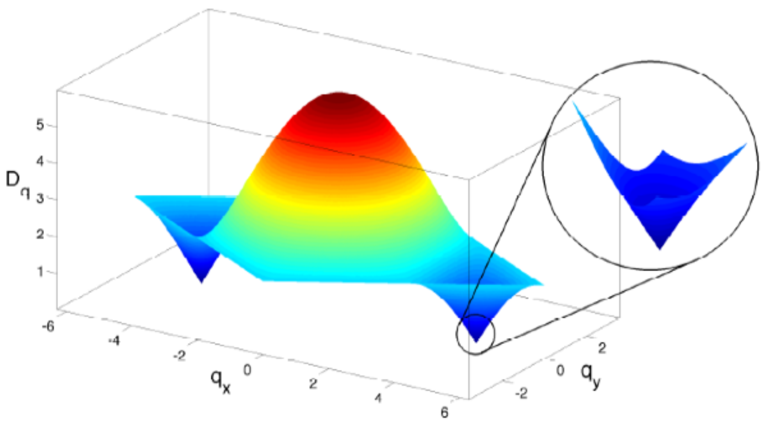
- Mapping spins to Majoranas:

$$\sigma_i^a = i c_i c_i^a, \qquad a = x, y, z$$

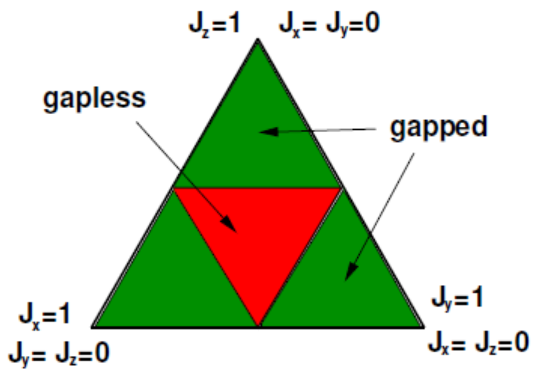
- Tight-binding Hamiltonian

$$H = -\sum_{a=x,y,z} J_a \sum_{\langle ij\rangle_a} i c_i \hat{u}_{\langle ij\rangle_a} c_j \text{ with } \hat{u}_{\langle ij\rangle_a} \equiv i c_i^a c_j^a$$

- Spectrum:



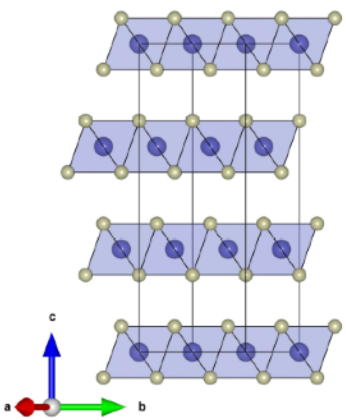
- Phase diagram:

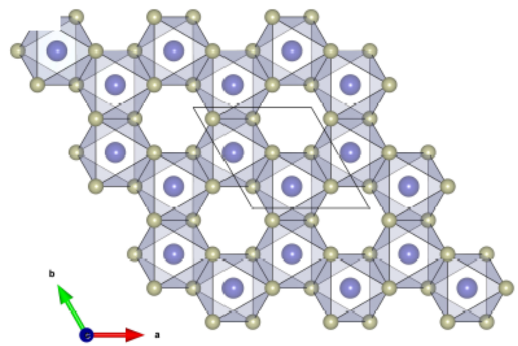


Kitaev‘s QSL: Candidate Materials

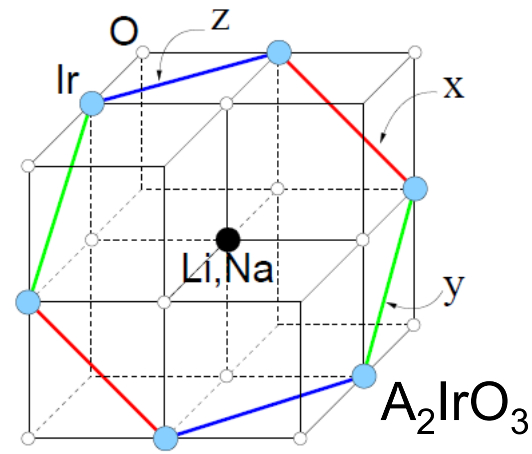
- Correlations between real and spin space?
  → Strong spin orbit interaction (e.g. Ir, Ru).

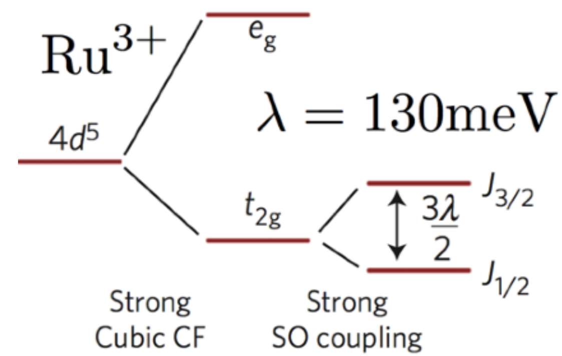
  Jackeli, Khaliullin 2009, Singh, Gegenwart 2010
  Modic et al. 2014, Takayama et al. 2015





Plumb et al. 2014





Many candidate materials and many developments

Motome, Nasu *JPSJ* 2020

Takagi, Takayama, Jackeli, Khaliullin, Nagler *Nature Review Physics* 2019

Hermann, Kimchi, JK *Annual Review CMP* 2018

Winter, Tsirlin, Daghofer, v.d.Brink, Singh, Gegenwart, Valenti *JPCM* 2017

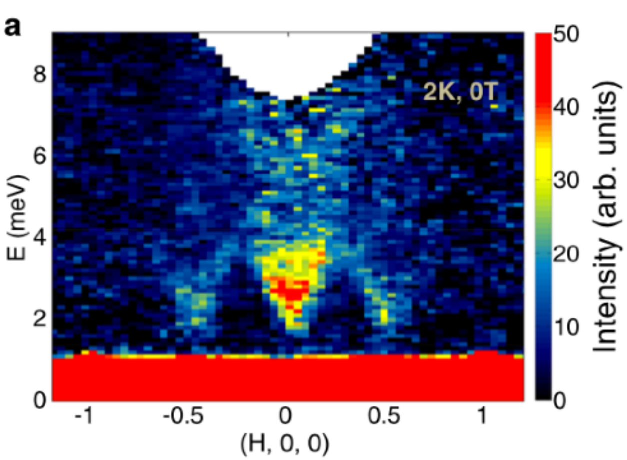
Kitaev's QSL: Candidate Materials

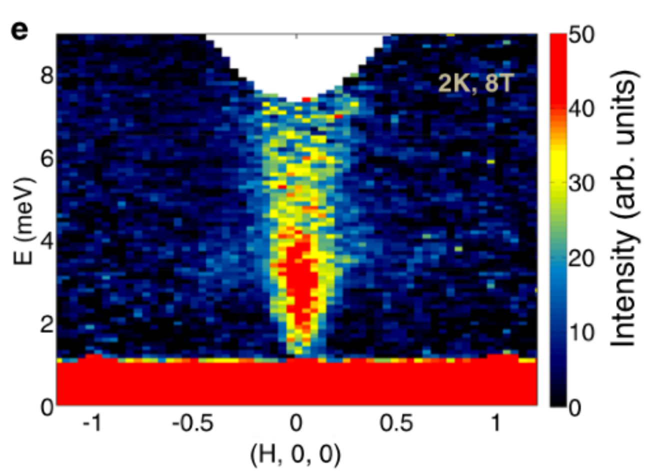
- Melt residual magnetic order of the '**proximate QSL**'

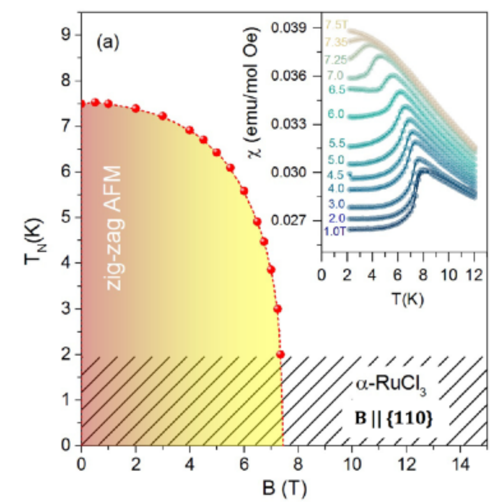
A. Banerjee, et al. Nature Materials 2016
A. Banerjee, et al. Science 2017

→ **Many tuning parameters**

magnetic field, pressure, intercalation, **thin film heterostructures**







A. Banerjee, et al., npj QM 2018

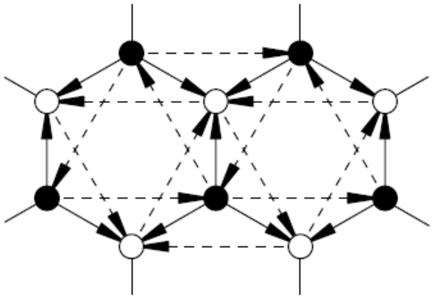
→ **Is there a magnetic field induced QSL?**

→ Half-integer quantized THE? Kasahara et al. Nature 559, 227-231 (2018)

Non-Abelian Kitaev QSL in a Field

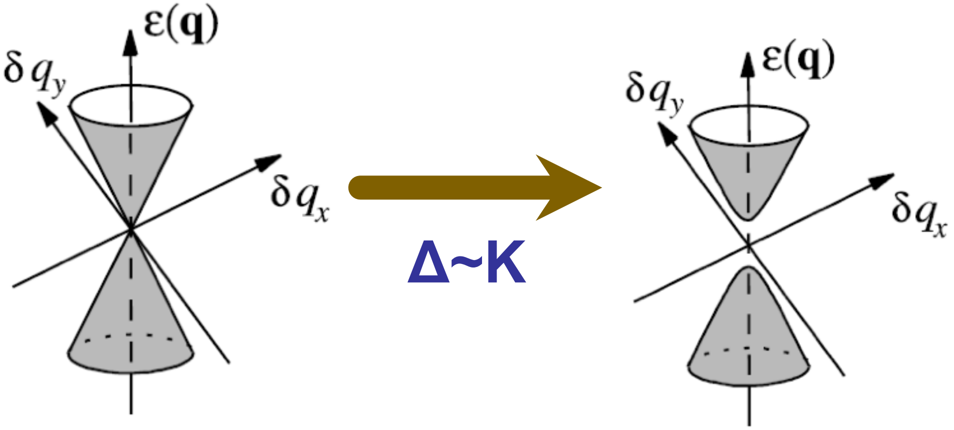
- Additional **three spin interaction** gives a **non-Abelian** QSL
  → term arises perturbatively in a magnetic field Kitaev 2006

$$H' = -K \underbrace{\sum_{\langle i,j\rangle_\alpha, \langle j,k\rangle_\beta}}_{\gamma \perp \alpha,\beta} \sigma_i^\alpha \sigma_j^\gamma \sigma_k^\beta$$



- Spectrum is gapped
  → Chern bands.

$$E(\mathbf{q}) = \sqrt{\xi_{\mathbf{q}}^2 + |\Delta_{\mathbf{q}}|^2}$$



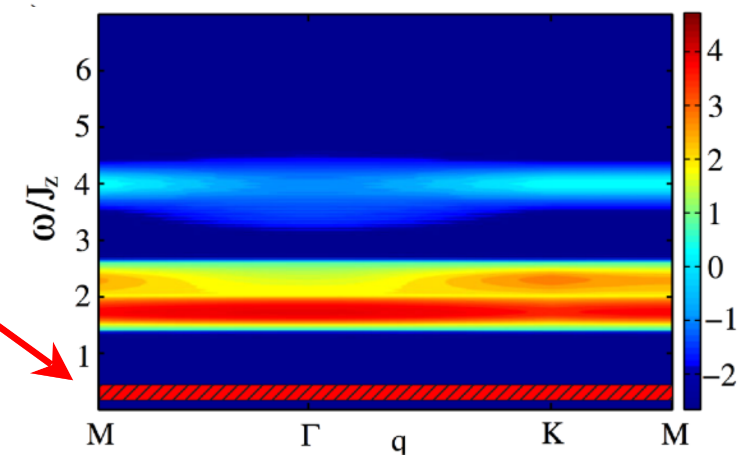
- **Now each flux σ binds a Majorana Fermion.**
  → Two Majorana fermions give one complex fermion **at energy $\varepsilon_0<\Delta$**

Bound states of fractionalized excitations

- Structure factor of Kitaev model in **a magnetic field**

  → **Flux-Majorana Bound State**

  JK, Kovrizhin, Chalker, Moessner, *PRB* (2015)

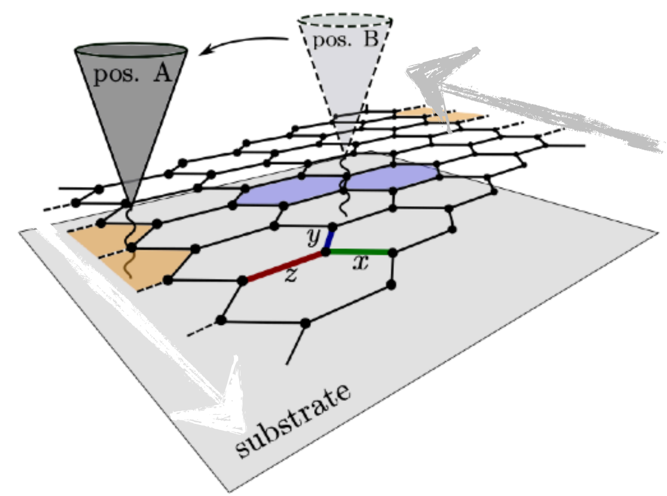


- Can we use spin-polarized STM for directly probing charge neutral excitations in real space?

  → Inelastic tunneling via magnetic layer

$$\frac{\partial I}{\partial V} = \frac{2e^2}{\hbar} \sum_{i,j,\alpha} t_1(\boldsymbol{r}-\boldsymbol{r}_i) t_1(\boldsymbol{r}-\boldsymbol{r}_j)\, c_{\alpha\beta} \int_0^{eV} d\omega\, \mathcal{S}_{ij}^{\alpha\beta}(\omega)$$

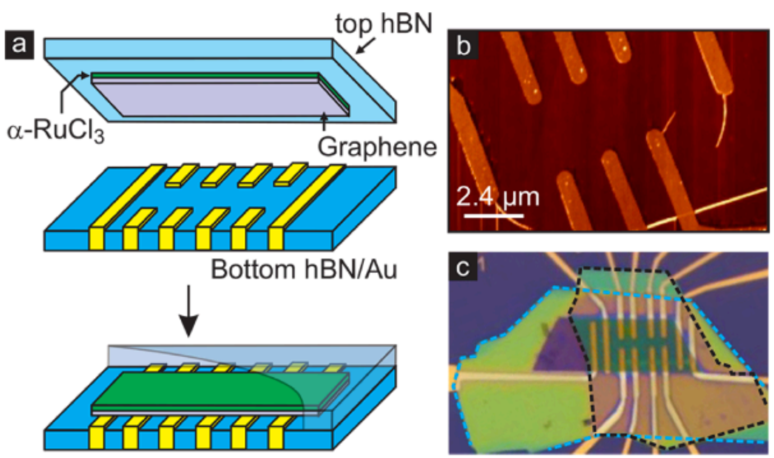
Feldmeier, Natori, Knap, JK, *arXiv:2007.07912*

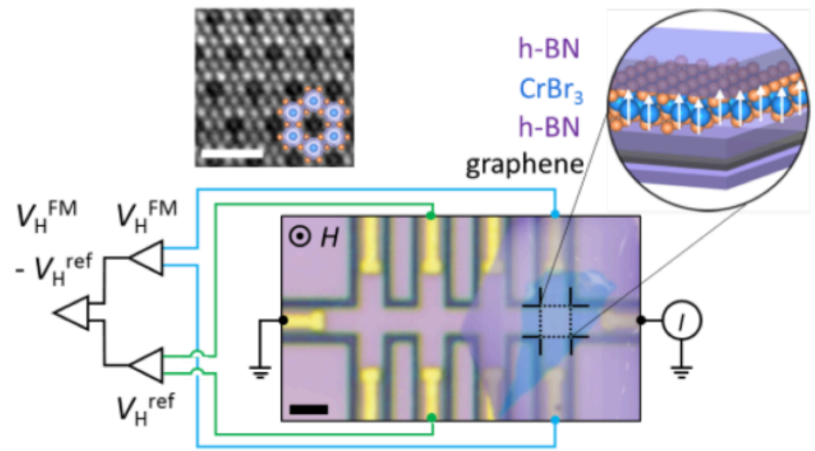


$RuCl_3$-graphene heterostructure

→ tuning parameter: **thin films and substrate engineering**

- **Single layer $RuCl_3$ on Graphene**





S. Mashadi, …, M. Burghard et al. Nano Lett. 19, 4659 (2019)
B. Zhou, … E.A. Henrikson PRB 100, 165426 (2019)

→ Use graphene transport as a sensor for QSL fluctuations?

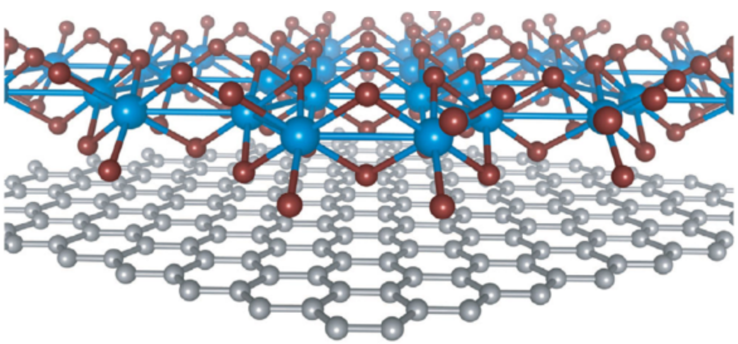
M. Kim, JK…, A.K. Geim, Nature Electronics (2019)

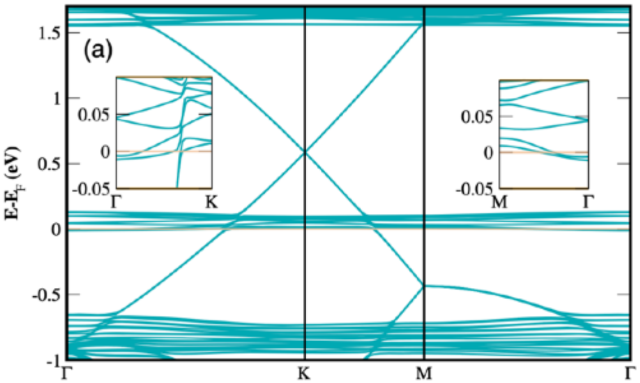
$RuCl_3$-graphene heterostructure

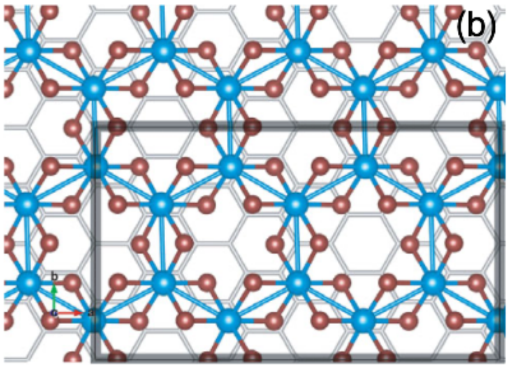
- **Single layer $RuCl_3$ on Graphene**

  → from ab-initio calculations to effective models

  Biswas, Winter, JK, Valenti PRL 123, 237201 (2019)







→ lattice mismatch causes local strain

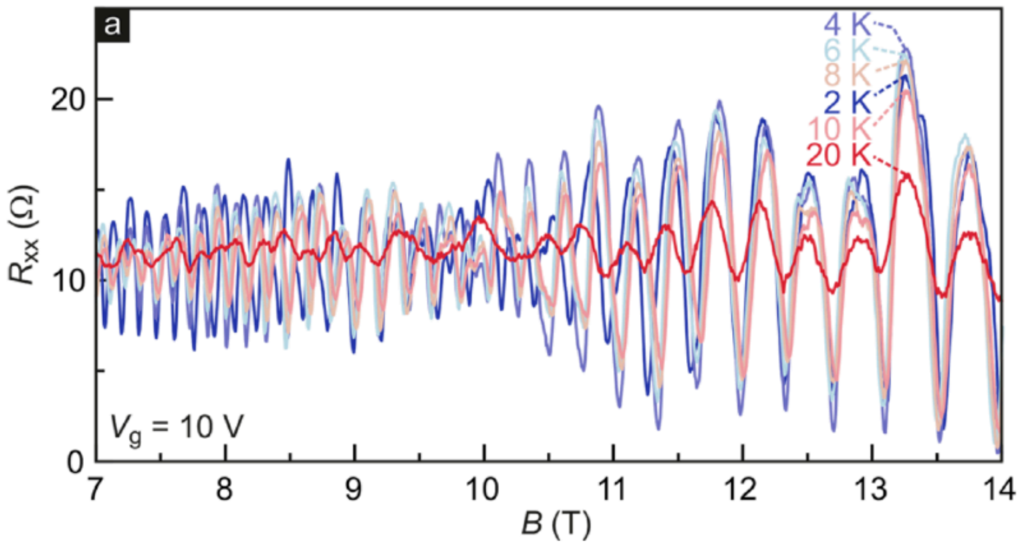
→ **increase of Kitaev interaction**

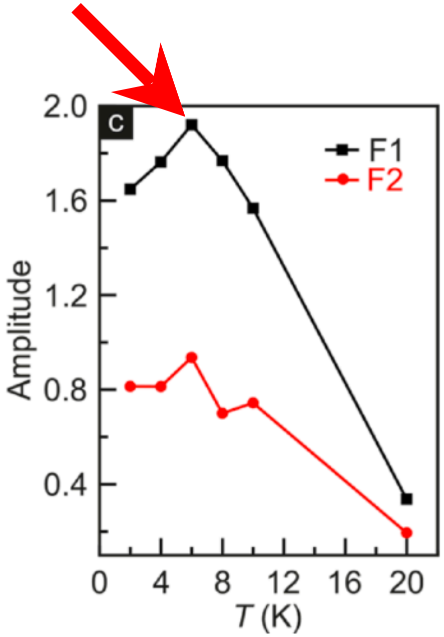
- **Charge transfer leads to electron (hole) doped $RuCl_3$ (graphene)**

  → recent observation of highly doped 2D interface

  Wang et al., arXiv:2007.06603; Rizzo et al., arXiv:2007.07147

Anomalous Quantum Oscillations





S. Mashadi, …, M. Burghard et al.
Nano Lett. 19, 4659 (2019)

Non-Lifshitz-Kosevich behaviour!

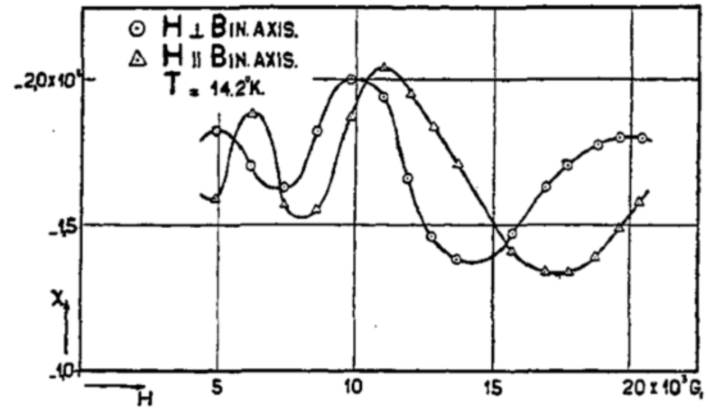
- Interaction between proximate QSL excitations and graphene?

Main question:

Are anomalous QO and non-LK behaviour related to QSL correlations? How to describe non-LK QO?

Recap: Quantum Oscillations

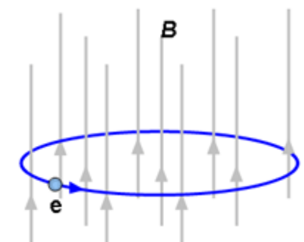
- Oscillation of magnetisation
  as a function of applied B-field dHvA 1930
  Landau 1930



→ experimental tool for measuring
properties of **metals** Lifshitz,Kosevich 1954

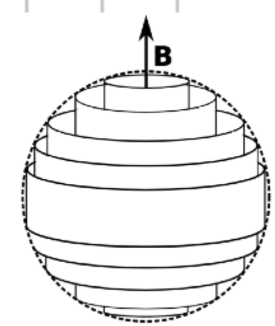
- Origin: Bohr-Sommerfeld quantisation of closed electron orbits

$$\oint \vec{p}\mathrm{d}\vec{r} = (n+\gamma)2\pi h = \frac{e}{c}\oint \vec{B}\mathrm{d}\vec{S} = \frac{e}{c}\Phi$$



- Quantised k-space area $Sl_B^2 = 2\pi(n+\gamma)$
  Onsager 1952

→ periodic oscillations $\Delta(\frac{1}{B}) = \frac{1}{B_{n+1}} - \frac{1}{B_n} = \frac{2\pi e}{\hbar c S_e}$



Lifshitz-Kosevich theory

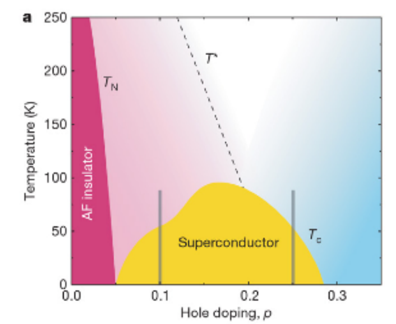
- **Universal T-dependence:** $$R(T) = \frac{\chi}{\sinh \chi}$$

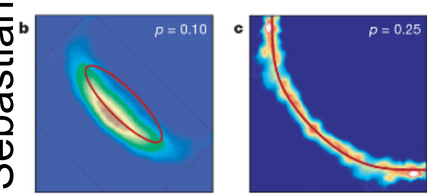
with $\chi = \frac{2\pi^2 T}{\hbar \omega_c}, \omega_c = \frac{eB}{m^* c}$

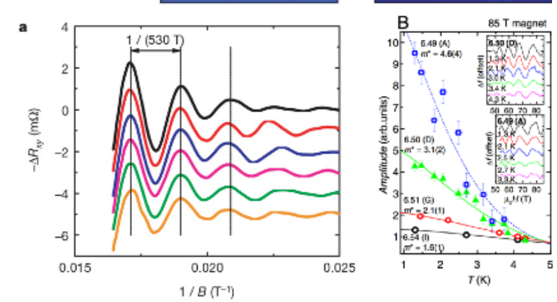
- **Measurement of effective mass** LK 1954

Ex.: Cuprates

Doiron-Leyraud Nature 2007
Sebastian PNAS 2009

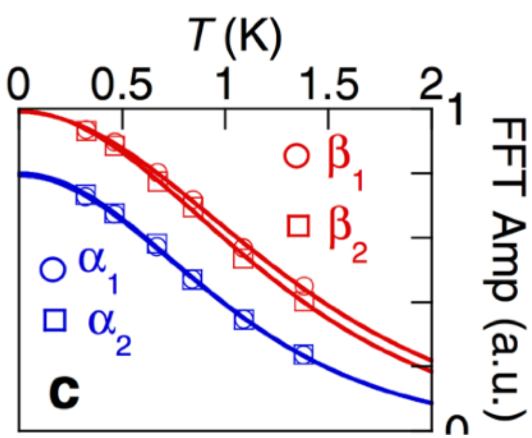






Ex.: LaFePO

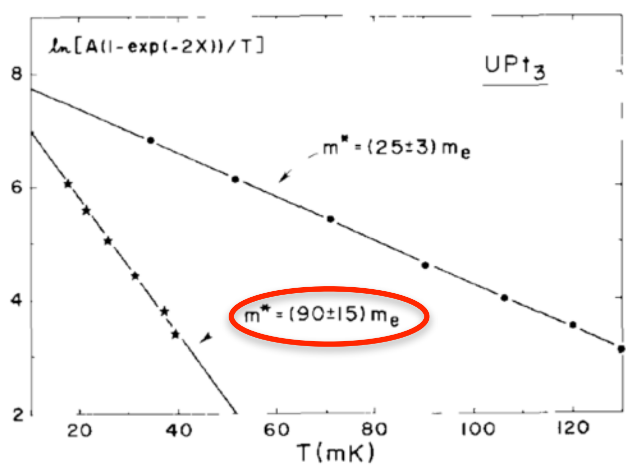
A. Coldea PRL 2008



→ mass enhancement ~2

Ex.: UPt$_3$ heavy fermions

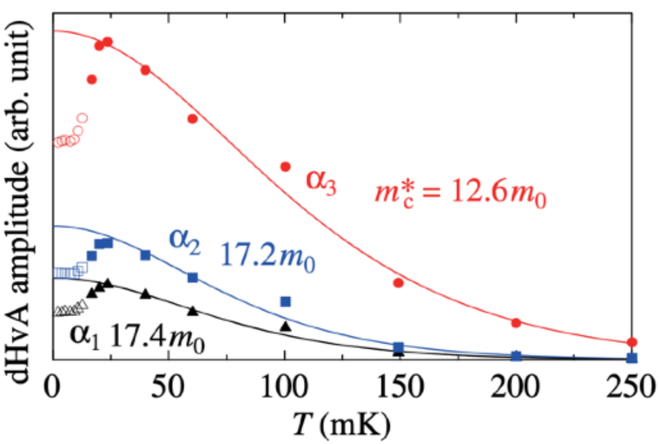
L. Taillefer JMMM 1987



A Triumph of Fermi Liquid theory

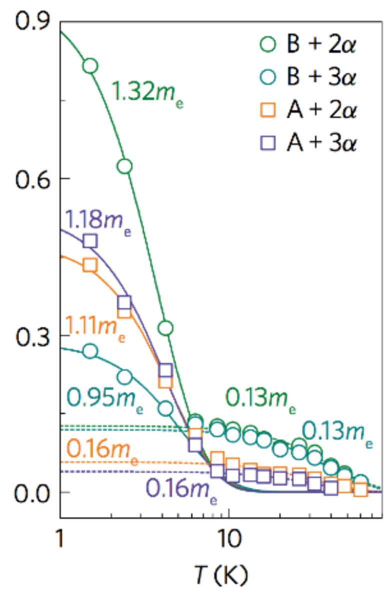
- **QO universally observed in metallic materials!**
  → Copper, Heavy Fermions, Cuprates, …

- **Very few exceptions to LK T-dependence:**



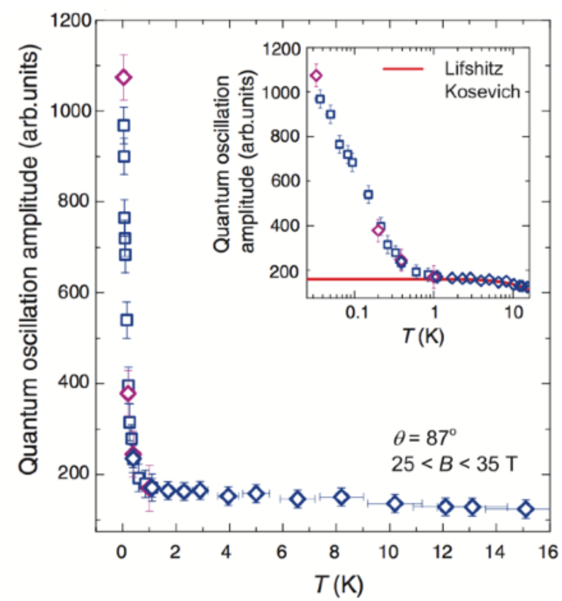
Ex.: $CeCoIn_5$

McCollam, et al. PRL 2005
Shishido, et al. PRL 2018



Ex.: ZrSiS

Pezzini, et al.
Nature Physics 2018



Ex.: **SmB_6** Kondo system

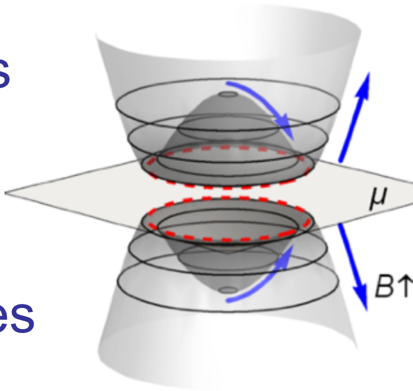
Tan, et al. Science 2015

Beyond LK ?

- **Anomalous QO in inverted (topological) insulators**

  → peculiar Landau level structure beyond semi-classics

  JK, N. R. Cooper, PRL (2015)
  Zhang, Song, Fa Wang PRL (2016); many others …



  → predicted in InAs/GaSb quantum well heterostructures

  JK, N. R. Cooper, PRL (2017)
  Z. Han, et al., PRL 123, 126803 (2019); D. Xiao, et al., PRL 122, 186802 (2019)

- **QO from fractionalized excitations in insulating magnets**

  → indirect coupling of the orbital magnetic field to spin interactions

  O. I. Motrunich, PRB 73, 155115 (2006)

  → orbital field effects in Mott insulators with strong SO coupling

  W. Natori, R. Moessner, JK, PRB 100, 144403 (2019)

  → QO from spinon Fermi surface in QSLs ?

  D. Chowdhury, I. Sodemann, and T. Senthil, Nature Comm. 9, 1 (2018)
  I. Sodemann, D. Chowdhury, and T. Senthil, PRB 97, 045152 (2018)

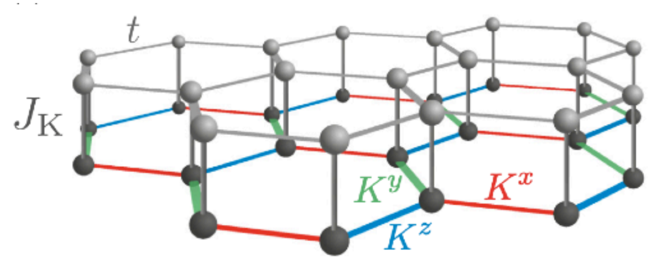
Kitaev-Kondo Model

- **Minimal model for the $RuCl_3$-Graphene heterostructure**

  Seifert, Meng, Vojta PRB (2018); Choi, Klein, Rosch, Y.B. Kim PRB (2018)

$$H = -K \sum_{\langle ij \rangle_\alpha} S_i^\alpha S_j^\alpha - t \sum_{\langle ij \rangle, \sigma} (c_{i,\sigma}^\dagger c_{j,\sigma} + h.c.) + J \sum_{i,\sigma,\sigma',\alpha} c_{i,\sigma}^\dagger \tau_{\sigma,\sigma'}^\alpha c_{i,\sigma'} S_i^\alpha$$

- **Rich phase diagram via Kondo coupling**



→ Fractionalized Majorana excitations may hybridise with the electronic excitations of the Graphene layer

→ **Effective description of the low-T heavy Fermi liquid phase**

Effective Model

- **Effective hybridisation between Majoranas and electrons**

$$H=\sum_{\boldsymbol{k},\sigma}\begin{pmatrix}c_{\boldsymbol{k},A,\sigma}\\ c_{\boldsymbol{k},B,\sigma}\\ f_{\boldsymbol{k},A,\sigma}\\ f_{\boldsymbol{k},B,\sigma}\end{pmatrix}^{\dagger}\begin{pmatrix}W & t\theta_{\boldsymbol{k}} & \frac{J}{2} & 0\\ t\theta^{*}_{\boldsymbol{k}} & W & 0 & \frac{J}{2}\\ \frac{J}{2} & 0 & 0 & \frac{K}{4}\theta_{\boldsymbol{k}}\\ 0 & \frac{J}{2} & \frac{K}{4}\theta^{*}_{\boldsymbol{k}} & 0\end{pmatrix}\begin{pmatrix}c_{\boldsymbol{k},A,\sigma}\\ c_{\boldsymbol{k},B,\sigma}\\ f_{\boldsymbol{k},A,\sigma}\\ f_{\boldsymbol{k},B,\sigma}\end{pmatrix}$$

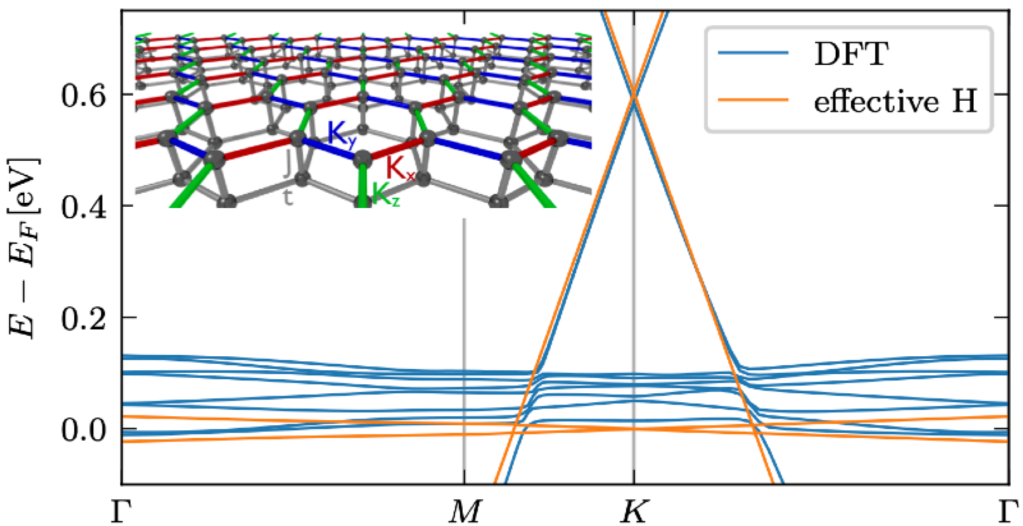
- **Microscopic model parameters from ab-initio calculations**

W~600meV

J~2meV

K~17meV << t~2.6eV

→ incommensurate lattice!



Anomalous QO

- **Exact Landau levels from low energy expansion**

$$E_l^{\xi=\pm1,\zeta=\pm1} = \frac{1}{2}\left(W + \xi(\omega_t + \omega_K)\sqrt{l} + \zeta\sqrt{\left(W + \xi(\omega_t - \omega_K)\sqrt{l}\right)^2 + J^2}\right)$$

- **QO from poles of Greens function** $G^{-1}_{\xi,\zeta}(\mathrm{i}\omega_n) = \mathrm{i}\omega_n - (E_l^{\xi,\zeta} - \mu)$

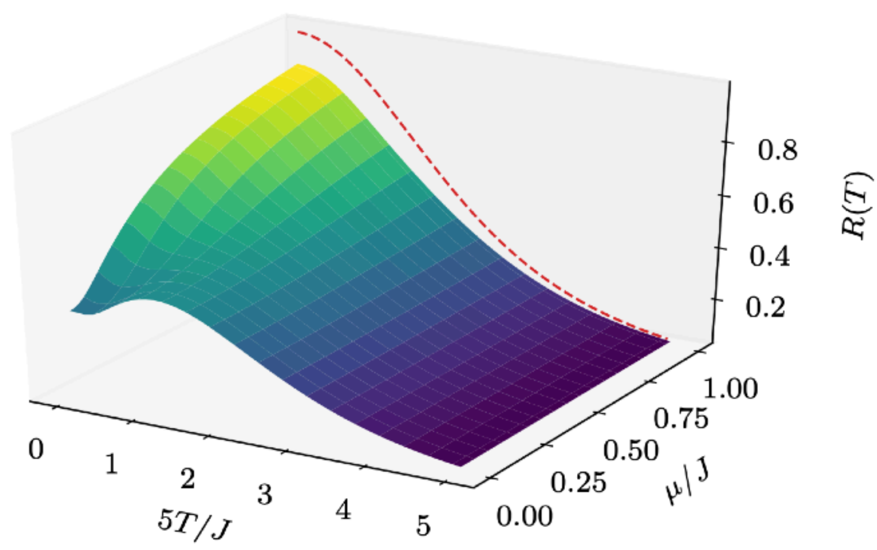
S. A. Hartnoll, D. M. Hofman, PRB 81 (2010)

- **New formula for anomalous QO**

$$M = -\frac{\partial \Omega_{\text{osc.}}}{\partial B_z} = -\frac{AW}{\phi_0 \pi}\sin\left(2\pi\left[\frac{W}{\omega_t}\right]^2\right)R(T)$$

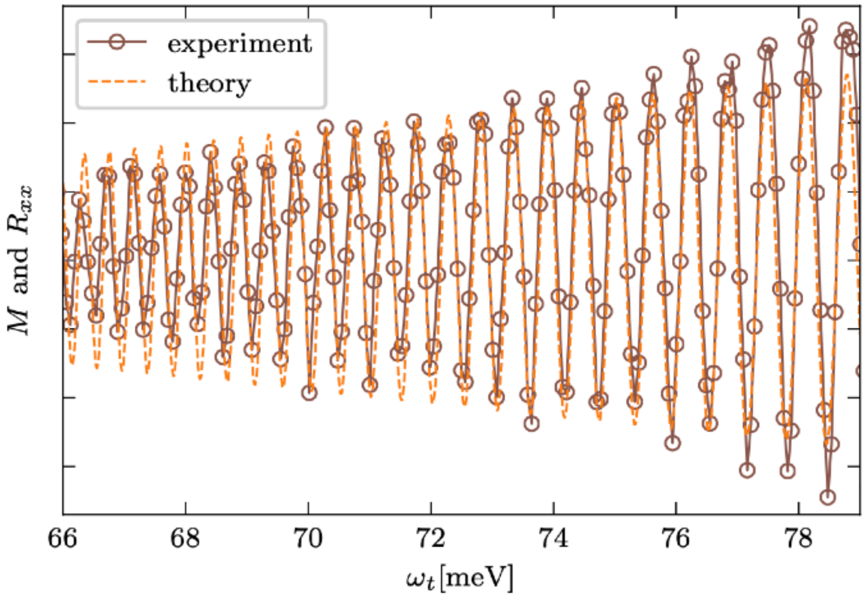
→ **Non-LK temperature decay**

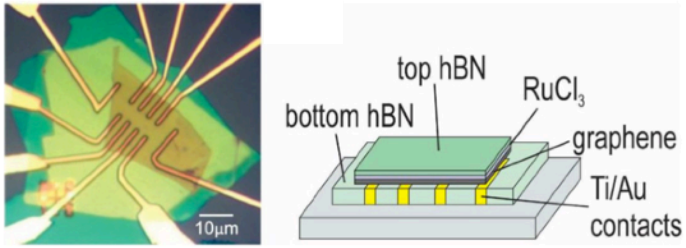
→ T_{max} set by Kondo scale J

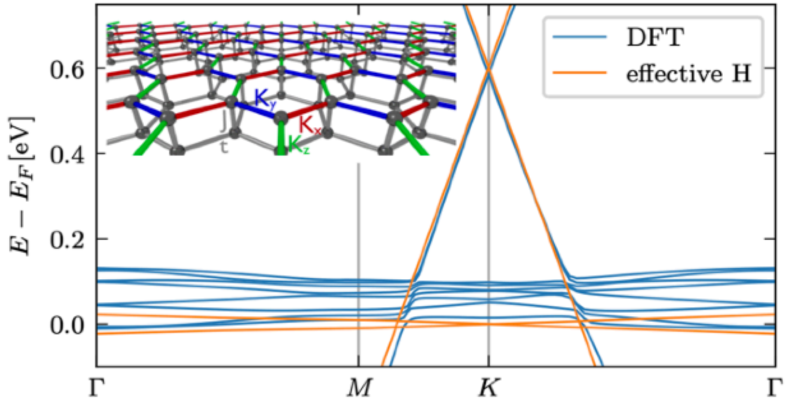


Anomalous QO in $RuCl_3$-Graphene

- **Ab-initio parameters**







→ W~600 meV from charge transfer accounts for the frequency

- **What about signatures of fractionalized QSL particles?**

→ charge neutral fermions acquire charge by effective hybridisation!

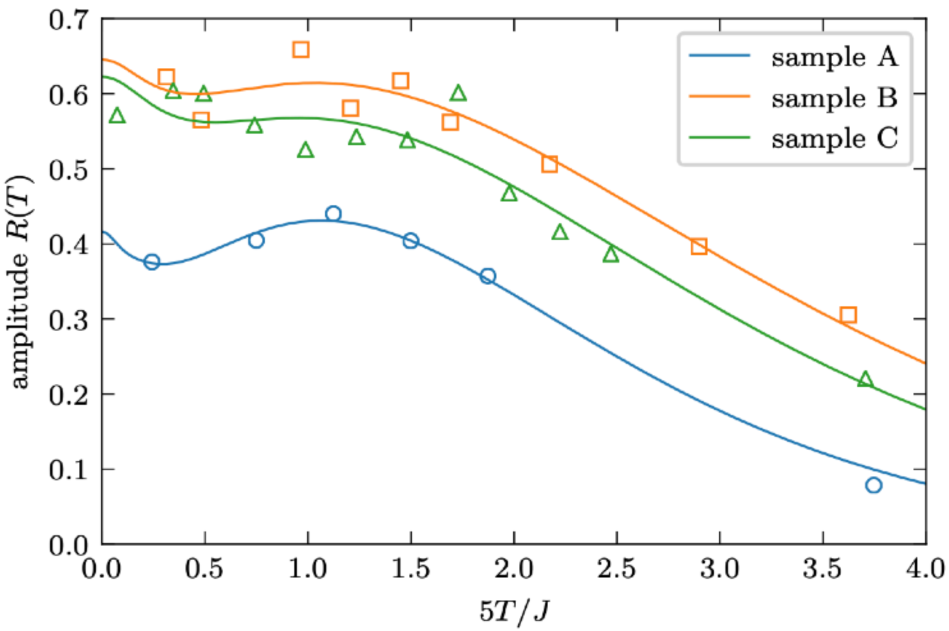
Anomalous QO in $RuCl_3$-Graphene

- **Non-Lifshitz-Kosevich temperature dependence**

$$R(T) = 2\chi \sum_{n=0}^{\infty} \mathrm{e}^{-2\chi\left(n+\frac{1}{2}\right)\Gamma\left(\frac{\mu}{J}, \frac{\omega_n}{J}\right)}$$

with $$\chi = 4\pi^2 \frac{TW}{\omega_t^2}$$

- **T_{max} set by Kondo coupling**



Interplay of itinerant Dirac electrons and fractionalized-excitations of the QSL may lead to non-LK behaviour!

→ What about scattering of spin fluctuations around T_N?

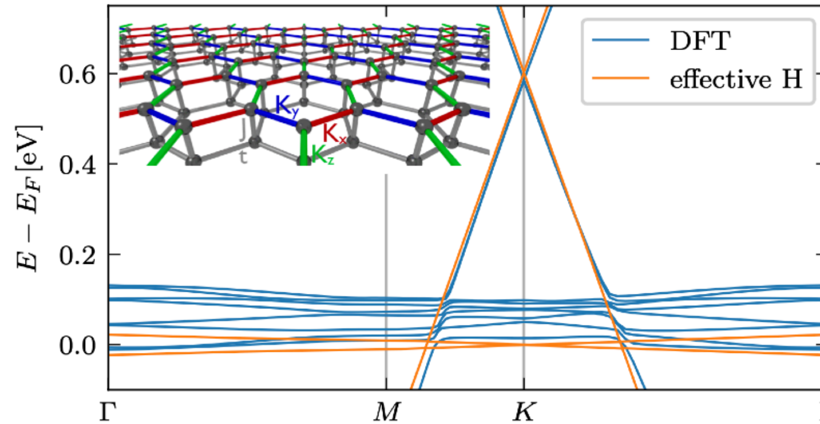
→ Test our scenario via pressure tuning, STM, other heterostructures …

Summary

- **Probing charge neutral excitations**

  → SP-STM as a local probe for charge neutral excitations

  Feldmeier, Natori, Knap, JK, *arXiv:2007.07912*



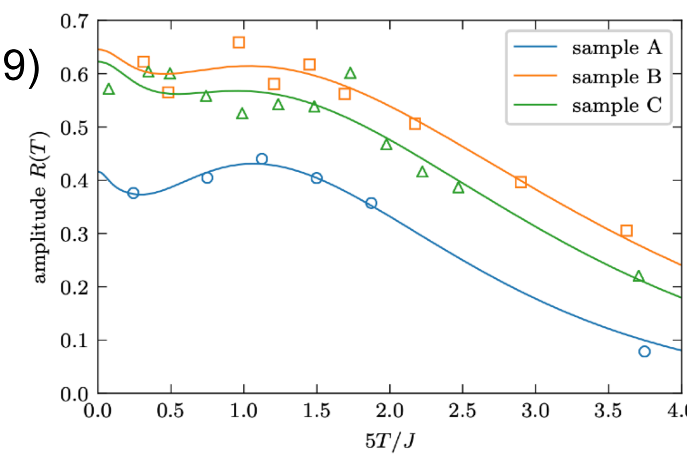
- **Kitaev heterostructures**

  → $RuCl_3$-graphene heterostructures

  Biswas, Winter, JK, Valenti PRL 123, 237201 (2019)

  → Interplay of electronic and fractionalized excitations

  → Anomalous QO and non-LK behaviour



Leeb, Polyudov, Mashhadi, Biswas, Valenti, Burghard, JK, *arXiv:2010.01649*

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Thank you!

Leeb, Polyudov, Mashhadi, Biswas, Valenti, Burghard, JK, *arXiv:2010.01649*

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Review Articles:

Fractionalization in Kitaev Quantum Spin Liquids
Hermanns, Kimchi, Knolle, *Annu. Rev. Condens. Matter Phys. 9 (2018)*

A Field Guide to Spin Liquids
Knolle, Moessner, *Annu. Rev. Condens. Matter Phys. 10 (2019)*