Engineering artificial quantum matter with twodimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



2D Materials 9 (2), 025010 (2022) 2D Materials 10 (2), 025026 (2023) Advanced Materials, 2311342 (2024)

Phys. Rev. Lett. 127, 026401 (2021) Nature 599, 582–586 (2021) Nano Letters 24 (14), 4272–4278 (2024)

17th European School on Molecular Nanoscience (ESMolNa2024), Spain, May 23rd 2024

Building quantum matter with artificial lattices

Atomic lattices



a = 0.5 nm

Nature Physics 12 (7), 656 (2016) Nature Physics 13 (7) 668 (2017) Science 335 (6065), 196-199 (2012)

Molecular lattices



a = 3 nm

Nature 408, 447–449 (2000) Nature Rev. Mat. 5 (2), 87-104 (2020) Nature 598, 287–292 (2021) Phys. Rev. Lett. 130, 100401 (2023)

Twisted 2D moire materials



a = 30 nm

Phys. Rev. Lett. 99, 256802 (2007) Nature Physics 6, 109–113 (2010) PNAS 108 (30) 12233-12237 (2011) Nature 556, 80–84 (2018)

The world of two-dimensional materials



Semimetal

Insulator BN



Superconductor NbSe₂

Ferroelectric



Semiconductor WSe₂



Quantum spin Hall insulator WTe₂



Multiferroic Nil₂



The flexibility of two-dimensional materials

They can be stacked

They can be rotated





These are unique features of two-dimensional materials

Upper graphene layer











One material, a zoo of electronic phases

Twisted bilayer graphene

Superconductivity



Nature 556, 43–50 (2018) Nature 600, 240–245 (2021)

Correlated insulators

Nano Lett. 18, 11, 6725-6730 (2018)

Quasicrystalline physics

Topological networks

Chern insulators



Science 365, 605-608 (2019)

Fractional Chern insulators



Nature 600, 439–443 (2021)

Nature 556, 80–84 (2018)

Science 361, 782-786 (2018)

A bilayer of a van der Waals material realizes a variety of widely different electronic states

Van der Waals magnetic materials

Ferromagnet, antiferromagnets



(proximal) Quantum spin-liquids



Crl₃, CrCl₃, CrBr₃

Nature 546, 270–273 (2017)

$RuCl_{3}$, 1T-TaS₂ 1T-TaS₂/1H-TaS₂

Nature 599, 582-586 (2021)

Nature Physics 17, 1154–1161 (2021)

Break time-reversal

Do not break time-reversal

Heavy-fermion Kondo insulators

Multiferroics



 Nil_2

Nature 602, 601–605 (2022)

Break time-reversal and inversion symmetry

Emergent excitations in van der Waals magnets

Magnons



Spinons



S=1 No charge

Instrumental for magnonics

S=1/2 No charge

Plan for today

Engineering and probing van der Waals multiferroics



2D Materials 9 (2), 025010 (2022) 2D Materials 10 (2), 025026 (2023) Advanced Materials, 2311342 (2024)

Engineering and probing van der Waals heavy fermion Kondo magnets



Phys. Rev. Lett. 127, 026401 (2021) Nature 599, 582–586 (2021) Nano Letters 24 (14), 4272–4278 (2024)

A reminder about magnetism

A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^{\dagger}c_{1\uparrow} + c_{0\downarrow}^{\dagger}c_{1\downarrow}] + \sum_{i} Uc_{i\uparrow}^{\dagger}c_{i\uparrow}c_{i\downarrow}^{\dagger}c_{i\downarrow} + h.c.$$

$$0 \qquad 1$$
Levels

The full Hilbert space at half filling is



A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^{\dagger}c_{1\uparrow} + c_{0\downarrow}^{\dagger}c_{1\downarrow}] + \sum_{i} Uc_{i\uparrow}^{\dagger}c_{i\uparrow}c_{i\downarrow}^{\dagger}c_{i\downarrow} + h.c.$$

The energies in the strongly interacting limit are $\,U\gg t$



A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^{\dagger}c_{1\uparrow} + c_{0\downarrow}^{\dagger}c_{1\downarrow}] + \sum_{i} Uc_{i\uparrow}^{\dagger}c_{i\uparrow}c_{i\downarrow}c_{i\downarrow} + h.c.$$

The low energy manifold is

Just one electron in each site for $~~U\gg t$

Local S=1/2 at each site

A reminder about magnetism, the origin of the Heisenberg model

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^{\dagger}c_{1\uparrow} + c_{0\downarrow}^{\dagger}c_{1\downarrow}] + \sum_{i} Uc_{i\uparrow}^{\dagger}c_{i\uparrow}c_{i\downarrow}^{\dagger}c_{i\downarrow} + h.c.$$

In the strongly correlated (half-filled) limit we obtain a Heisenberg model

$$\mathcal{H} = J\vec{S}_0 \cdot \vec{S}_1$$

$$J \sim \frac{|t|^2}{U}$$

The lattice Heisenberg model

For a generic lattice and a generic interacting model, the Hamiltonian is

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

In those generic cases, the exchange couplings can be positive or negative



Antiferromagnetic coupling

 $J_{ij} < 0$

Ferromagnetic coupling

Spin-orbit coupling introduces anisotropic couplings

$$\mathcal{H} = \sum_{ij} J_{ij}^{\alpha\beta} S_i^{\alpha} S_j^{\beta}$$

The classical Heisenberg model

Ferromagnetism

Antiferromagnetism

Frustrated magnetism





The quantum Heisenberg model

What is the ground state of the quantum Heisenberg Hamiltonian?

The ground state is unique, and does not break time-reversal

$$|GS\rangle = \frac{1}{\sqrt{2}}[|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle] \qquad \langle \vec{S}_i \rangle = 0$$

The state is maximally entangled

A quantum magnet is the macroscopic version of the previous state

$$\langle \vec{S}_i \rangle = 0$$

Classical and quantum magnets

Ferromagnet, antiferromagnets & helimagnets



They break time-reversal symmetry <u>Classical magnets</u> *Quantum spin liquids and heavy ferrmion Kondo magnets*



 $\langle \vec{S}_n \rangle = 0$

They no not break time-reversal symmetry Quantum entangled magnets

Van der Waals multiferroic magnets

Plan for 2D multiferroics

Multiferroicity in Nil₂



2D Materials 9 (2), 025010 (2022) Advanced Materials, 2311342 (2024)

Artificial multiferroicity by twisting 2D magnets



2D Materials 10 (2), 025026 (2023)

Behind the scenes

Intrinsic multiferroics, Nil₂

(theory and experiment)

















P. Liljeroth



2D Materials 9 (2), 025010 (2022) Advanced Materials, 2311342 (2024)

Artificial multiferroics, twisted CrBr₃ bilayers

(theory)

A. Fumega



2D Materials 10 (2), 025026 (2023)

Origin and visualization of multiferroicity in Nil₂















H. Gonzalez Herrero

S. Kezilebieke

P. Liljeroth







2D Materials 9 (2), 025010 (2022) Advanced Materials, 2311342 (2024)

Monolayer Nil₂



Frustrated magnetic Heisenberg model



Multiferroicity in monolayer Nil₂



Probe of magnetic order Magneto-optic Kerr effect

Probe of ferroelectric order Second harmonic generation

Nature 602, 601-605 (2022)

The impact of spin-orbit coupling in magnetic ordering

In the presence of spin-orbit coupling, new terms can appear in the Hamiltonian



In Nil₂, the non-collinear order is promoted by isotropic exchange In Nil₂, the ferroelectricity is created by inverse antisymmetric exchange

The origin of ferroelectricity in Nil₂

Magnetically driven ferroelectric order

$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$$

$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e} \cdot \mathbf{S})(\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}_0))} \mathbf{M}(\mathbf{r}_0)$$



The emergence of non-collinear order is drives ferroelectricity

The origin of ferroelectricity in Nil₂



Wavevector of the spin spiral

The impact of spin-orbit coupling in the electronic structure



Spin-orbit coupling introduces corrections of 100 meV close to the Fermi level

There is a major role of the halide close to the Fermi level

Ferroelectric correction to the electronic density



We will examine the ferroelectric density correction $\delta \rho = \rho[\vec{q_1}] - \rho[-\vec{q_1}]$

Ferroelectric correction to the electronic density



In the absence of SOC, no ferroelectric density appears

Ferroelectric correction to the electronic density



In the presence of SOC, a ferroelectric density appears both in Ni and I

The ferroelectric force



The forces between the two magnetic configuration account for the ferroelectric displacement

The ferroelectric force



The dominant contribution to the ferroelectric force comes from iodine
Modulated multiferroicity in monolayer Nil₂

The ferroelectric order has actually a modulation in real space



 $\mathbf{P} =$

Visualizing multiferroicity in monolayer Nil₂

Modulated electrostatic potential



Advanced Materials, 2311342 (2024)

Visualizing multiferroicity in monolayer Nil₂

Experiment



Theory



 $\Lambda q(-\sin(2\mathbf{q}\cdot\mathbf{r}),\sin^2(\mathbf{q}\cdot\mathbf{r}),0)$

Ferroelectricity from modulated band offset

Band offset (experiment)





Controlling multiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

Advanced Materials, 2311342 (2024)

Engineering artificial multiferroics with twisted van der Waals materials



A. Fumega



2D Materials 10 025026 (2023)

Twisted 2D trihalides





The local stacking determines the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Mixed FE/AF domains

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism

arXiv:2204.01636

Twisted 2D magnetic materials

Non-collinear magnetism and multiferroic order appear due to the moire





Stacking, magnetic order and electric polarization



Stacking, magntic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



Ferroelectric electronic density

$$\delta \rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

Ferroelectric force

$$\boldsymbol{\mathcal{F}}_{\alpha} = \frac{1}{2} (\mathbf{F}_{\alpha}[\mathbf{q}_1] - \mathbf{F}_{\alpha}[\mathbf{q}_2]),$$

The ferroelectric electronic correction



Ferroelectric electronic density $\delta \rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$

The ferroelectric electronic correction emerges in the region with non-collinear magnetization

It becomes stronger the heavier the halide

The ferroelectric force

Magnetic configuration

Ferroelectric force

$$\boldsymbol{\mathcal{F}}_{\alpha} = \frac{1}{2} (\mathbf{F}_{\alpha}[\mathbf{q}_1] - \mathbf{F}_{\alpha}[\mathbf{q}_2]),$$

The ferroelectric force becomes Stronger for heavier halides

Electrically controllable skyrmions in twisted magnets



The electric polarization directly couples to an out-of plane electric field

$$\mathcal{H}_E = rac{1}{2} \sum_{ij} \mathbf{E} \cdot \mathbf{P}_{ij}$$

Electrically controllable skyrmions in twisted magnets



Skyrmions at finite interlayer bias



Van der Waals heavyfermion Kondo magnets

The world of heavy fermion compounds

We have a rich family of bulk heavy-fermion compounds

CeCoIn₅ Science 375, 6576, 76-81 (2021)



UTe₂ Nature 579, 523–527 (2020)



- Strongly correlated matter, dominated by Kondo lattice physics
- Unique system to realize quantum criticality and unconventional superconductivity
- Found in rare-earth compounds, such as UTe₂ or UPt₃

Searching for new heavy fermion compounds

We have a rich family of bulk heavy-fermion compounds

CeCoIn₅ Science 375, 6576, 76-81 (2021)



UTe₂ *Nature 579, 523–527 (2020)*



Can we realize heavy-fermion physics with two-dimensional materials?





Phys. Rev. Lett. 127, 026401 (2021) Nature 599, 582–586 (2021) Phys. Rev. Research 3, 043173 (2021) Phys. Rev. B 106, L041116 (2022) Phys. Rev. Lett. 129, 047601 (2022) Nature 616, 61–65 (2023) Nature 625, 483–488 (2024) Nano Letters 24 (14), 4272–4278 (2024)

Behind the scenes

Heavy fermions in twisted graphene trilayers





Phys. Rev. Lett. 127, 026401 (2021)

Heavy fermions in twisted dichalcogenide bilayers



S. Ganguli M. Amini





G. Chen



V. Vaňo





P. Liljeroth



Nature 599, 582–586 (2021)

Building an artificial heavy fermion state





Lattice of Kondo impurities



f-electrons in rare-earth compounds

Dispersive electron gas

s/p/d-electrons in rare-earth compounds

Building an artificial heavy fermion state

Conduction electrons form Kondo singlets with the impurities

Kondo-lattice model

Associated with Kondo lattice physics:

- Colossal mass enhancement of electrons
- Quantum criticality
- Unconventional (topological) superconductivity

Basics of heavy fermion physics



Science 332.6026, 196-200 (2011)

The Kondo problem



The Kondo lattice problem

Lattice of Kondo impurities



Dispersive electron gas



Both ingredients coupled through Kondo coupling

The Kondo lattice problem



Solving the Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^{\dagger} c_{j\sigma} + \text{H.c} \right) + J_K \sum_{j,\alpha\beta} \left(c_{j\beta}^{\dagger} \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Replace the spin sites by auxiliary fermions

$$S_{\alpha\beta}(j) \sim f_{j\alpha}^{\dagger} f_{j\beta} - \frac{n_f(j)}{N} \delta_{\alpha\beta}$$

This makes the effective Hamiltonian an "interacting" fermionic Hamiltonian

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - J_K \sum_{j,\alpha\beta} \left(c_{j\beta}^{\dagger} f_{j\beta} \right) \left(f_{j\alpha}^{\dagger} c_{j\alpha} \right)$$

Solving the Kondo lattice problem

Now we decouple the fermions with a mean-field approximation

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - J_K \sum_{j,\alpha\beta} \left(c_{j\beta}^{\dagger} f_{j\beta} \right) \left(f_{j\alpha}^{\dagger} c_{j\alpha} \right)$$

Obtaining a quadratic Hamiltonian

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_{K} \sum_{\mathbf{k},\alpha} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

Electronic structure of the Kondo lattice problem



Electronic structure of the Kondo lattice problem



Electronic structure of a rare-earth van der Waals heavy-fermion material CeSil

Monolayer CeSil



Ultrathin heavy-fermion isolated in Nature 625, 483–488 (2024)

Heavy-fermion (gapped) electronic structure



Nano Letters 24 (14), 4272–4278 (2024)



Aline Ramires



Phys. Rev. Lett. 127, 026401 (2021)

Top view







Top view



Band structure



Top view



Electronic structure in the presence of an electric bias



Low energy model



Electronic structure in the presence of an electric bias


Kondo lattice model in the presence of interactions



A heavy fermion regime appears when the Kondo coupling dominates over the exchange coupling

 $|J/J_{K}| < 1$

Kondo lattice model in the presence of interactions

Interactions of the Kondo lattice model



Electric control of the Kondo regime



A heavy fermion regime appears when the Kondo coupling dominates over the exchange coupling

 $|J/J_K| < 1$

Heavy-fermions in twisted graphene trilayers

Heavy-fermion (Doniach) phase diagram

Electric control of the Kondo regime



The full phase diagram of the heavy fermion system can be explored in a single twisted graphene, by tuning the system with an electric bias

An experimental signature of heavy-fermion physics: spin-triplet superconductivity





Heavy-fermions in a van der Waals dichalcogenide heterostructure



V. Vaňo



G. Chen







S. Ganguli

S. Kezilebieke

P. Liljeroth







Nature 599, 582–586 (2021)

Heavy-fermions in dichalcogenide bilayers

Triangular lattice of local magnetic moments



Two-dimensional electron gas





1T-TaS₂

Moment formation in 1T-TaS₂

Charge-density wave reconstruction, leading to a localized orbital in a $\sqrt{13}$ imes $\sqrt{13}$ unit c





Strong interactions give rise to local moment formation

Effectively a S=1/2 Heisenberg model in a triangular lattice

PNAS 114.27 (2017) Phys. Rev. X 7, 041054 (2017) Phys. Rev. B 96, 195131 (2017)

Heavy-fermions in dichalcogenide bilayers

Interlayer coupling creates the Kondo coupling

Intralayer exchange between local moments





The bilayer realizes all the ingredients for heavy-fermion physics

Experimental signatures of heavy-fermion physics

Kondo physics in the magnetic latticeGap opening in the metallic layer

Both signatures can be probed with scanning tunnel microscopy by growing two heterostructures

Probing the Kondo peak

Probing the heavy-fermion gap





Grid spectroscopy of 1T-1H



Zero bias (Kondo) peak appearing with the CDW periodicity

0 mV

Kondo physics in the magnetic lattice

Sample

Differential conductance

The system shows a Kondo peak when the local moments are probed

Temperature and field dependence consistent with Kondo

Kondo physics in the magnetic lattice

The Kondo peak follows the expected temperature dependence

Heavy-fermion gap in the 2D electron gas

Temperature and field dependence consistent with a heavy-fermion gap

Heavy-fermion gap in the 2D electron gas

STM 2D electorn gas (1H-TaS2) Spin lattice (1T-TaS2) Substrate

Heavy-fermion gap in the 2D electron gas

The gap shows a temperature dependence beyond thermal broadening, expected from a heavy-fermion gap

The phase diagram van der Waals heavy-fermions

Controlling the heavy-fermion state

Of course, we can use the typical knobs of bulk heavy fermion compounds

(pressure, chemical doping, etc)

Most importantly, we can exploit the full tunability of van der Waals materials

Gating

Science 306, 5696, 666-669 (2004)

Twist engineering

Nature Reviews Materials 6, 201–206 (2021)

Nature 499, 419-425 (2013)

Allowing to independently control Kondo coupling, exchange coupling, doping and electronic dispersion

Open-source software for van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

Bulk

K-path [2π]

Berry curvature

Max

205

Quantum Lattice: open source interactive interface for tight binding modeling

https://github.com/joselado/quantum-lattice

Quantum matter in van der Waals materials school

Emergent quantum matter in artificial two-dimensional materials

University of Jyväskylä
 31st Jyväskylä
 Summer School

Schedule:

- Session 1: Introduction to 2D materials
- Session 2: Superconductivity in 2D materials
- Session 3: Magnetic 2D materials
- Session 4: Moire electronic states and twisted van der Waals heterostructures
 Session 5: Topological states in 2D materials

https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ

- Session 1: Introduction to 2D materials
- Session 2: Superconductivity in 2D materials
- Session 3: Magnetic 2D materials

Recordings available at

- Session 4: Moire electronic states and twisted van der Waals heterostructures
- Session 5: Topological states in 2D materials

Take home

Van der Waals materials provide a new platform to engineer exotic matter, including multiferroics and heavy-fermion Kondo lattices

Thank you

Funding from

ANE AND AATOS

ERKKO FOUNDATION

2D Materials 9 (2), 025010 (2022) 2D Materials 10 (2), 025026 (2023) Advanced Materials, 2311342 (2024) Phys. Rev. Lett. 127, 026401 (2021) Nature 599, 582–586 (2021) Nano Letters 24 (14), 4272–4278 (2024)