Title: Delafossite layered metals: intriguing physics in the high purity limit

Date: May 25, 2017 10:00 AM

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Abstract: In this talk I will introduce a relatively little-studied but intriguing family of metals, the delafossite series of layered oxides ABO2 in which the A site is occupied by Pd or Pt, and the B site by a transition metal. For reasons that are not perfectly understood, these materials have amazingly high electrical conductivity, with mean free paths of hundreds of angstroms (longer than even elemental copper or silver) at room temperature, growing to tens of microns at low temperatures. The electronic structure that yields these properties is in one way very simple, with a single half filled conduction band, but in another sense very rich, because the nearly free electrons originate mainly from the (Pt,Pd) layers in the crystal structure, while the adjacent transition metal oxide layers host Mott insulating states to which the conduction electrons also have some coupling. My group is interested in the delafossites for a number of reasons. Firstly, they are possible hosts for electronic transport at the crossover between ballistic and hydrodynamic regimes, which we investigate by fabricating size-restricted microstructures using focused ion beam techniques. As layered materials that can be cleaved at low temperatures, they are also well suited to study by angle resolved photoemission spectroscopy, and host a variety of interesting surface states in addition to a simple single-band bulk electronic structure. I will discuss our findings on non-magnetic PdCoO2, PtCoO2 and PdRhO2 and magnetic PdCrO2.

University of St Andrews

Max Planck Institute for Chemical Physics of Solids

Physics opportunities in ultra-pure delafossite oxide metals

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Collaborators

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Max Planck Institute for Chemical Physics of Solids, Dresden
University of St Andrews
UC Berkeley

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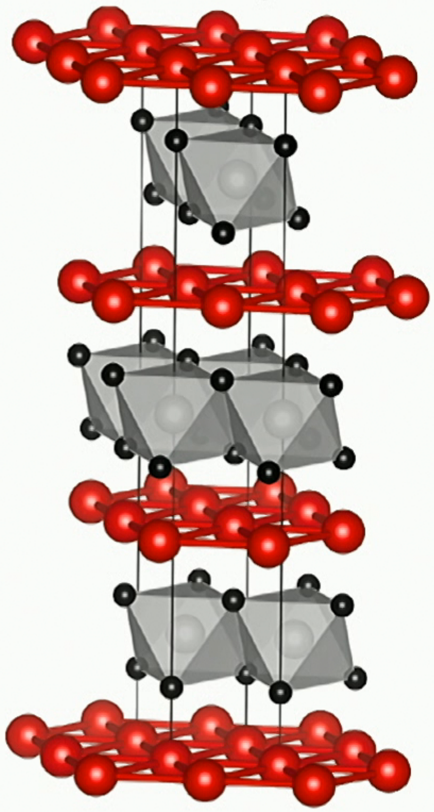
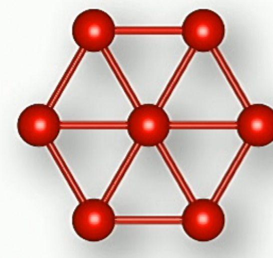
Contents

Metallic delafossites – layered metals based on triangular lattices

ABO_2



A sites:
- Pt, Pd
- Triangular lattice



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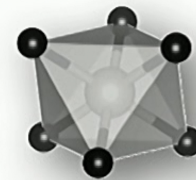

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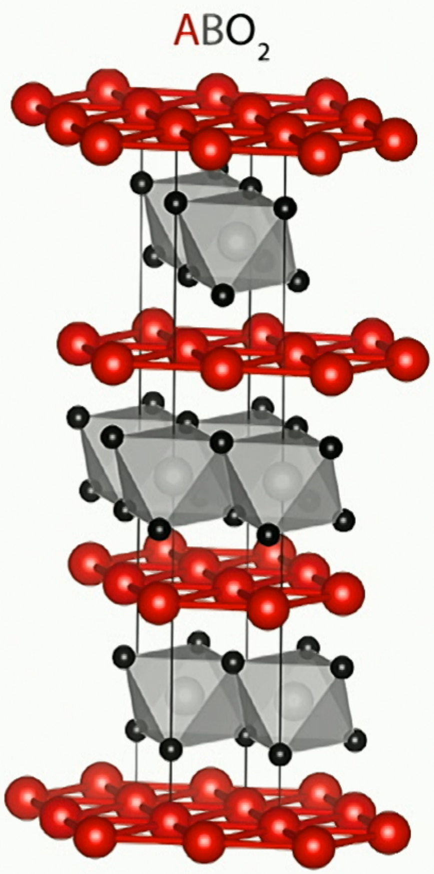

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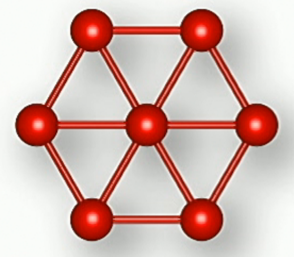

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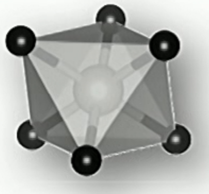

Nearly free electrons flowing in A site layers with strongly 2D conduction

Strong correlations in B site layers (U ~ 4 eV for Co, Cr, ~ 2 eV for Rh).

Metallic delafossites – layered metals based on triangular lattices

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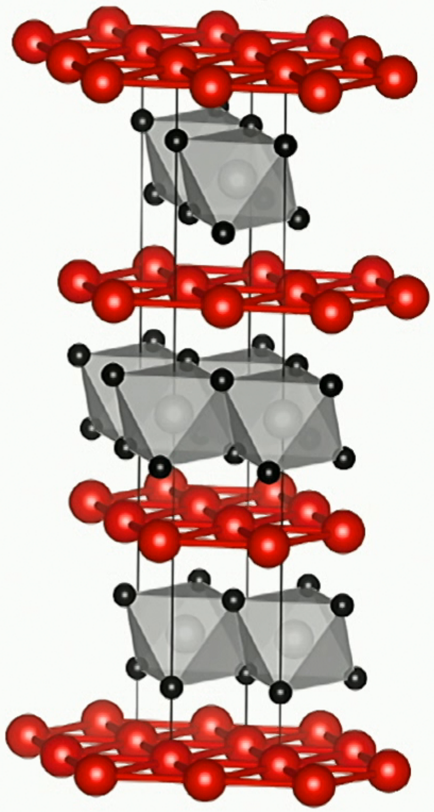

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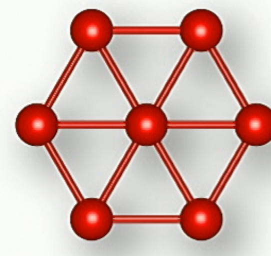

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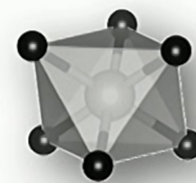

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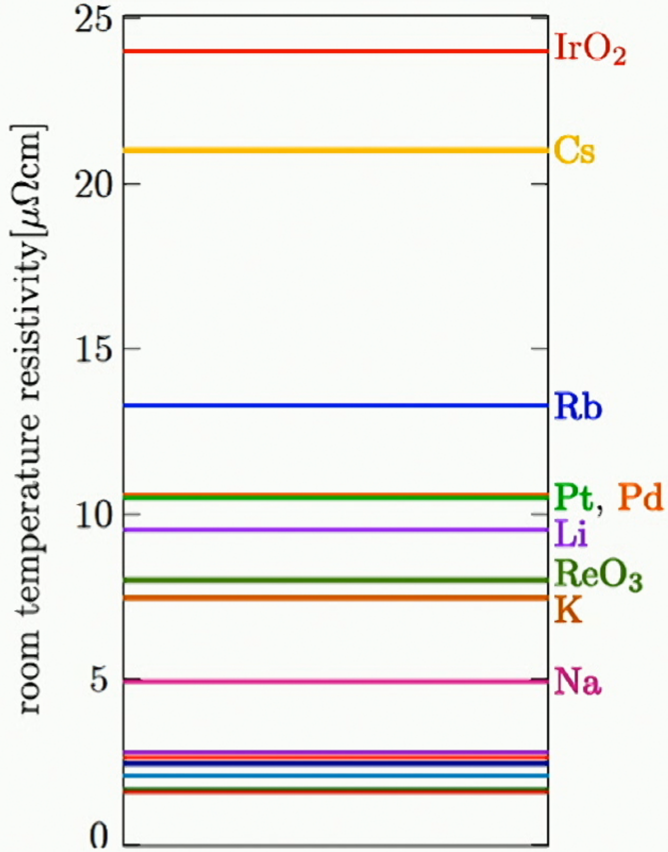
CrO_2 layer Mott insulating with spin 3/2 Cr^{3+}; Co^{3+} and Rh^{3+} in non-magnetic low spin configuration.

$PdCoO_2$ and $PtCoO_2$: record-breaking conductivity



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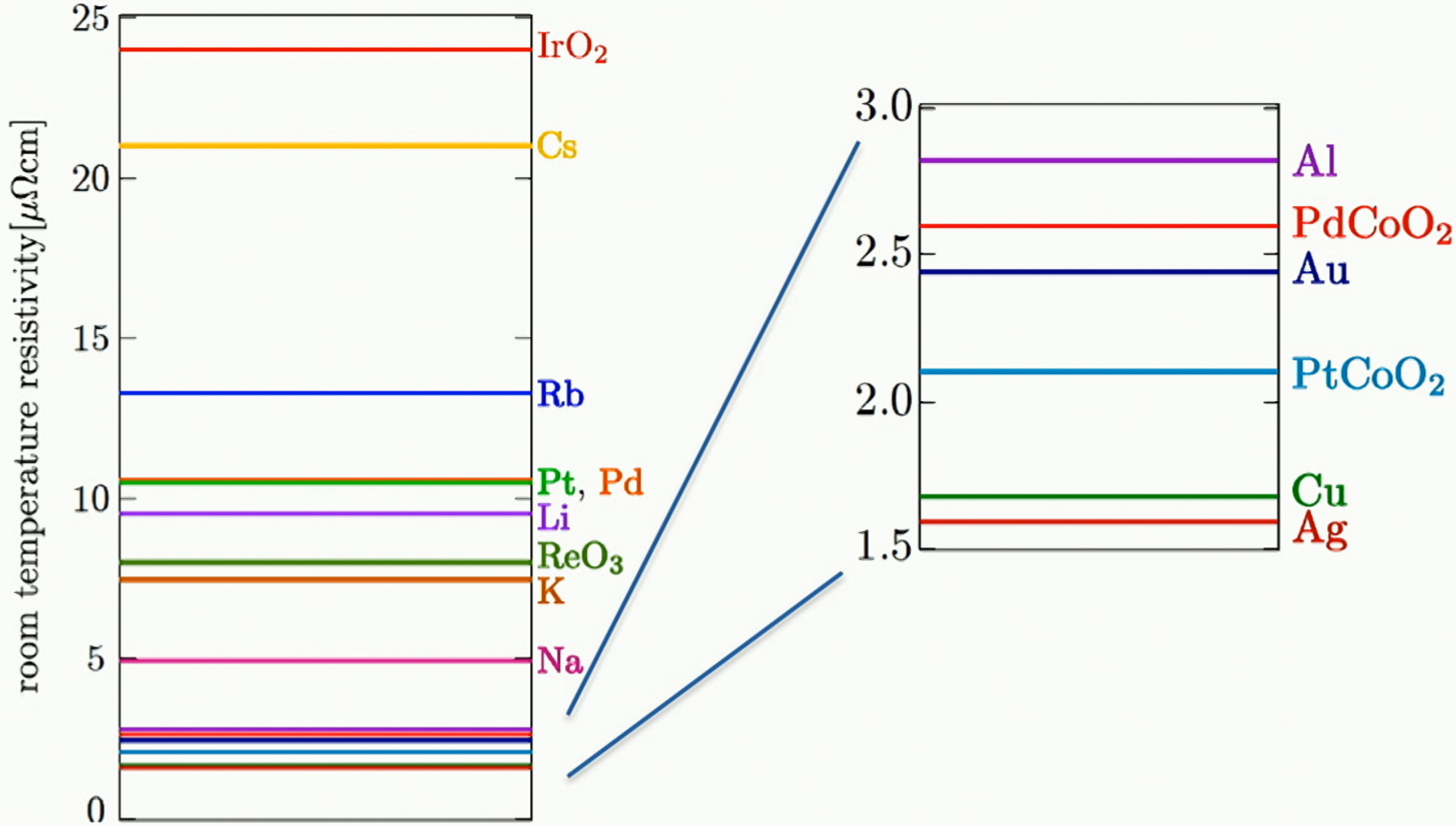

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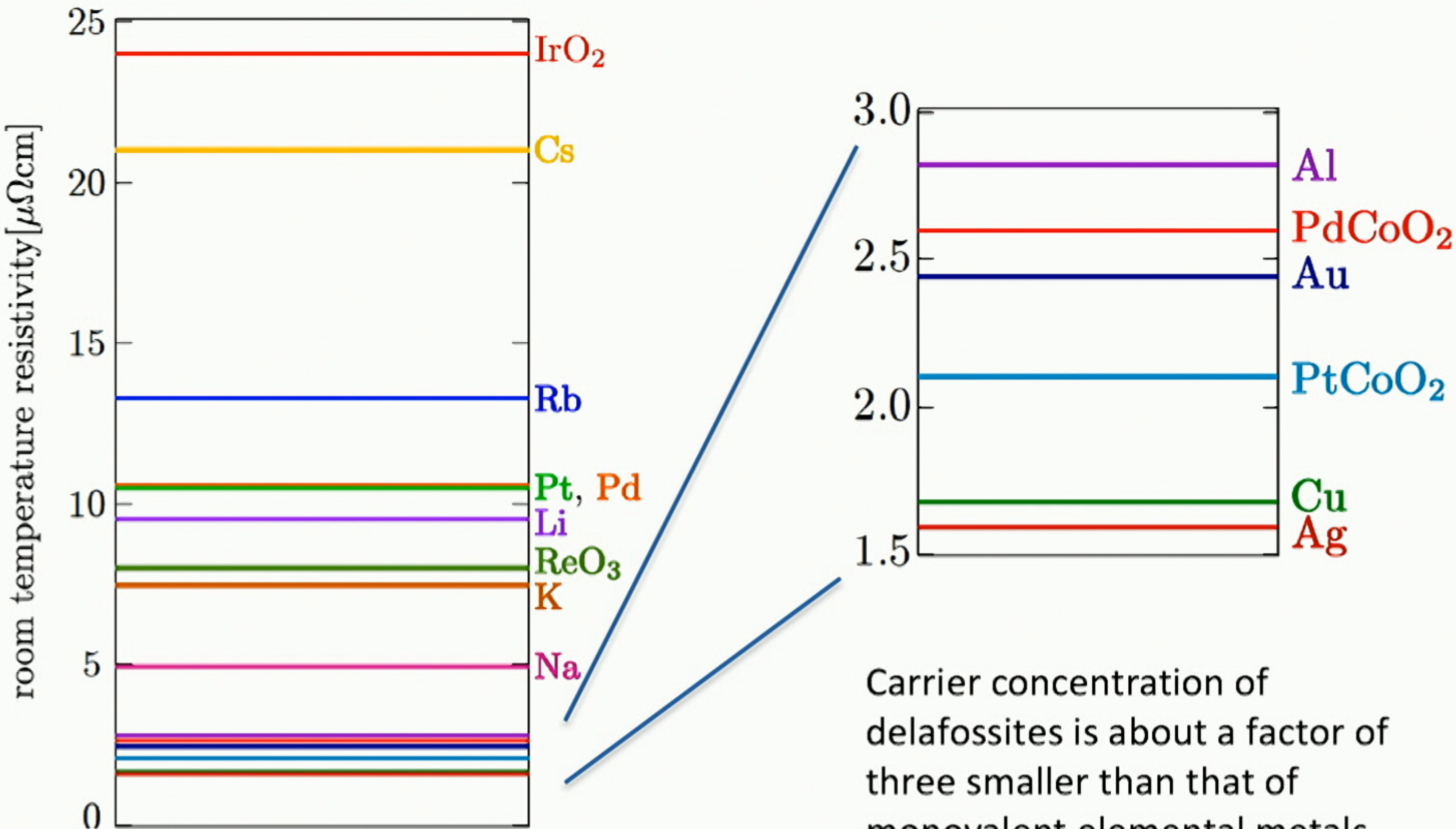
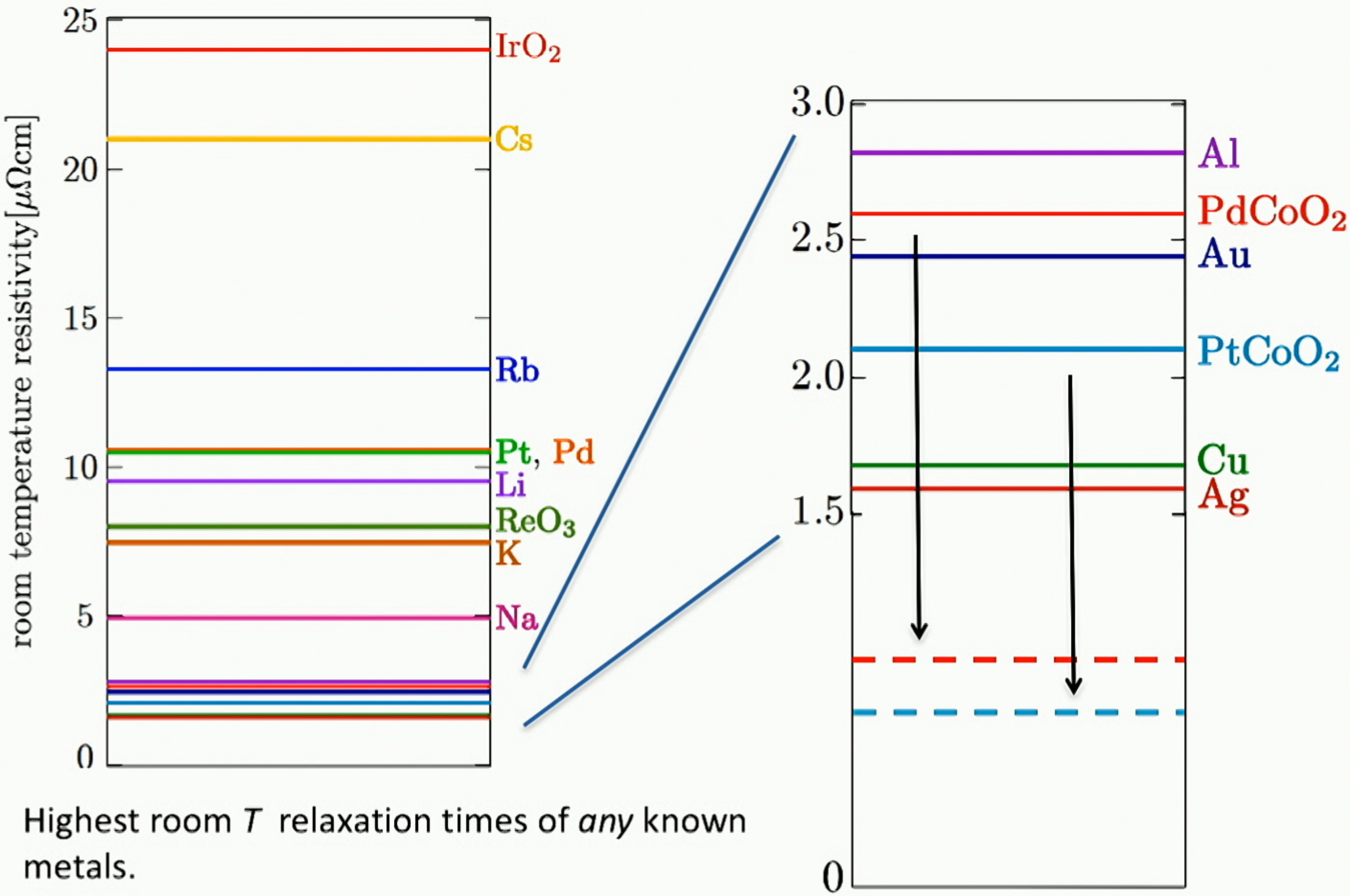


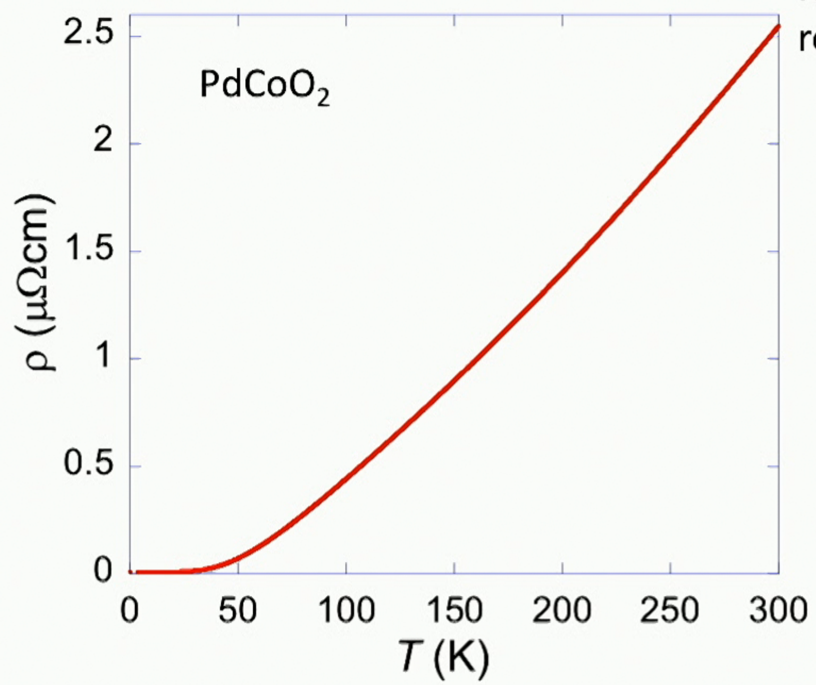
Carrier concentration of delafossites is about a factor of three smaller than that of monovalent elemental metals

$PdCoO_2$ and $PtCoO_2$: record-breaking conductivity



Highest room T relaxation times of *any* known metals.

Enormous low temperature conductivity and huge mean free paths



Room temperature resistivity about 2.5 μΩcm

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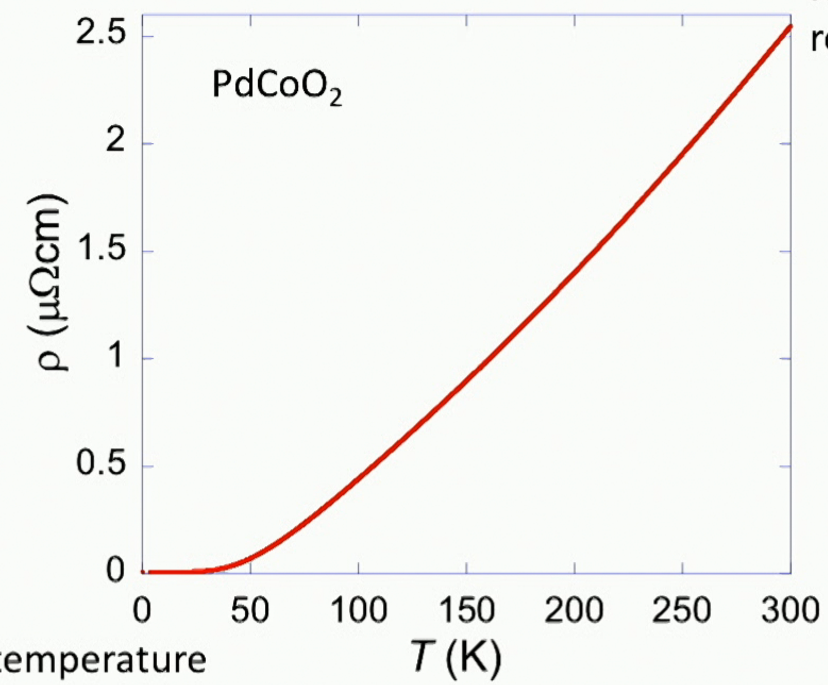
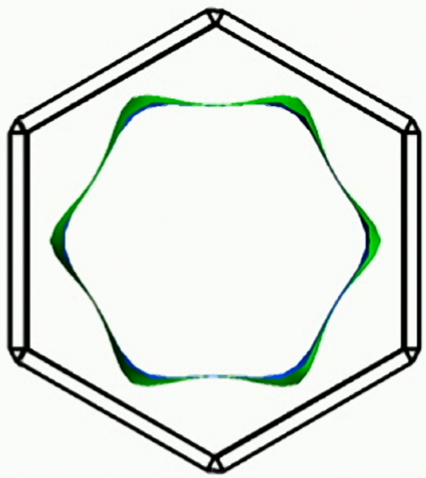


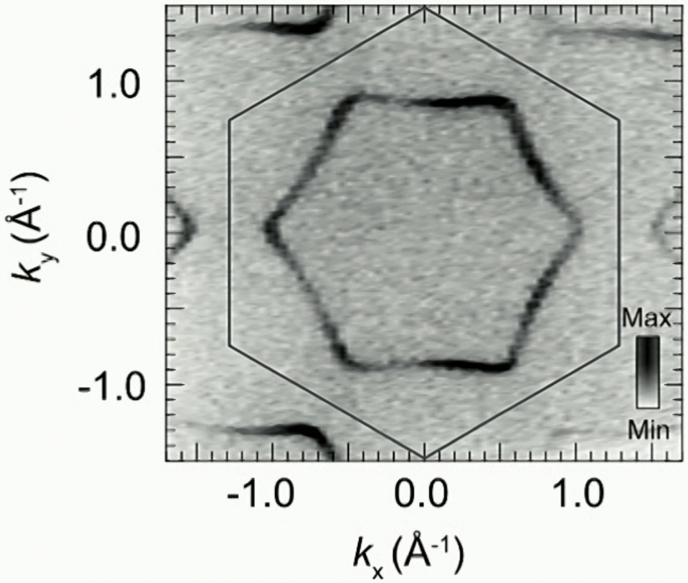
Room temperature resistivity about 2.5 μΩcm

Low temperature resistivity a few nΩcm: mean free paths of tens of microns!

Bulk Fermi surfaces of $PdCoO_2$ & $PtCoO_2$ from calculation and ARPES



*K.P. Ong, J. Zhang, J.S. Tse and P. Wu Phys. Rev. B **81**, 115120 (2010)*



*P. Kushwaha, V. Sunko et al., Science Advances **1**, 1500692 (2015).*

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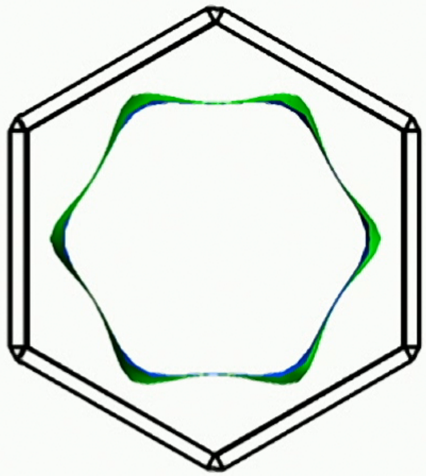

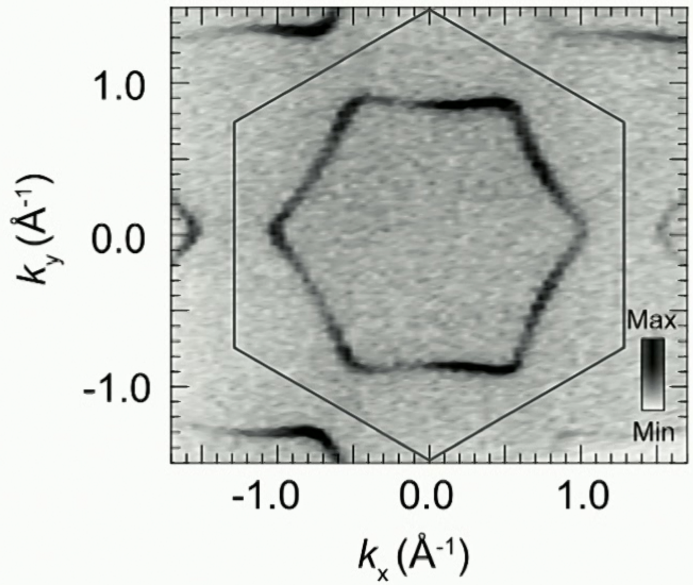

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Extremely simple electronic structure – only one band crosses the Fermi level.

Combined ARPES – dHvA now performed on $PdCoO_2$, $PtCoO_2$ and $PdCrO_2$.

*C.W. Hicks et al., Phys. Rev. Lett. **109**, 116401 (2012)*
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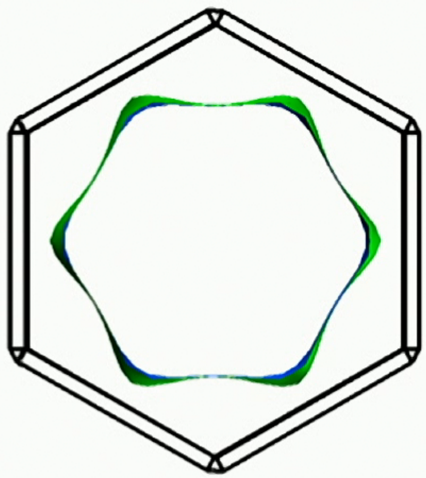

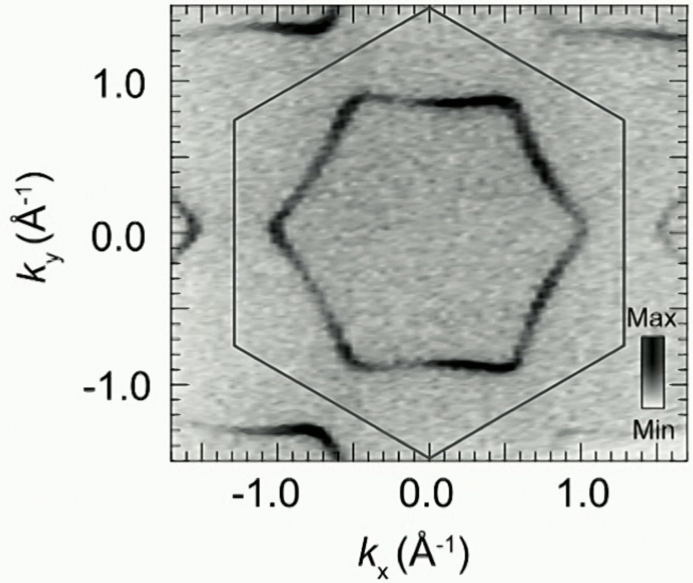

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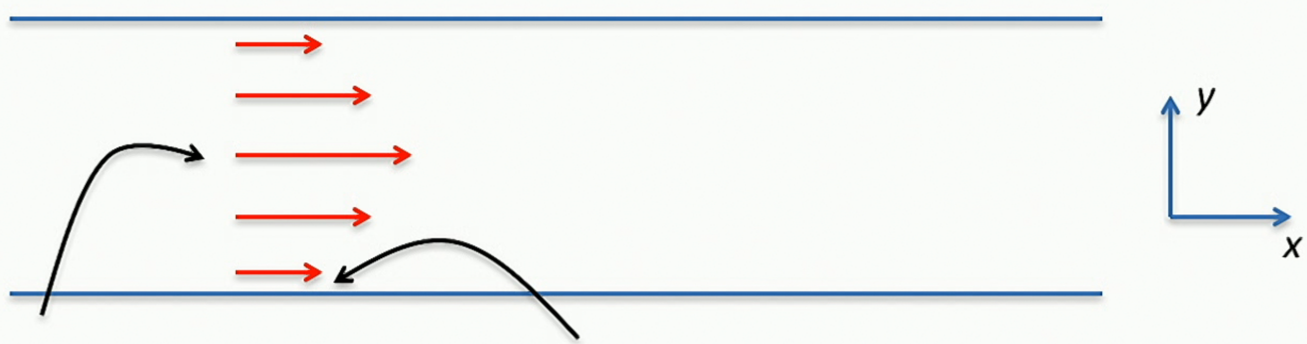
Bulk experiment on delafossites – search for electron hydrodynamics

What happens when we drive a classical fluid through a ‘2D pipe’ by applying a pressure gradient along *x*?



Fluid velocity is greater in the middle of the channel than at the edges

The only transfer of momentum to the outside world is at the edges

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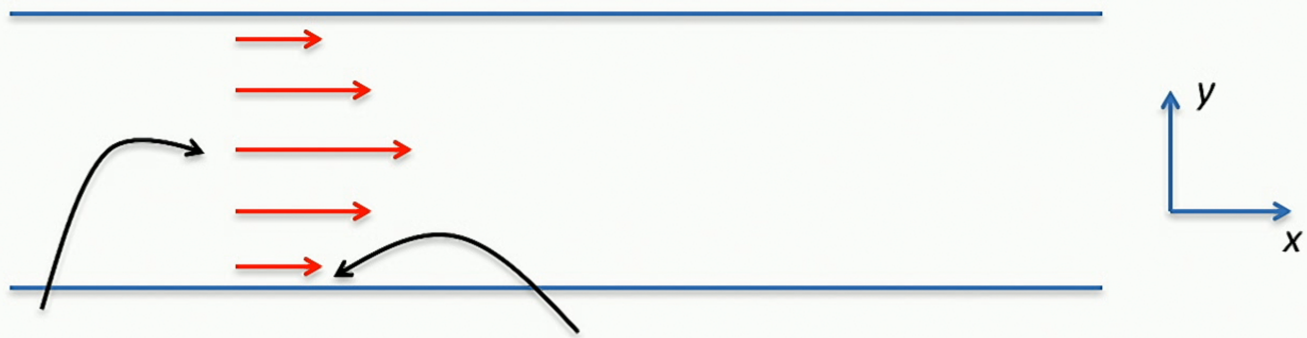

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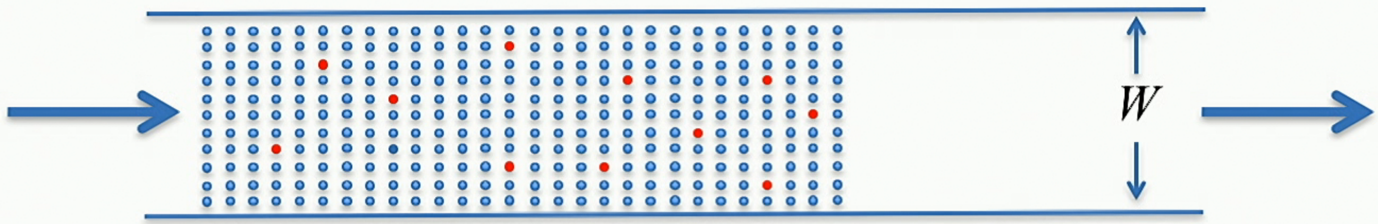
Counterintuitively for most condensed matter physicists, *η is proportional to the fluid particles' internal scattering time* τ

Why is electron hydrodynamics a challenge experimentally? Electrons flowing in a standard solid are *far* from the hydrodynamic regime



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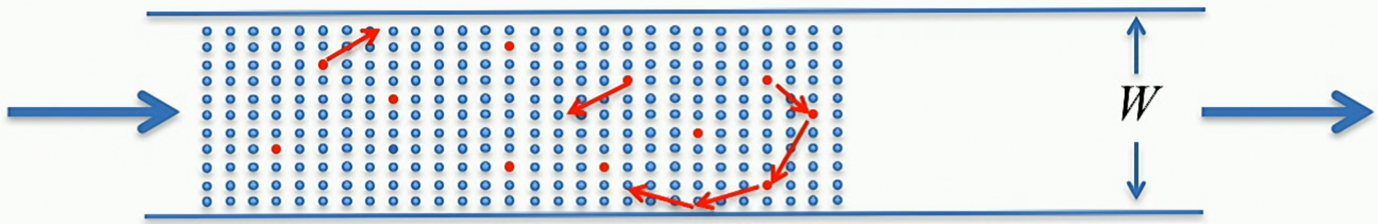
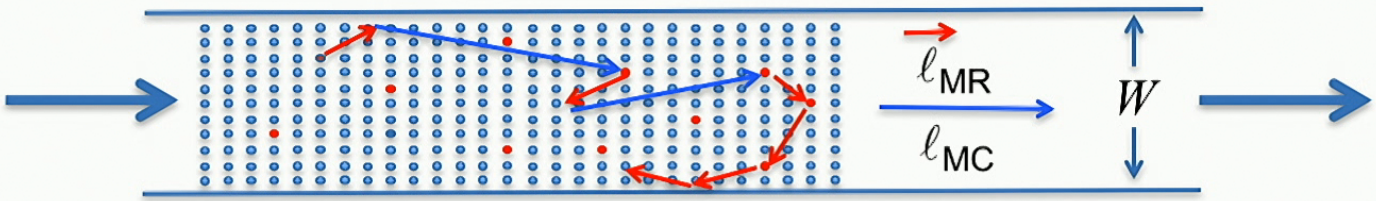


- Unlike the fluid in the empty tube, electrons in solids have many ways of making collisions in the bulk that relax the momentum to the solid.
- 'Squalid state physics' – 99.9999% of materials we work with are so dirty that electronic hydrodynamic corrections are negligible.
- In the standard theories of metals they are ignored altogether.

The 0.00001%

*R.N. Gurzhi, JETP **44**, 771 (1963); Usp. Fiz. Nauk **94**, 689 (1968)*

Key point introduced by Gurzhi: In solids, hydrodynamic effects can be parameterised in terms of the relationship between the three length scales: momentum relaxing mfp ℓ_{MR}, momentum conserving mfp ℓ_{MC} and sample dimension (here *W*).



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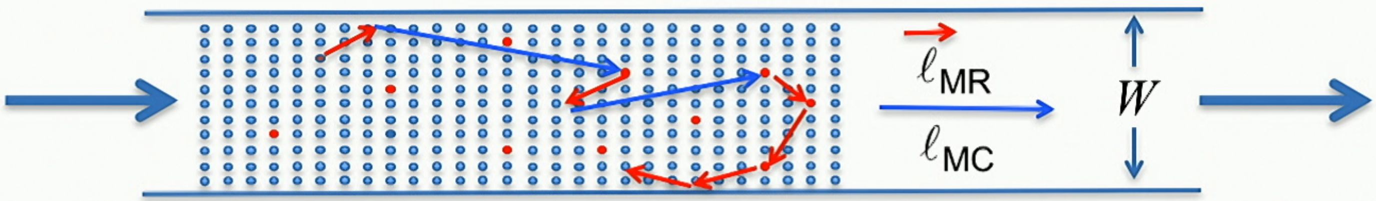

$\ell_{MR} << \ell_{MC}, W$

Standard ohmic theory applies;
R is determined entirely by solid resistivity ρ and usual geometrical factors

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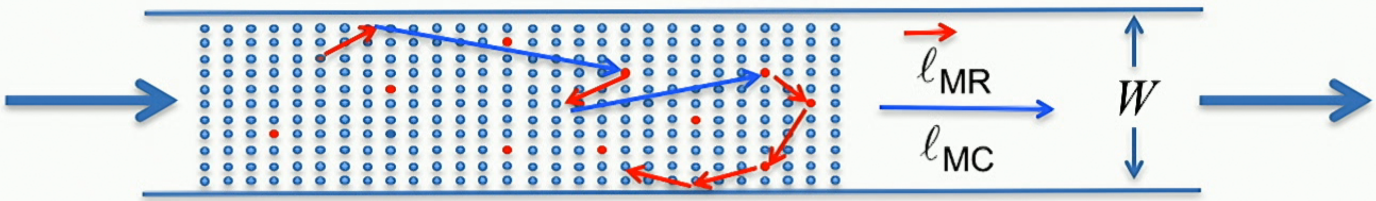

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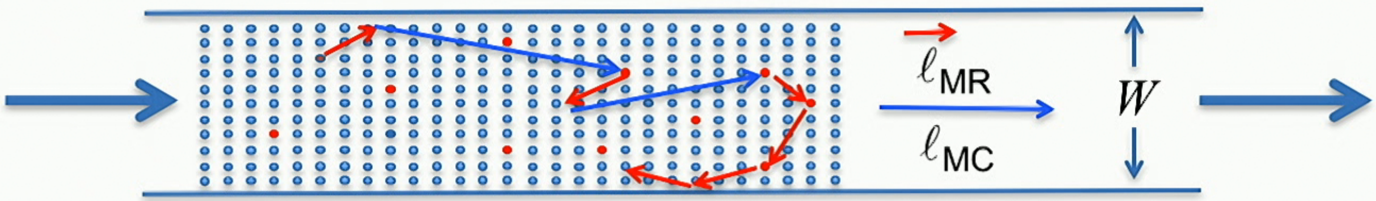

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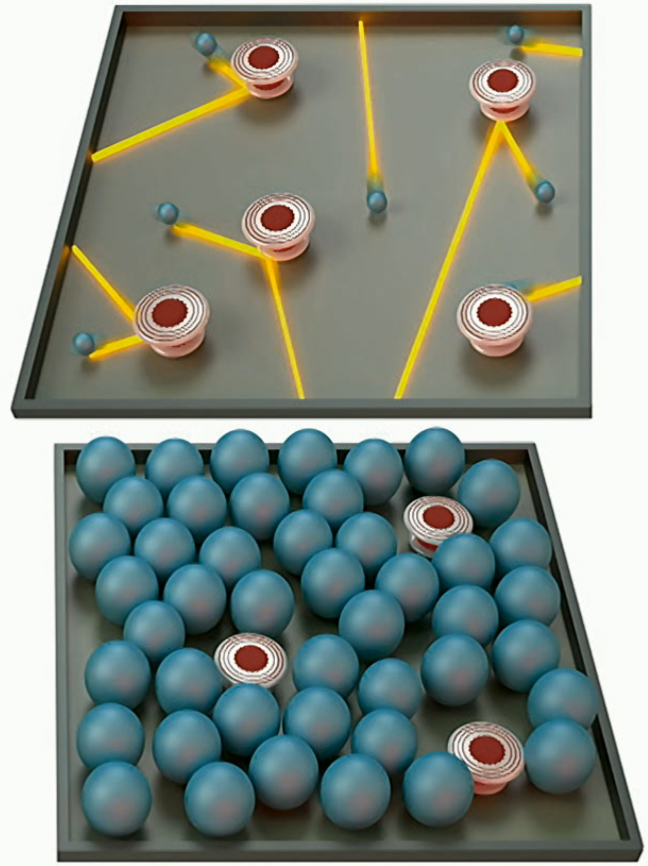
Tentative experimental observation : *Z.-Z. Yu et al., Phys. Rev. Lett. **52**, 368 (1984)*

Hydrodynamics experiments reported in 2016

Negative local resistance caused by viscous electron backflow in graphene *D. A. Bandurin, I. Torre, R. Krishna Kumar, M. Ben Shalom, A. Tomadin, A. Principi, G. H. Auton, E. Khestanova, K. S. Novoselov, I. V. Grigorieva, L. A. Ponomarenko, A. K. Geim, M. Polini, Science **351**, 1055 (2016)*

Observation of the Dirac fluid and the breakdown of the Wiedemann-Franz law in graphene *J. Crossno, J. K. Shi, K. Wang, X. Liu, A. Harzheim, A. Lucas, S. Sachdev, P. Kim, T. Taniguchi, K. Watanabe, T. A. Ohki & K. C. Fong, Science **351**, 1058 (2016)*

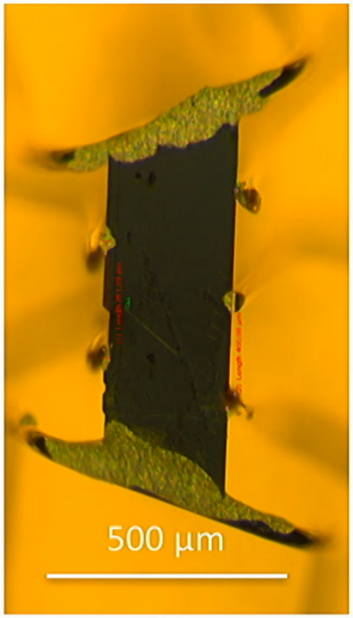
Evidence for hydrodynamic electron flow in $PdCoO_2$ *P.J.C. Moll, P. Kushwaha, N. Nandi, B. Schmidt and A.P. Mackenzie, Science **351**, 1061 (2016)*



Commentary on graphene and $PdCoO_2$ papers *J. Zaanen, Science **351**, 1058 (2016)*

Focused ion beam sculpting – a world of new possibilities

20th century
crystal mounting



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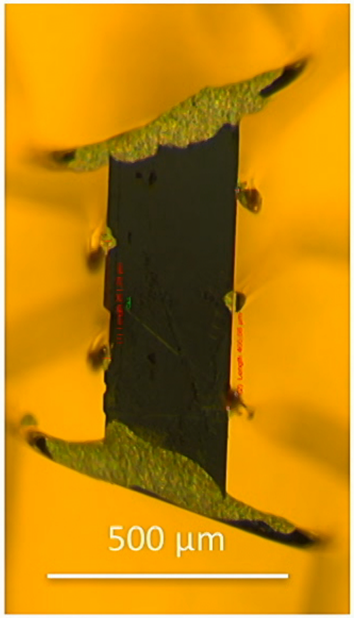
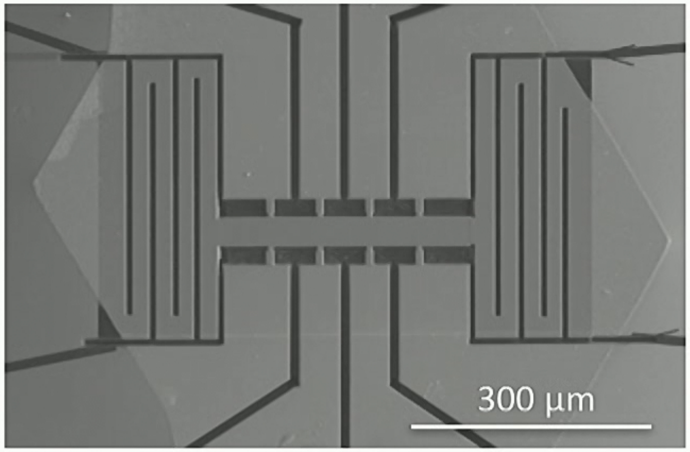


The 21st century approach



Both 'devices' made by N. Nandi

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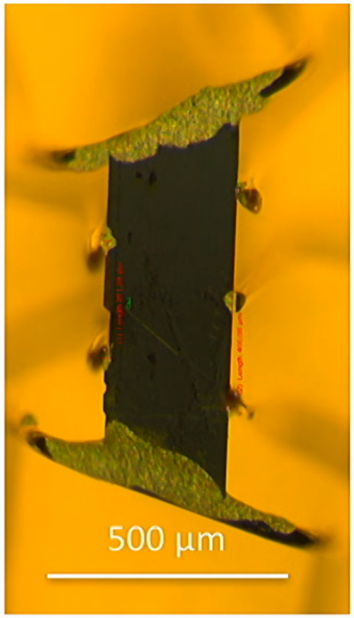

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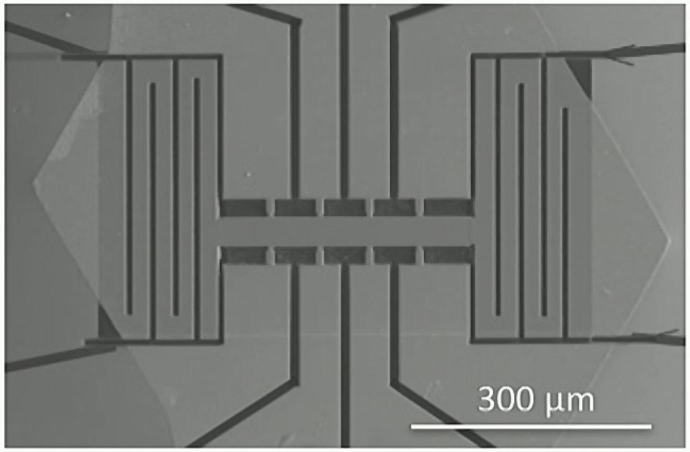

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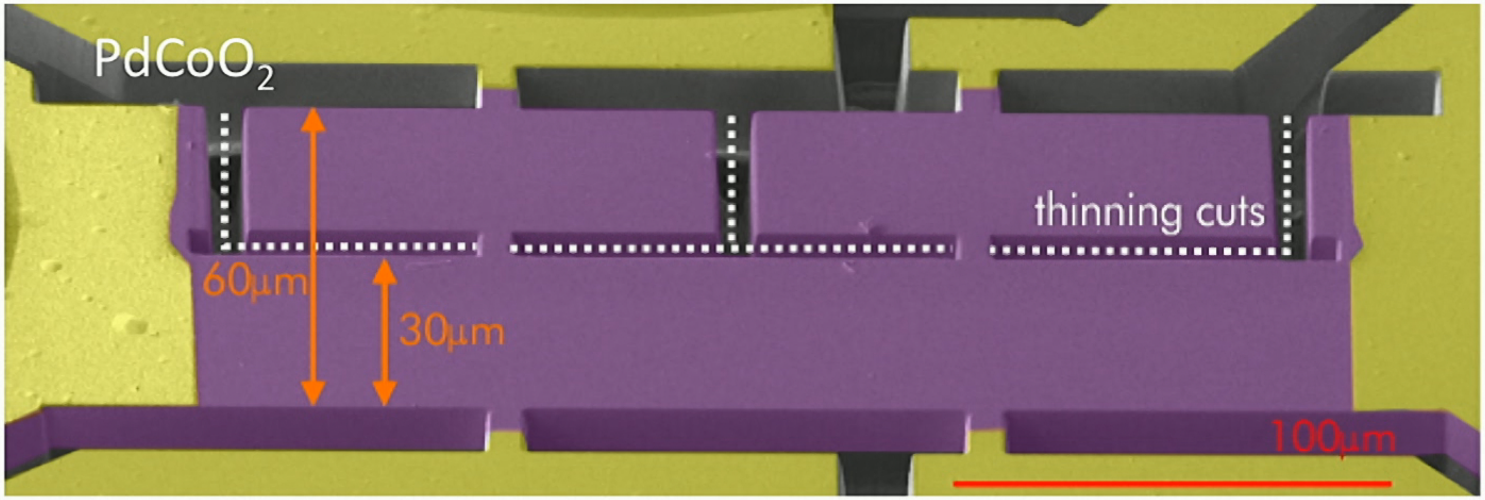
For a review of the state-of-the-art see *P.J.W. Moll, preprint, to appear in Annual Reviews of Condensed Matter Physics*

Search for signatures of hydrodynamic flow in well-defined microstructures



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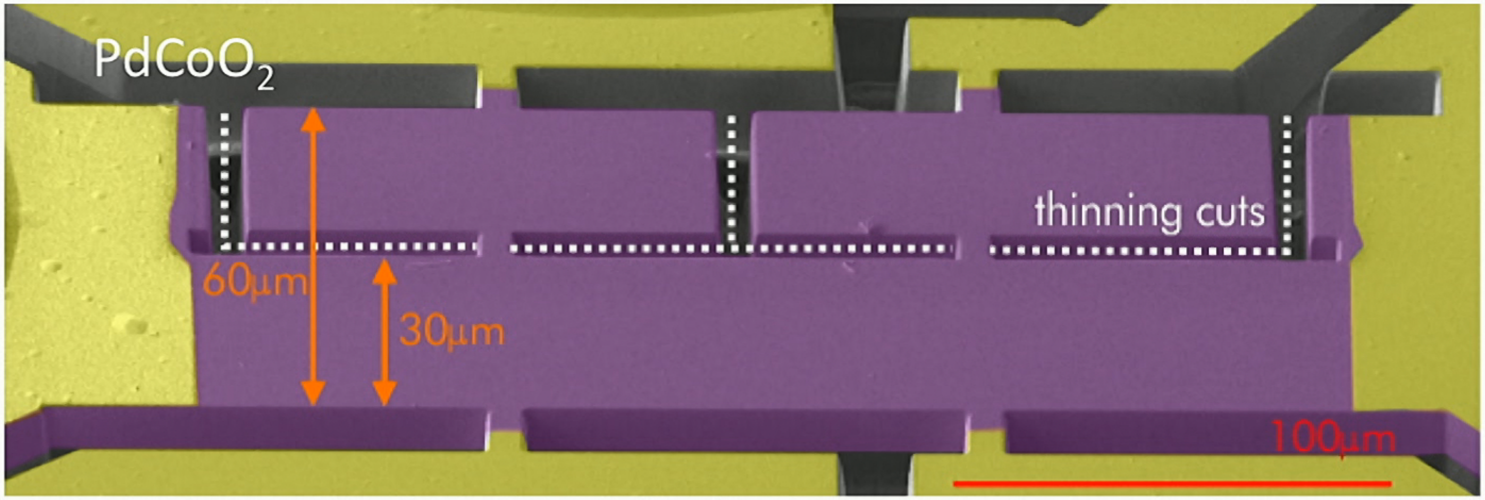
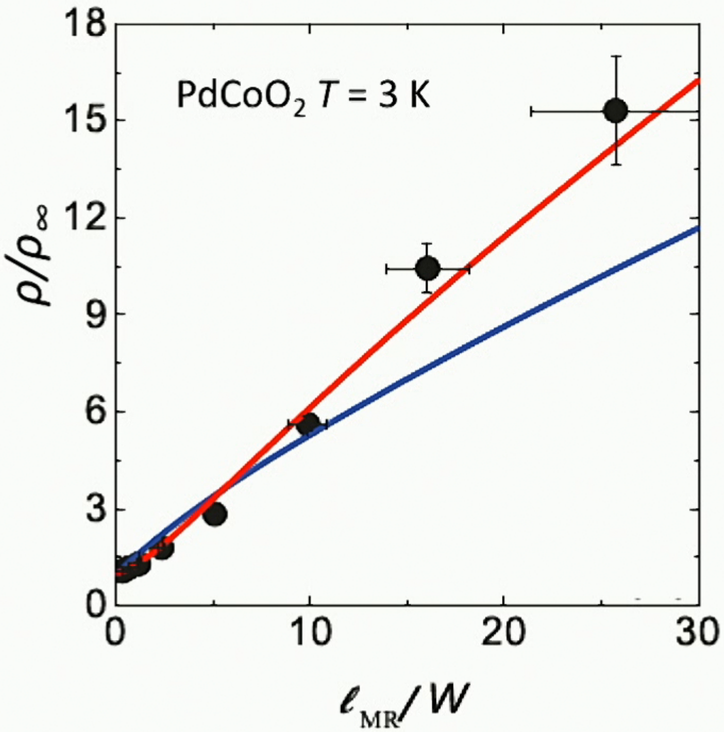


Experiment: Successively narrow the channel in factors of 2, measuring the resistance after every step.

*P.J.W. Moll, P. Kushwaha, N. Nandi, B. Schmidt and A.P. Mackenzie, Science **351**, 1061 (2016)*

Width dependence of channel resistance analysed using Boltzmann theory including momentum-conserving collisions



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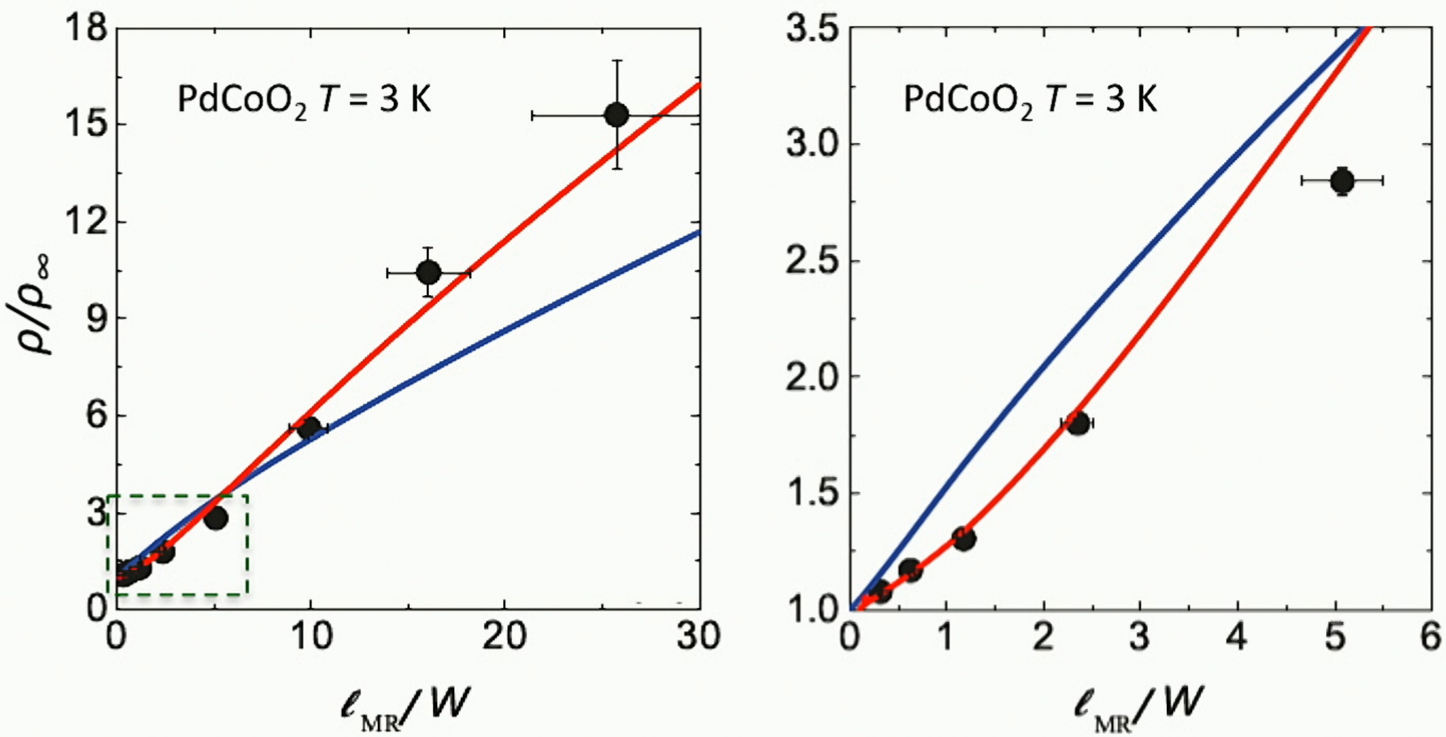

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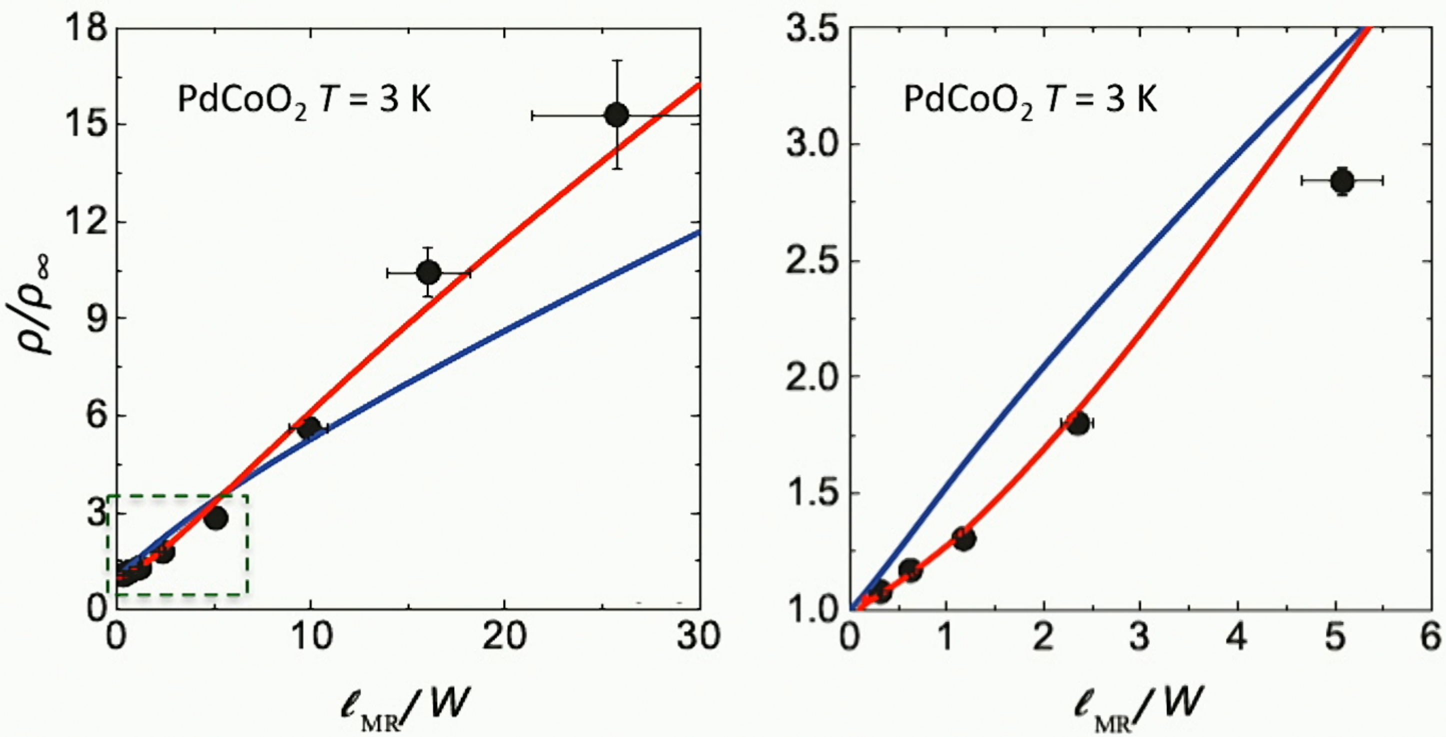

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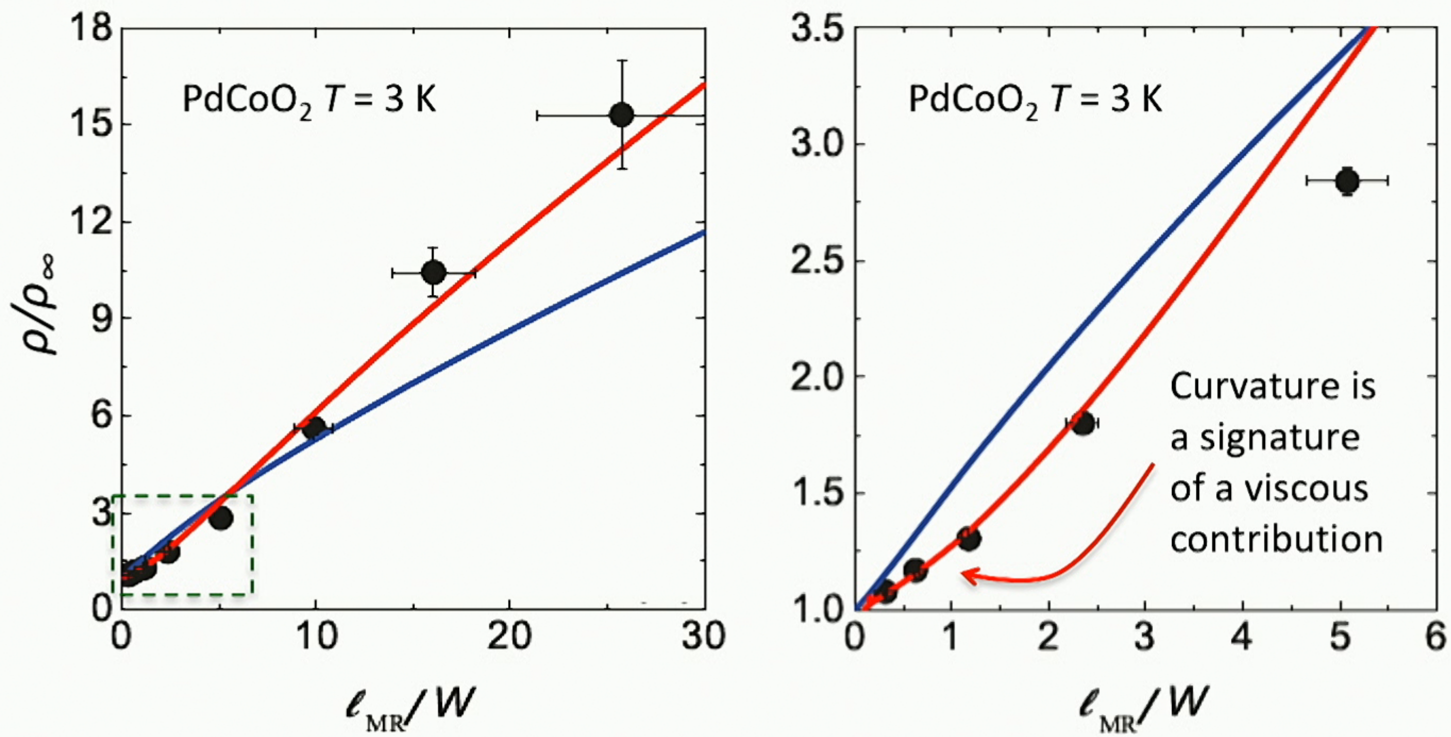

Theoretical approach of this kind first used in *M.J.M de Jong & L.W. Molenkamp, Phys. Rev. B* ***51****, 13389 (1995)*

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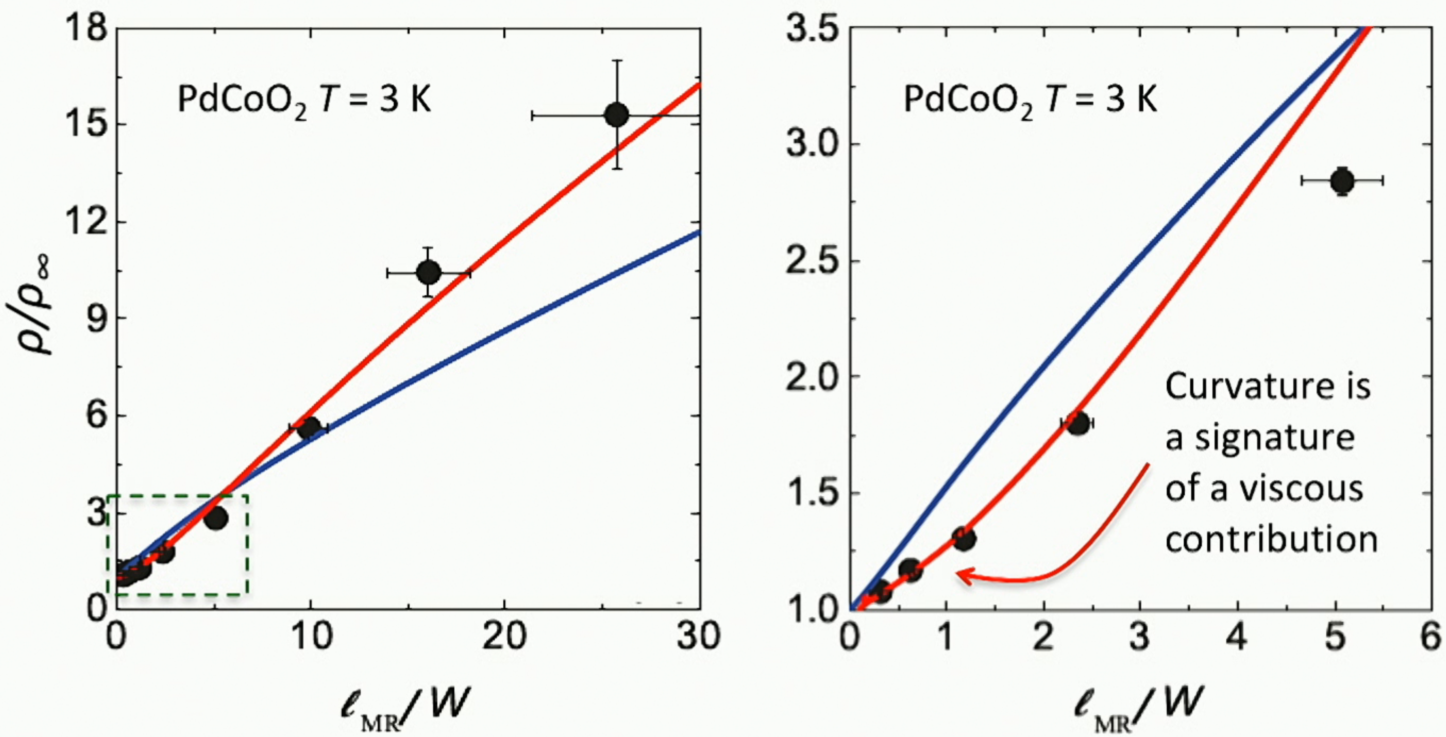

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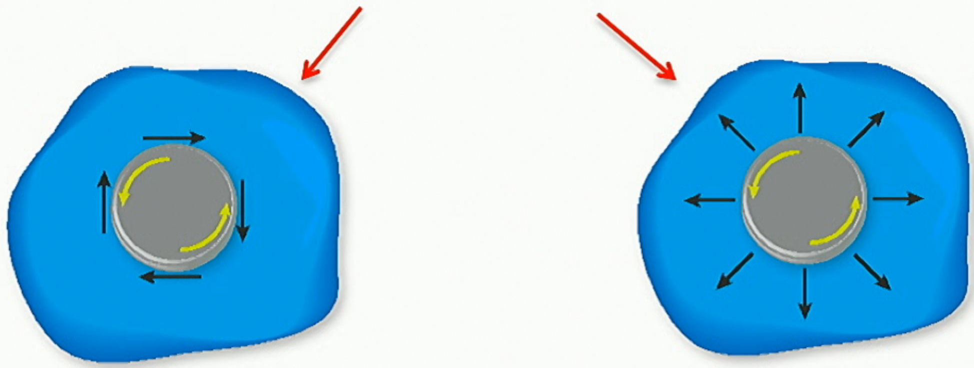
Next step – extend electron hydrodynamic theory to include magnetic field

Now *two* viscosities to consider: standard shear viscosity and Hall viscosity

$$\partial_t \vec{v} = \eta_{xx} \nabla^2 \vec{v} + \eta_{xy} \nabla^2 \vec{v} \times \vec{z}$$



η_{xy} only exists when time reversal symmetry is broken. Of considerable interest in gapped topological systems. See e.g. *T. Hughes et al., Phys. Rev. D* ***88****, 025040 (2013)*

Discussed recently for electrons using classical hydrodynamic theory e.g.
P.S. Alekseev, Phys. Rev. Lett. ***117****, 166601 (2016)*
S. Ganeshan and A. G. Abanov, arXiv:1703.04522

More useful approach for comparison with real materials – add magnetic field to the extended Boltzmann theory

Ohmic: $\ell_{MR} << W, \ell_{Mc}$ Hydrodynamic: $\ell_{MC} << W << \ell_{MR}$ Ballistic: $W << \ell_{MR}, \ell_{MC}$

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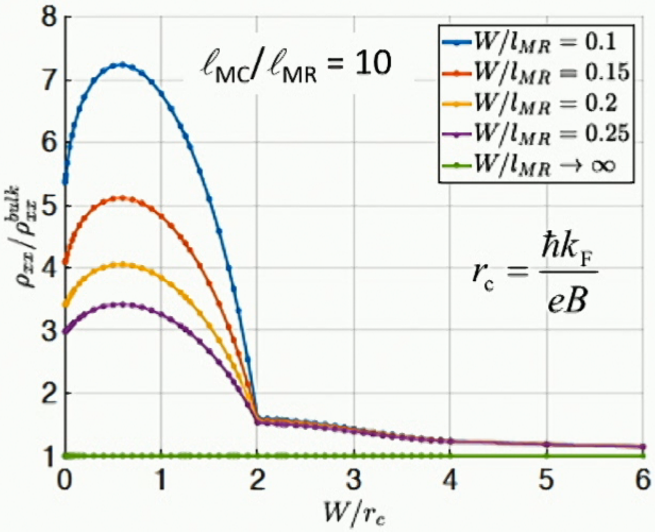

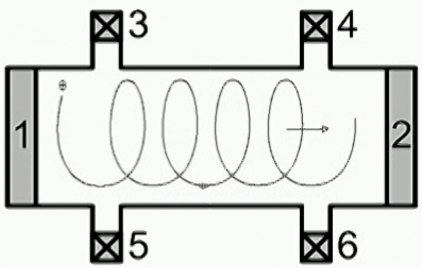

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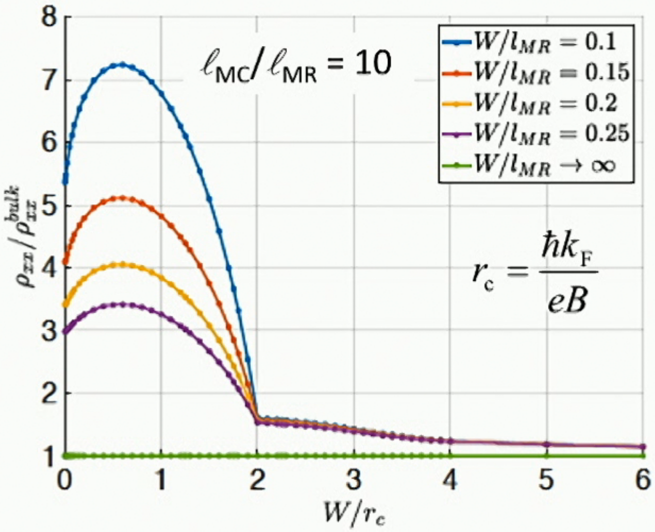

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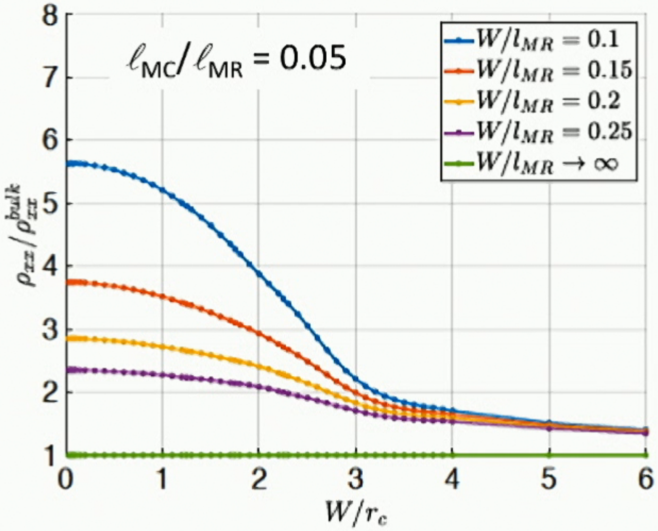
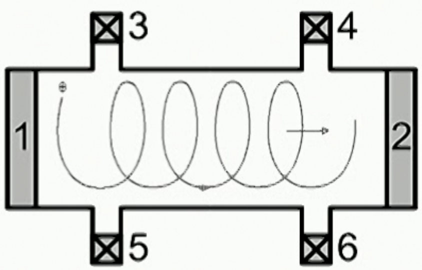
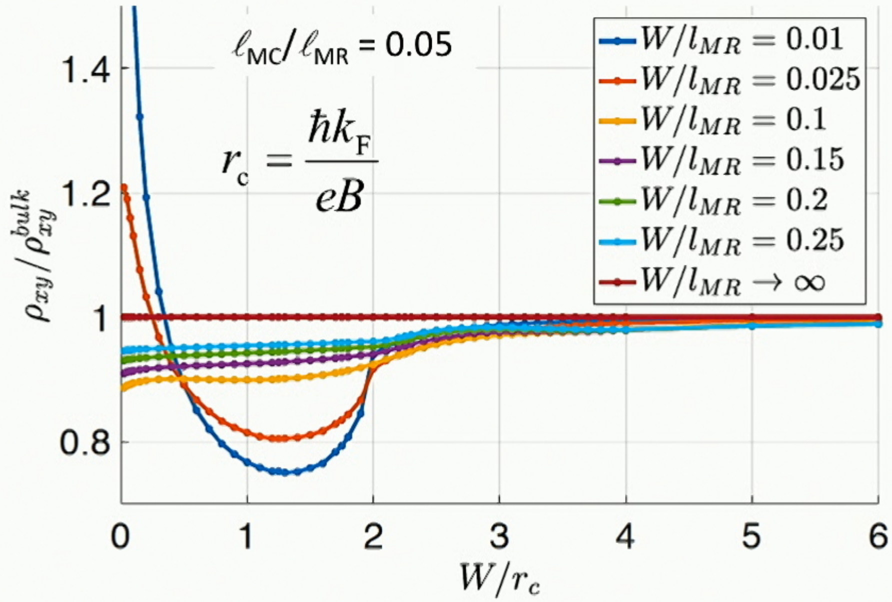




Hall resistivity distinguishes clearly between the three regimes

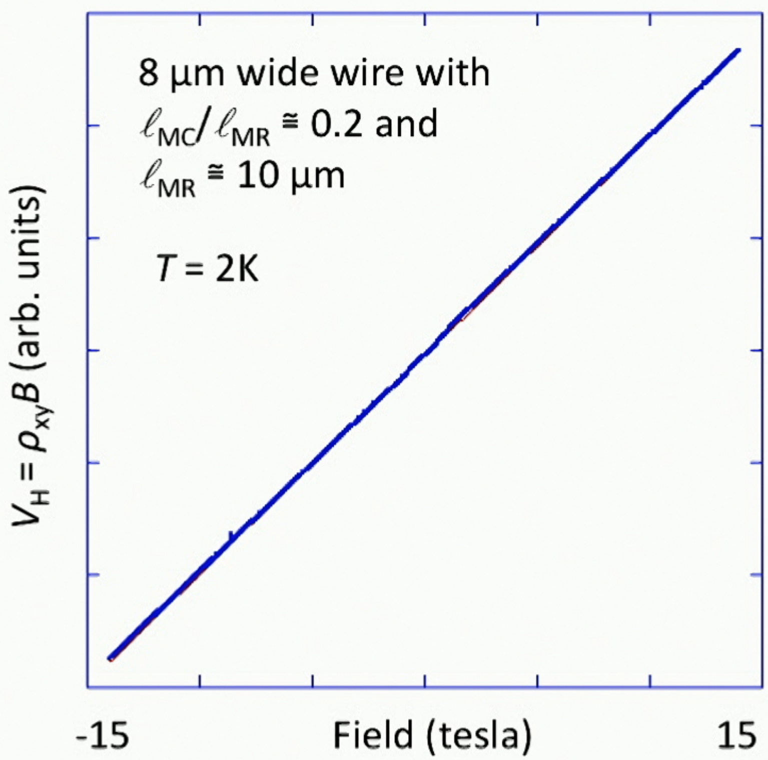


Thomas Scaffidi,
UC Berkeley

Ohmic, hydrodynamic and then ballistic behaviour as W/r_c is reduced from infinity.

T. Scaffidi, N. Nandi, B. Schmidt, A.P. Mackenzie and J.E. Moore, arXiv:1703.07325, to appear in Phys. Rev. Lett.

Work in progress on $PtCoO_2$



Now working on samples controlled to exquisite precision, mounting evidence for first successful direct measurements of Hall viscosity.

N. Nandi, S. Khim, P. Kushwaha, T. Scaffidi, M. König, B. Schmidt, P.J.W. Moll, J.E. Moore and A.P. Mackenzie, to be published

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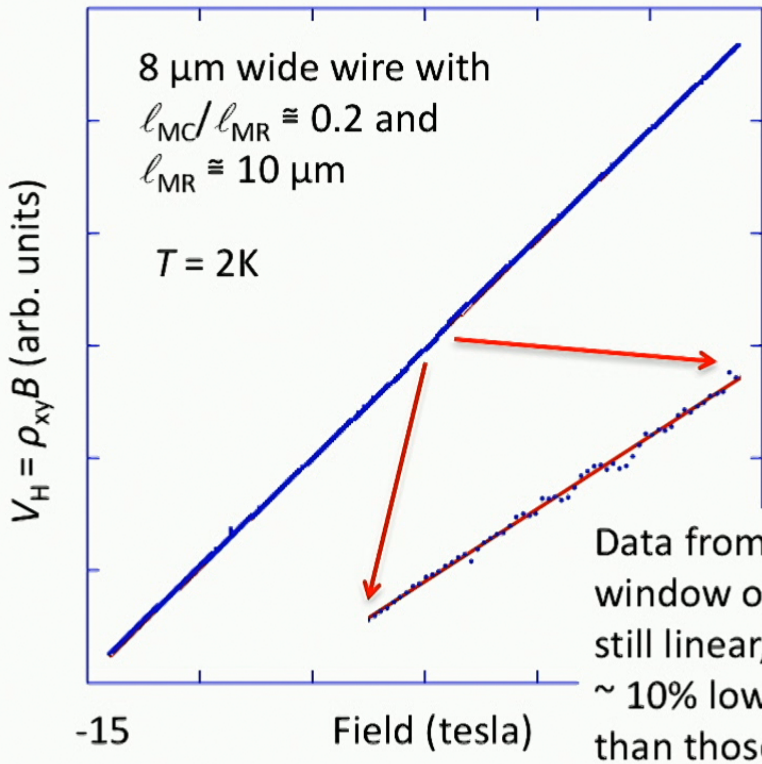


Data from a narrow window of ± 0.1 T are still linear, but with ~ 10% lower gradient than those at high fields

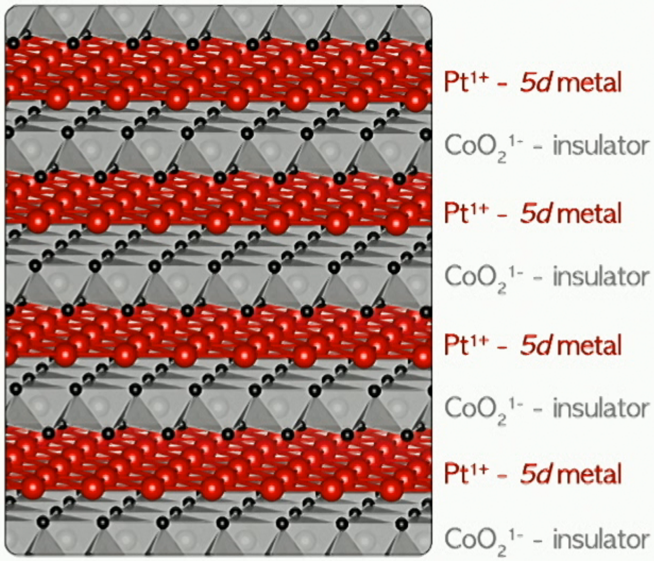
Now working on samples controlled to exquisite precision, mounting evidence for first successful direct measurements of Hall viscosity.

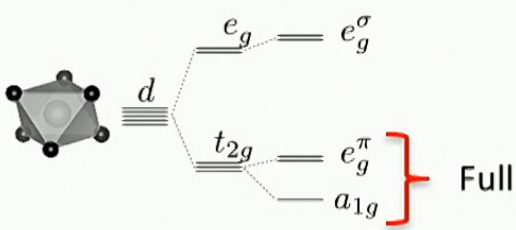
Nabhanila Nandi

N. Nandi, S. Khim, P. Kushwaha, T. Scaffidi, M. König, B. Schmidt, P.J.W. Moll, J.E. Moore and A.P. Mackenzie

Surface states in delafossites

Bulk





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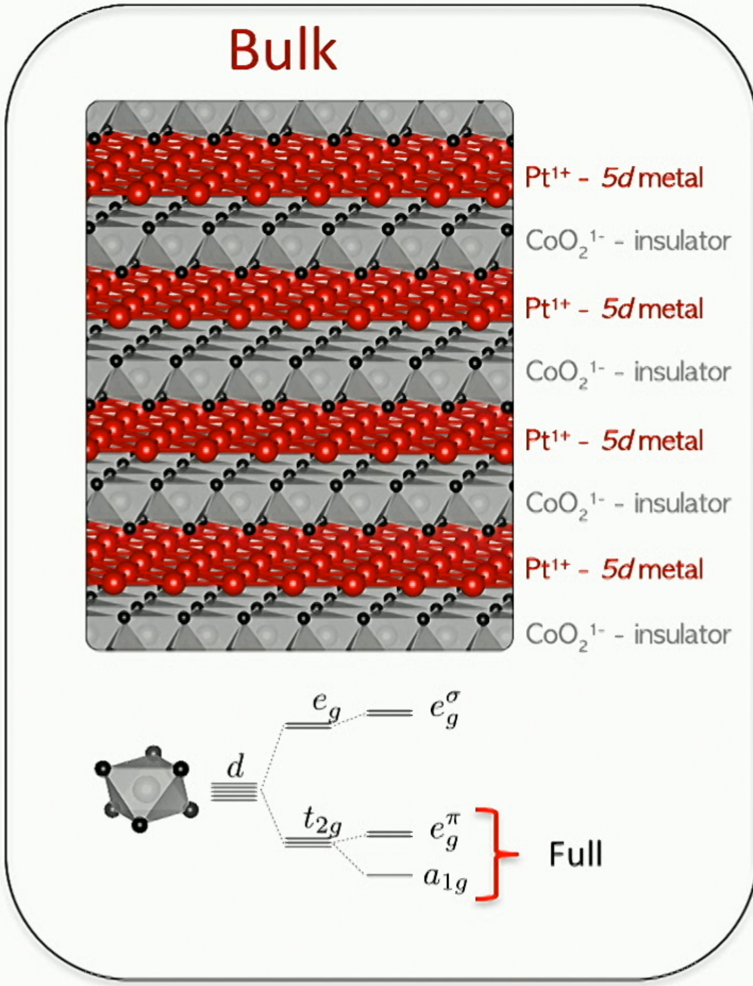
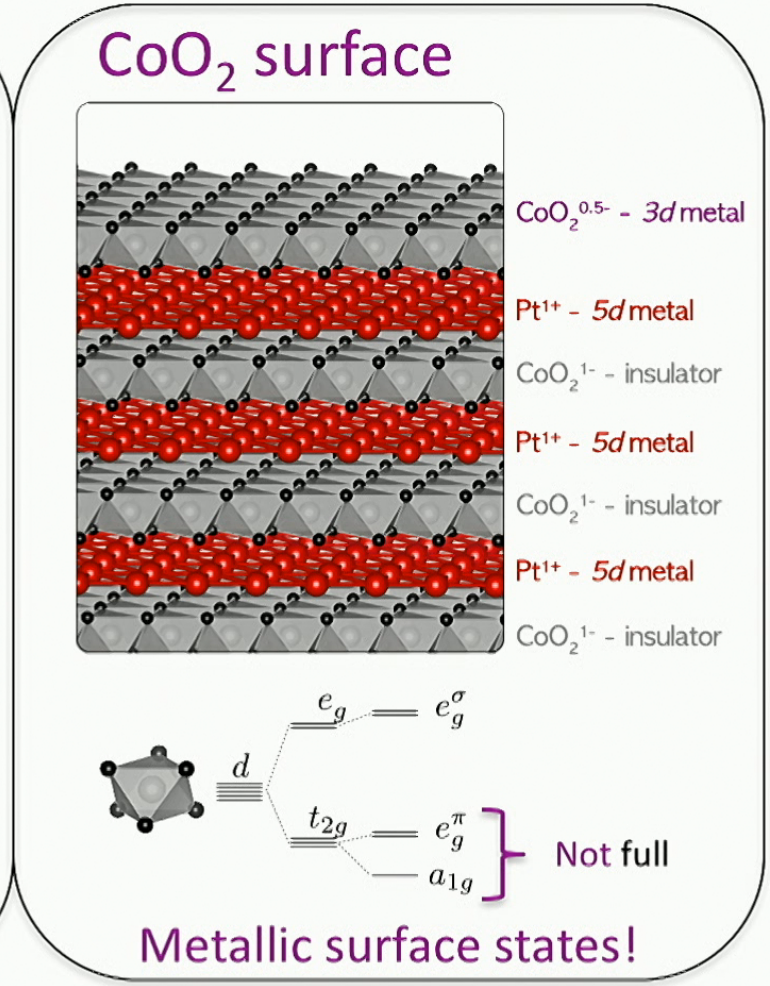
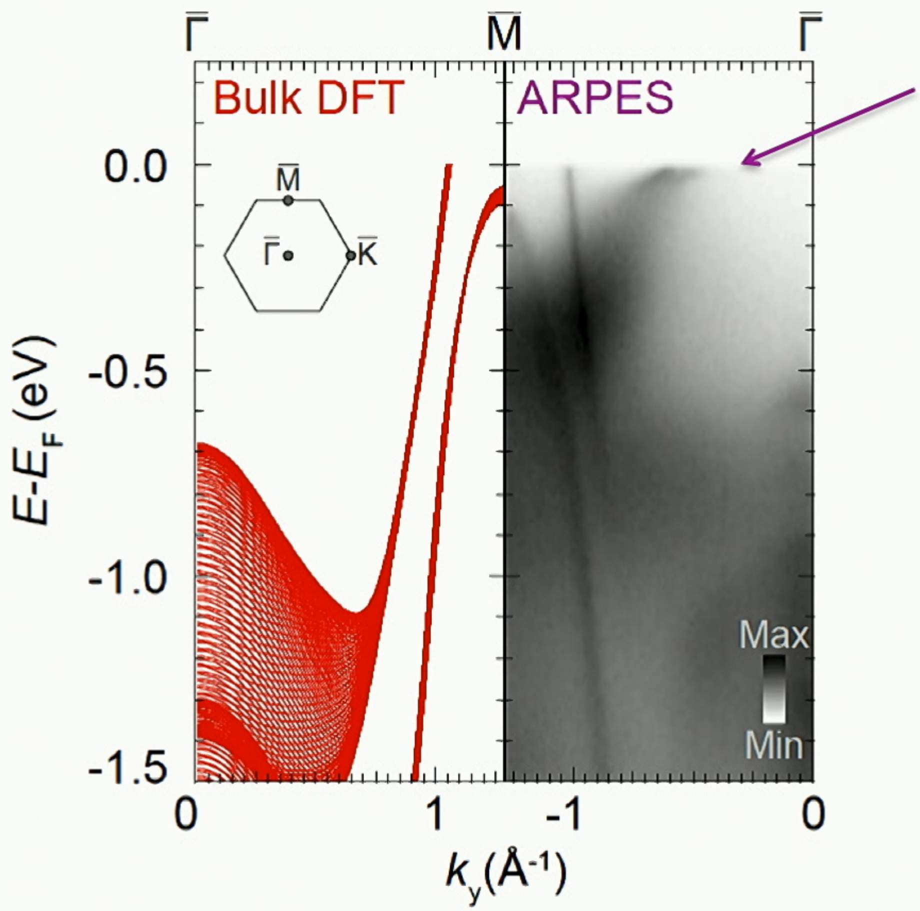




Typical ARPES data from $PtCoO_2$



- States not expected in the bulk at any k_z

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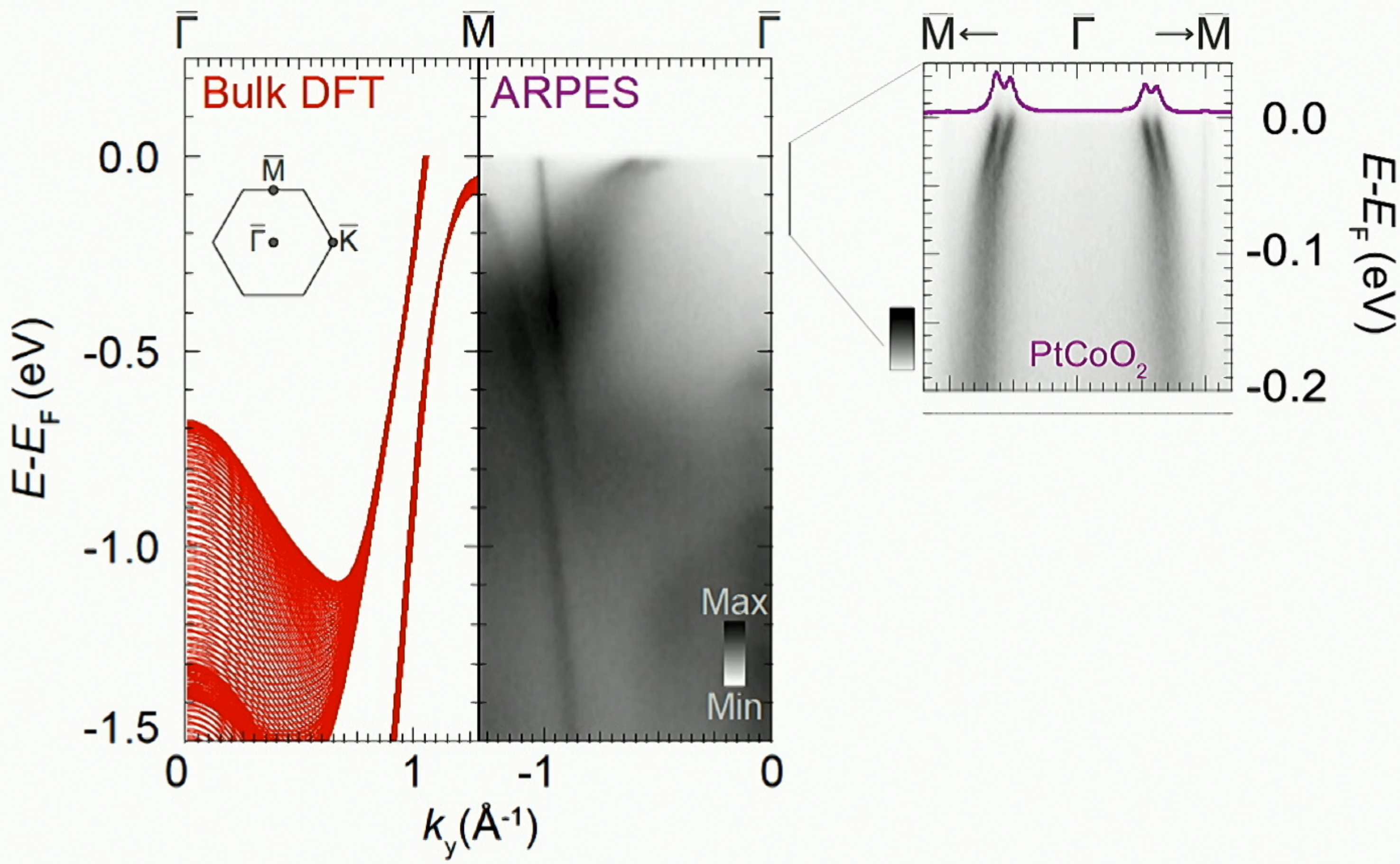
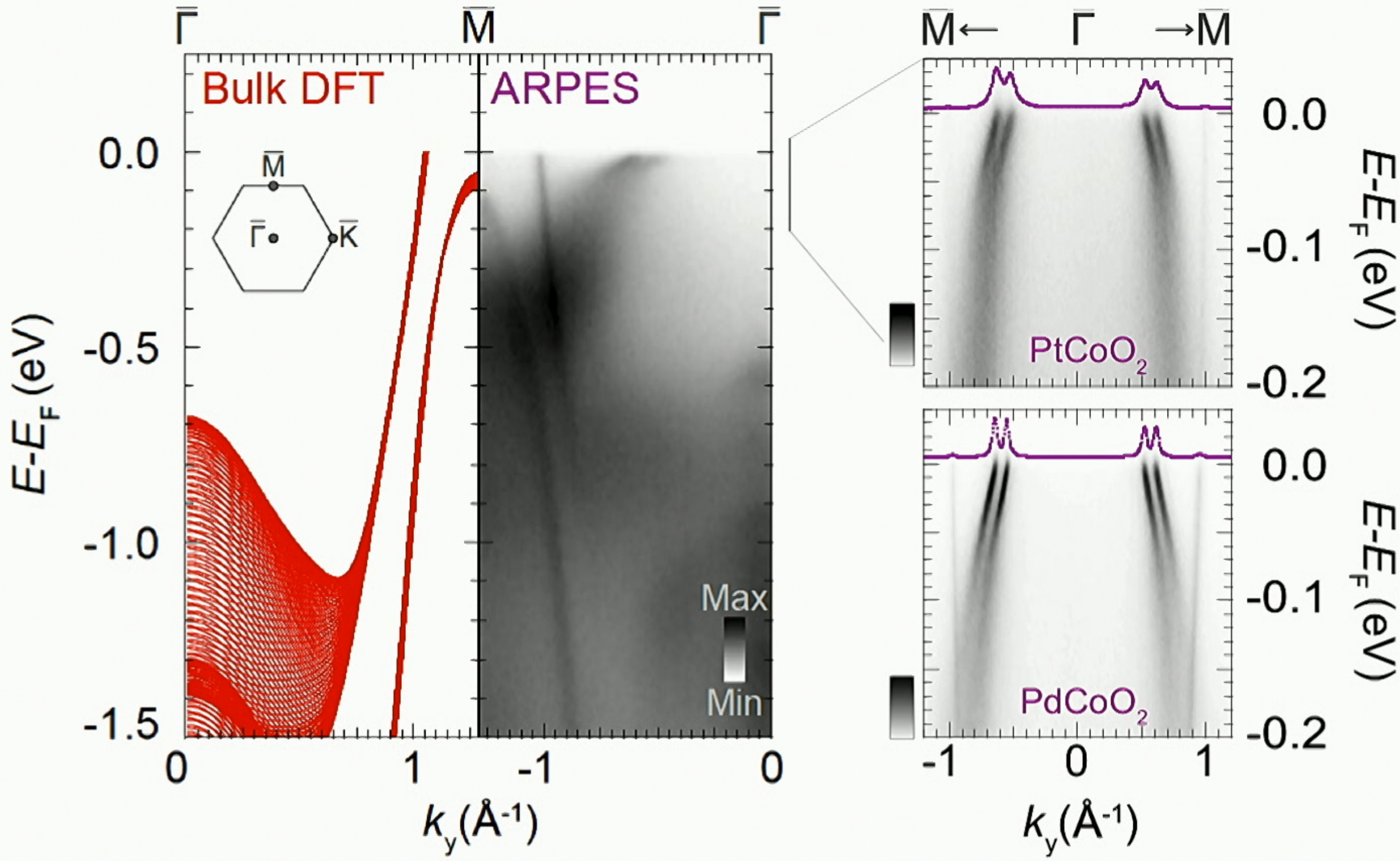
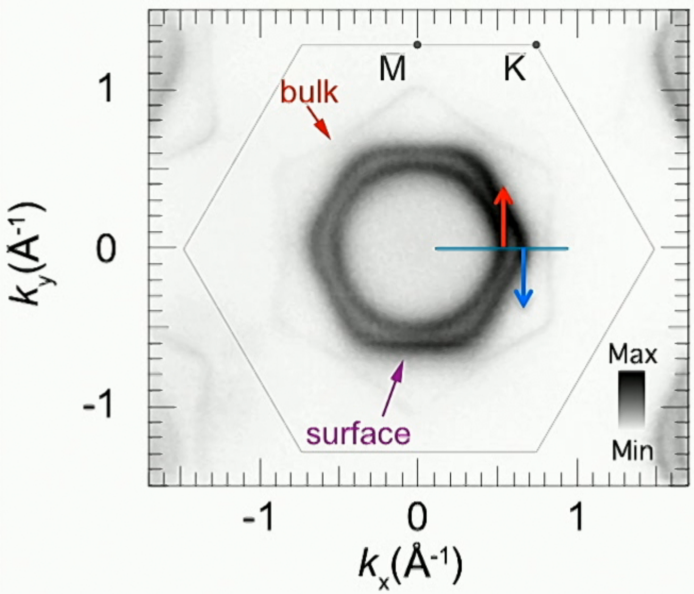


Similar data obtained in $PdCoO_2$: Pt/Pd spin-orbit irrelevant



Fermi surfaces of bulk and surface bands from experiment and density functional theory calculation



Experiment:

- Two spin - polarised Fermi surfaces (FS)
- Chiral in-plane spin texture

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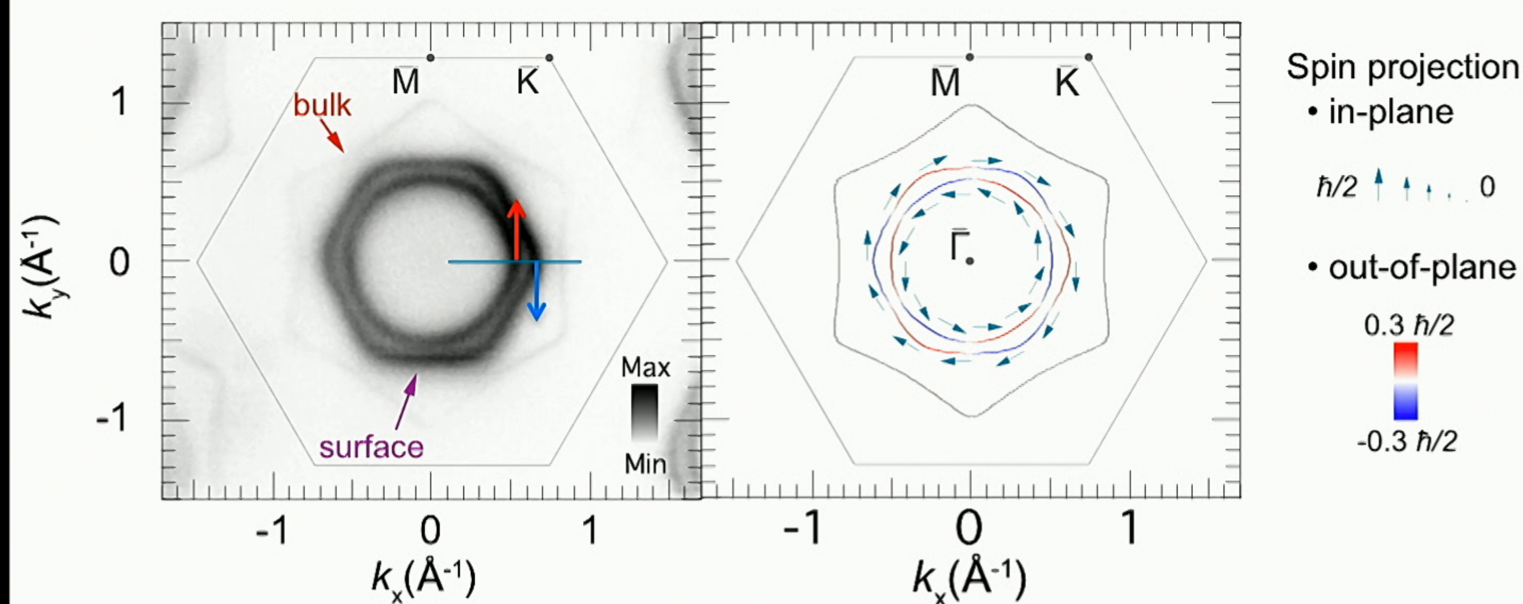


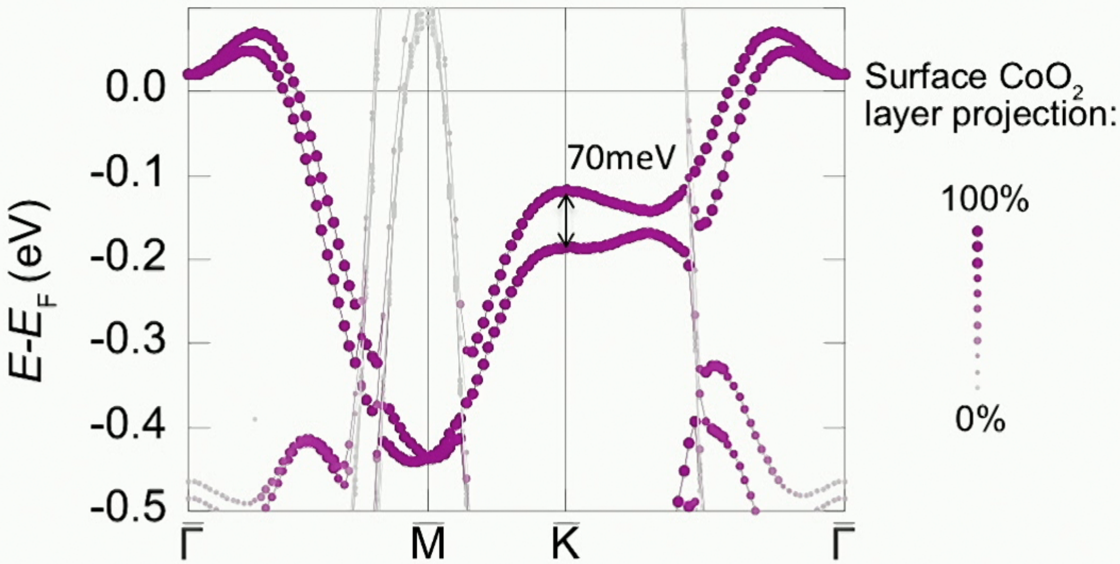
Experiment:

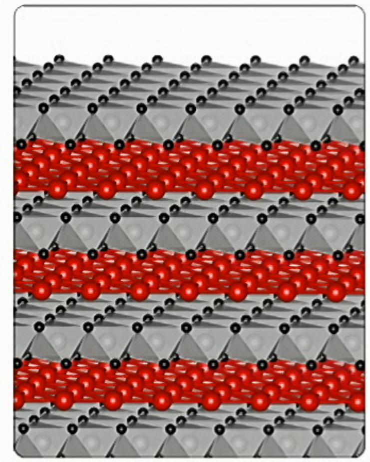
- Two spin - polarised Fermi surfaces (FS)
- Chiral in-plane spin texture

CoO_2–terminated slab calculation:

- Agrees with experiment
- Strong Rashba-like spin component
- Experiment and DFT both give a large split – why?

Surface state DFT bandstructure





Spin-polarised bands have their origin almost entirely in from the top CoO_2 layer .

Spin-splitting reaches as much as 70 meV – the full atomic value – at the K point.

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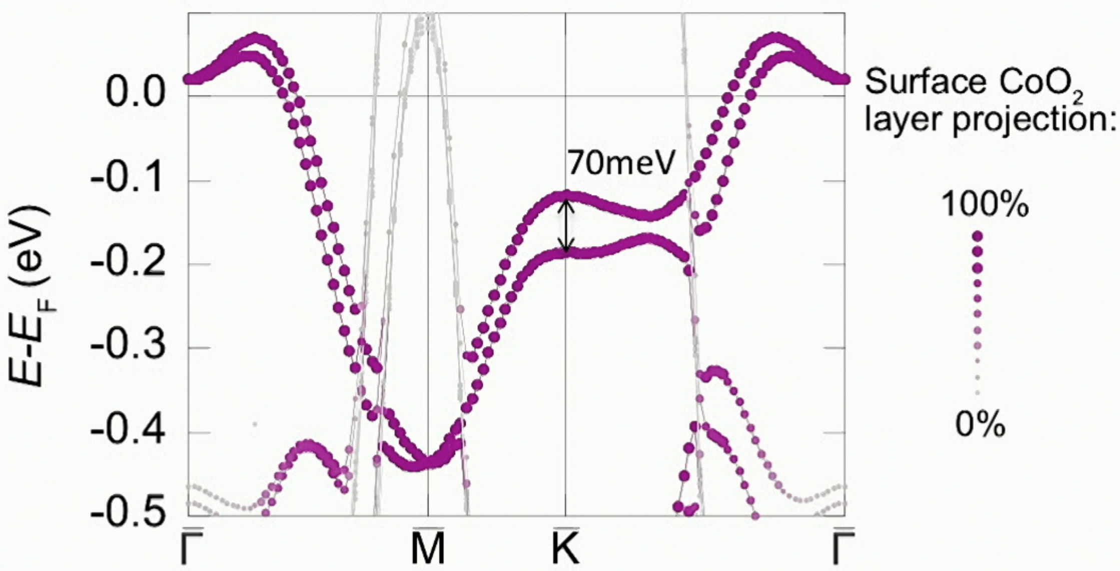

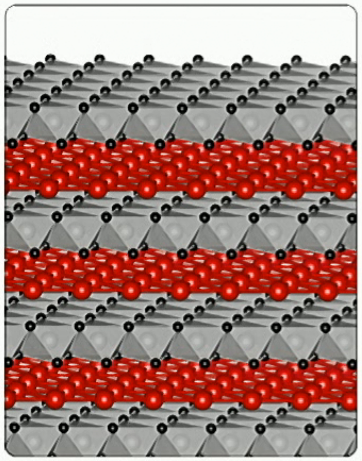

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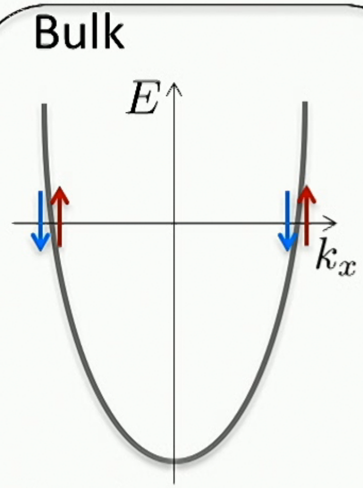
In (all) other known cases of Rashba-like splitting, the observed split is only a small fraction of the atomic value. What is going on here?

Spin-split surface states require both spin-orbit coupling and inversion symmetry breaking



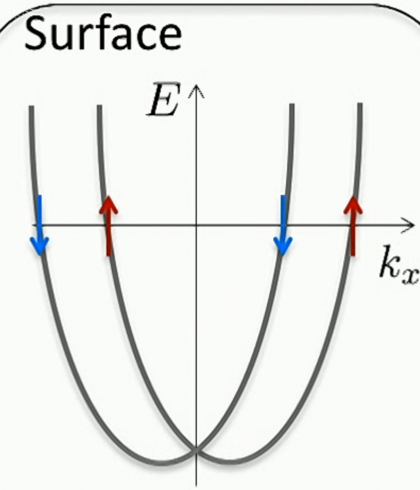
Inversion symmetry:

$$E(k,\uparrow) = E(-k,\uparrow)$$

Time reversal:

$$E(k,\uparrow) = E(-k,\downarrow)$$

Spin degeneracy



Time reversal:

$$E(k,\uparrow) = E(-k,\downarrow)$$

Spin splitting

Relative energy scales of the two effects is crucial

Veronika Sunko

Necessary ingredients:

1. Spin orbit coupling (SOC)
   - energy scale: E_{SOC}
2. Inversion symmetry breaking (ISB)
   - energy scale: E_{ISB}

Relative energy scales of the two effects is crucial

Strong ISB

No ISB,
no SOC

Weak ISB

No ISB,
no SOC

Orbital angular momentum (OAM): ↓↑, spin angular momentum (SAM): ↓↑

*cf. Kim, B., Kim, P., Jung, W., Kim, Y., Koh, Y., Kyung, W., Park, J., Matsunami, M., Kimura, S., Kim, J.S., Han, J.H., Kim, C., 2013. , Phys. Rev. B **88**, 205408; Park, S.R., Kim, C., 2015., Special issue on electron spectroscopy for Rashba spin-orbit interaction 201, 6–17.*

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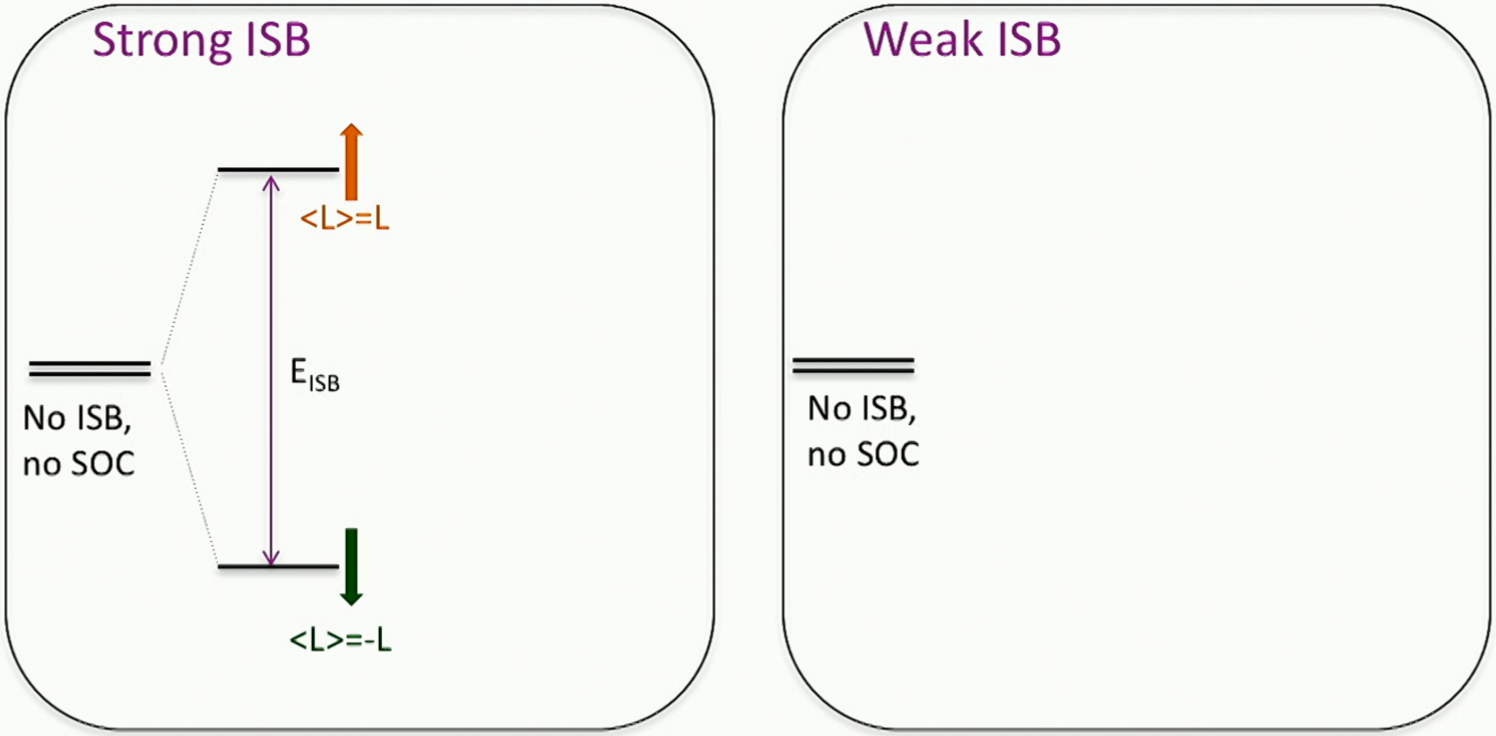

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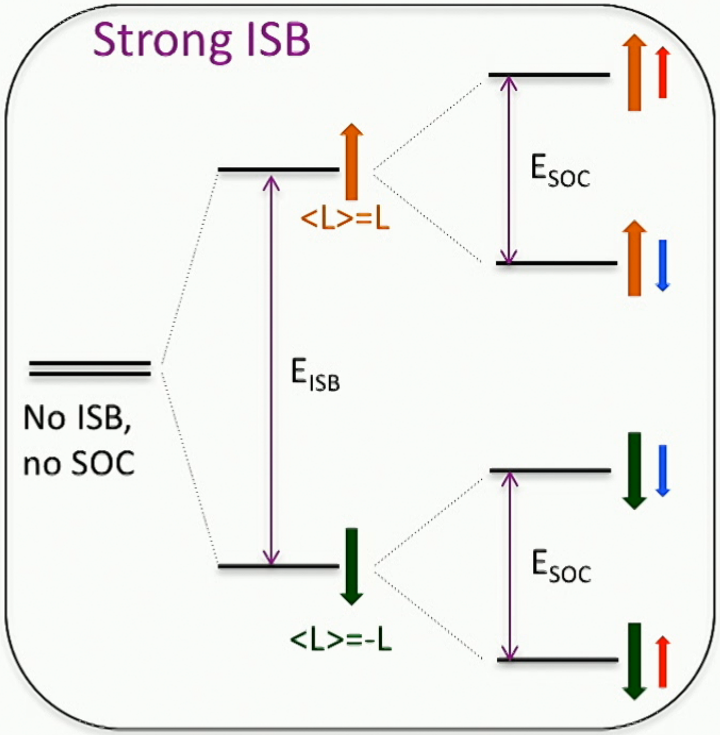

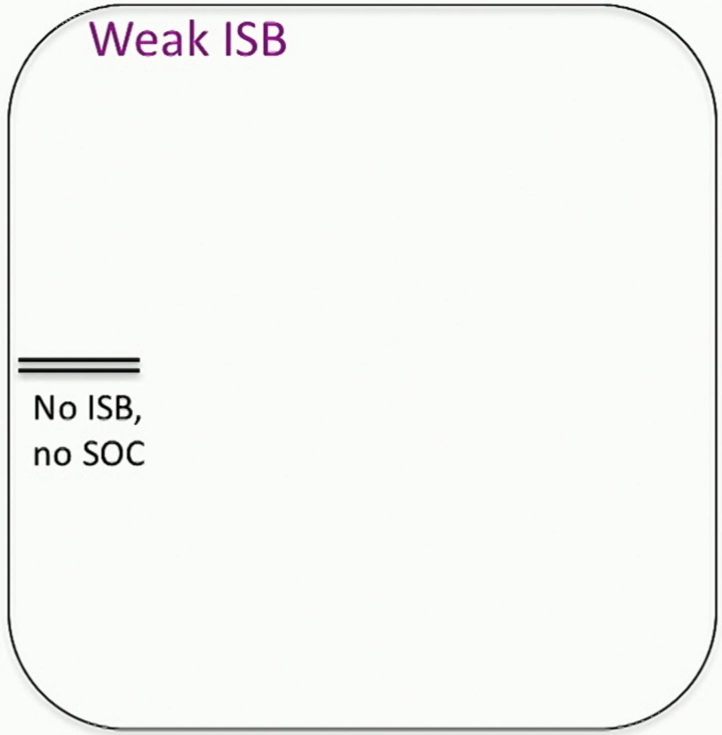

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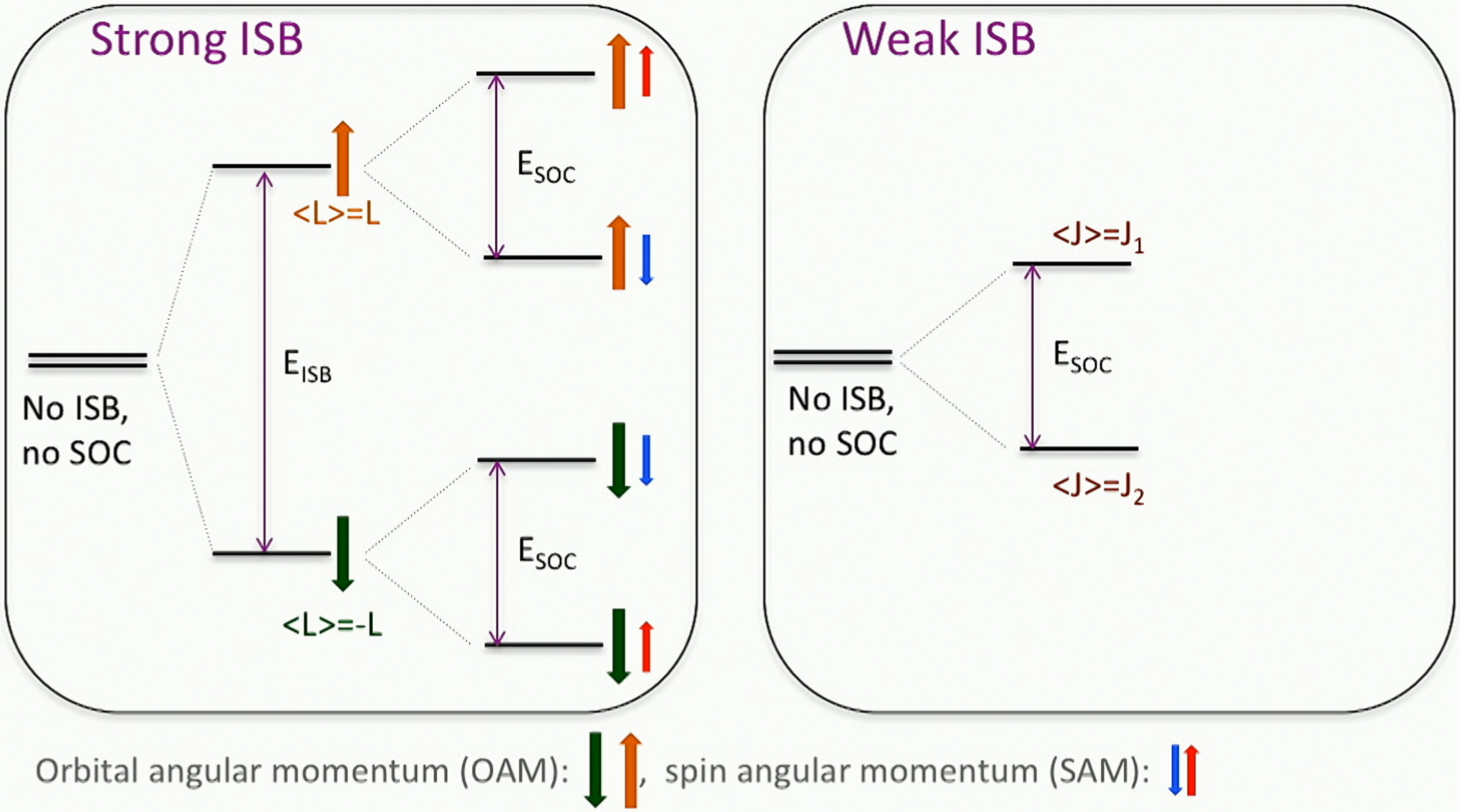

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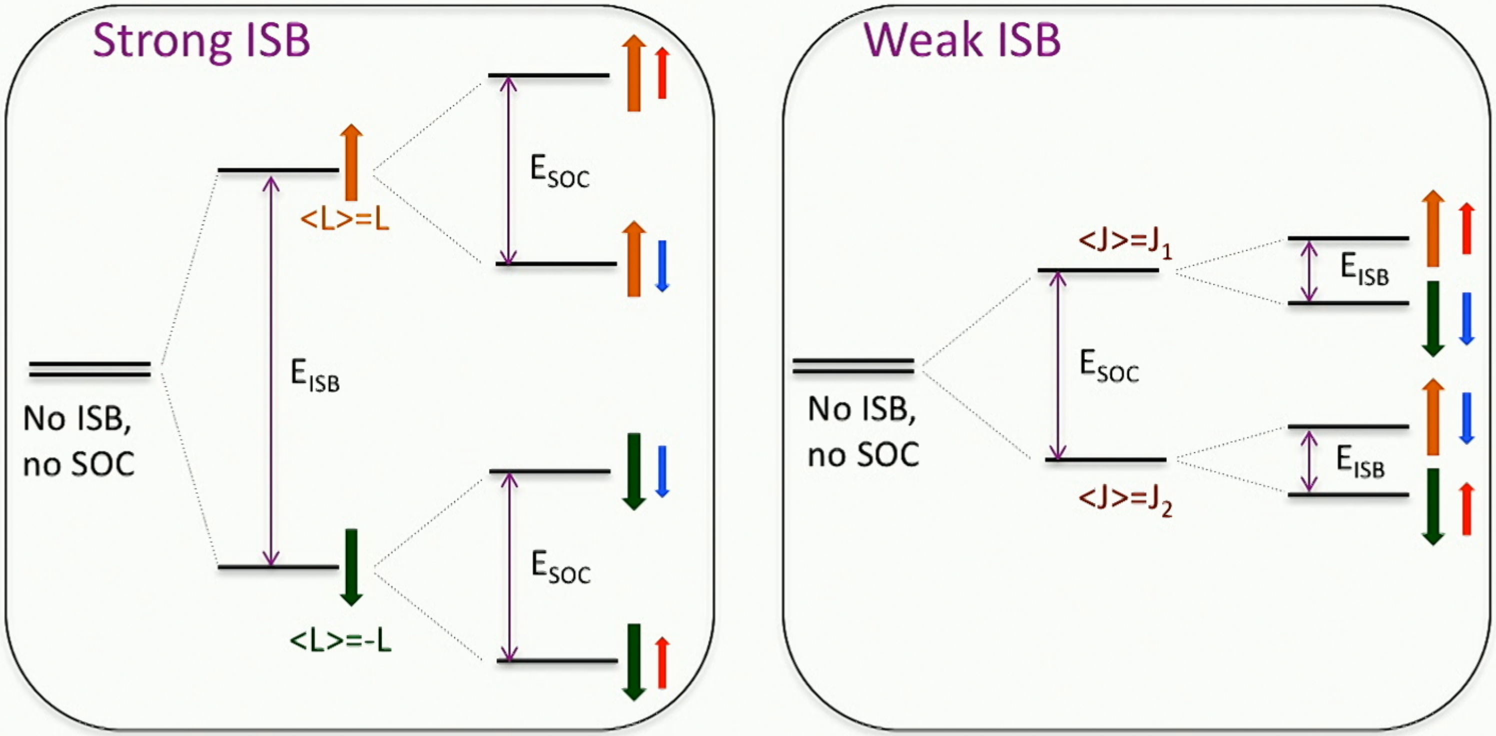

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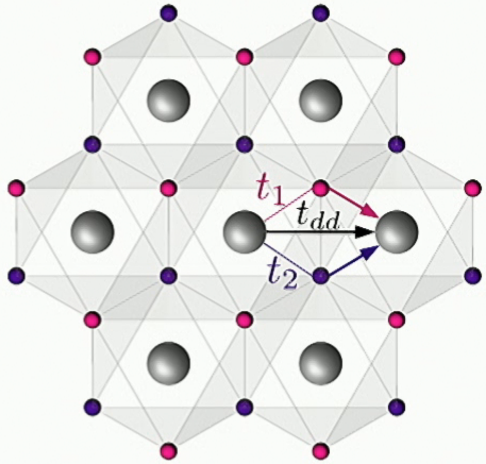
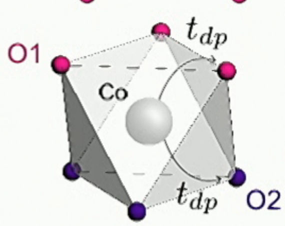
Extremely strong ISB in delafossites because of asymmetric Co-O-Co hopping in the 'side-on' octahedra

CoO_2 layer: View from the top





Effective Co-Co hopping

- Through O1
- Through O2

ISB: Different probabilities!

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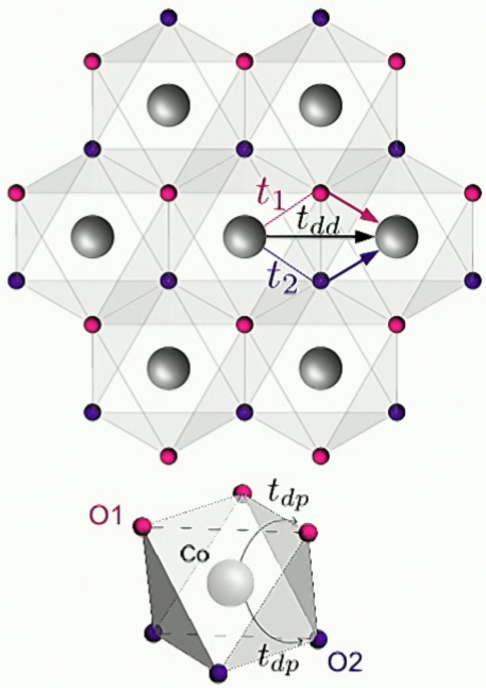

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ISB: Different probabilities!

Energies estimated from DFT :

- Average hopping: $t \approx 360 meV$
- Relative ISB: $\alpha_{ISB} = \frac{t_1 - t_2}{t_1 + t_2} \approx 42\%$
- Absolute ISB : $E_{ISB} = \alpha_{ISB} t \approx 150 meV$

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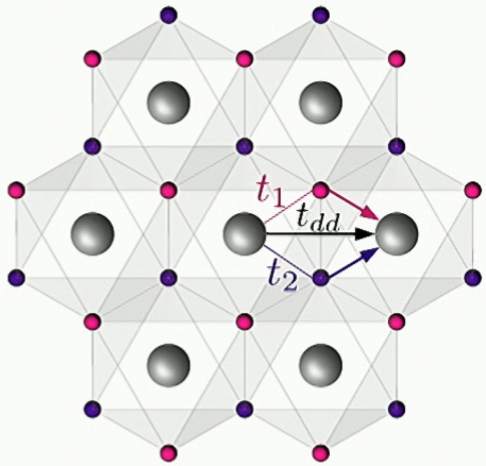

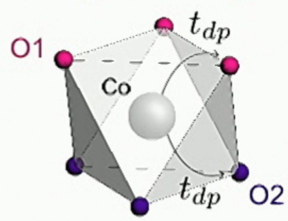

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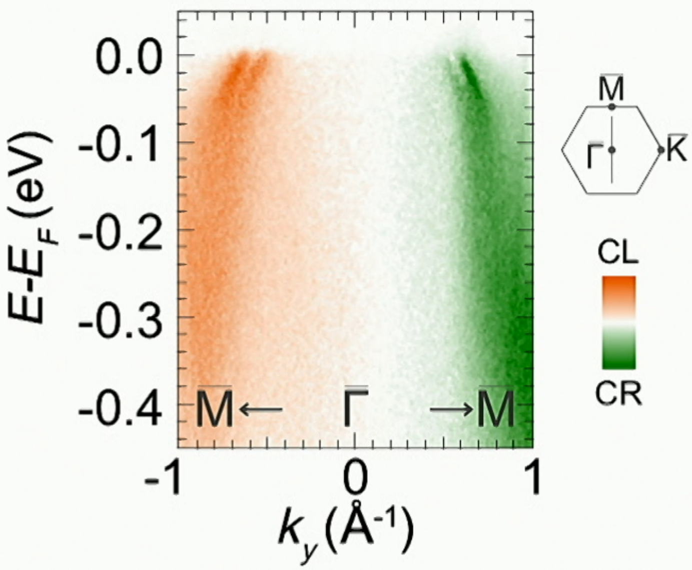
Evidence that $PtCoO_2$ and $PdCoO_2$ are in the strong ISB limit so the huge splitting can be post-facto understood.

Check – what about the orbital vs spin texture?

- **Model:** same sign of orbital angular momentum on the two spin split branches if inversion symmetry breaking is the dominant scale

- **Experimental probe:** circular dichroism photoemission *(Park, J.-H. Phys. Rev. B **85**, 195401, 2012)*



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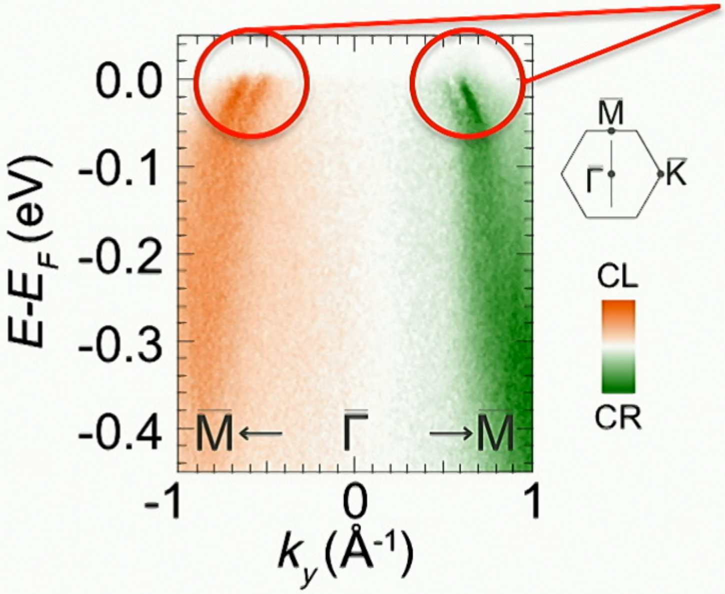

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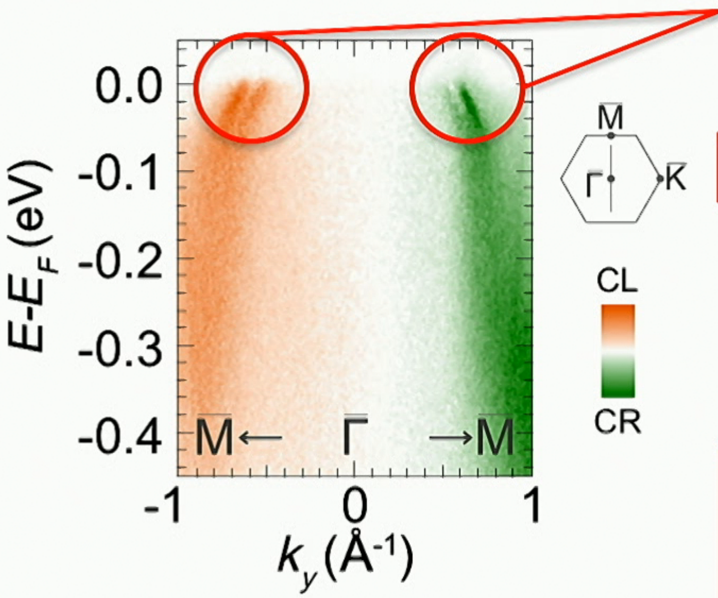


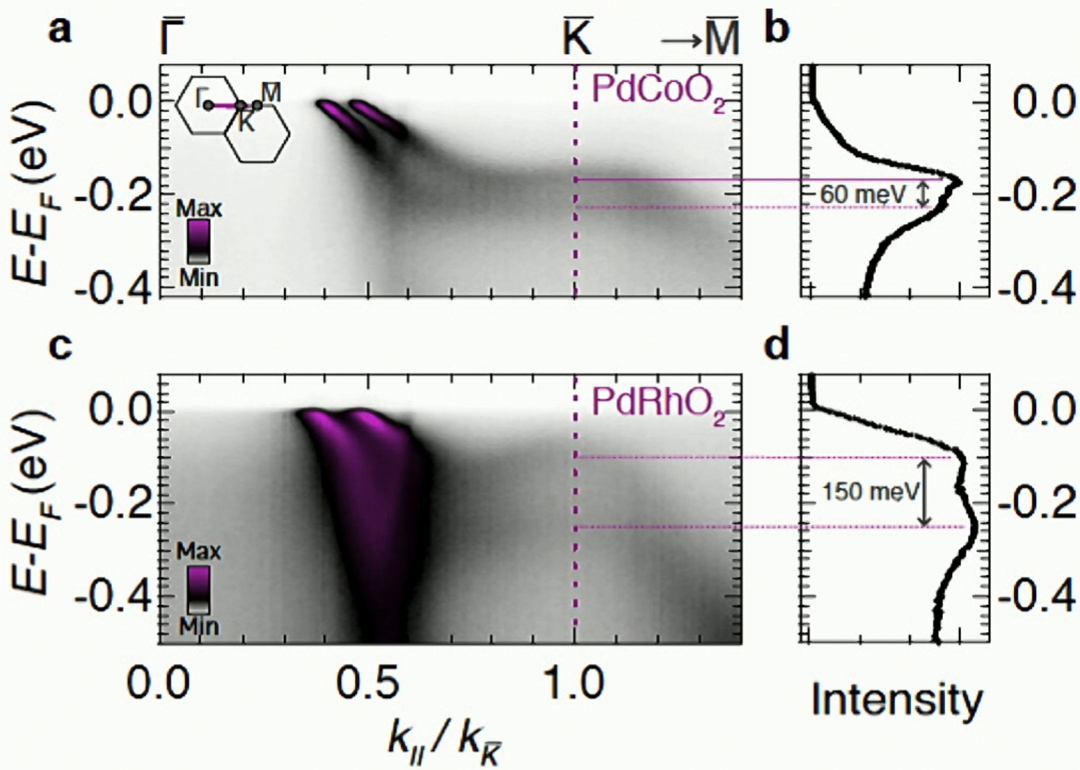
As expected, dichroism reverses on opposite sides of the surface bands, but it is always the same for both the inner and outer band, even though their spins are opposite.

Good evidence that the model is correct.

Important prediction: inversion symmetry breaking will scale with bandwidth, so will increase on changing Co for a heavier transition metal.

Rashba-like split should therefore also scale up according to the larger spin orbit coupling of the new transition metal.

Grow and study $PdRhO_2$



Spin-split does indeed increase in line with the ratio of the atomic SOC strengths of Co and Rh. New possibilities for surface and interface engineering.

V. Sunko, H. Rosner, P. Kushwaha, S. Khim, F. Mazzola, L. Bawden, O.J. Clark, J.M. Riley, D. Kasinathan, M.W. Haverkort, T.H. Kim, M. Hoesch, J. Fujii, I. Vobornik, A.P. Mackenzie and P.D.C. King, preprint

University of St Andrews

Max Planck Institute for Chemical Physics of Solids

Conclusions

1. Delafossites combine purity and the simplicity of a single half-filled conduction band in a layered 'natural heterostructure' between nearly free and strongly correlated layers.
2. In bulk, they will allow us to reach entirely new regimes, particularly accessible in microstructures.
3. They are ideal for investigation by angle-resolved photoemission, and both surface- and bulk-state spectroscopy will likely be highly fruitful.