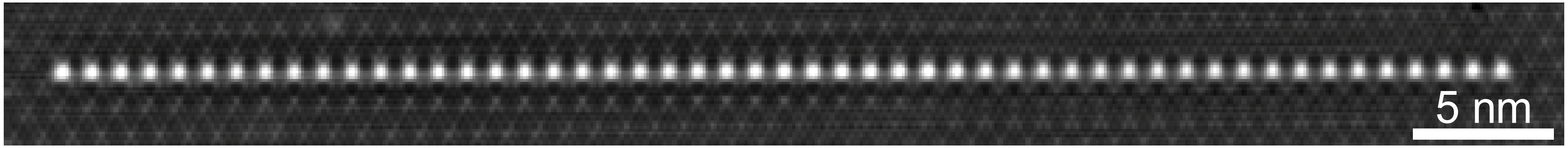
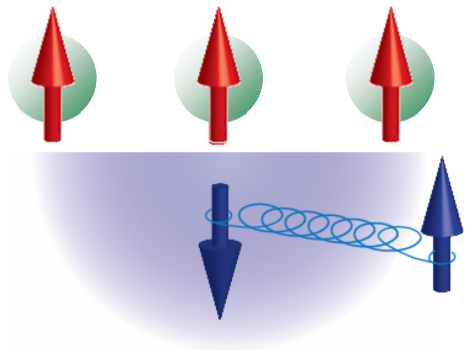
Building and investigating magnetic adatom chains on superconductors atom by atom

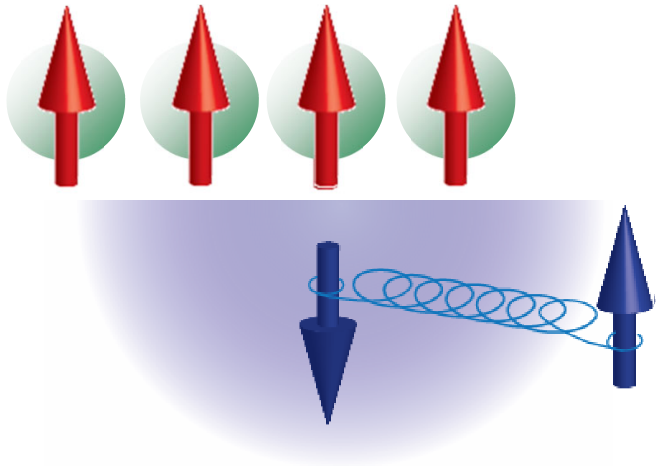


Eva Liebhaber, Lisa Rütten, Gaël Reecht, Katharina Franke

Theory:
Sergio Acero Gonzalez, Jacob Steiner,
Felix von Oppen

Samples:
Sebastian Rohlf, Kai Rossnagel (Kiel)





Fundamentals of local spin-superconductor interactions

Quantum spin magnetism in chains

- Effect of local exchange coupling?

- Coupling of YSR states?
- Topological superconductivity?

Yu-Shiba-Rusinov states: classical model

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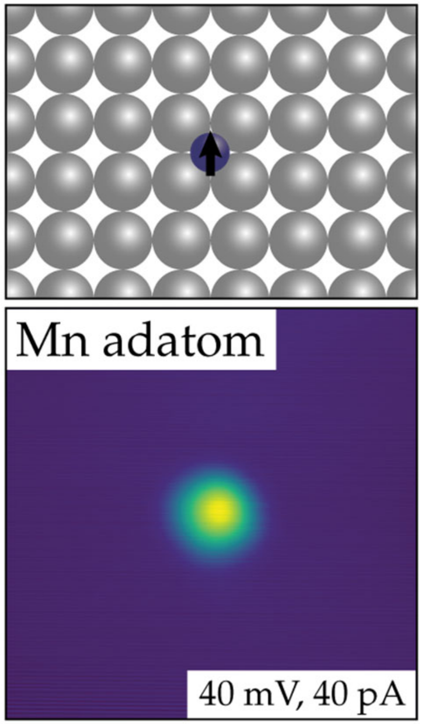
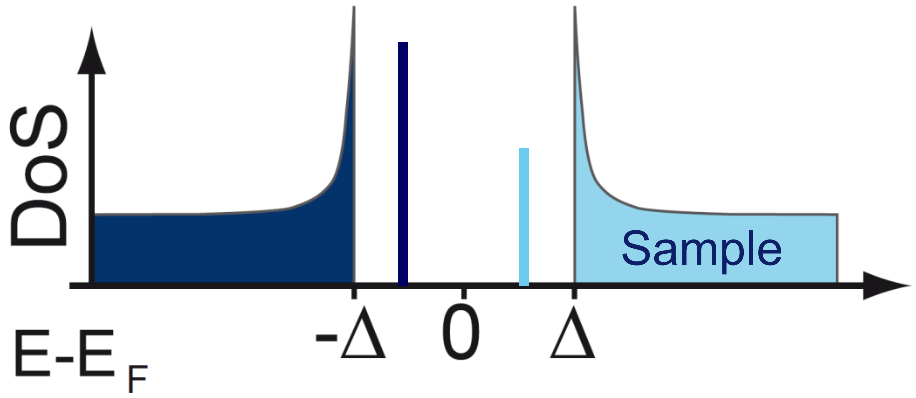


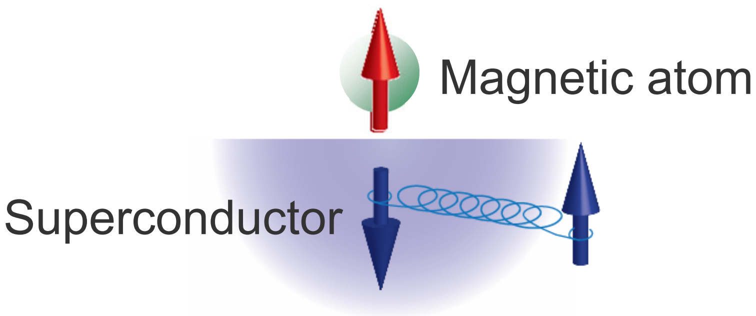


► Spins exchange scatter at a magnetic center:

$$\epsilon_S = \Delta \frac{1 - A^2 + B^2}{\sqrt{4A^2 + (1 - A^2 + B^2)}}$$

$$A = \frac{\pi}{2} J\, S\, \nu_0 \qquad B = \pi\, K\, \nu_0$$

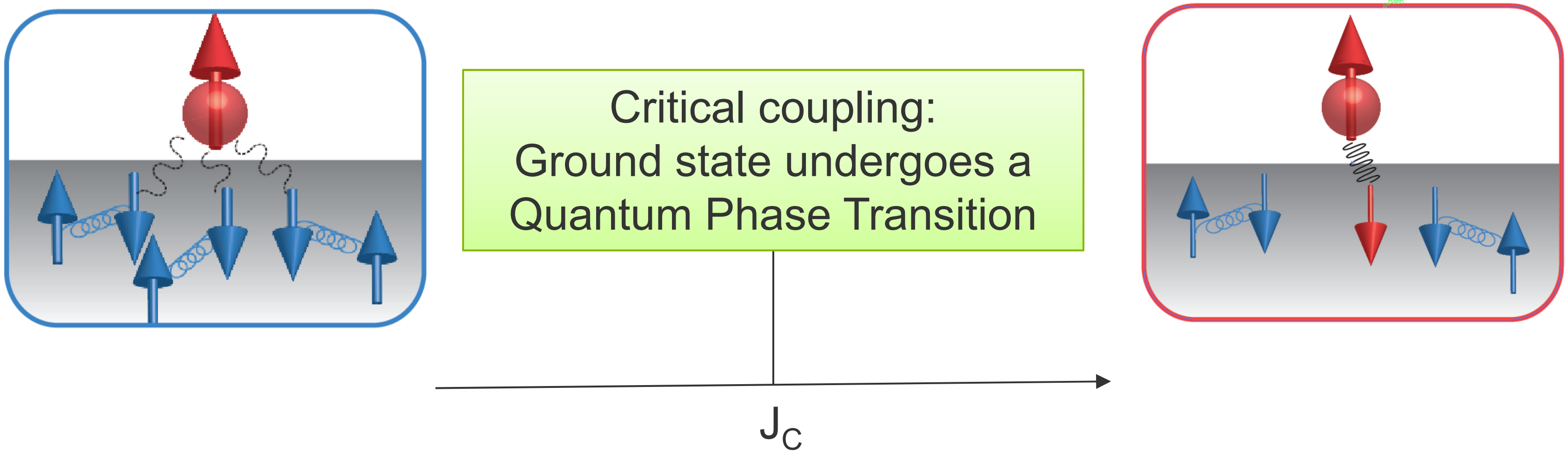
► Yu-Shiba-Rusinov states



L. Yu, Acta Phys. Sin. 21, 75 (1965)
H. Shiba, Prog. Theor. Phys. 40, 435 (1968)
A.I. Rusinov, JETP Lett. 9, 85 (1969)

Yu-Shiba-Rusinov State

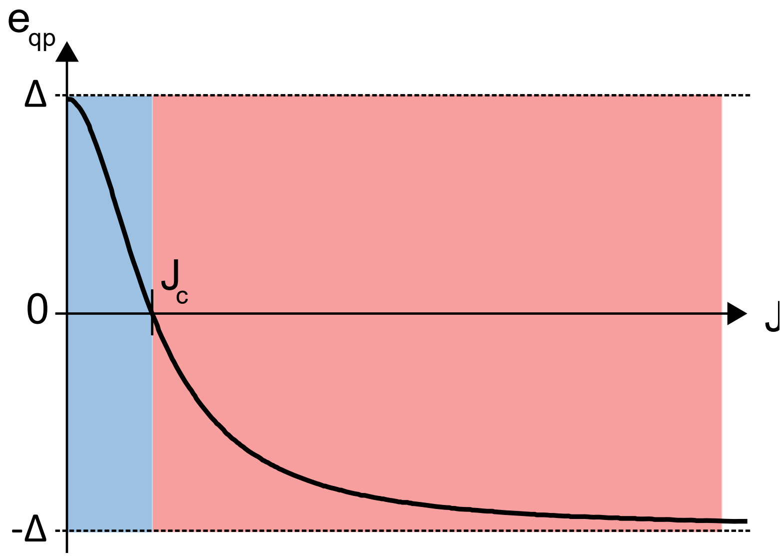
► Competition with superconducting order: two possible ground states



► Energy dependence of e_{qp} (quasi-particle energy)

$$e_{qp} = \Delta \frac{1-(\pi\nu_0 JS)^2}{1+(\pi\nu_0 JS)^2}$$

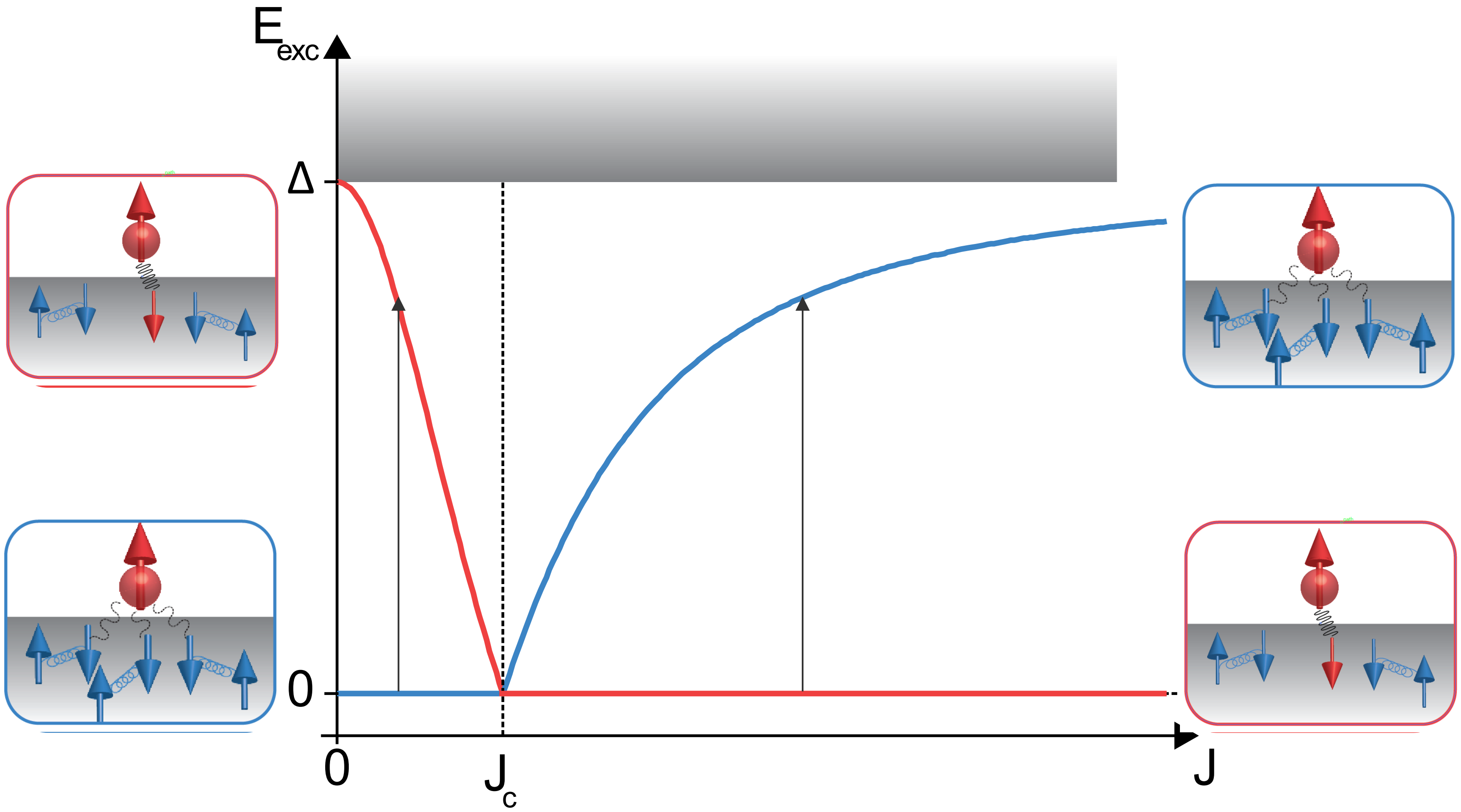
- $e_{qp} > 0$: weak coupling
- $e_{qp} < 0$: bound quasiparticle



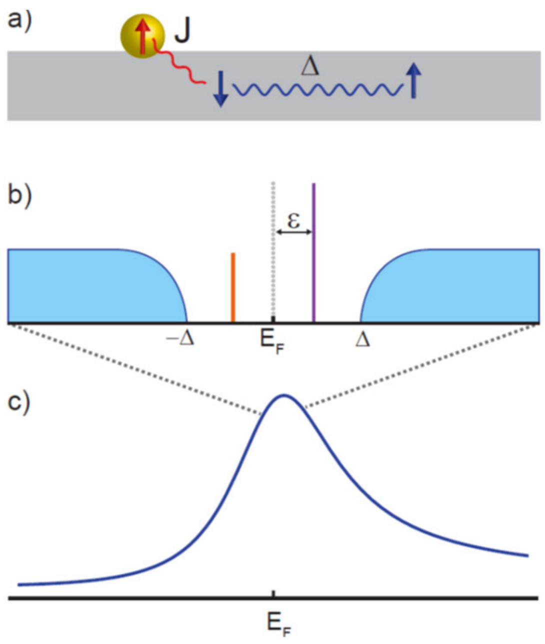
L. Yu, *Acta Phys. Sin.* **21**, 75 (1965)
H. Shiba, *Prog. Theor. Phys.* **40**, 435 (1968)
A.I. Rusinov, *JETP Lett.* **9**, 85 (1969)

Yu-Shiba-Rusinov State

► Energy of the excitation: $e_b = \pm\Delta \frac{1-(\pi\nu_0 JS)^2}{1+(\pi\nu_0 JS)^2}$



Quantum spins on superconductors



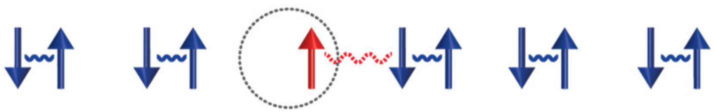
- YSR states within superconducting gap

$$\varepsilon = \Delta \frac{1 - a^2}{1 + a^2} \qquad a \approx \frac{\pi\Delta}{4\,k_B T_K} \ln \frac{4\,k_B T_K}{\pi\Delta} e$$

  - YSR energy scales mainly with T_K
  - YSR asymmetry depends on potential scattering

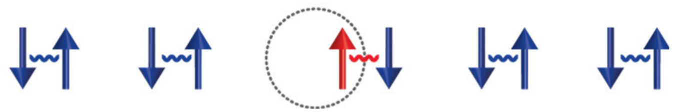
- Kondo resonance outside of the gap

ground state: $S = 1/2$



"free spin"

ground state: $S = 0$

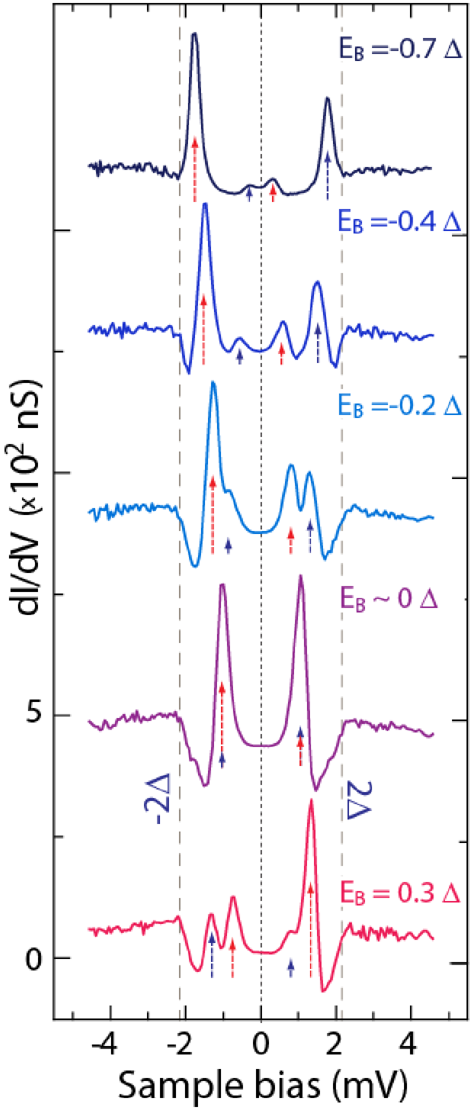


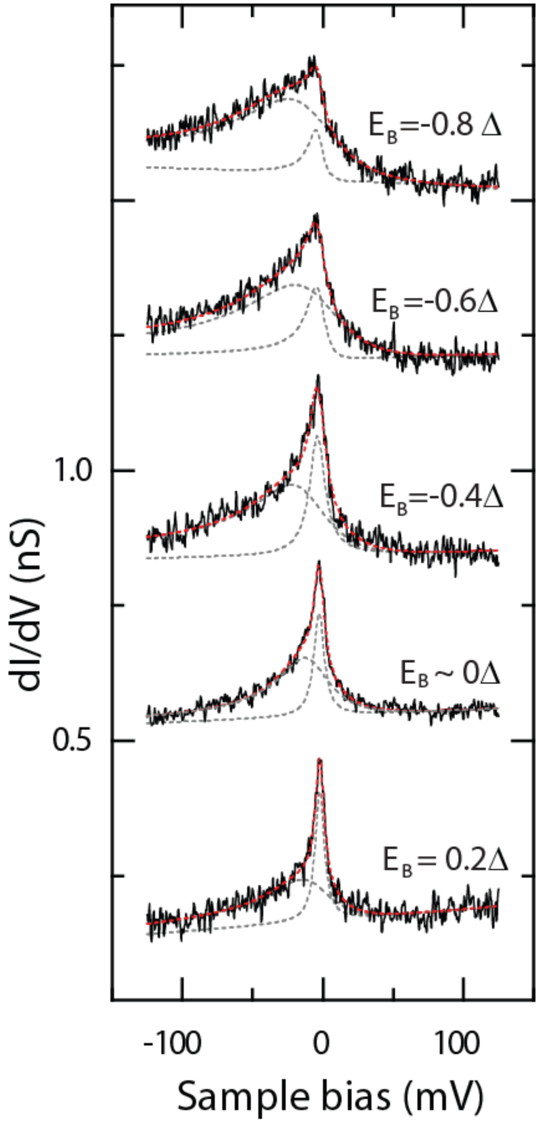
"screened spin"

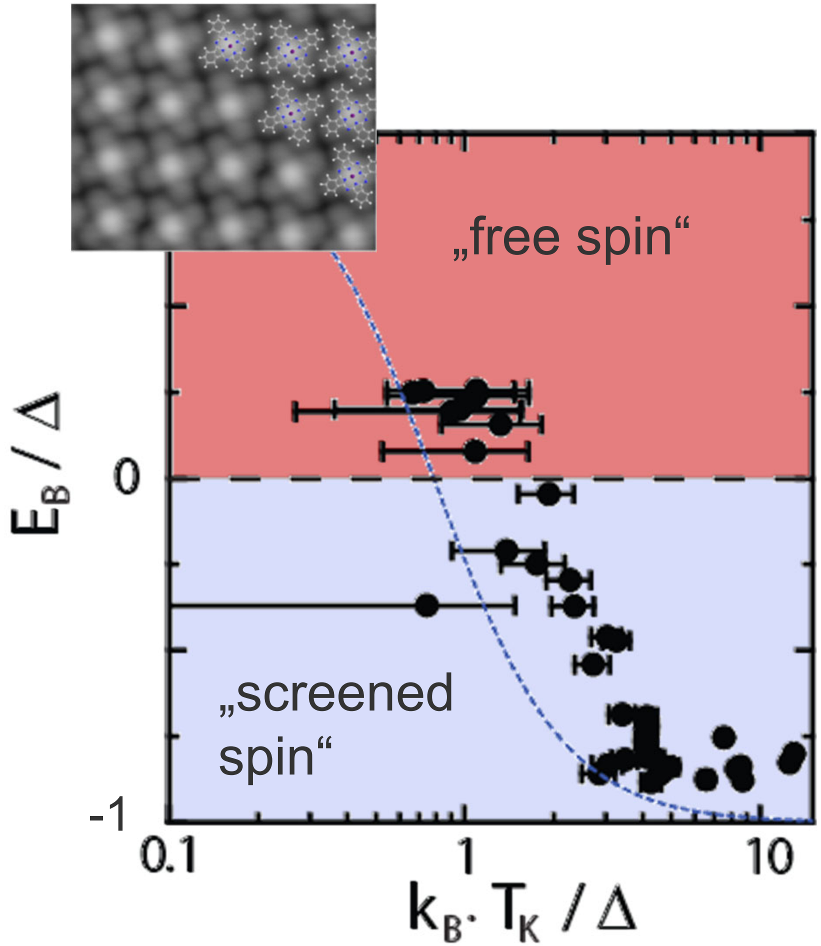
Original YSR papers:
L. Yu, Acta Phys. Sin. 21, 75 (1965)
H. Shiba, Prog. Theor. Phys. 40, 435 (1968)
A.I. Rusinov, JETP Lett. 9, 85 (1969)

B.W. Heinrich, et al. Prog. Surf. Sci. 93, 1 (2018)

Kondo screening vs. YSR bound states







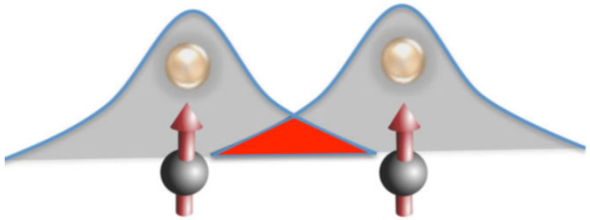
- Correlation of YSR and Kondo
- Quantum phase transition

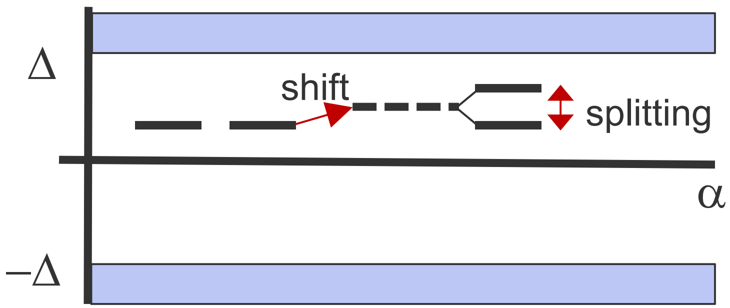
Franke, Schulze, Pascual, Science, 332, 940 (2011)

T. Matsuura, Prog. Theor. Phys. 57, 1823 (1977)

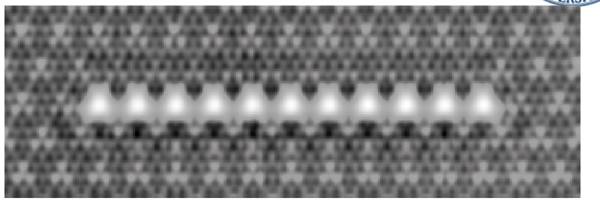
See also: A. Odobesko et al., Phys. Rev. B 102, 174504 (2020)

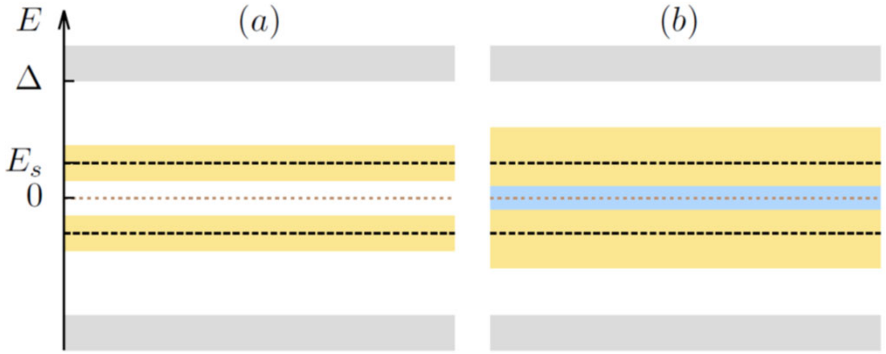
Coupling of YSR states





Yao et al., PRB 90 (2014), Flatte et al., PRB 61 (2000), Yao et al., PRL 113 (2014), Hoffman et al., PRB 92 (2015)





Pientka et al., Phys. Rev. B 88, 155420 (2013)
Pientka et al., Phys. Scr. 014008 (2015)

Design from YSR states:

- Dilute magnetic adatoms
- Coupling of YSR states
- YSR bands crossing Fermi level
  - possibly topological gap opening

Outline

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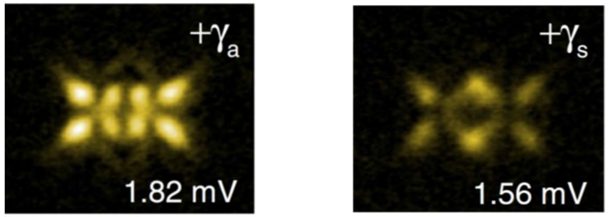


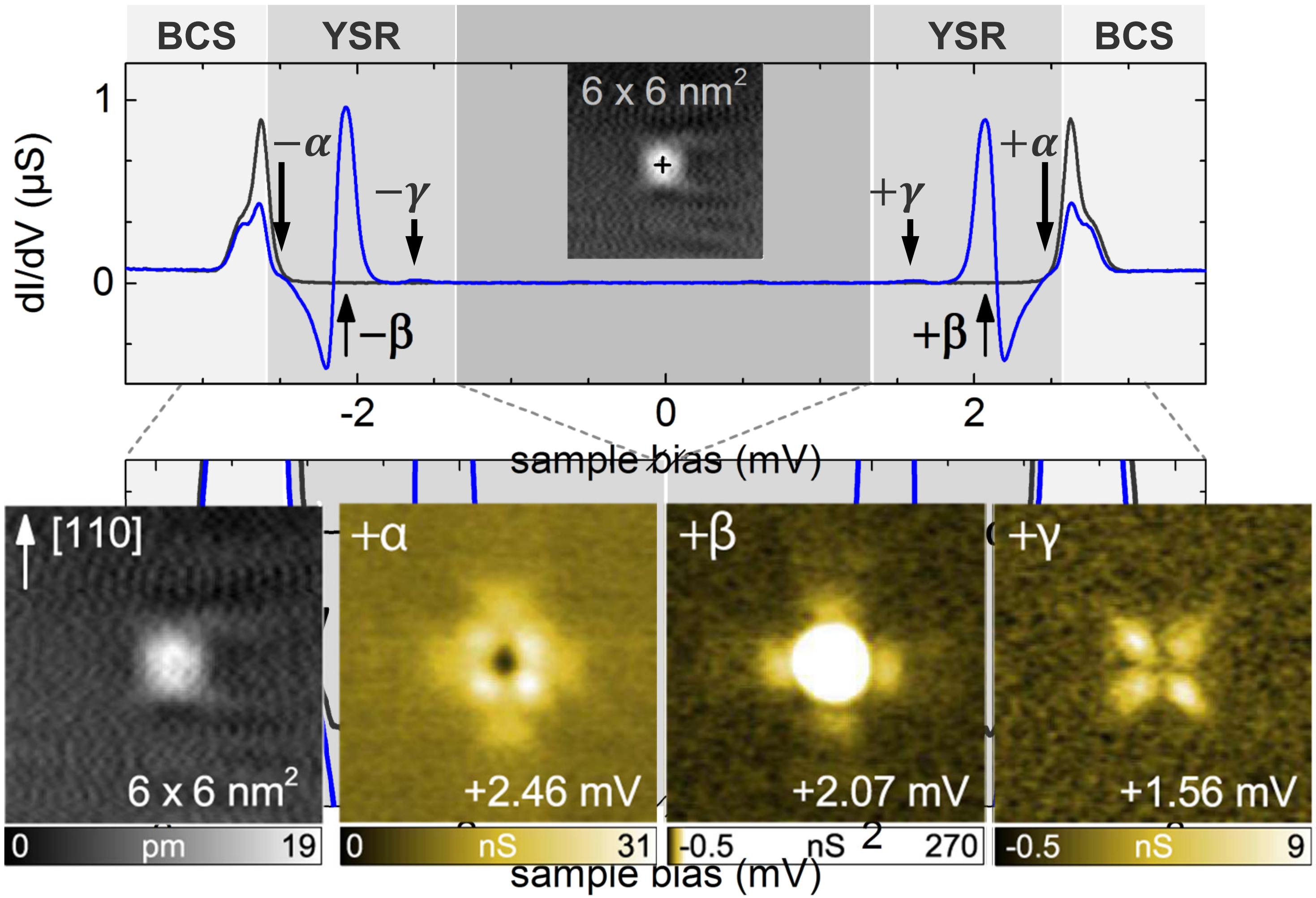
- Hybridisation of YSR states (on Pb)

- Interaction of magnetic atoms on $NbSe_2$

- Evolution of YSR states in atomic chains on $NbSe_2$

- Influence of charge-density wave on magnetic chains

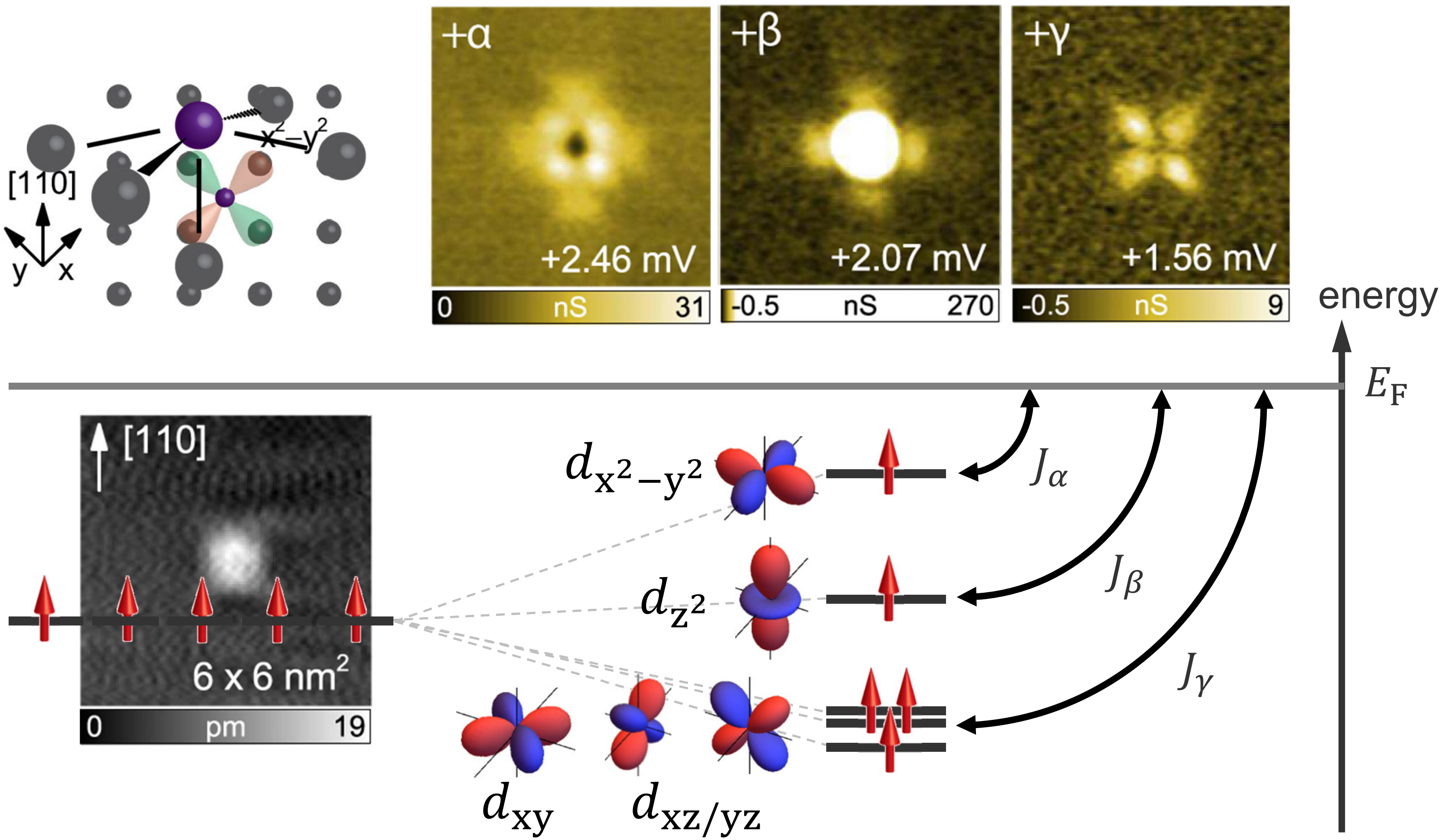
Manganese adatoms on Pb(001)



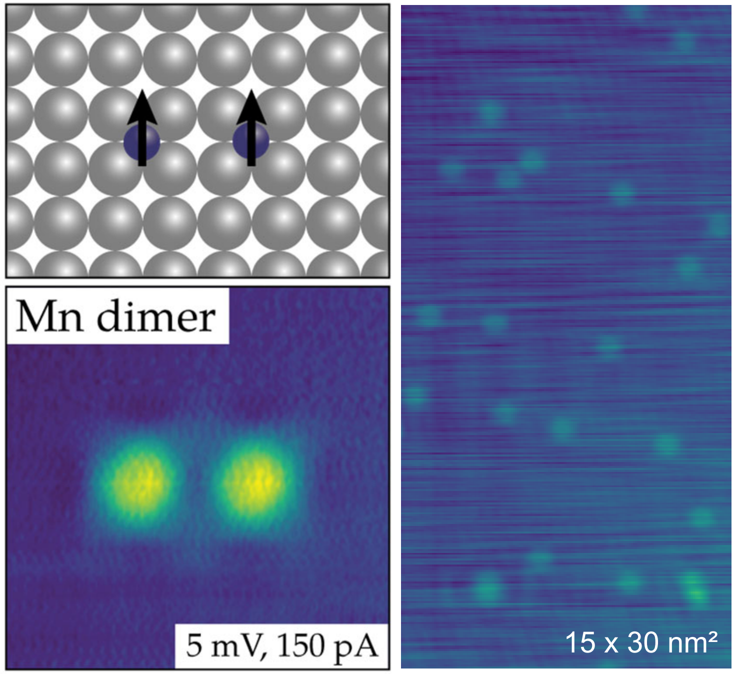
- 3 pairs of YSR resonances

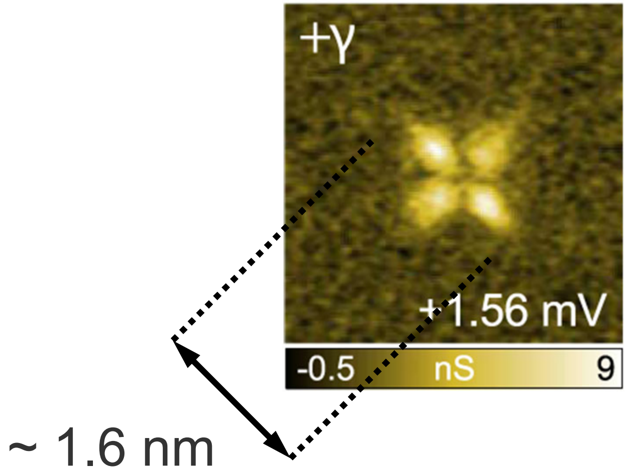
M. Ruby et al., Phys. Rev. Lett. 117, 186801 (2016)

Pb(001): origin of multiple YSR states



M. Ruby et al., Phys. Rev. Lett. 117, 186801 (2016)





- large lateral extension:

$$\Psi(r) \propto \frac{1}{k_{\mathrm{F}} r} \mathrm{e}^{-r/\xi}$$

Theoretical predictions of coupled YSR states: Yao *et al.*, PRB **90** (2014), Flatte *et al.*, PRB **61** (2000), Yao *et al.*, PRL **113** (2014) Hoffman *et al.*, PRB **92** (2015)
More recent experiments on dimers: Kezilebieke *et al.*, Nano Lett. **18** (2018), Choi, et al., PRL **120** (2018), Beck *et al.*, Nat Comm. **12** (2021), Ding et al., PNAS **118** (2021); QDs: Grove-Rasmussen *et al.*, Nat. Commun. **9** (2018)

Splitting of YSR states in dimers

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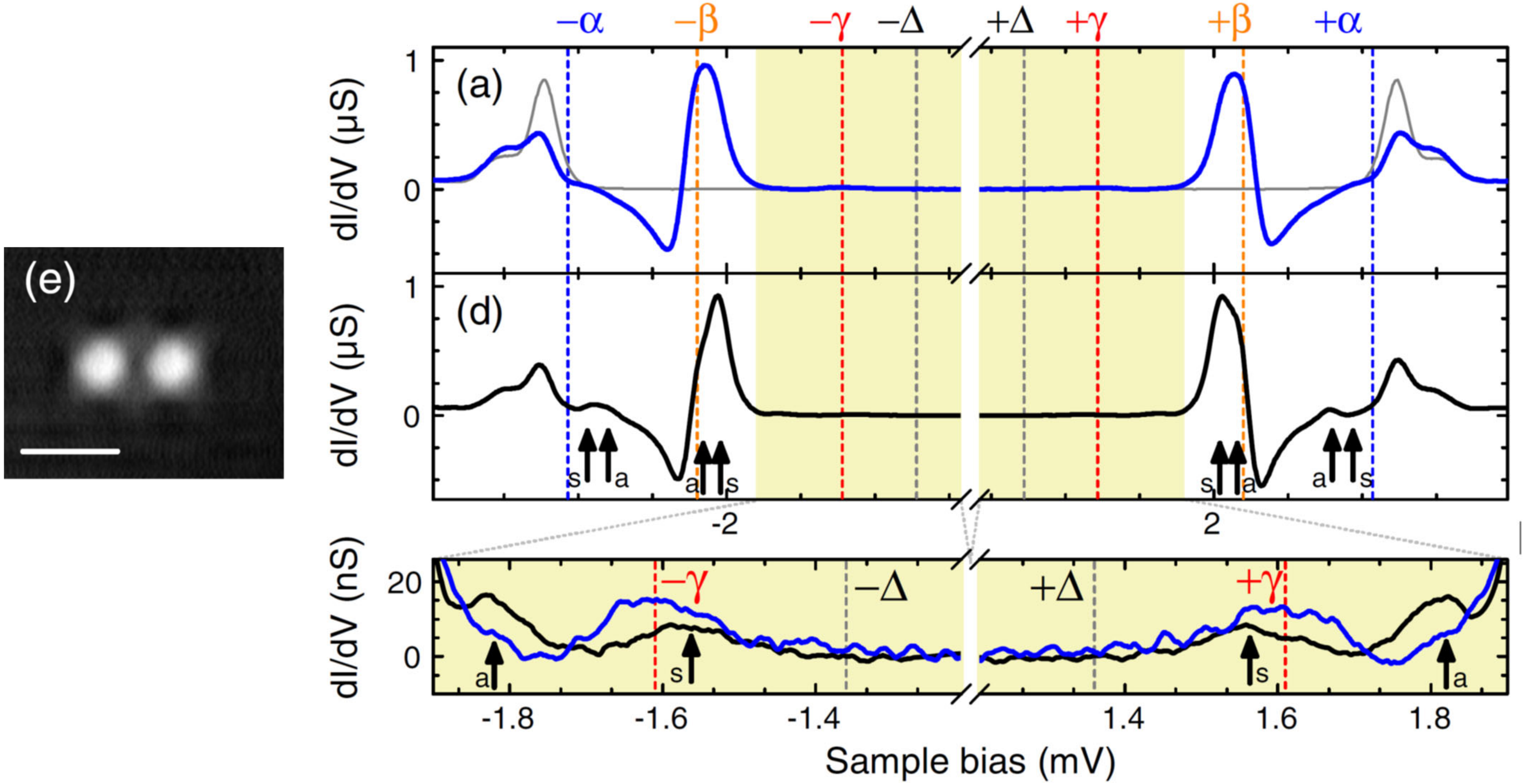
Dimer axis along [110]



M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

Orbital structure

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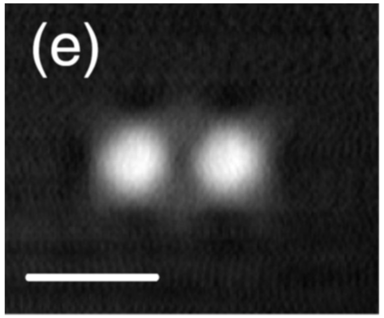
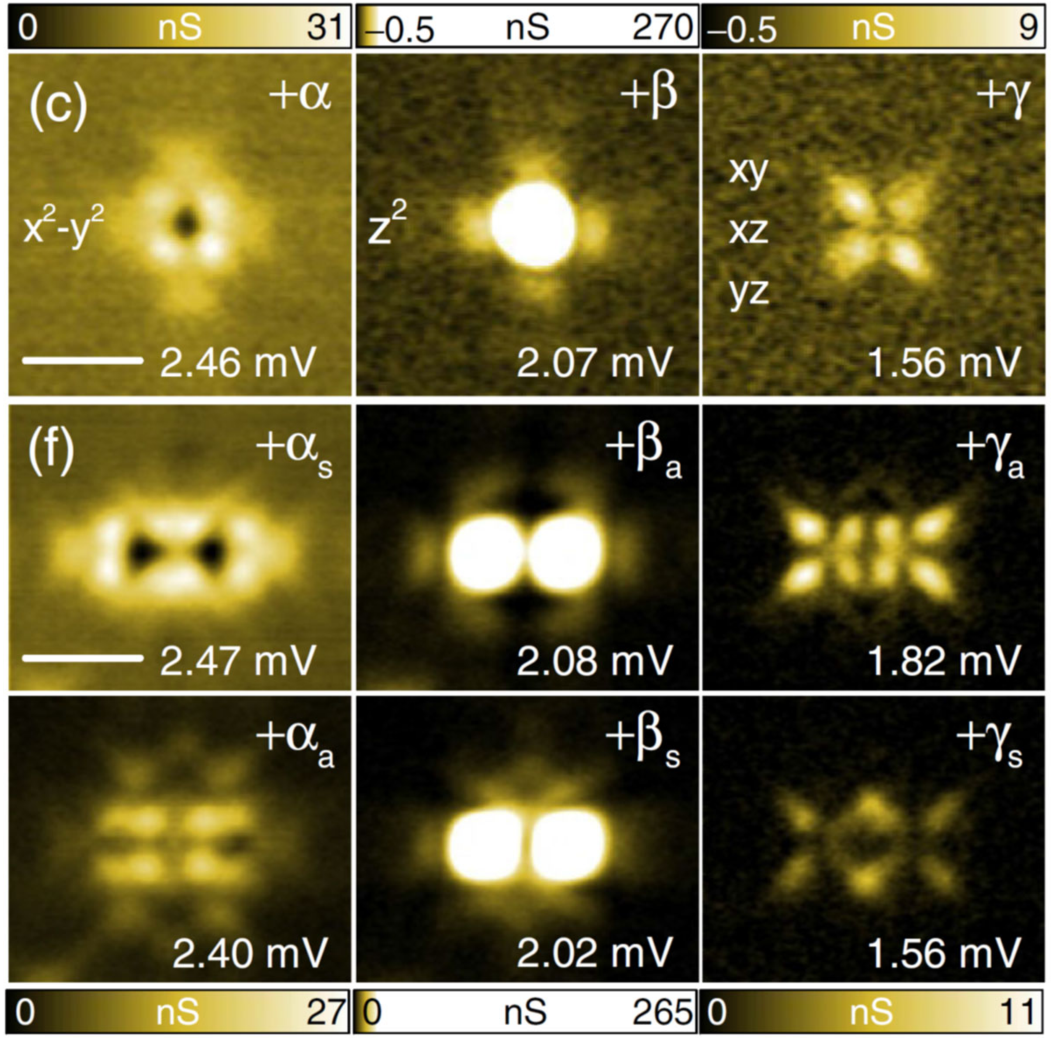
Dimer axis along [110]



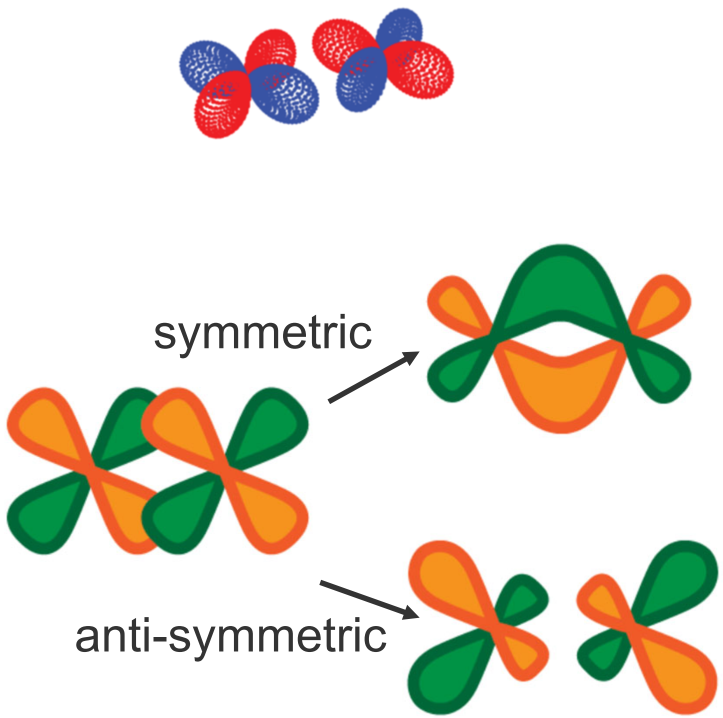


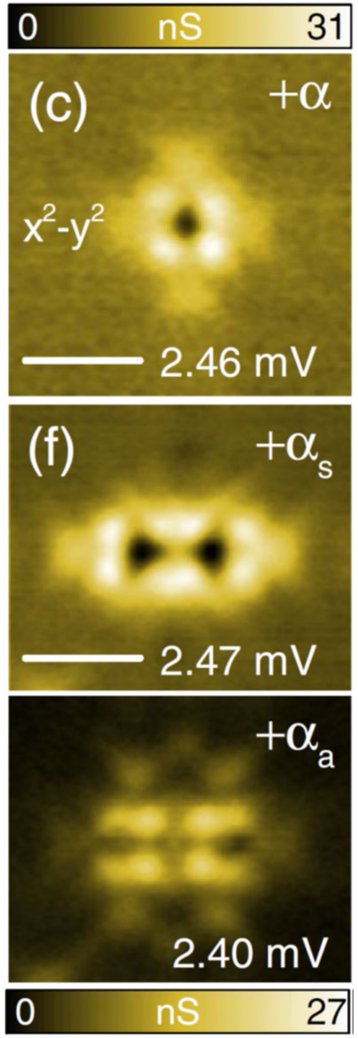
M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

α resonances

Dimer axis along [110]

α: $d_{x^2-y^2}$



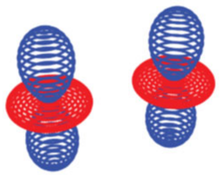


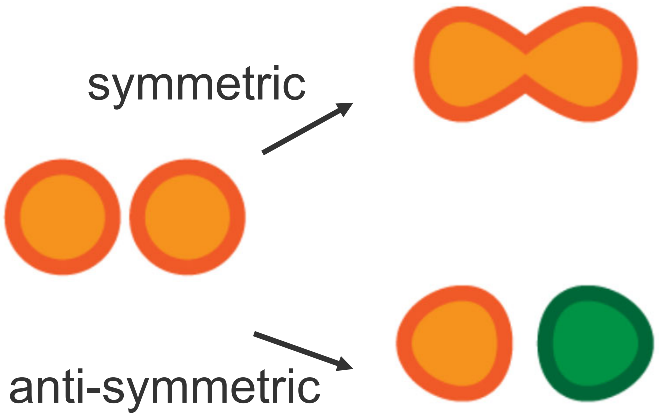
M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

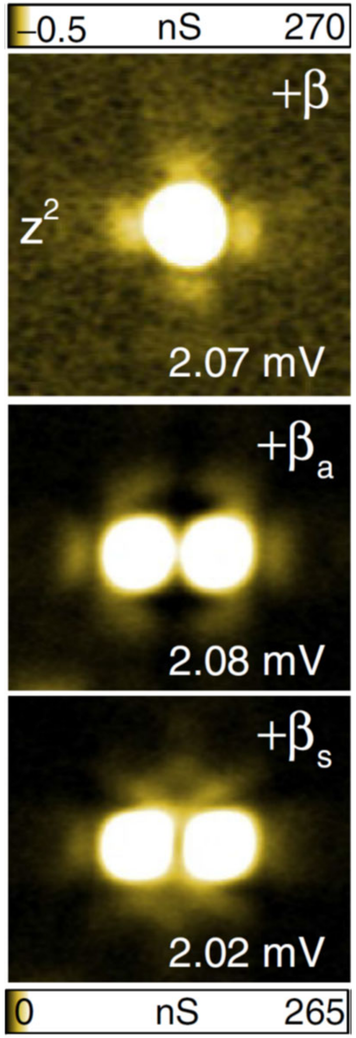
β resonances

Dimer axis along [110]

β: d_{z^2}



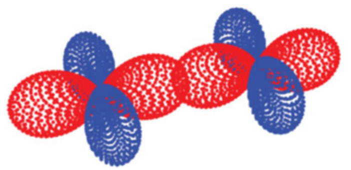




M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

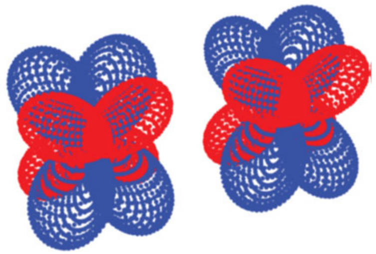
γ resonance

d_{xy}

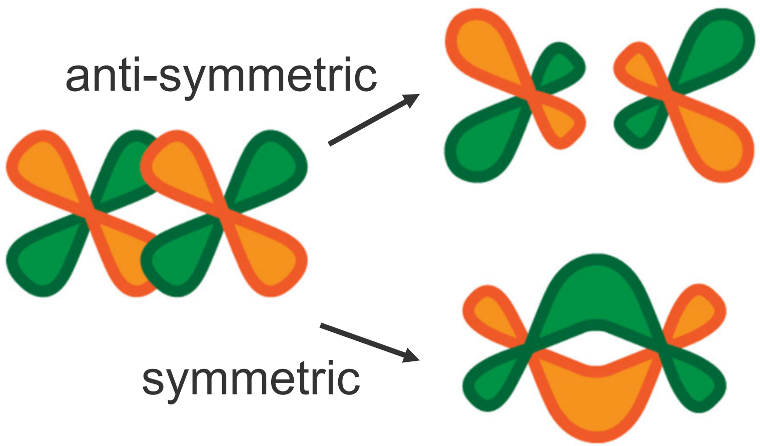


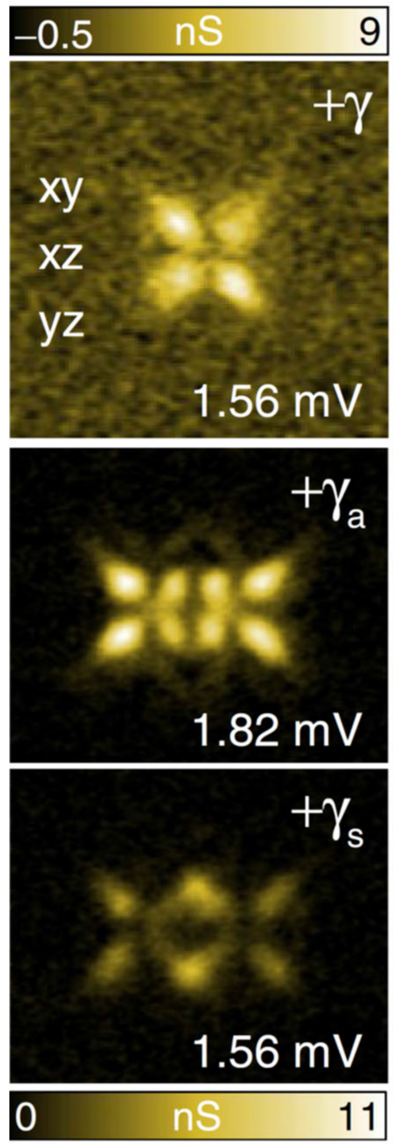
not visible

$d_{xz,\,yz}$



equal splittings of (anti) symm. combinations → 2 dimer resonances





M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

Hybridization of YSR states in dimers

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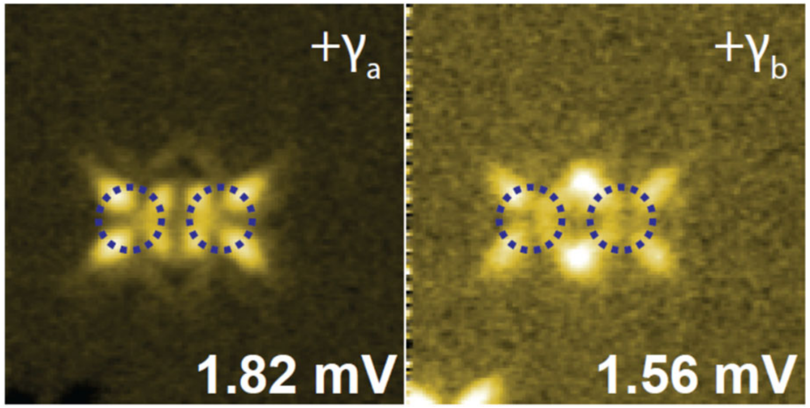
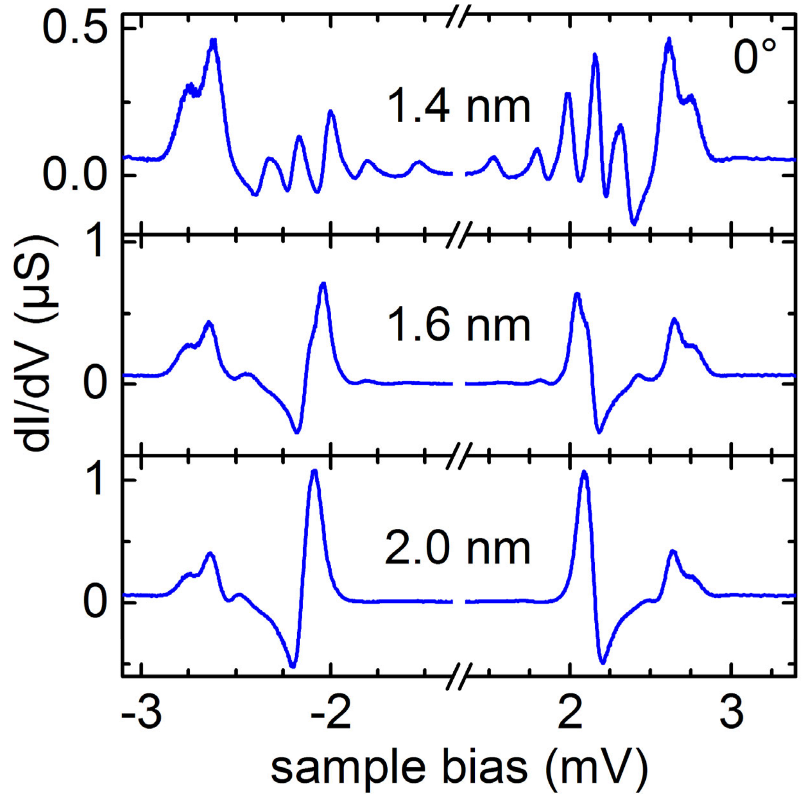




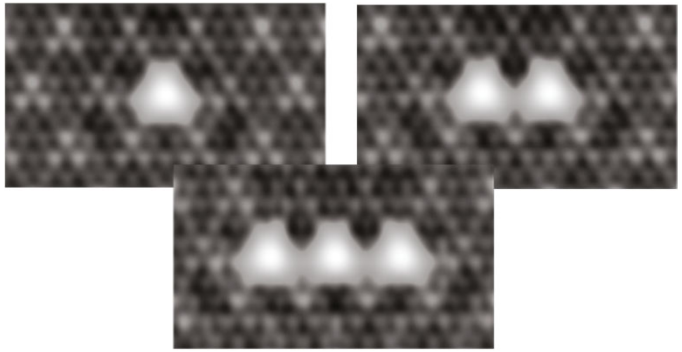
► symmetric and anti-symmetric character of split Shiba states

► strength of splitting depends on distance and orientation

M. Ruby et al., Phys. Rev. Lett. 120, 156803 (2018)

Outline

► Hybridisation of YSR states (on Pb)



► Interaction of magnetic atoms on $NbSe_2$

► Evolution of YSR states in atomic chains on $NbSe_2$

► Influence of charge-density wave on magnetic chains

Quasi-2D superconductor

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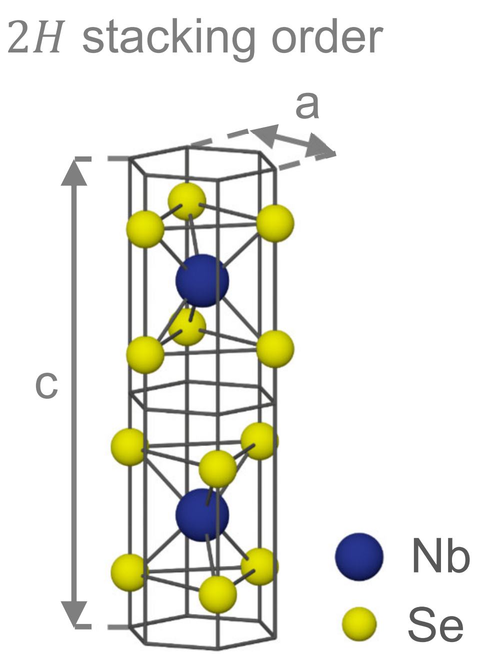
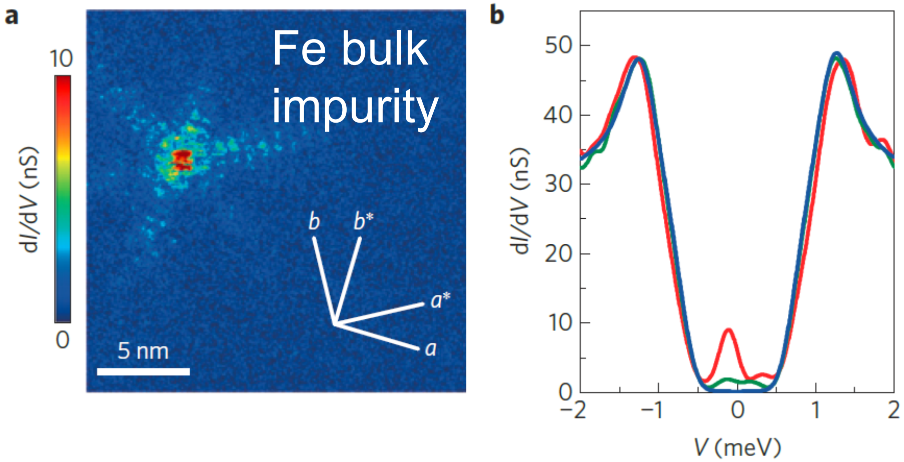
- van der Waals stacked layers of $NbSe_2$



- critical temperature: T_c=7.2 K

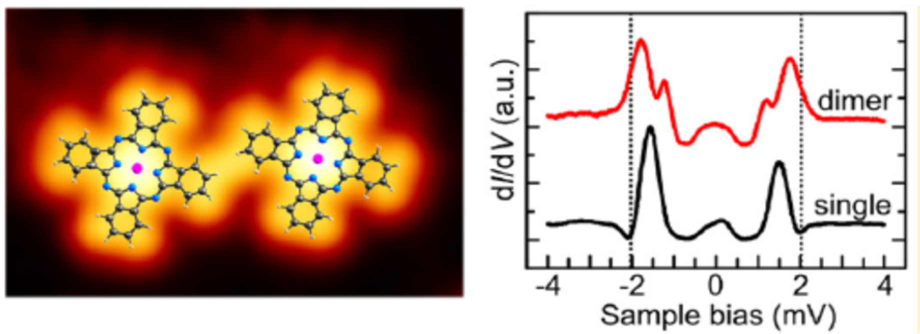
Previous YSR states in 2H-$NbSe_2$



Ménard, et al., Nature Phys. 11, 1013 (2015)

- Bulk Fe impurities create long-range YSR states
- Decay with $1/\sqrt{r}$

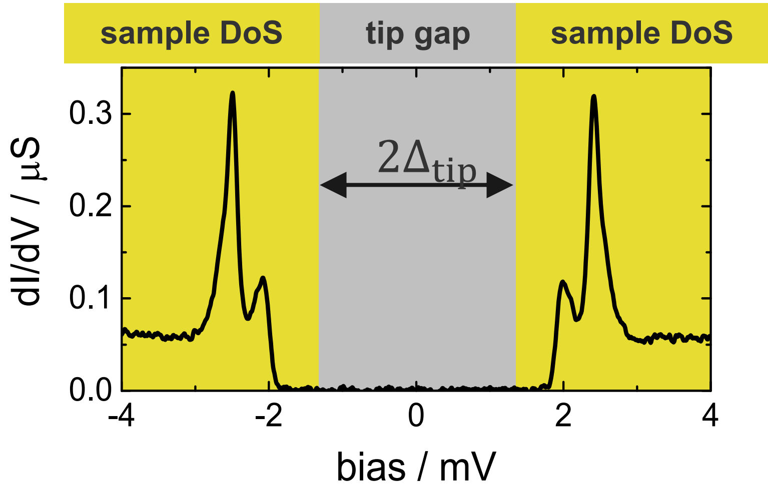
CoPc

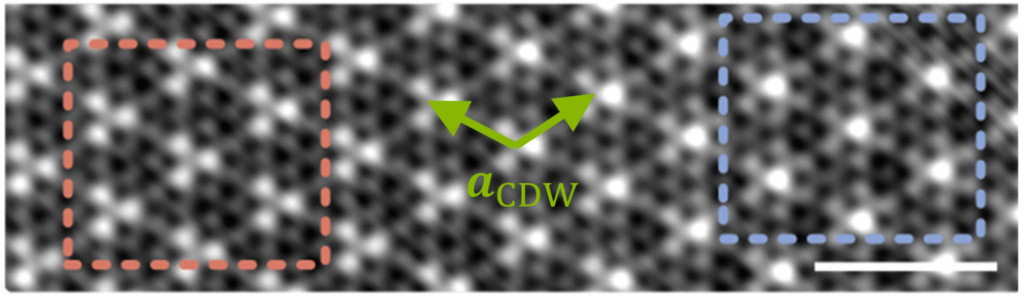


Kezilebieke, et al., Nano Lett. 18, 2311 (2018)

- Magnetic molecules coupled via YSR states

$2H$-$NbSe_2$ - substrate





superconductivity

- $T_{SC} \approx 7.2\mathrm{K}$
- multiband/anisotropic superconductor

charge density wave

- $T_{CDW} \approx 33\mathrm{K}$
- **roughly** 3×3 – superstructure

STS using superconducting Pb tips at $T \approx 1.1\mathrm{K}$

↳ energy shifted by $\pm\Delta_{tip}$

Smooth transition between different regions of CDW

$2H$-$NbSe_2$ – charge density wave

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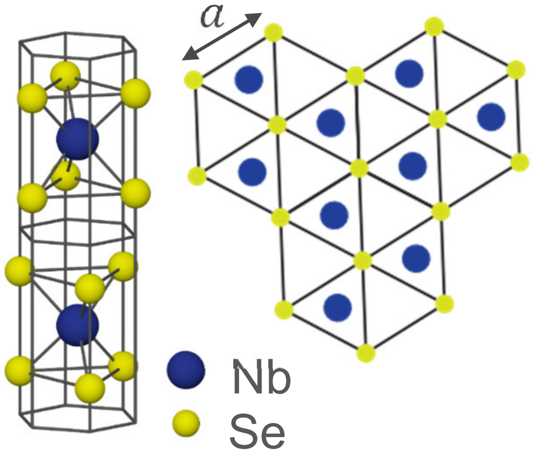
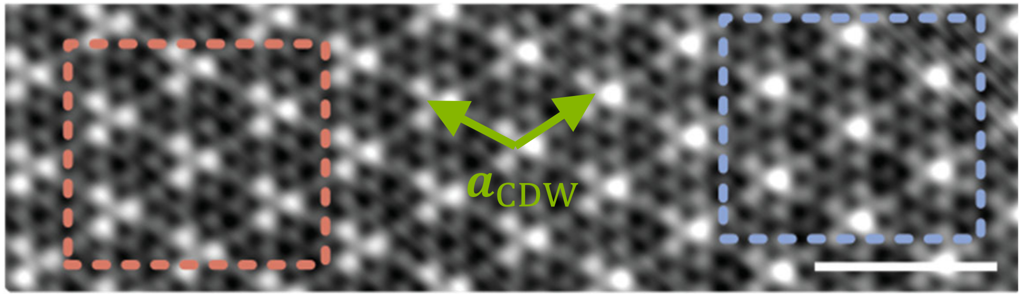
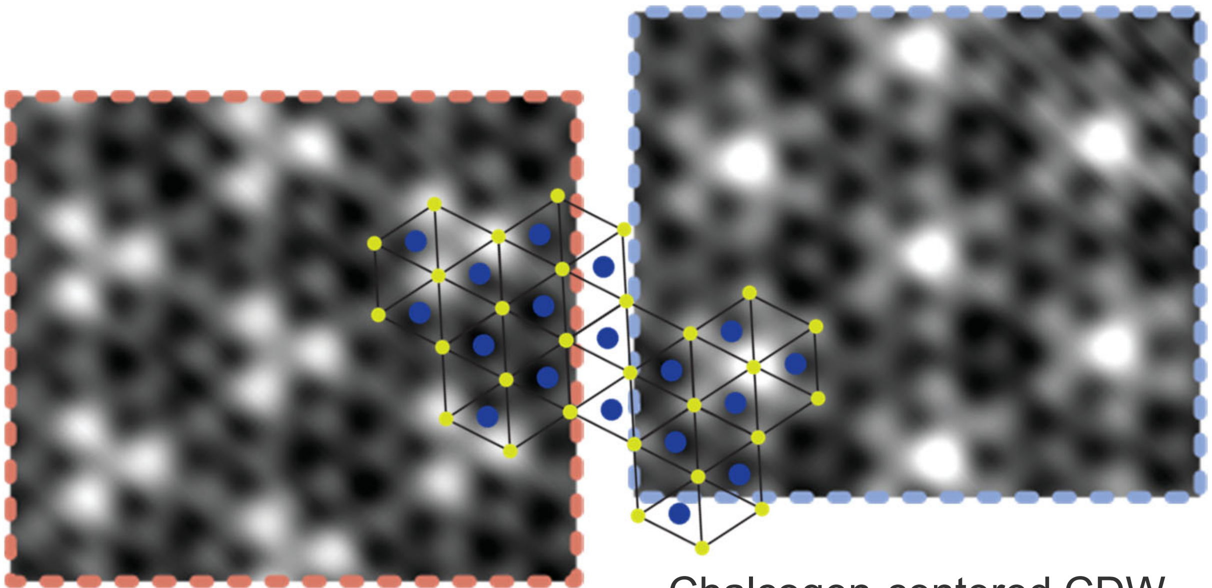


- Modulation of YSR states?
- Template for YSR chains?

Smooth transition between different regions of CDW



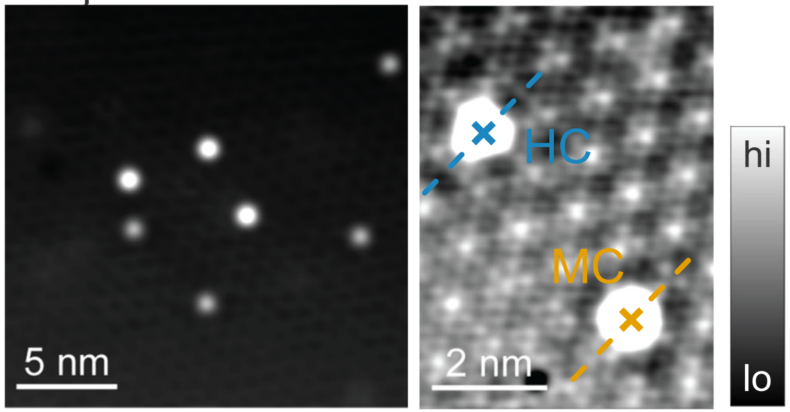


Chalcogen-centered CDW

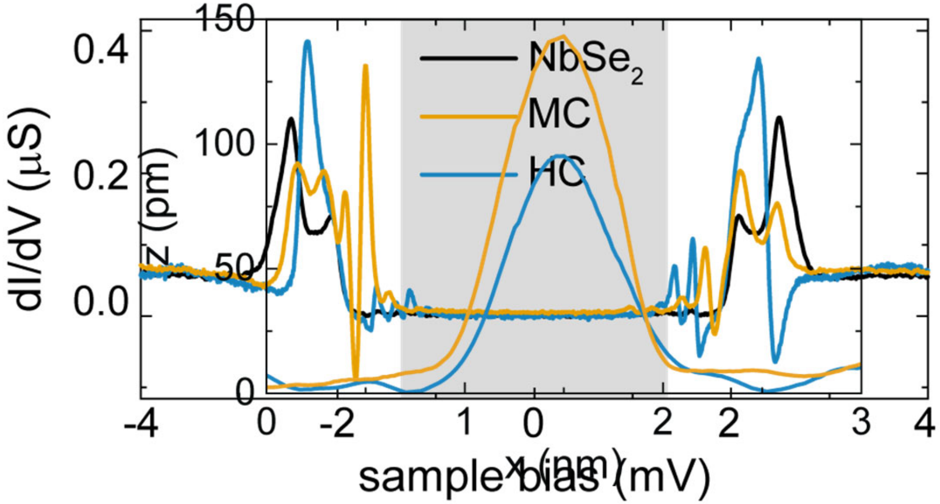
Hollow-centered CDW

$2H$-$NbSe_2$ + Fe atoms

Evaporation of Fe atoms at $T < 12\mathrm{K}$

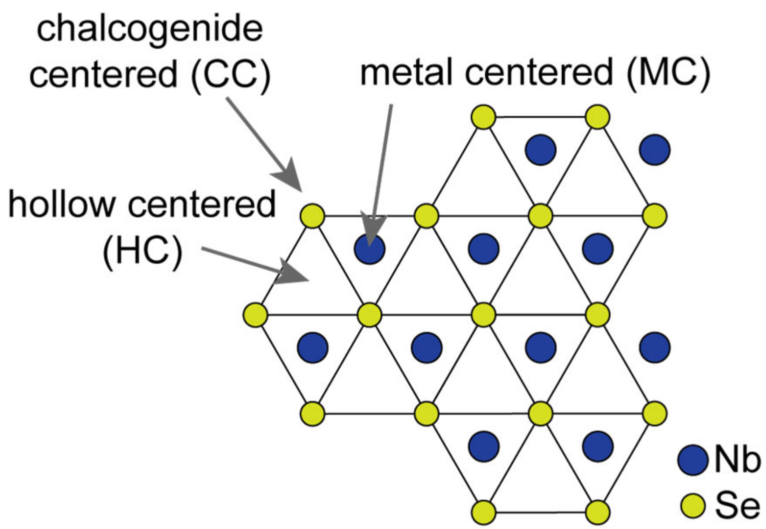


500mV, 80pA   4mV, 200pA



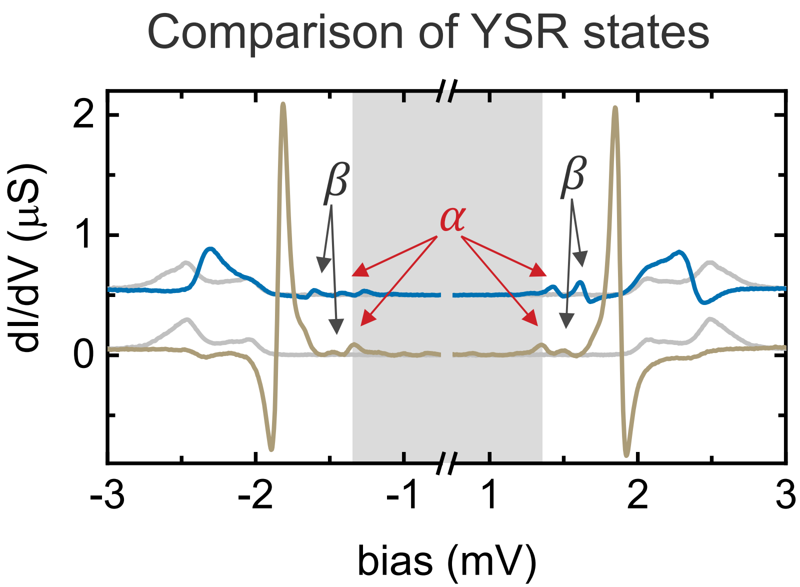
- different apparent heights
- adsorption in HC and MC
- both species show several YSR states

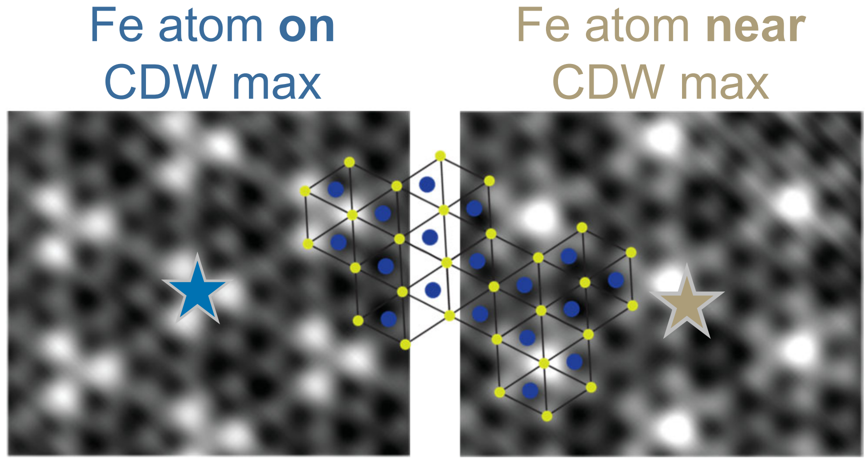
  (unpaired e^- in d-orbitals)



E. Liebhaber, S. Acero Gonzalez, et al., Nano Lett. 20, 339 (2020)

YSR states on $2H$-$NbSe_2$



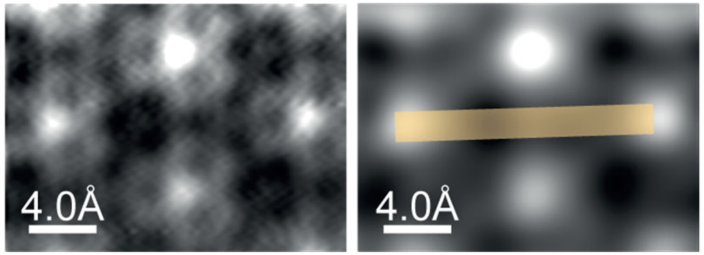


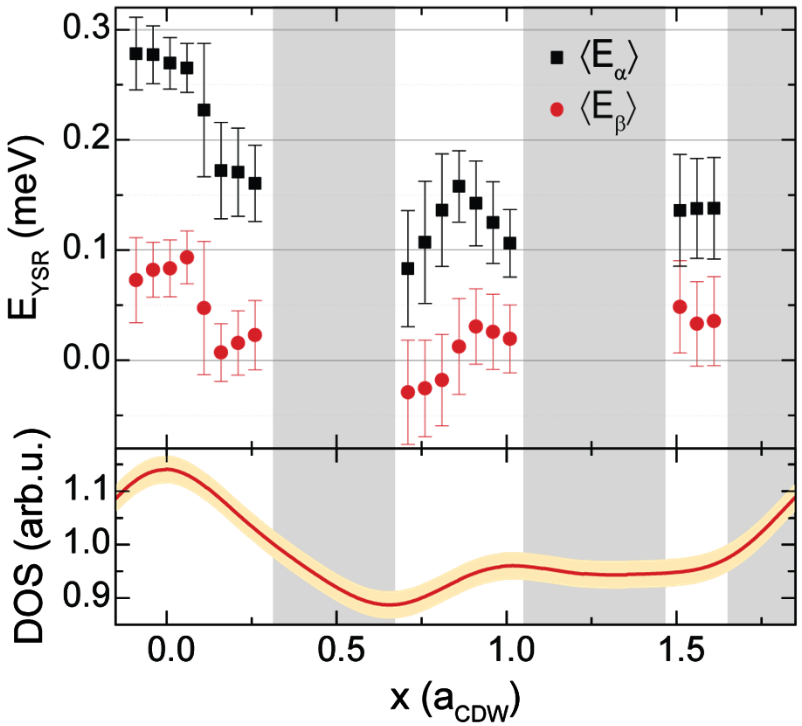
- YSR states depend on CDW

E. Liebhaber, S. Acero Gonzalez, et al., Nano Lett. 20, 339 (2020)

Tracking the energy shift

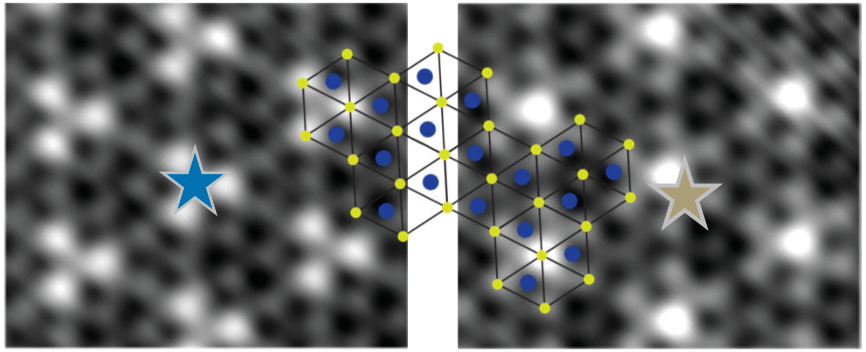
Const. Z dI/dV map ($V_{\text{bias}} = 0$, T = 8 K)

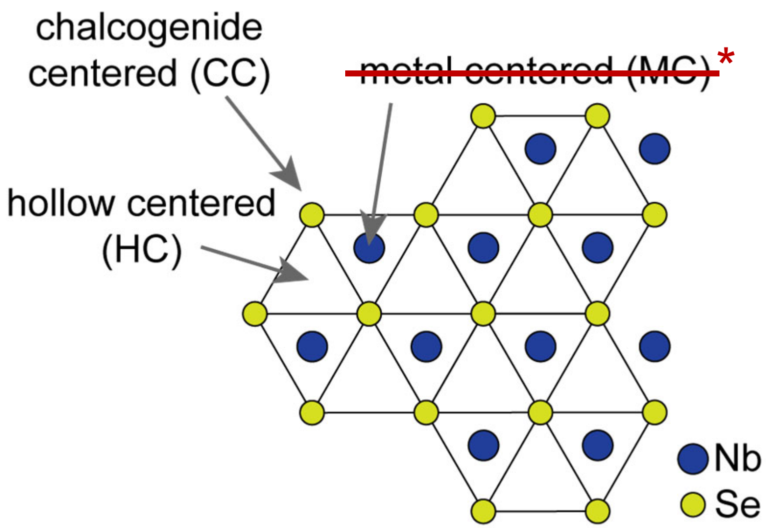




Fe atom **on** CDW max

Fe atom **near** CDW max



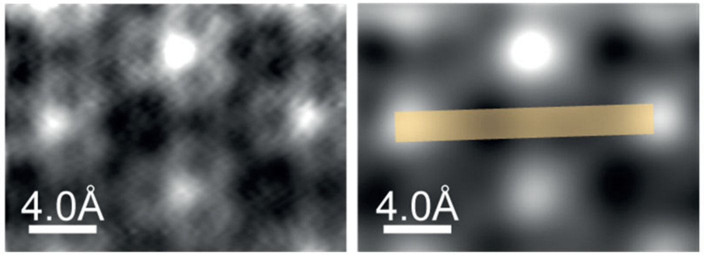


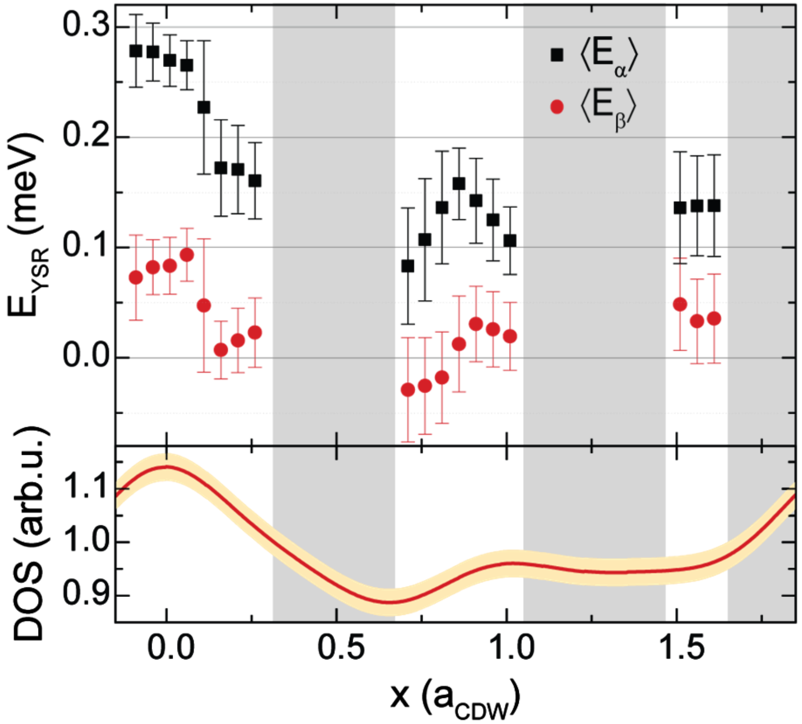
*Gye, Oh, Yeom – *Phys. Rev. Lett.* **122**, 016403 (2019)

E. Liebhaber, S. Acero Gonzalez, et al., Nano Lett. 20, 339 (2020)

Tracking the energy shift

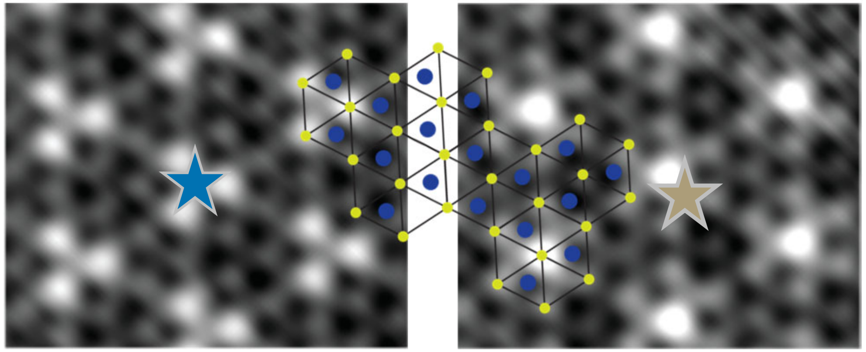
Const. Z dI/dV map ($V_{\text{bias}} = 0$, $\text{T} = 8$ K)





Fe atom **on** CDW max

Fe atom **near** CDW max



Yu-Shiba-Rusinov bound states:

$$\epsilon_S = \Delta \frac{1 - A^2 + B^2}{\sqrt{4A^2 + (1 - A^2 + B^2)}}$$

$$A = \frac{\pi}{2} J\, S\, \nu_0$$

$$B = \pi\, K\, \nu_0$$

- YSR energy scales with CDW

E. Liebhaber, S. Acero Gonzalez, et al., Nano Lett. 20, 339 (2020)

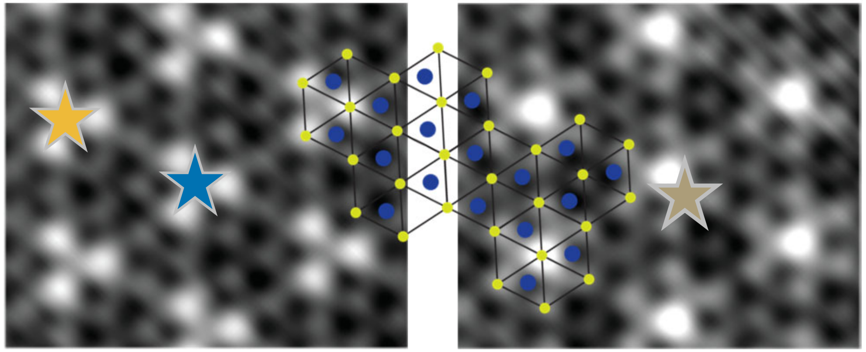
Choice of adsorption sites for chains

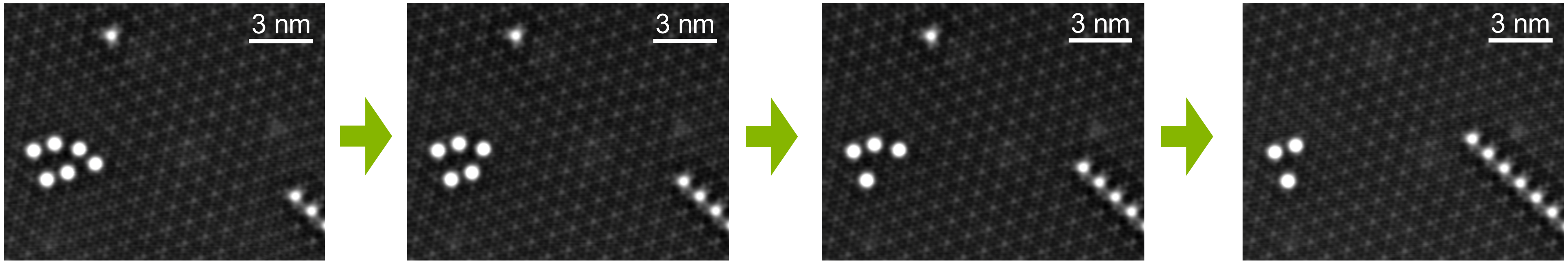
Freie Universität Berlin

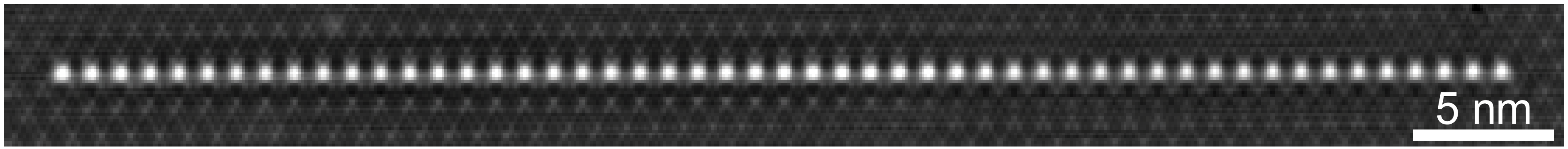
- Assembly of chains by atom manipulation
- Placement of atoms in 3a distance: similar w.r.t. CDW
- Dilute limit of coupling

Fe atom **on** CDW max

Fe atom **near** CDW max

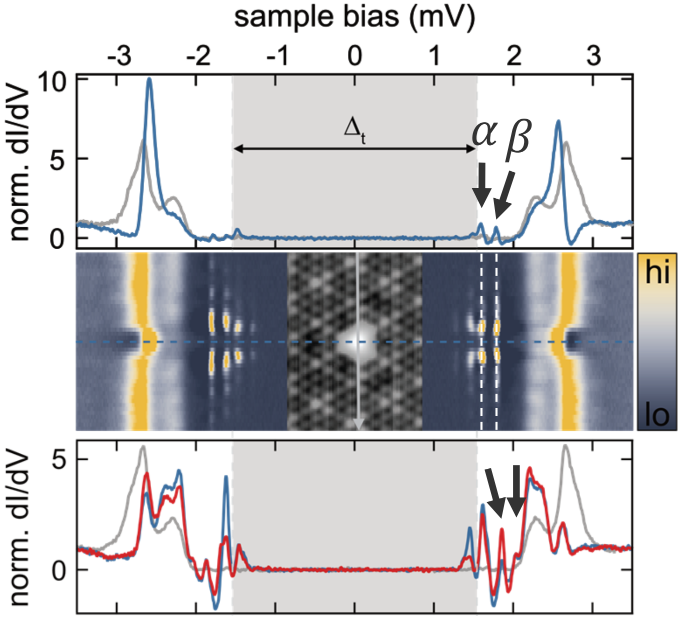


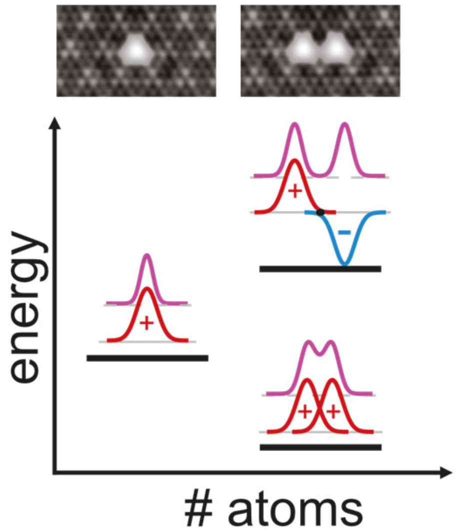


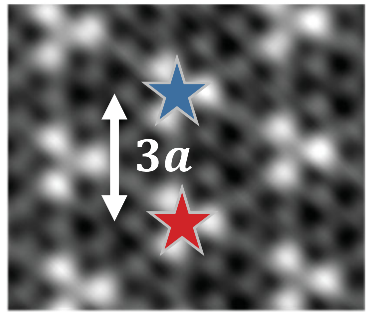


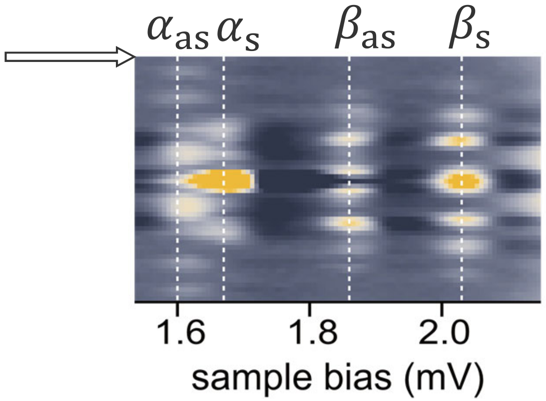
Theoretical predictions of coupled YSR states: Yao *et al.*, PRB **90** (2014), Flatte *et al.*, PRB **61** (2000), Yao *et al.*, PRL **113** (2014) Hoffman *et al.,* PRB **92** (2015)
Theoretical YSR chains: Pientka, *et al.*, Phys. Rev. B **88** (2013), Phys. Scr. 014008 (2015)
Experiments on dimers: Kezilebieke *et al.,* Nano Lett. **18** (2018), Choi, et al., PRL **120** (2018), Beck *et al.*, Nat Comm. **12** (2021), Ding et al., PNAS **118** (2021); QDs: Grove-Rasmussen *et al.,* Nat. Commun. **9** (2018)

YSR hybridization





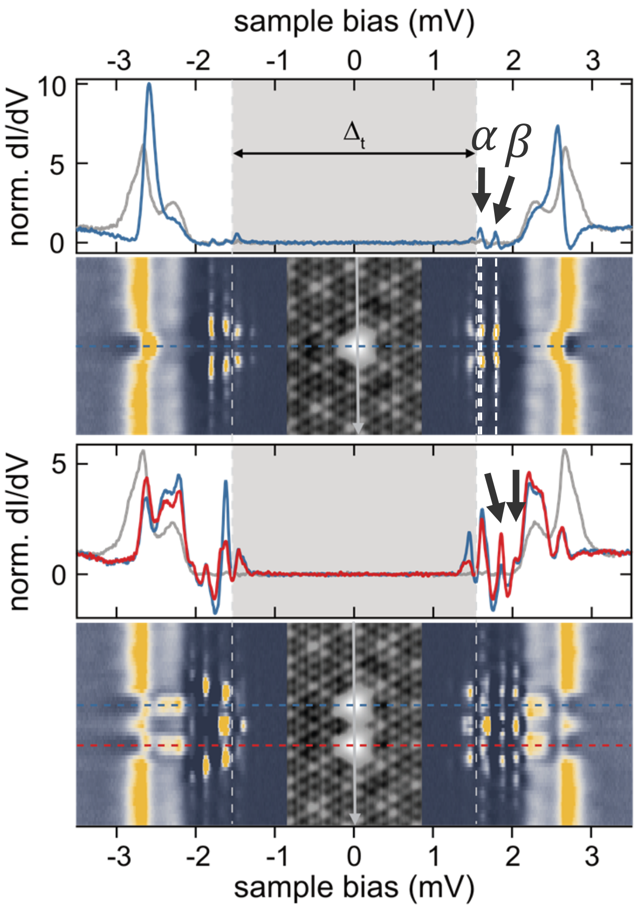


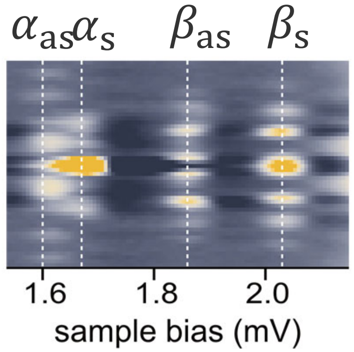


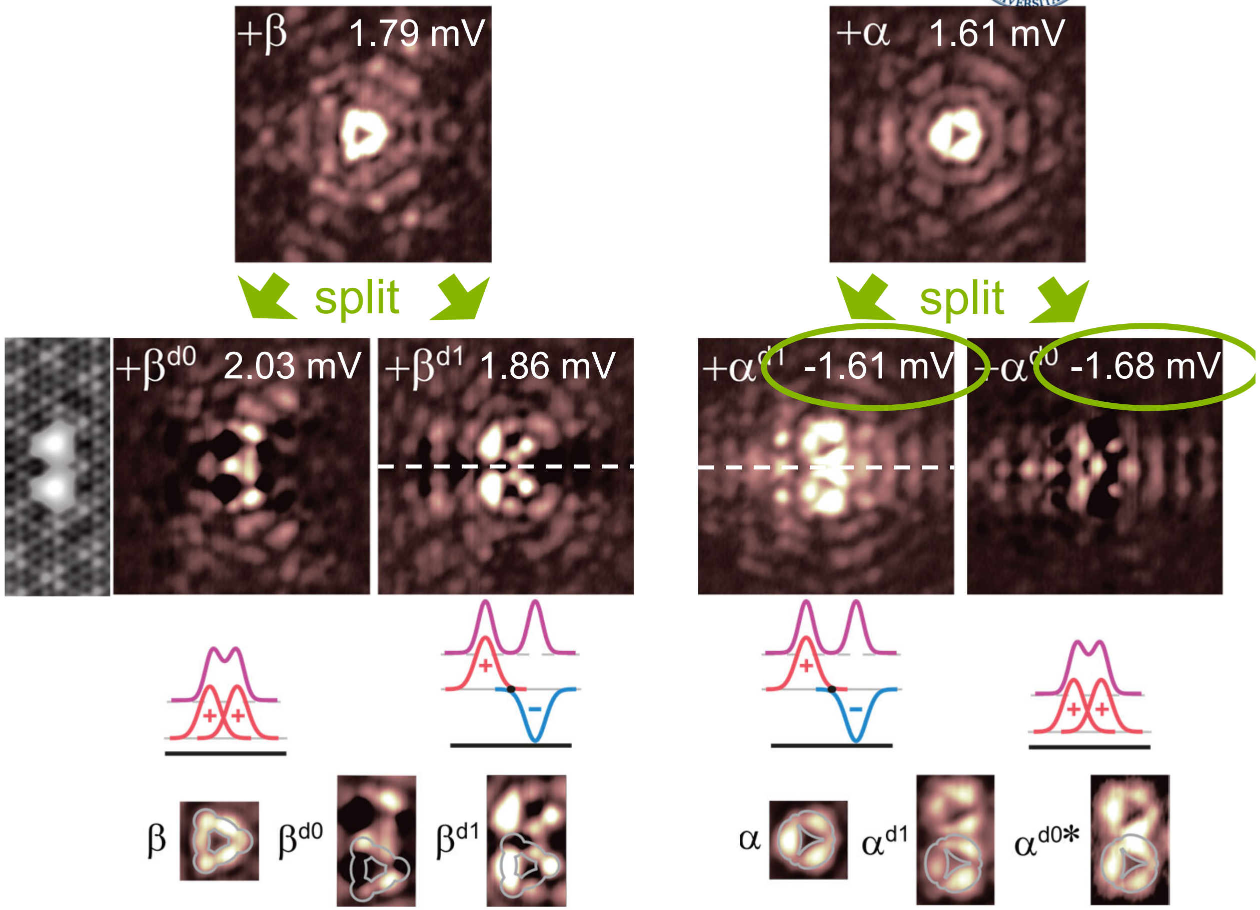
- Doubling of YSR resonances

→ Hybridization in dimers with spacing of $3a \approx a_{CDW}$

YSR hybridization







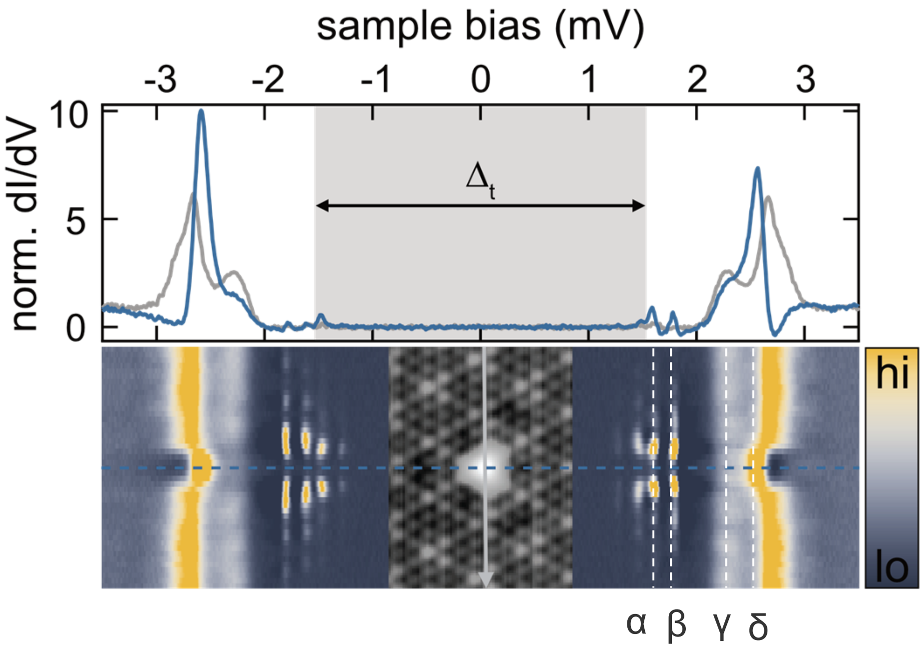
- Symmetric and antisymmetric wave functions

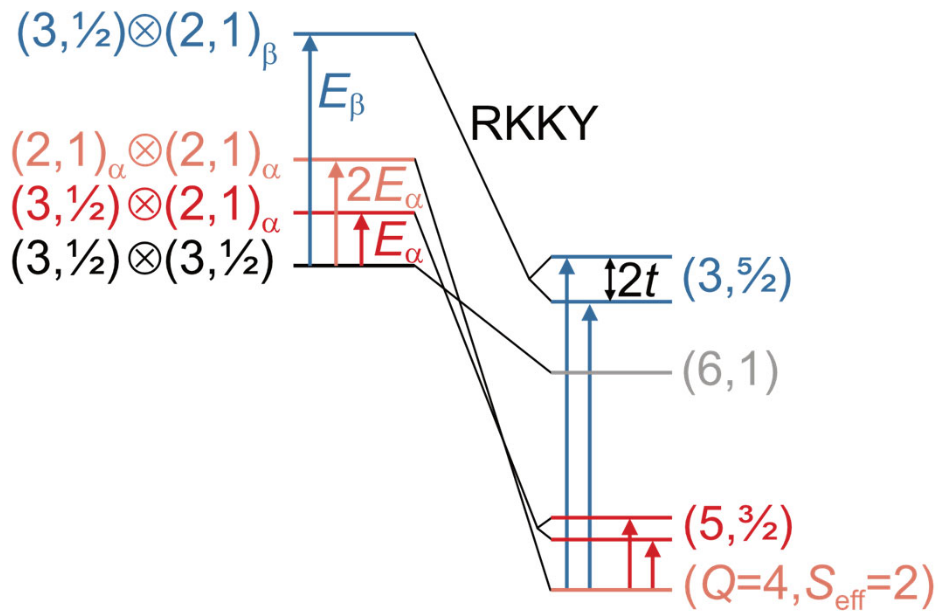
→ Hybridization in dimers with spacing of $3a \approx a_{\mathrm{CDW}}$

- Shift of α resonance through Fermi level

→ Quantum phase transition

RKKY interaction





Fe monomer:

- Four YSR resonances

→ S=2

- α-, β-, γ- resonances screened
- δ-resonance unscreened

→ S=1/2

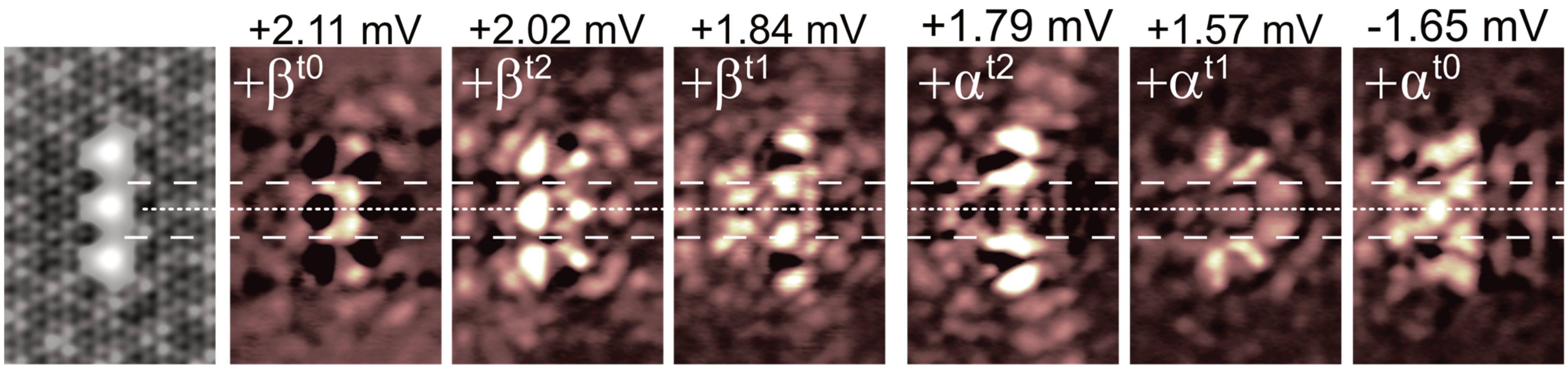
Fe dimer:

- α-resonance undergoes QPT to unscreened state due to RKKY

→ Coupling of two S=1 atoms

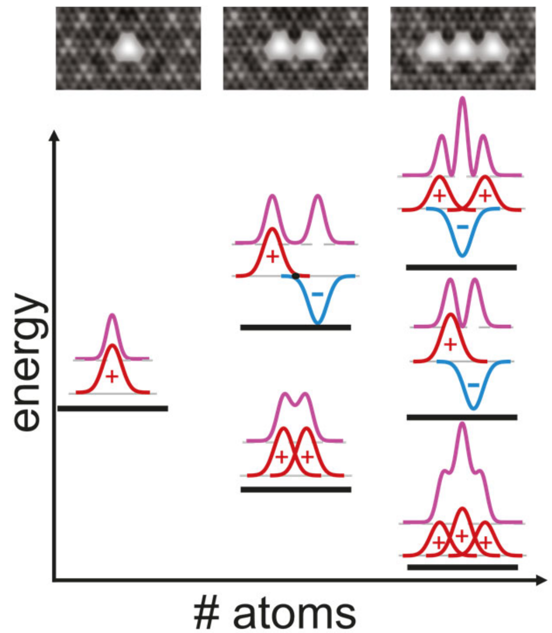
→ S=2 dimer

Fe trimer

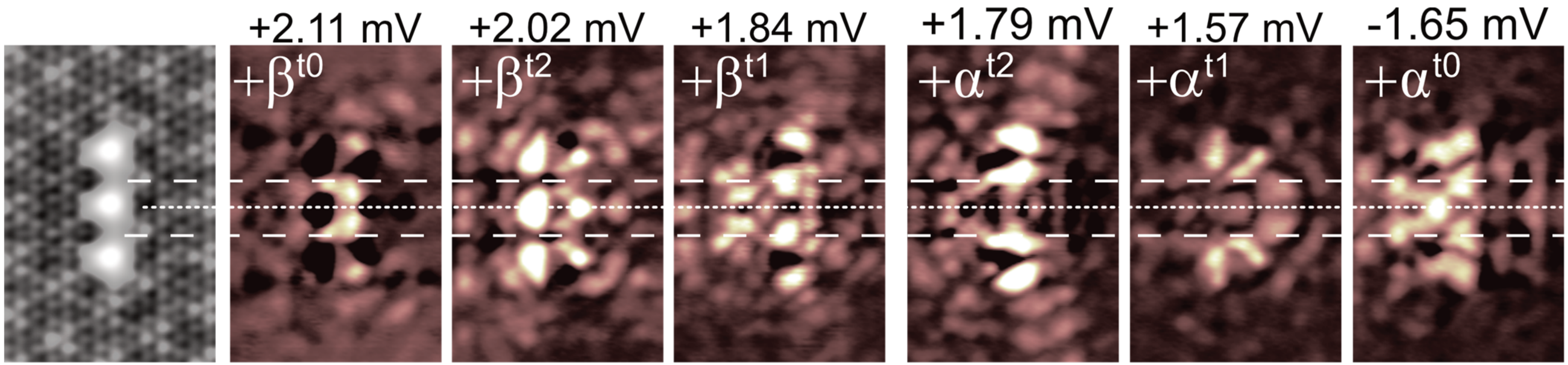


Fe trimer:

- Each YSR resonance splits into three

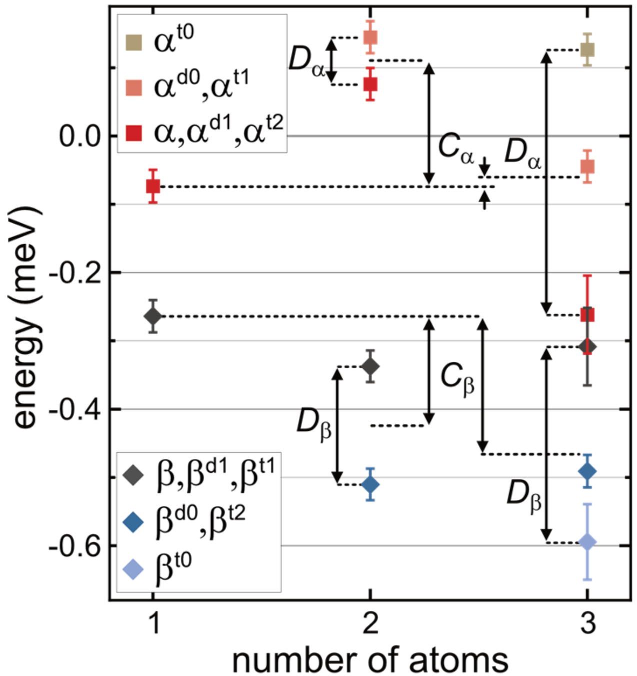


Fe trimer



Fe trimer:

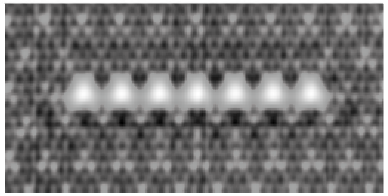
- Each YSR resonance splits into three
- Hybrid α states overlap with Fermi level



Outline

► Hybridisation of YSR states (on Pb)

► Interaction of magnetic atoms on $NbSe_2$

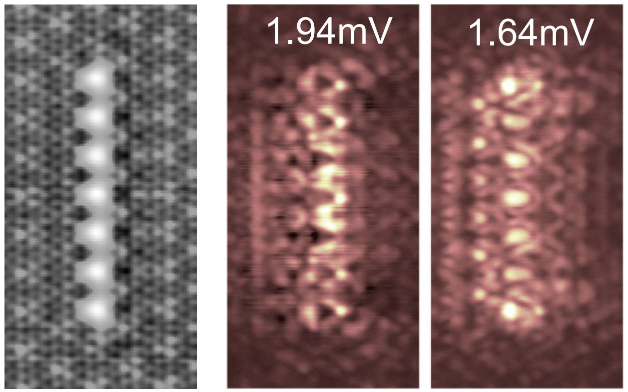


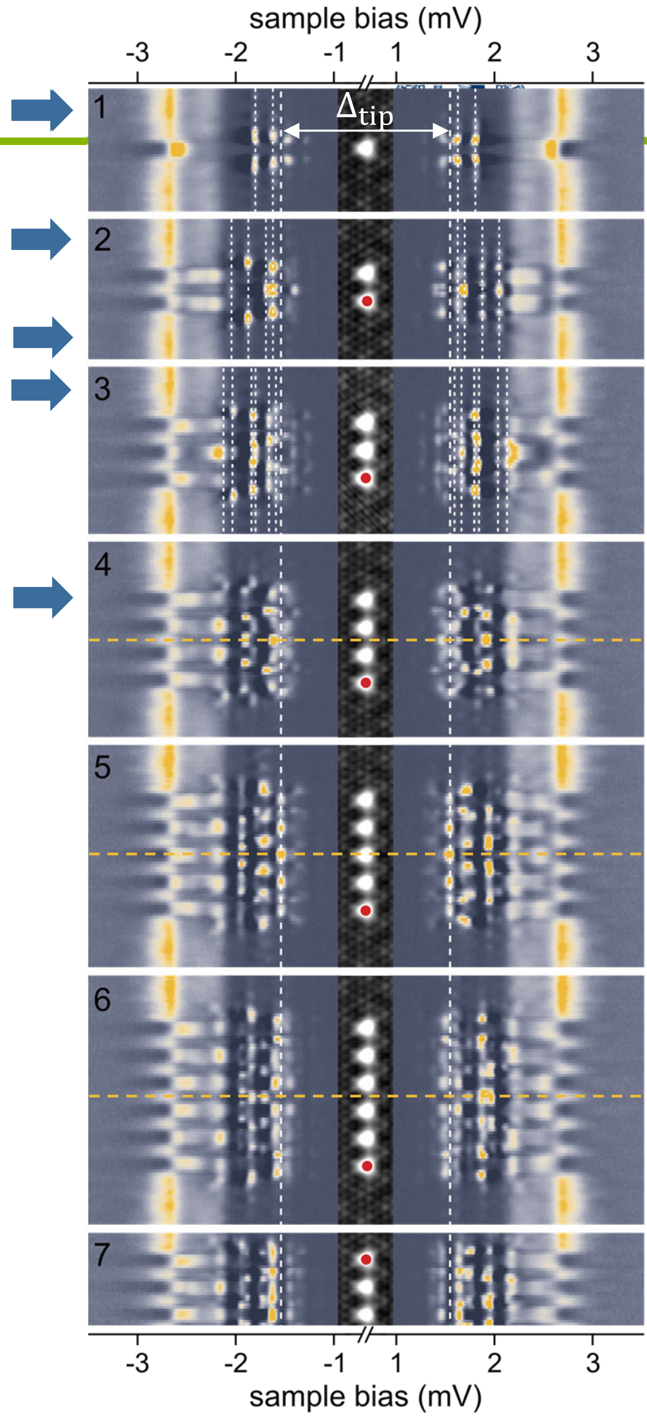
► Evolution of YSR states in atomic chains on $NbSe_2$

► Influence of charge-density wave on magnetic chains

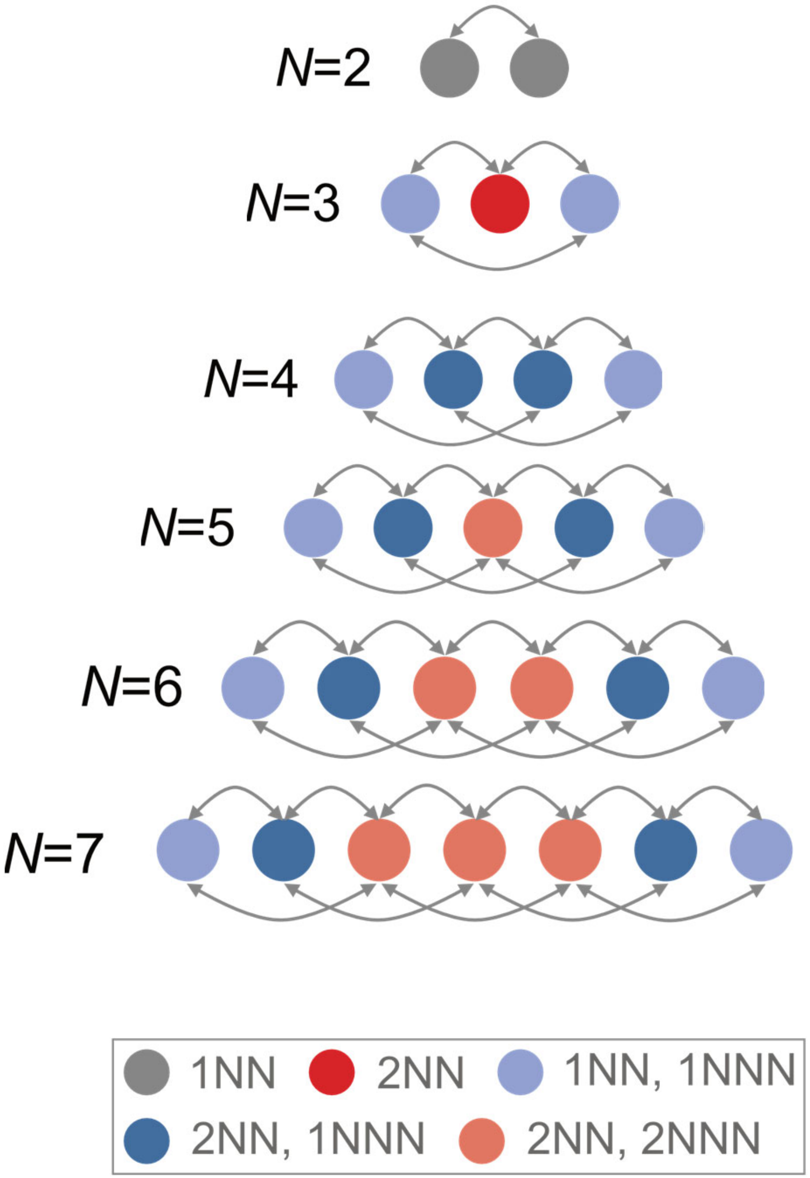
Formation of YSR bands

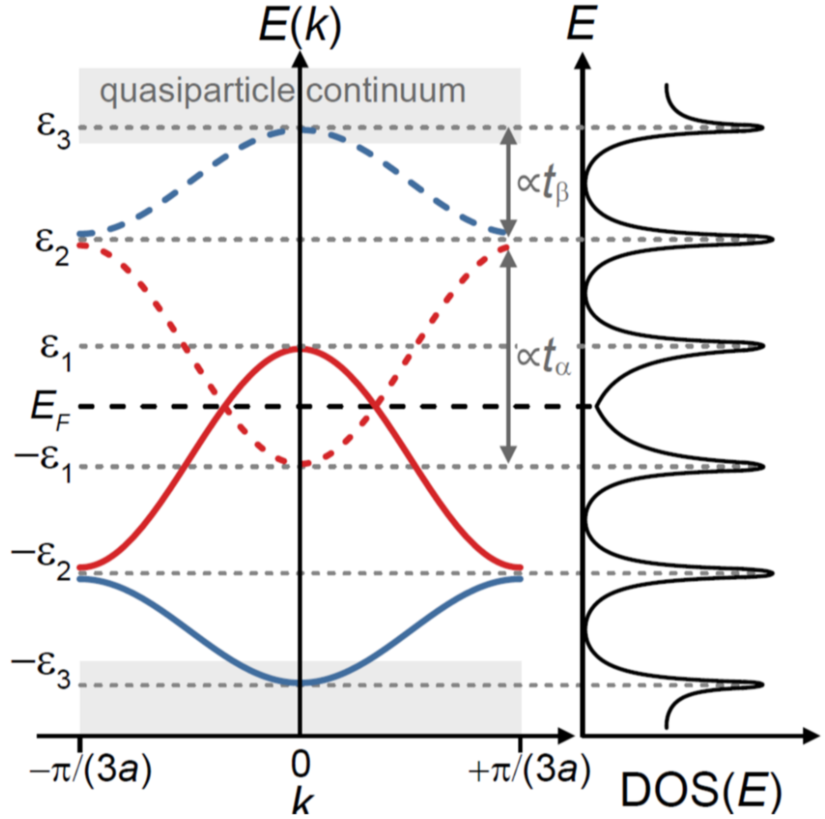
- all atoms on CDW maximum
- increasing number of hybrid states
- horizontal mirror symmetry
- delocalized over whole chain
- stabilization at specific energies





YSR bands





Fe chains:

- Formation of bands
- Van Hove singularities
- α band crossing Fermi level

Outline

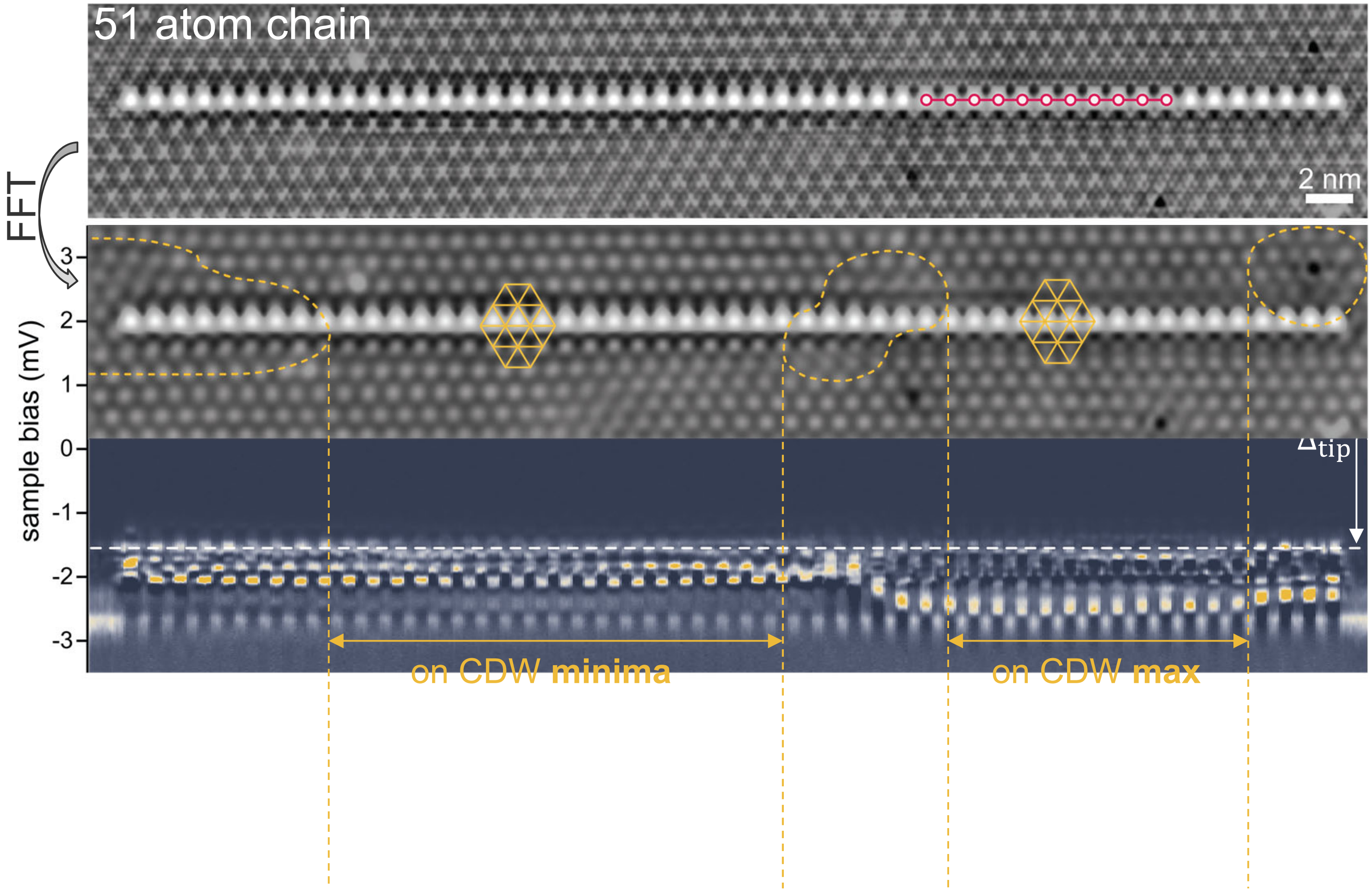
► Hybridisation of YSR states (on Pb)

► Interaction of magnetic atoms on $NbSe_2$

► Evolution of YSR states in atomic chains on $NbSe_2$

► Influence of charge-density wave on magnetic chains

Structures ≫ a_{CDW}



- rich energy variations along chain
- CDW smoothly transforms in background

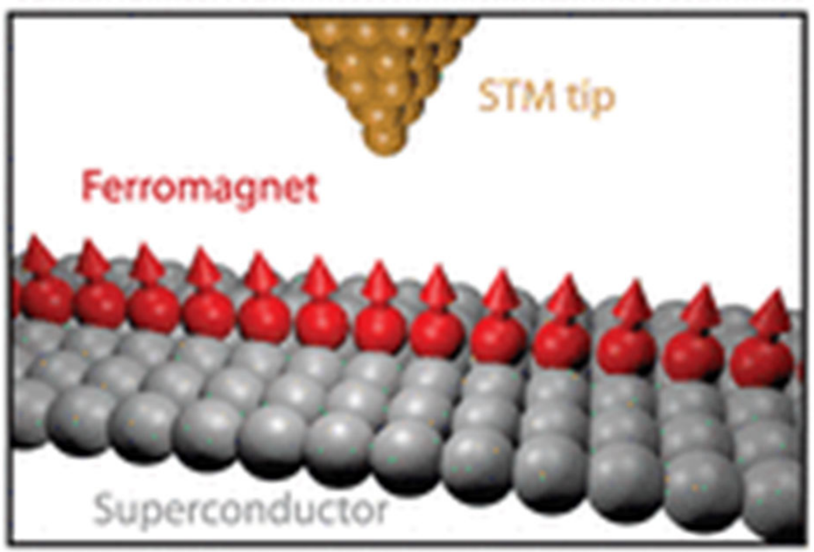
► band bending along chain due to incommensurate CDW

► two independent subchains

Magnetic chains on superconductors

Versatile platform for:

- ▶ Topological superconductivity
- ▶ Majorana states

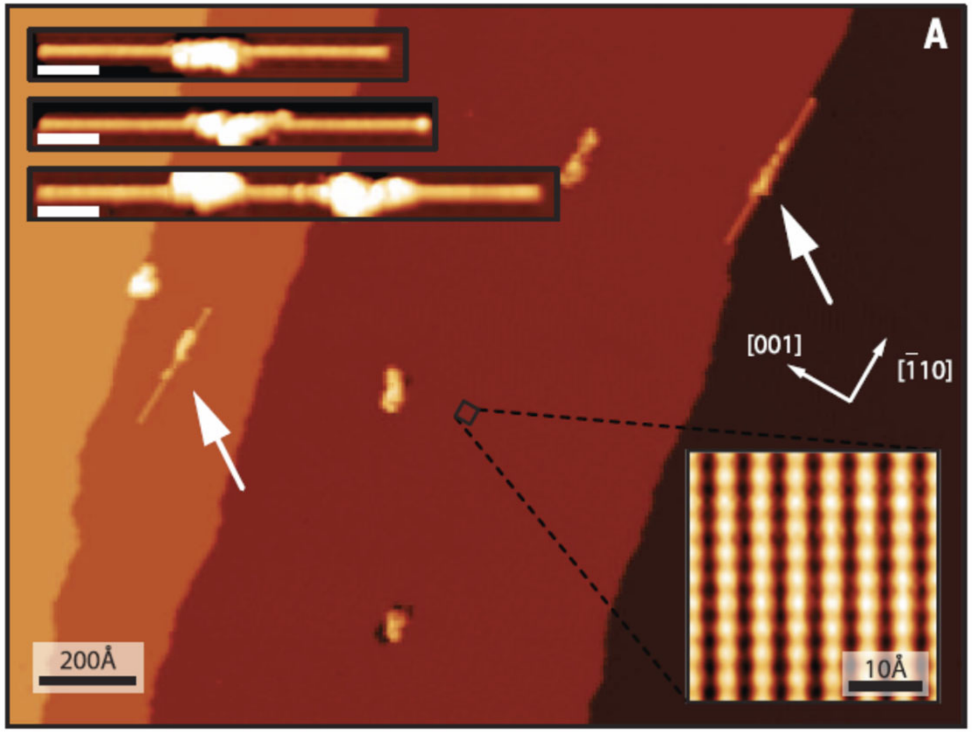


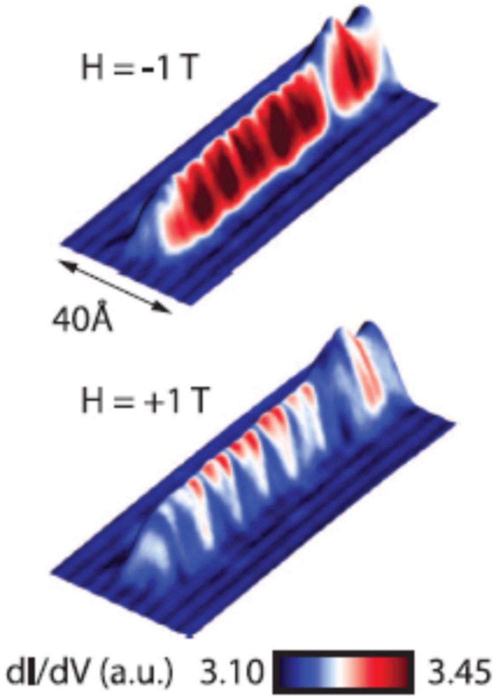
Nadj-Perge *et al.*, Science 346, 6209 (2014)

Magnetism of Fe chain on Pb(110)

Nadj-Perge *et al.*, Science 346, 6209 (2014):

Observation of Majorana fermions in ferromagnetic atomic chains on a superconductor

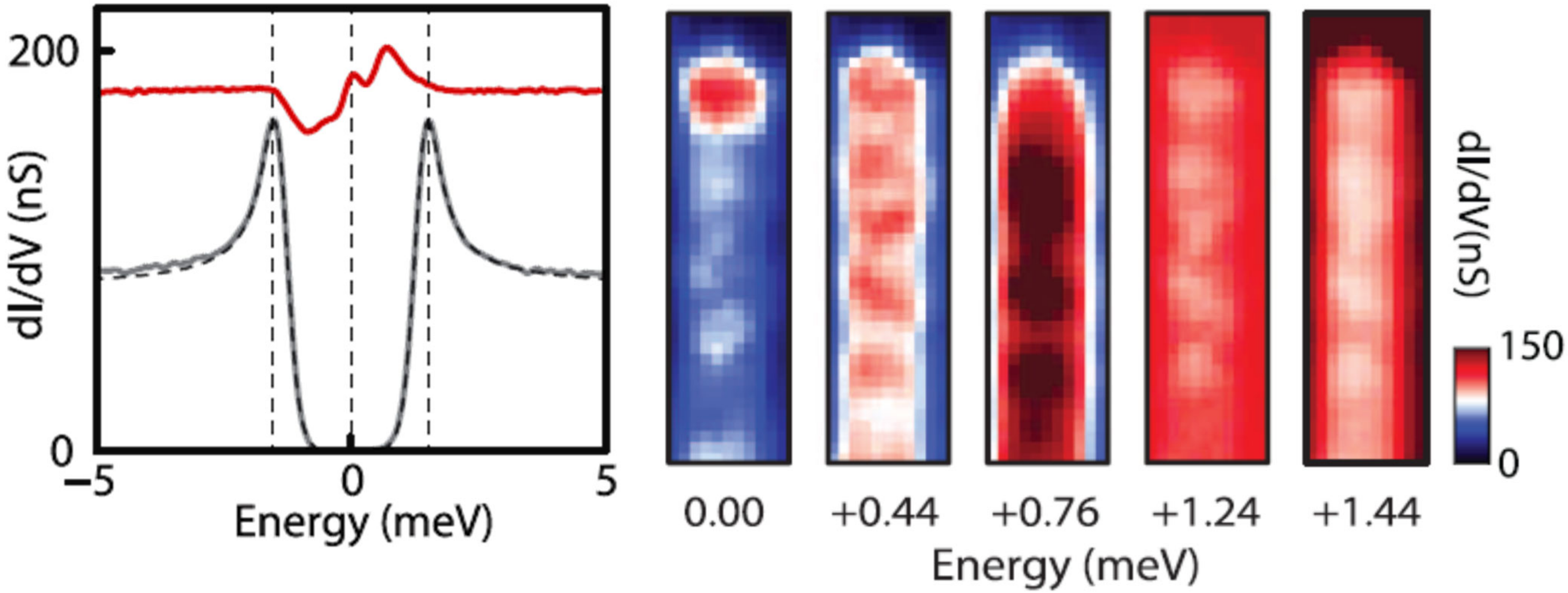




- Ferromagnetic spin structure along Fe chain

Sub-gap structure in Fe chains

Nadj-Perge *et al.,* Science 346, 6209 (2014):



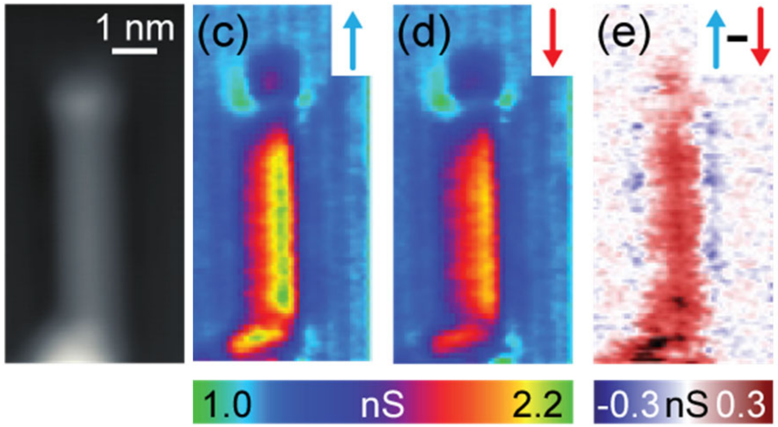
- Peak at zero bias
- Localized at chain end
- Interpretation as Majorana states
- Topological gap 200-300 µeV

See also:
Exp:
Ruby, et al., Phys. Rev. Lett. 115, 197204 (2015)
Pawlak, et al., npj Quantum Information 2, 16035 (2016)
Kim, et al., Sciences Adv. 4, eaar5251 (2018)
...

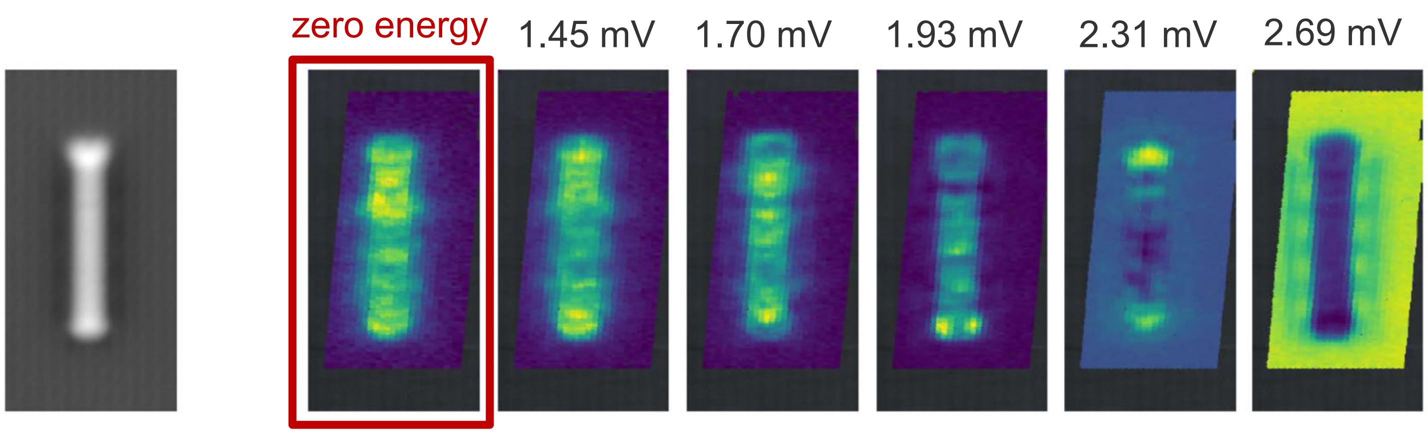
See also:
Theory:
Nadj Perge, et al., Phys. Rev. B 88, 020407 (2013)
Pientka, et al., Phys. Rev. B 88, 155420 (2013)
Li, et al., Phys. Rev. B 90, 235422 (2014)
...

Co chains on Pb(110): no Majorana modes

- Ferromagnetic Co chain



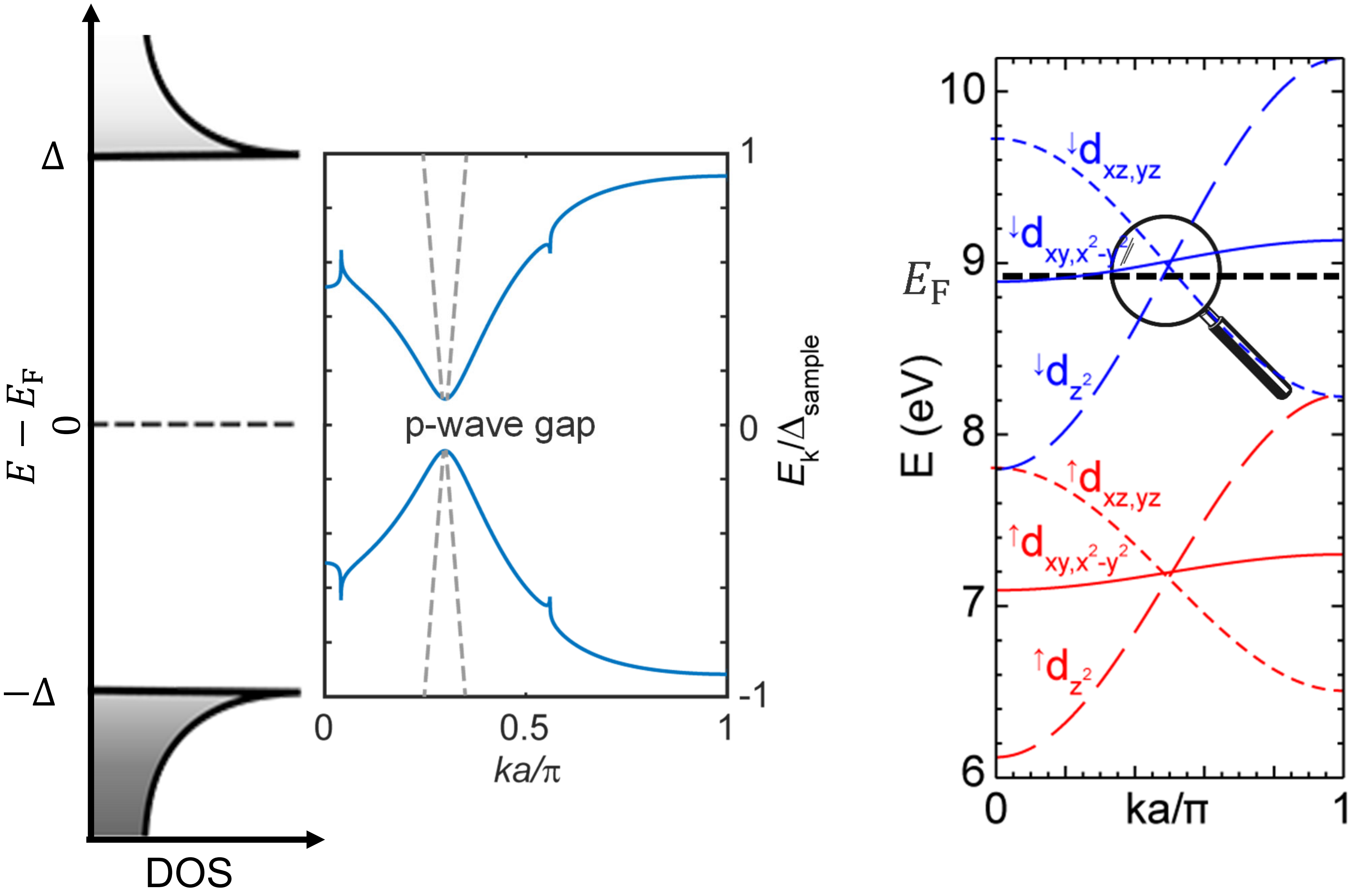
- Sub-gap signal



- No signature of Majorana modes

M. Ruby, et al., Nano Lett. 17, 4473 (2017)

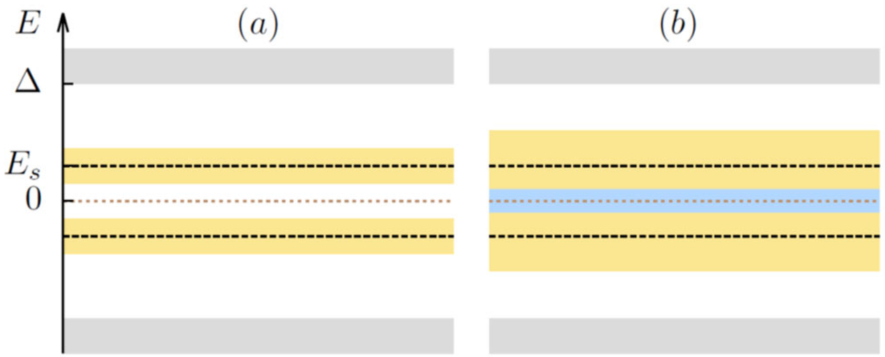
Transition metal chains on Pb(110)



Design of topological states

So far:

- Majorana states in ferromagnetic chains with direct exchange coupling

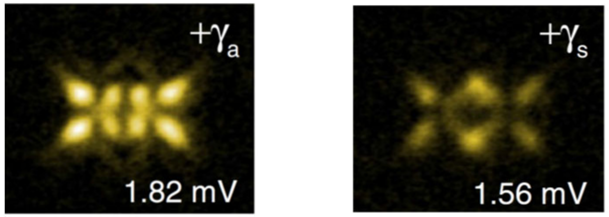


Pientka, et al., Phys. Rev. B 88, 155420 (2013)
Pientka, et al., Phys. Scr. 014008 (2015)

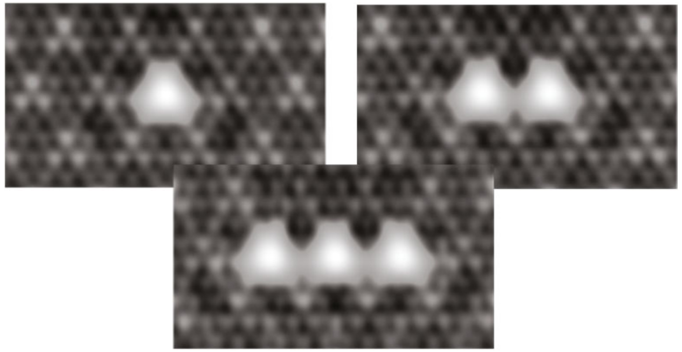
Design from YSR states:

- Dilute magnetic adatoms
- Coupling of YSR states
- YSR bands crossing Fermi level
  - possibly topological gap opening, but not detected for Fe on $NbSe_2$

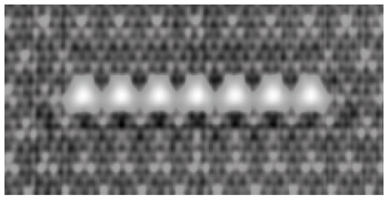
Conclusions



- Hybridisation of YSR states:
  - Symmetric and anti-symmetric linear combination



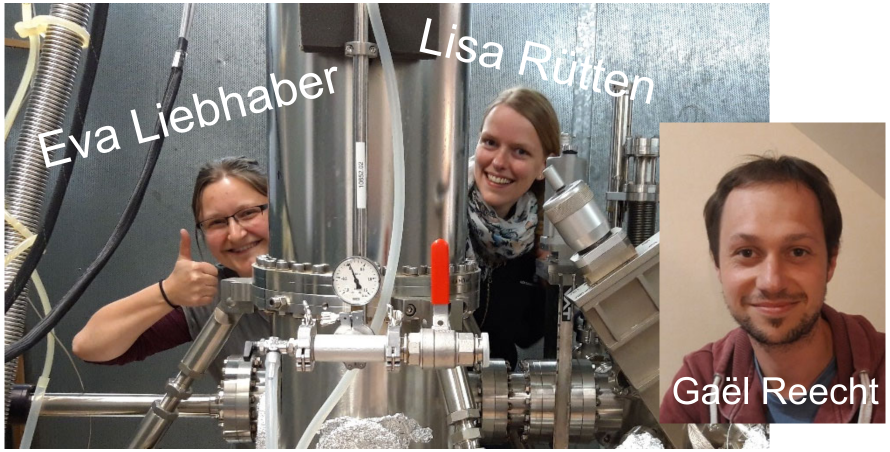
- RKKY interactions and hybridisation
  - Inducing quantum phase transition
  - Quantum spin description of interactions

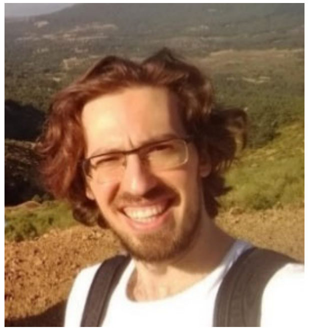


- Formation of YSR bands

- Band bending and different domains due to charge-density wave

Thanks!





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Nils Bogdanoff, Giada Franceschi, Jennifer Hartfiel, Eva Liebhaber, Christian Lotze, Bharti Mahendru, Gaël Reecht, Lisa Rütten, Rika Simon, Idan Tamir, Martina Trahms, Sergey Trishin, Ali Yazdani, Asieh Yousofnejad

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