Ultrafast filling of an electronic pseudogap in $[LaS]_{1.2}VS_2$

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FemtoARPES

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Optical measurements

Outline

$[LaS]_{1.2}VS_2$: a « strange » insulator

⇒ Resistive switching under applied electric pulses

ARPES investigation of the electronic structure

⇒ A very strong temperature dependence of the Fermi level « pseudogap »

Manipulating the pseudogap by pump-probe experiments

⇒ Ultrafast filling of the pseudogap
⇒ Evidence for very strong electron-phonon coupling

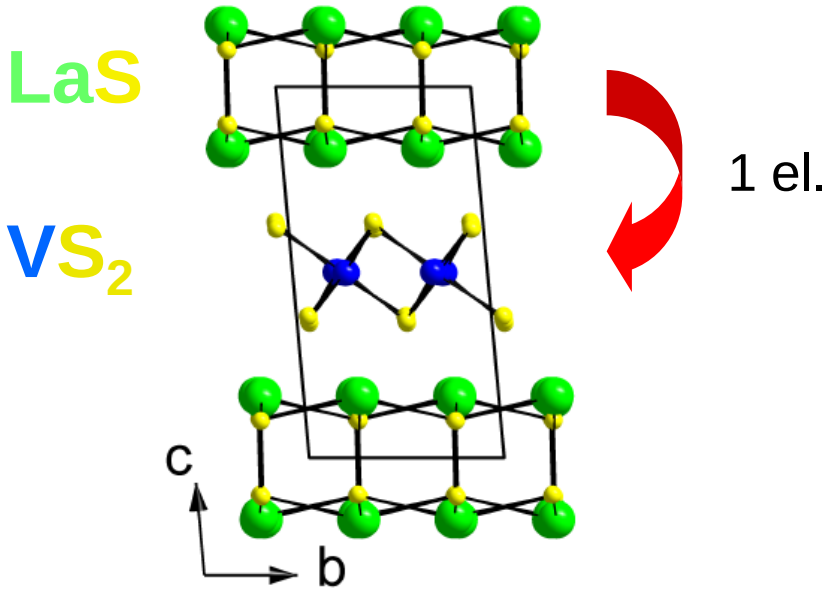
$[LaS]_{1.2}VS_2$ appears to be at the frontier between cluster and solid behaviors

$[LaS]_{1.2}VS_2$: a « misfit » structure

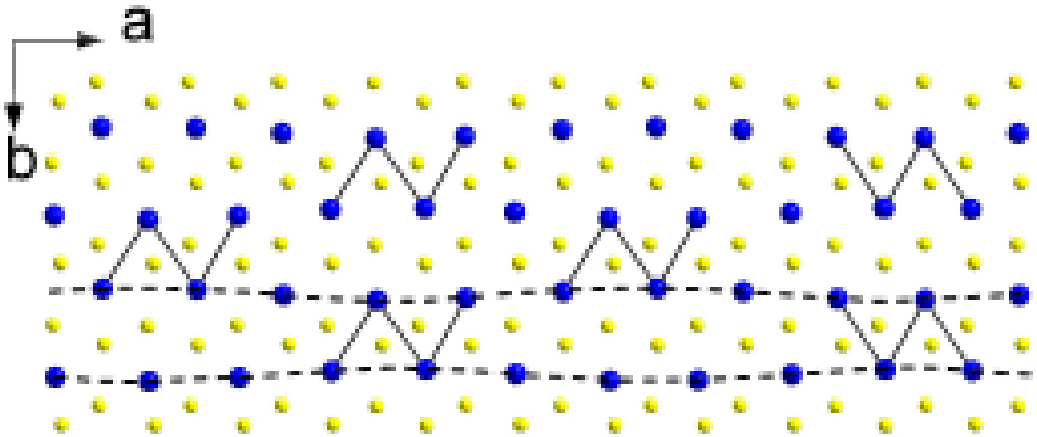
LaS and VS_2 are incommensurate along a.

=> approximate supercell :

$3*a_{LaS} = 5*a_{VS2} = 17Å$



Distorted triangular V planes



Modulation of V position with :

$$q = \frac{a_{VS2}}{a_{LaS}} a_{VS2}^{*} = 1{,}1Å^{-1}$$

=> V-V (min) = 2,92Å
V-V (max) = 3,73Å

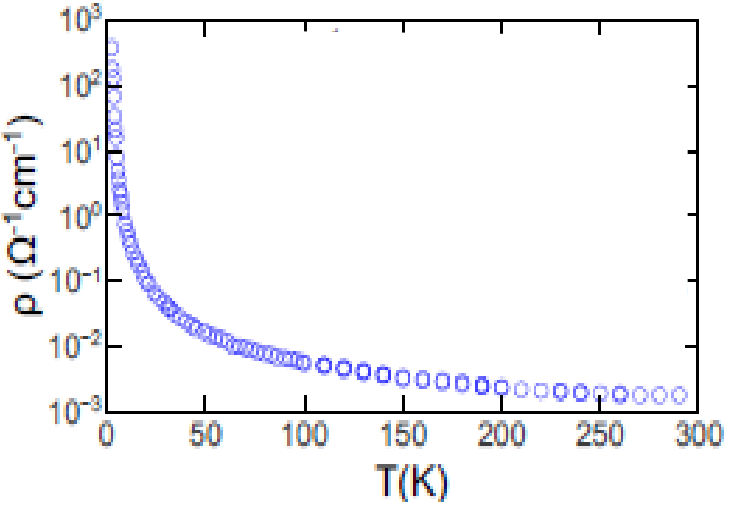
L. Cario et al., Mat Res Bul 2005

A « strange » insulator ?

- 2.2 electrons in the V 3d t_{2g} orbitals

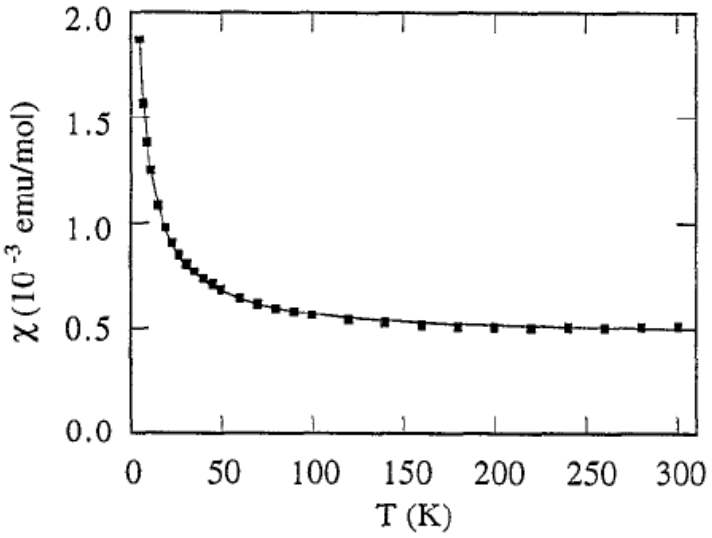
Resistivity

~ semiconducting-like ?



Susceptibility

~ Pauli-like ?



L. Cario et al., Synthetic Metals 1999

A « strange » insulator ?

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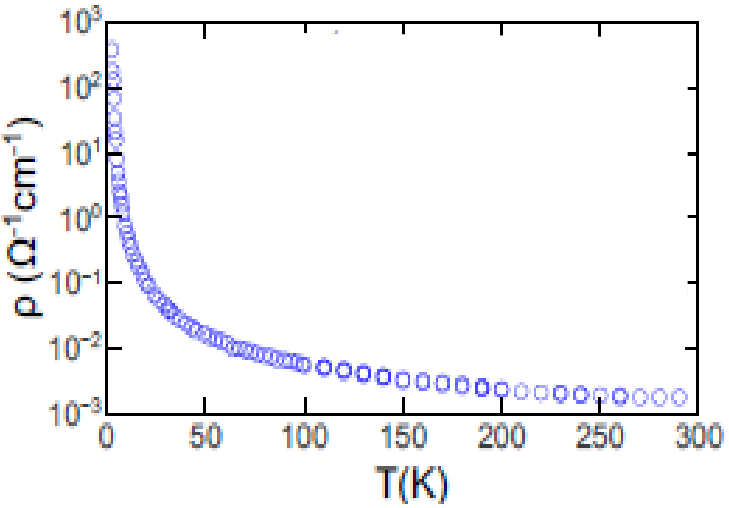
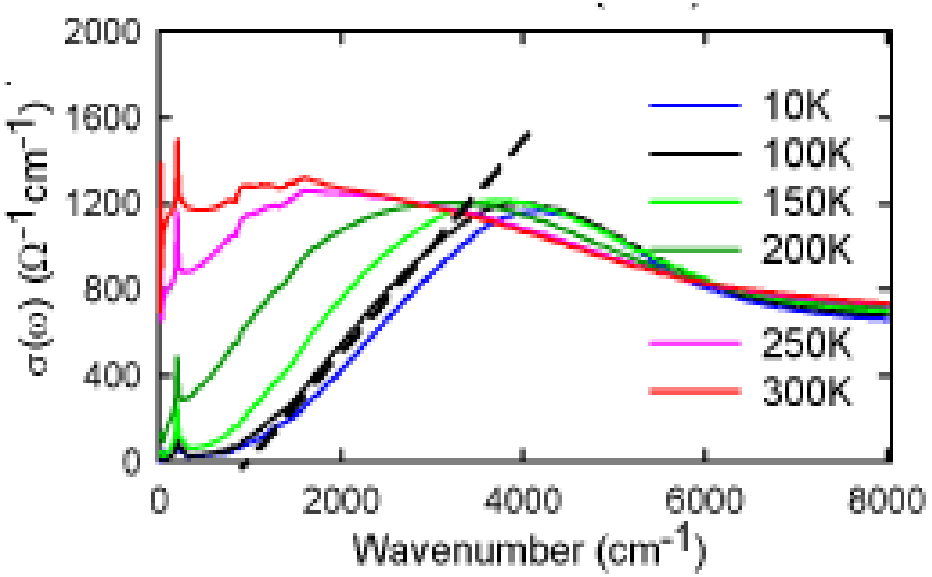


Optical conductivity

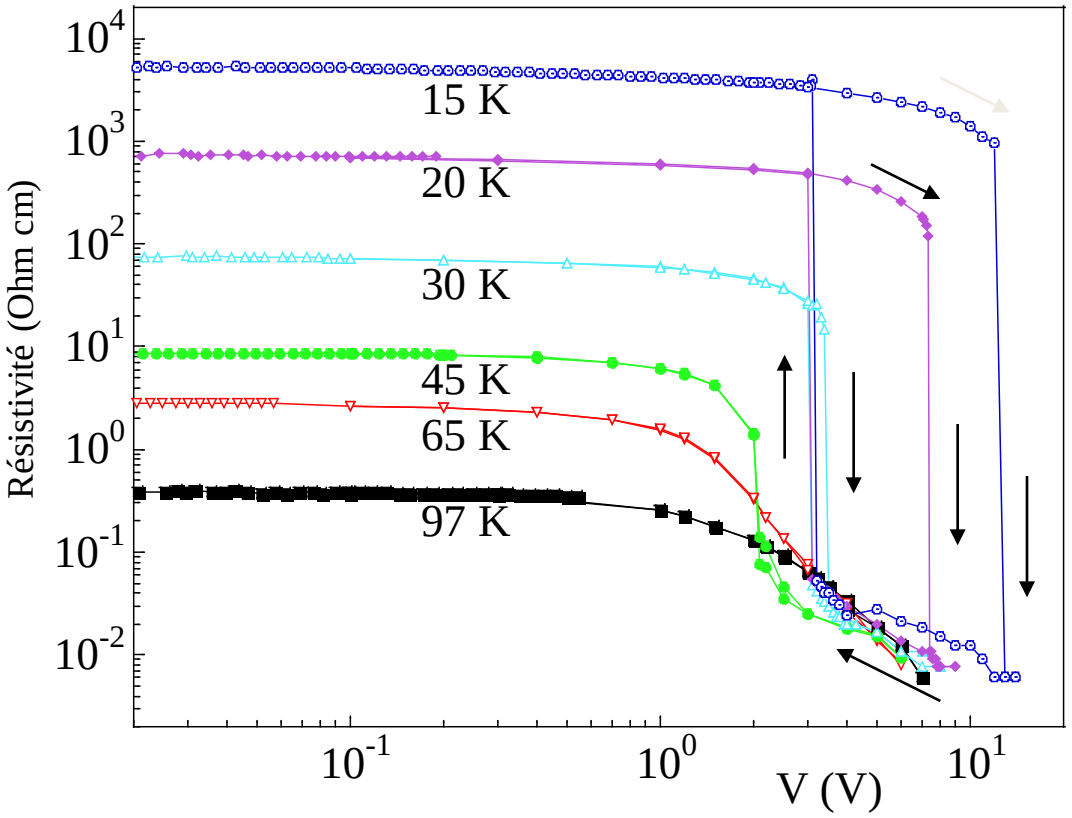
Unexpectedly strong T dependence !



V. Ta Phuoc et al., submitted

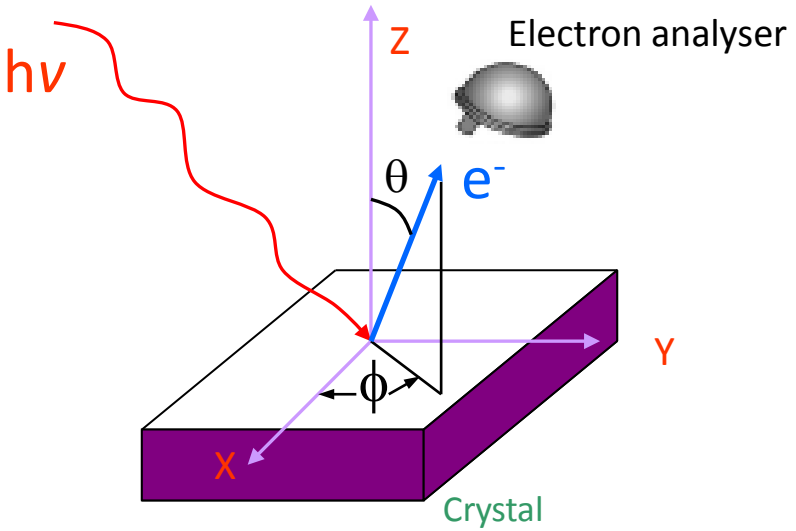
Resistive switching under applied electric pulses

Resistivity can change up by 5 order of magnitudes !



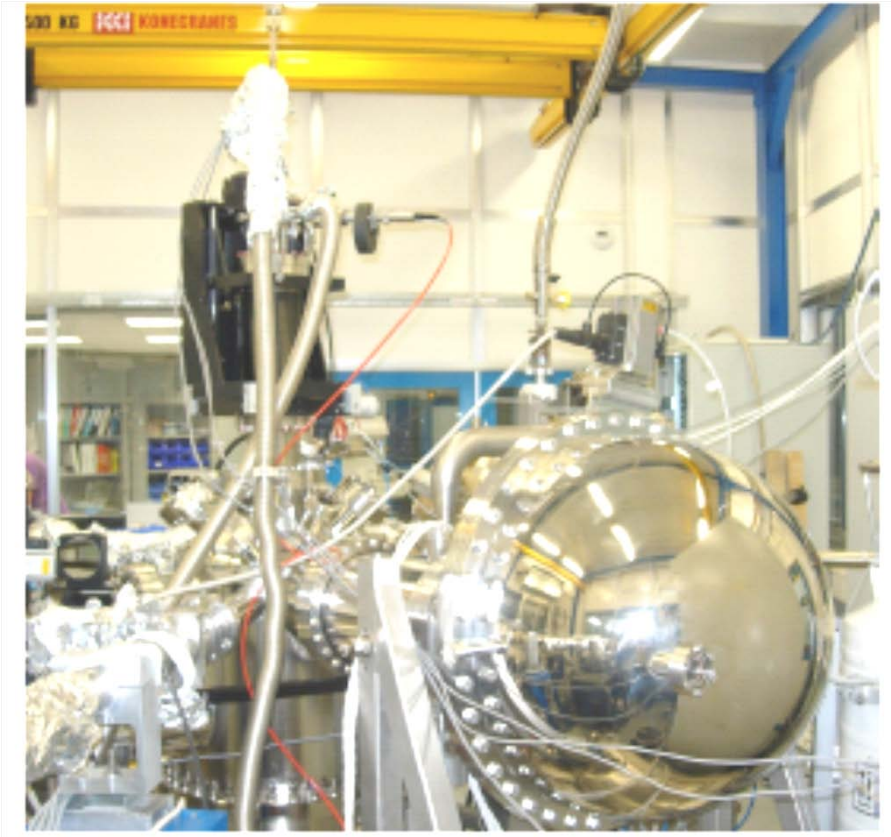
L. Cario et al., Phys. Rev. B 2006

ARPES view of the electronic structure



$$E_{kin} = h\nu - W - \left|E_B\right|$$

$$\hbar \mathbf{k}_{\parallel} = \sqrt{2mE_{kin}}\sin\theta$$

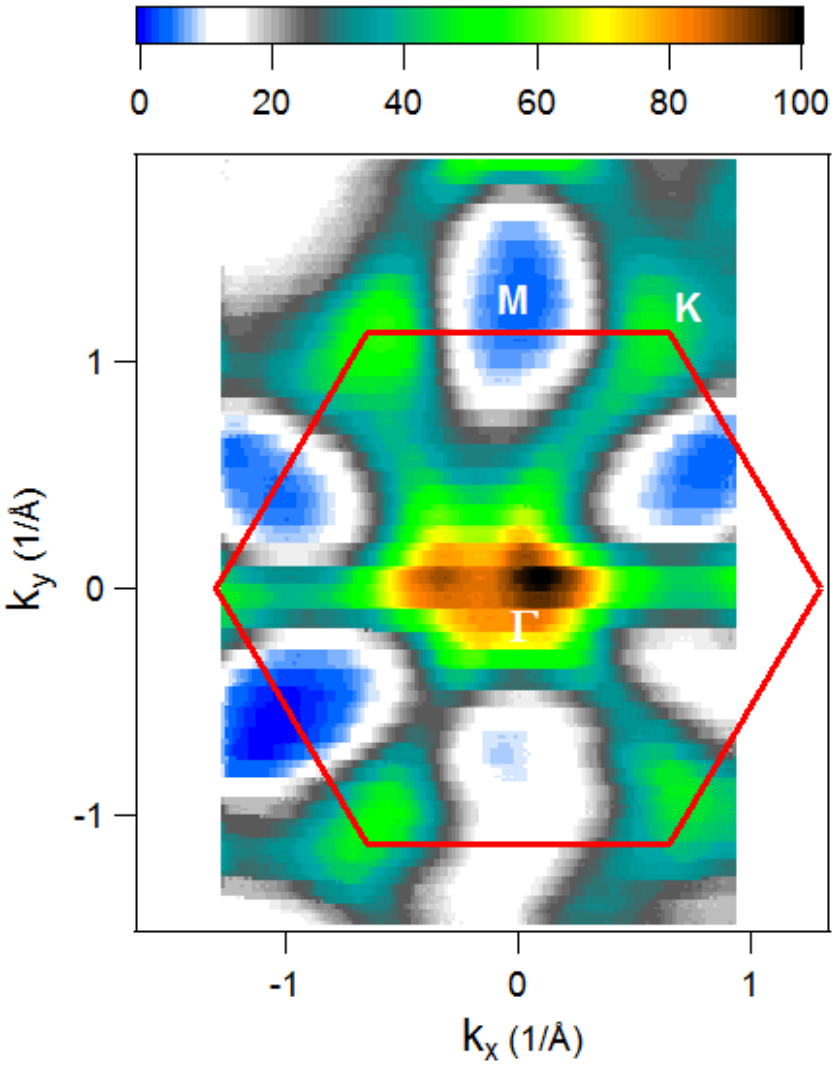


CASSIOPEE beamline, SOLEIL synchrotron

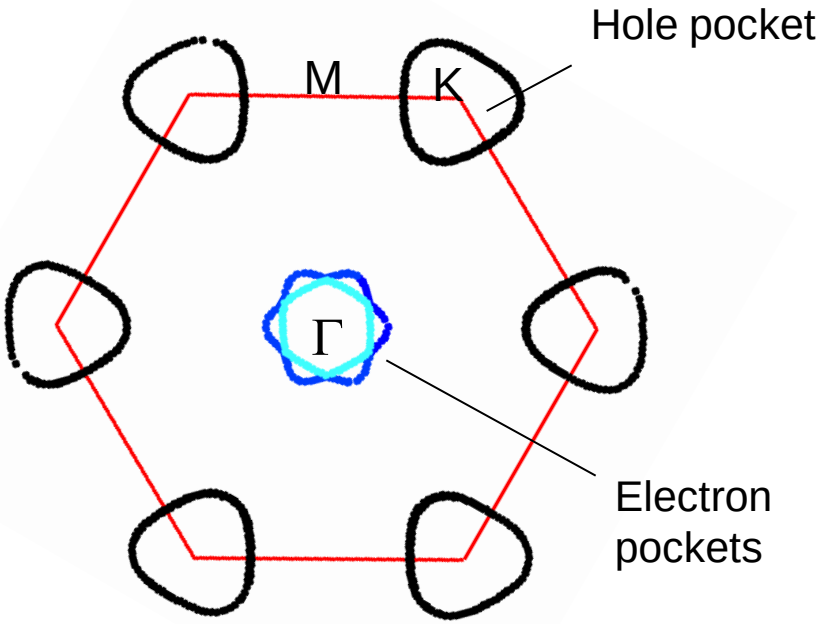
=> 10-1000eV

=> high energy and angular resolution

Fermi Surface (300K, 96eV)

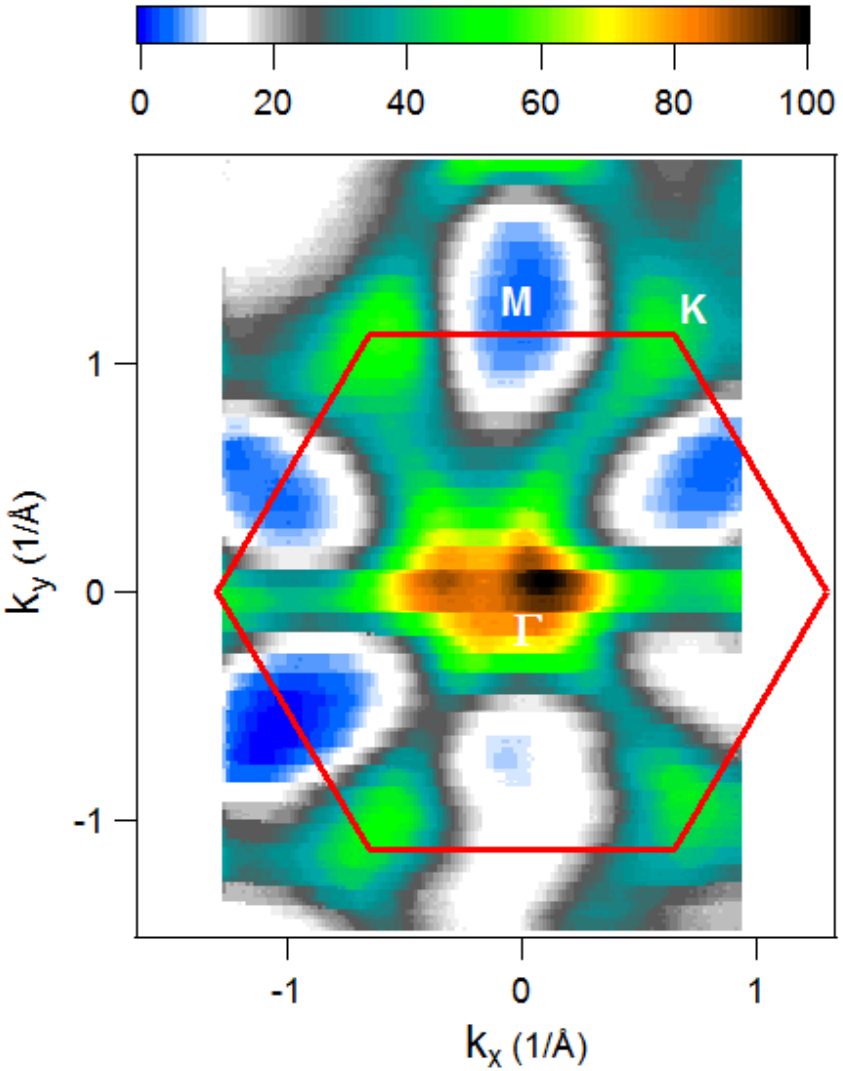


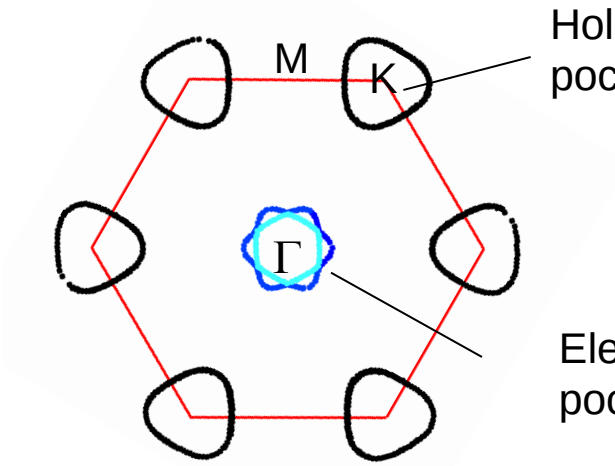
Calculated FS in $LiVS_2$ (d^2)



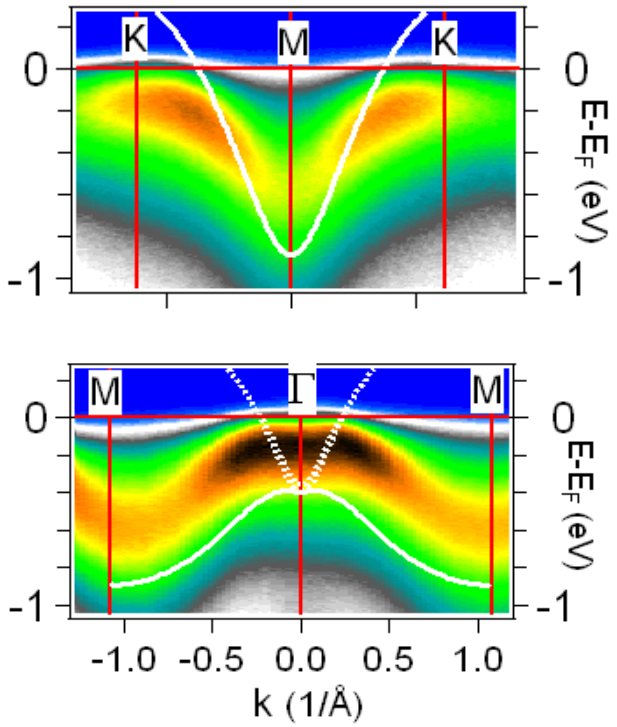
=> 6-fold symmetry expected for a triangular lattice

Fermi Surface & band structure

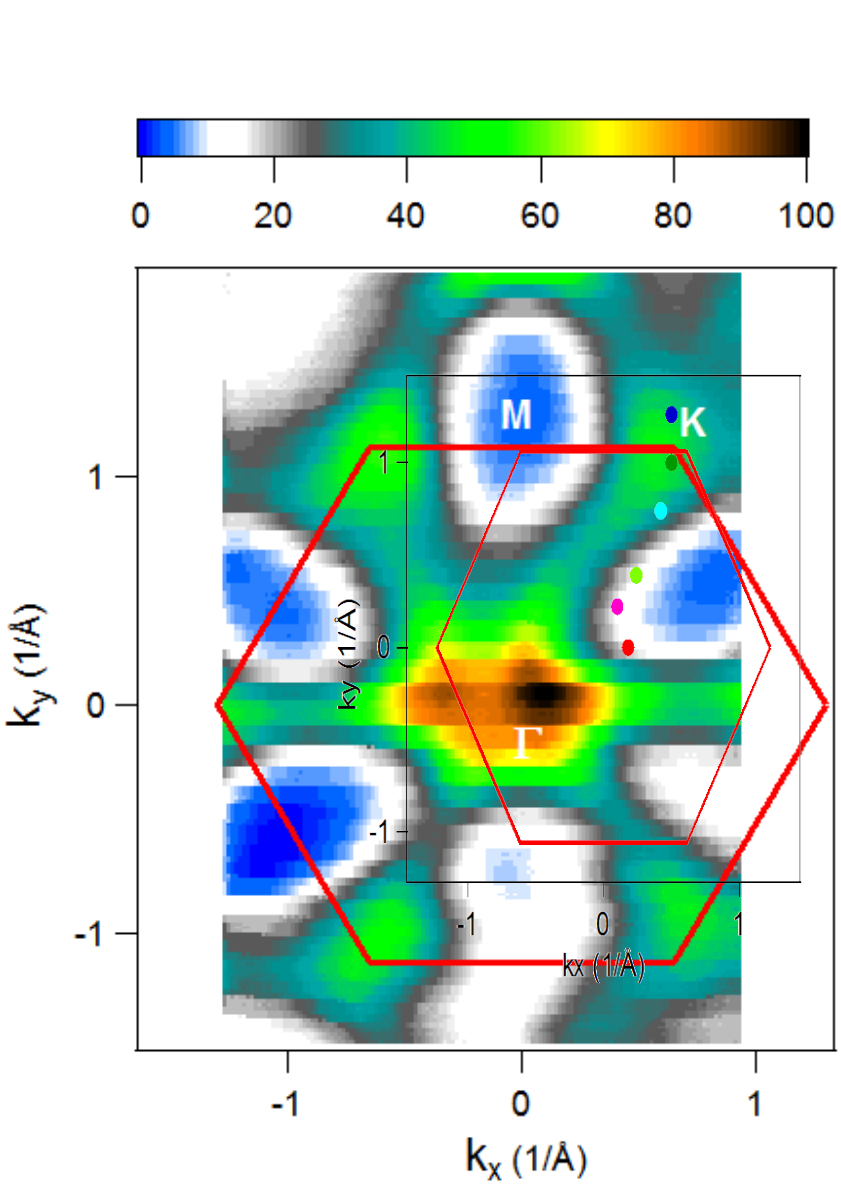


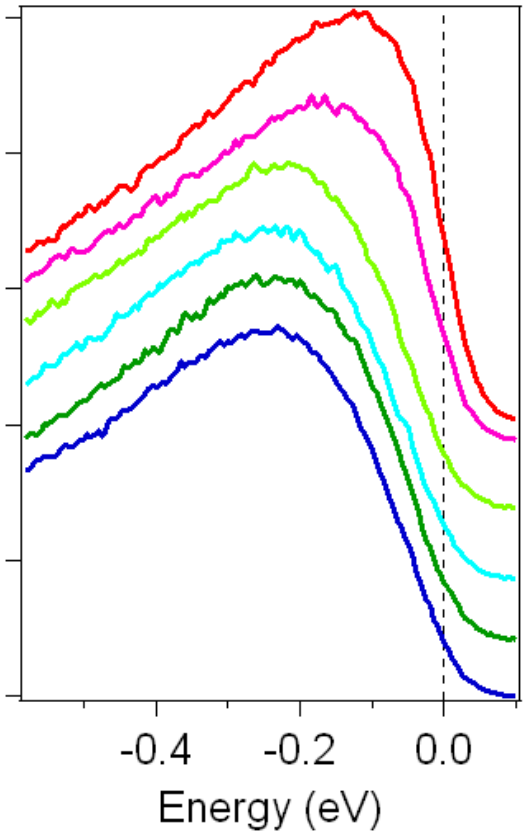


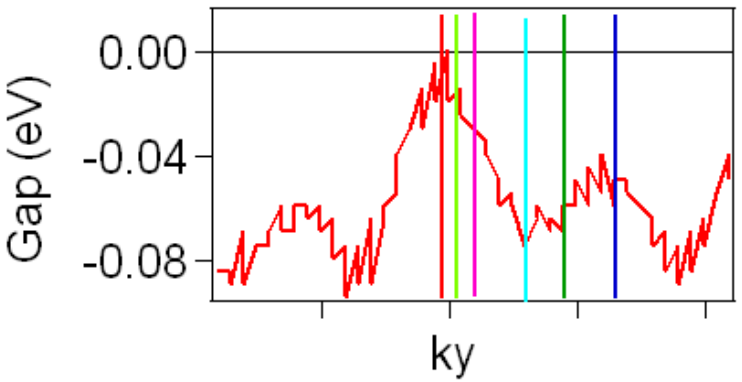
Calculation for $LiVS_2$



Pseudogaps along the FS (300K)

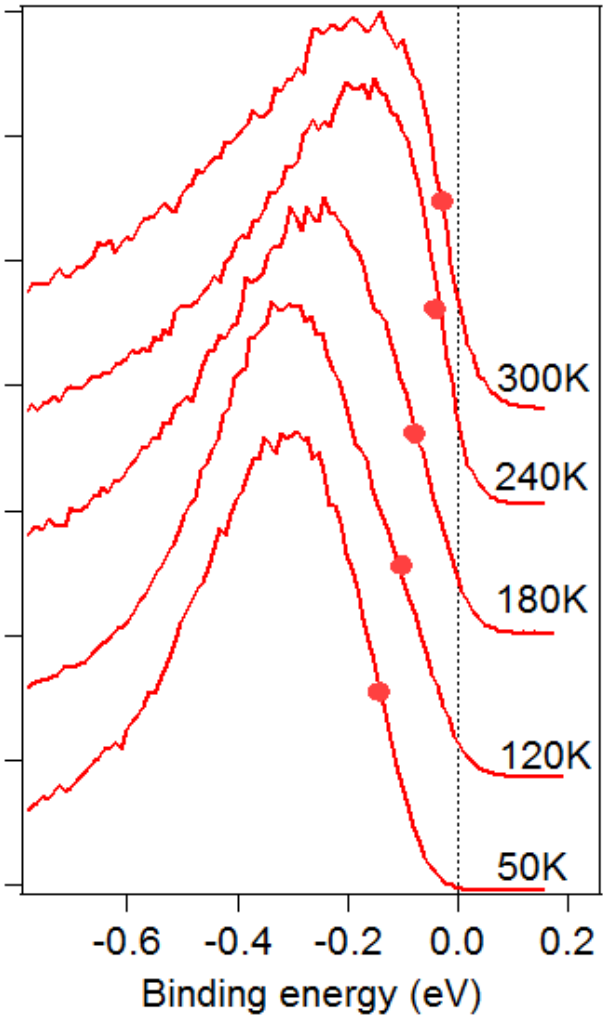






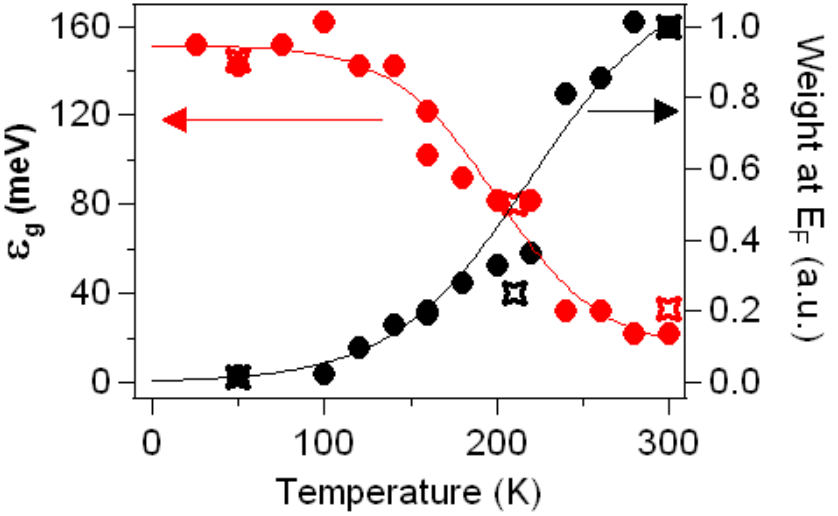
Evolution with temperature (at Γ)

ARPES spectra as a function of T

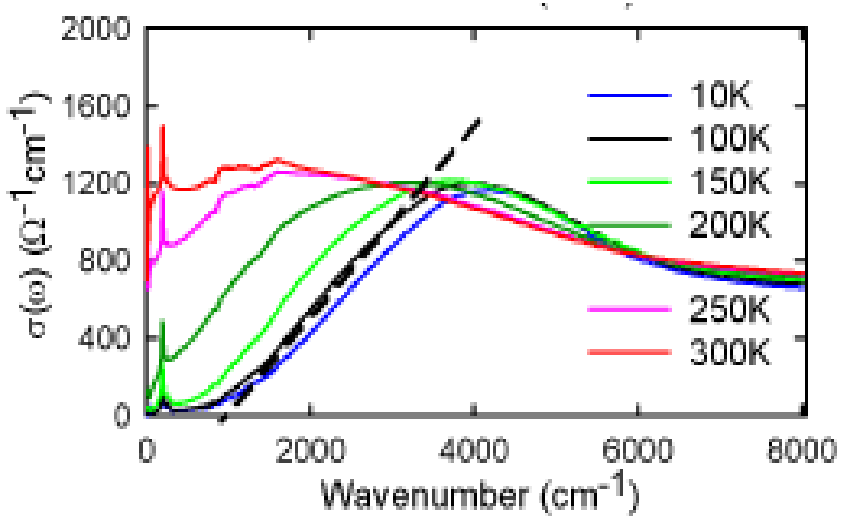


First angle integrated study :
Ino, Fujimori et al., PRB 2004

Gaps and spectral weight at E_F

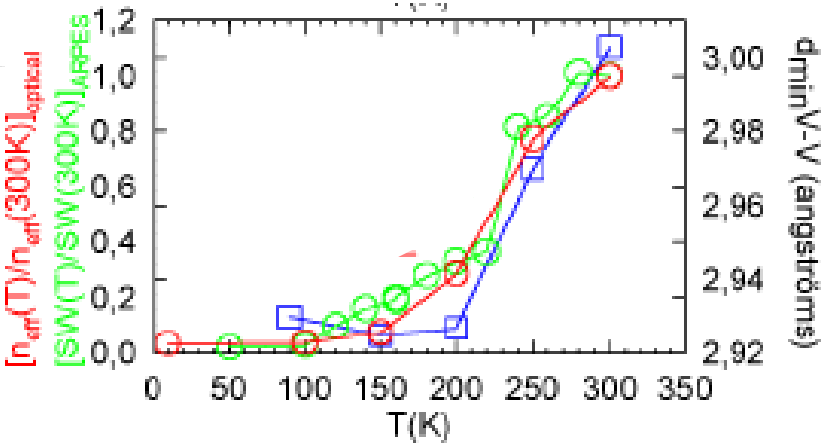


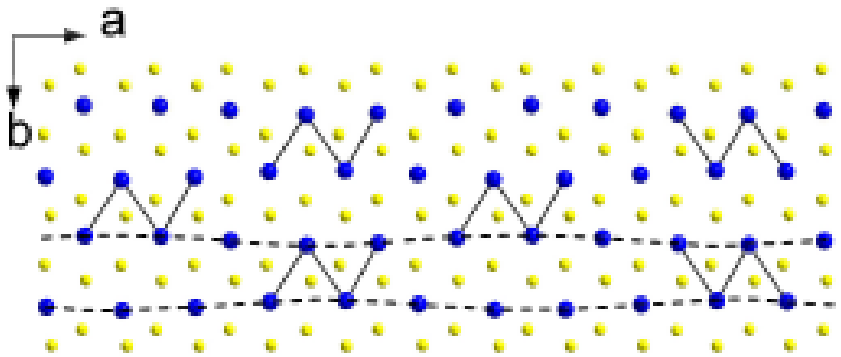
Optical conductivity



Direct connection between $n(E_F)$ and structure

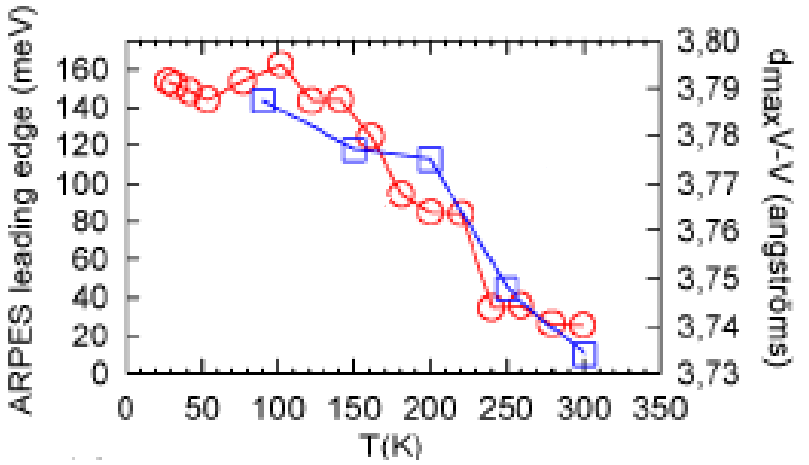
ARPES and optic spectral weight scale with the distorsion

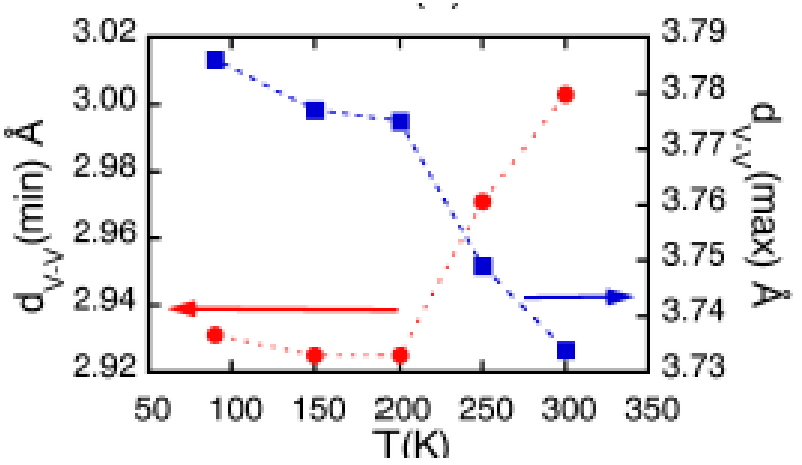




The modulation amplitude increases with decreasing T

ARPES pseudogap scales with the distorsion



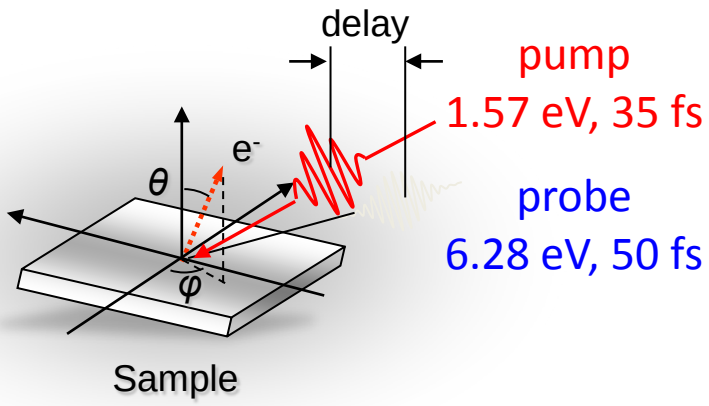


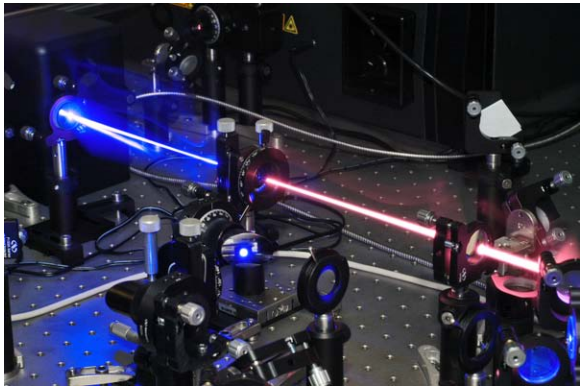
V. Ta Phuoc et al., submitted

Probing the dynamic of the pseudogap formation

How are structural and electronic degrees of freedom linked in this system ?

FemtoARPES setup

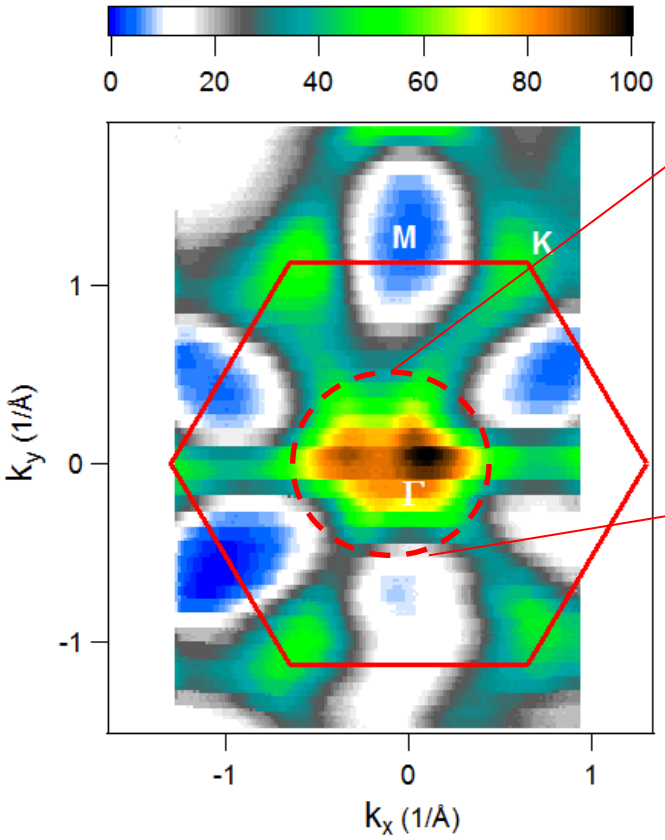




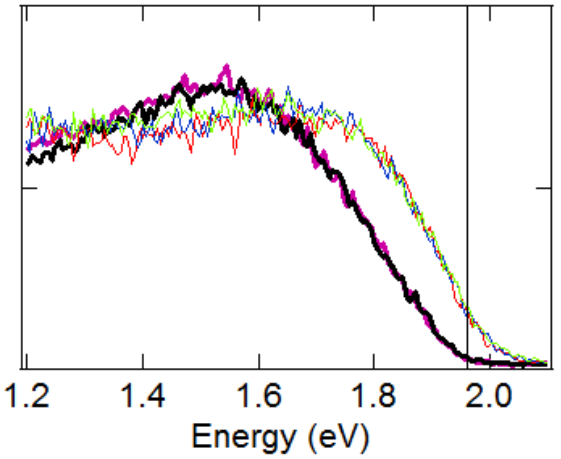
J. Faure *et al.*, *Rev. Sci. Intrum.*, 2012

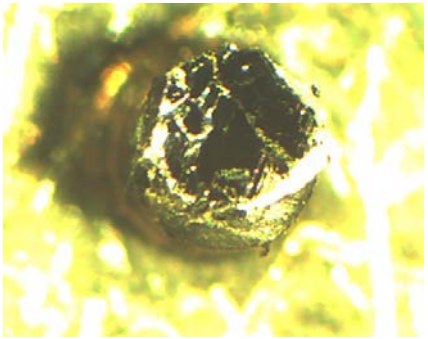
Laser ARPES of $[LaS]_{1.2}VS_2$ (6.28eV)

FS measured at 100eV

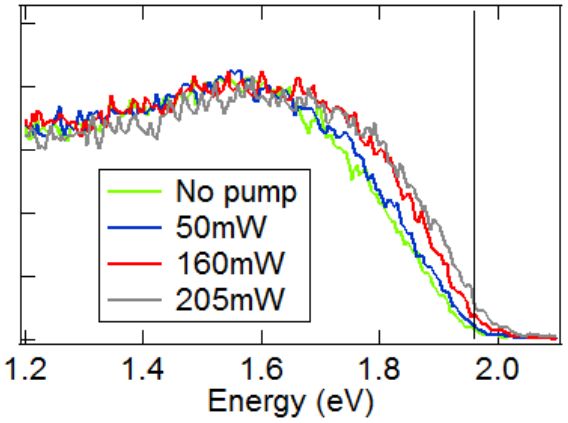


Opening of the pseudogap between 300 and 40 K observed at 6eV





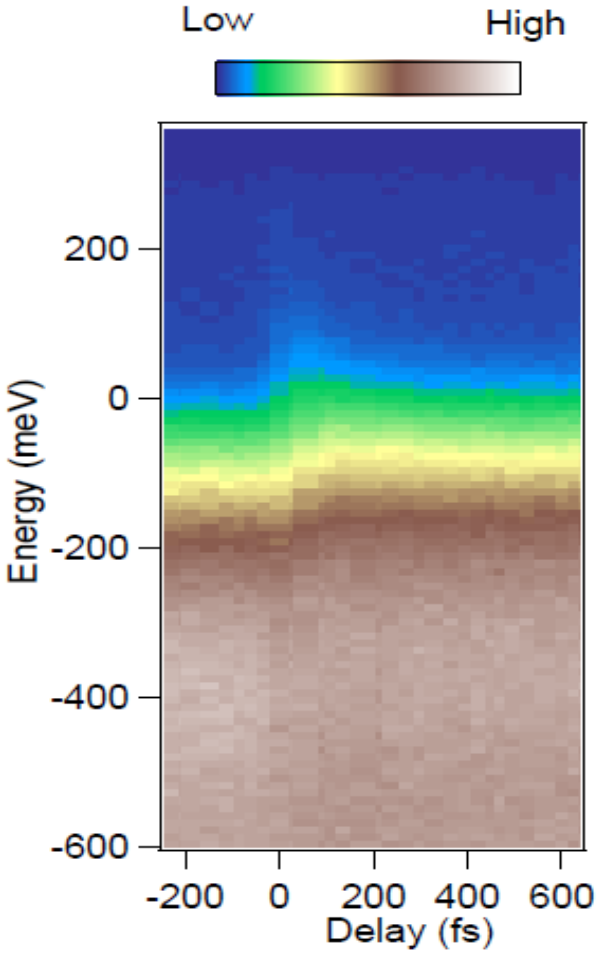
Heating due to laser pumping



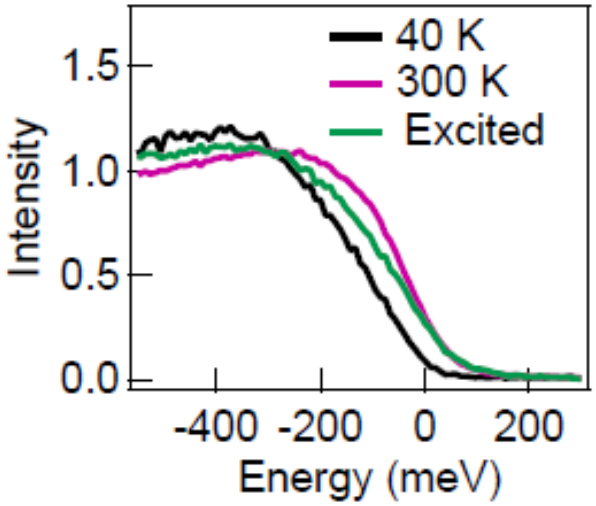
=> Cut down repetition rate

Ultrafast filling of the pseudogap

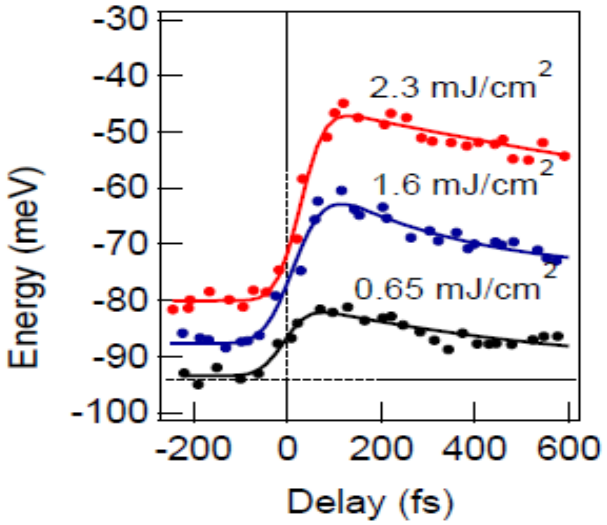
Sample at 40K excited by laser pulse of 2.3 mJ/cm^2 fluence



Excited spectra (green) compared to high and low T



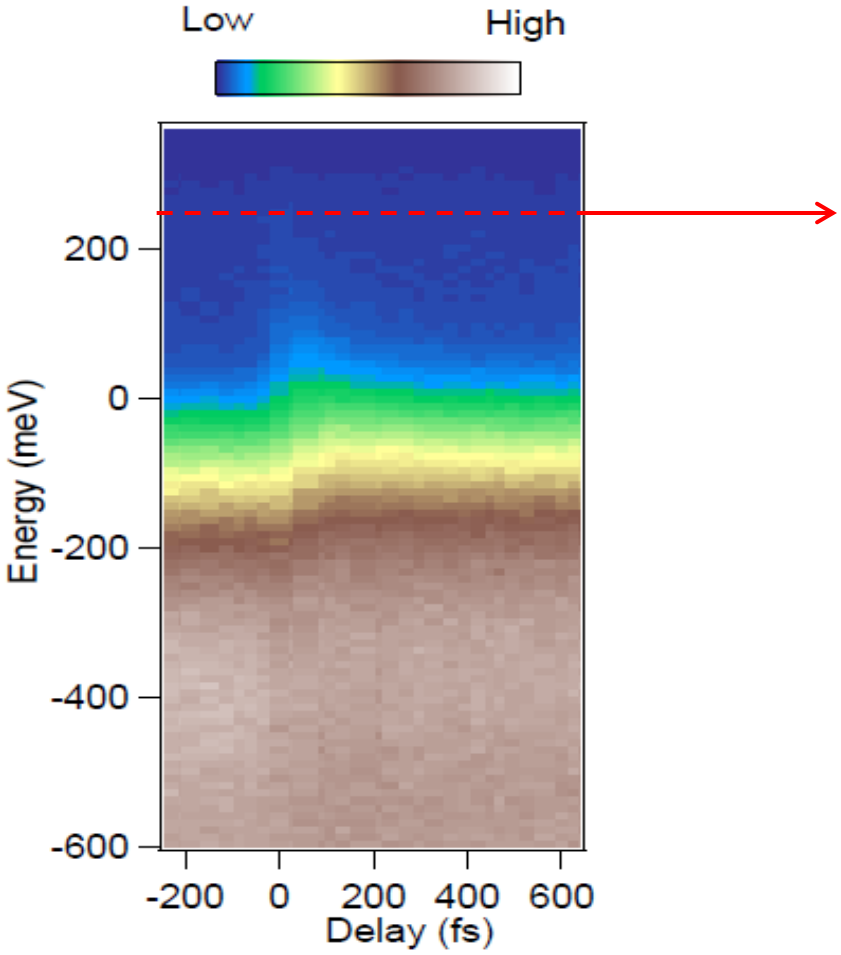
Leading edge vs delay



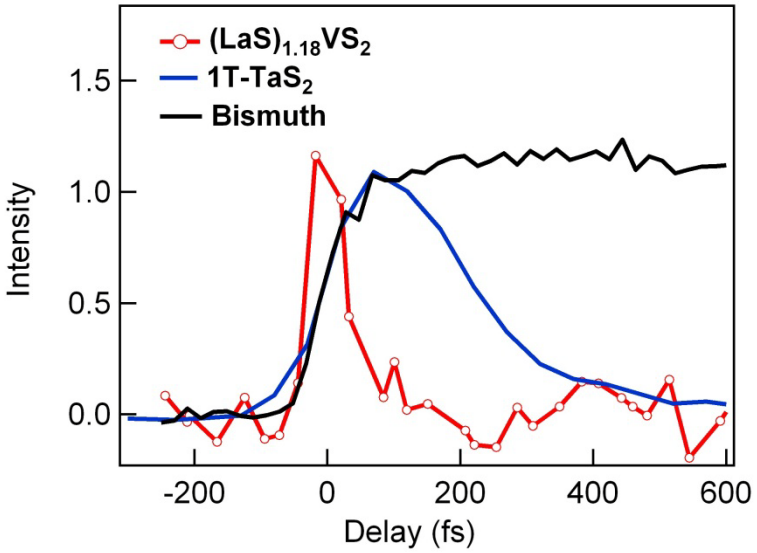
The pseudogap fills up in less than 80fs, close to the experiment temporal resolution

Ultrafast filling of the pseudogap

Sample at 40K excited by laser pulse of 2.3 mJ/cm^2 fluence



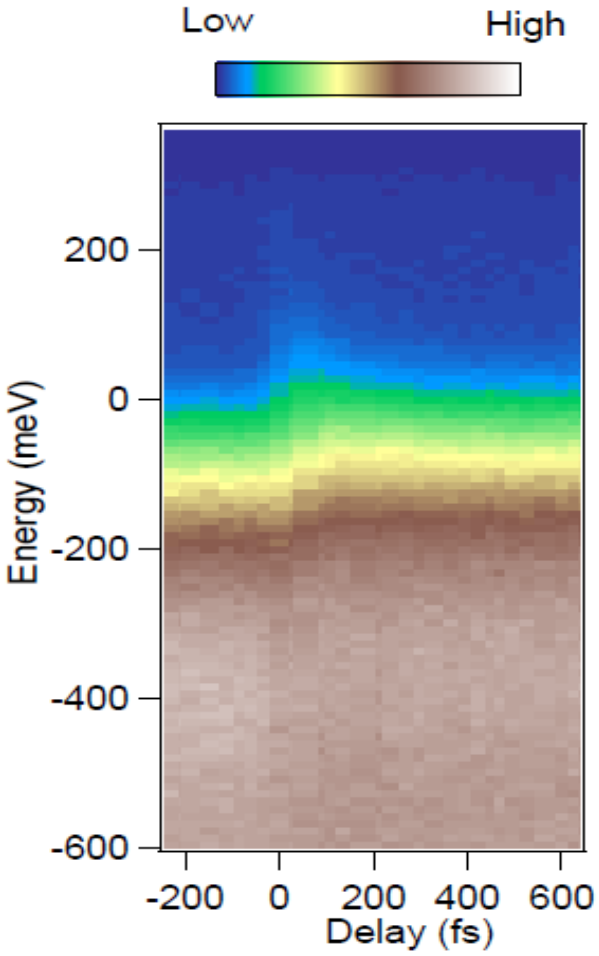
Comparaison of hot electron decay rate in different systems



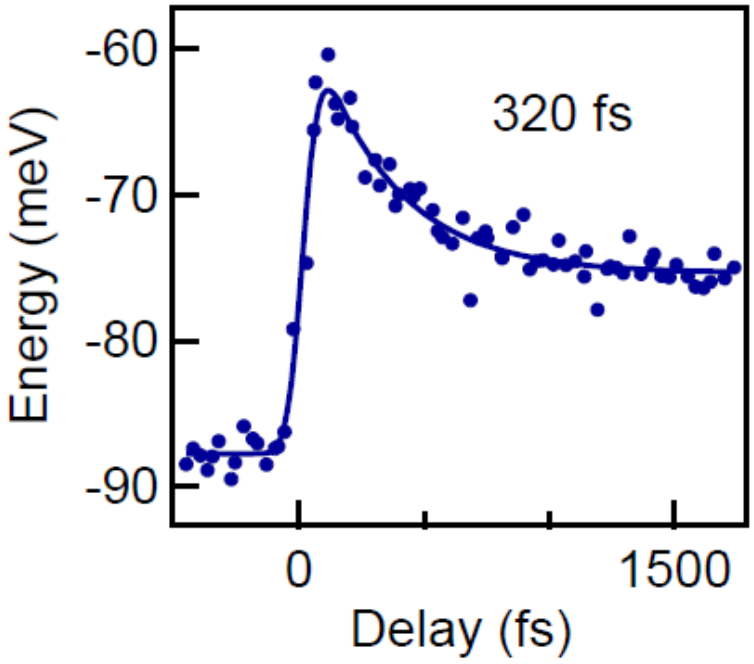
Hot electrons release their energy to phonon modes in less than 80fs.
=> very strong electron-phonon coupling

Ultrafast filling of the pseudogap

Sample at 40K excited by laser pulse of 2.3 mJ/cm^2 fluence

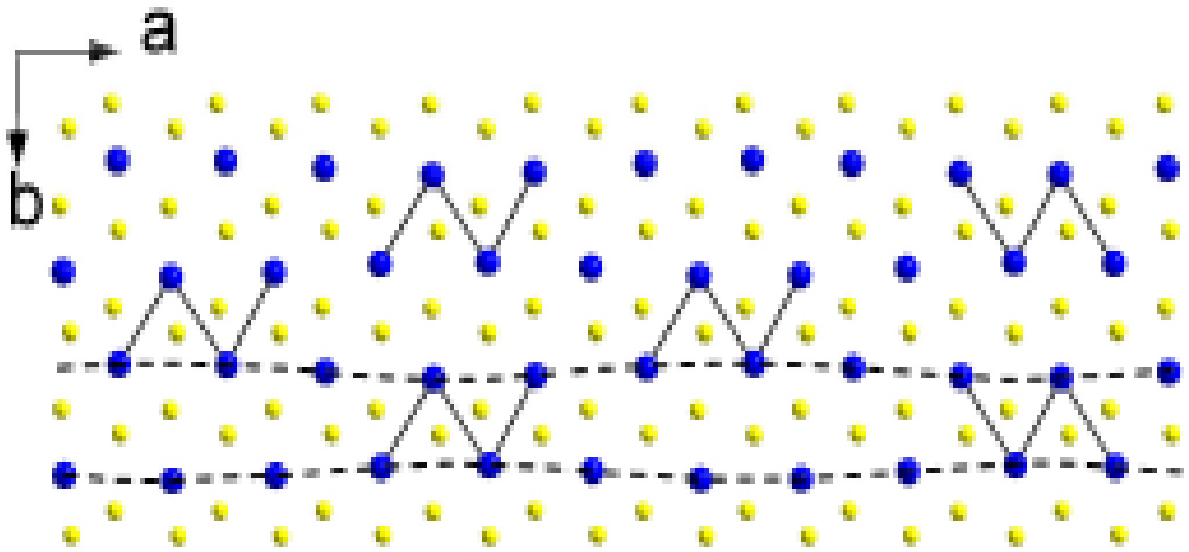


Recovery of the pseudogap



After closing the gap, the system is still far from thermal equilibrium.

Why is $[LaS]_{1.2}VS_2$ insulating ?



2.2 el. / V

- Band gap

  Incommensurate structure : it's hard to make a band insulator !

  Why such a fast relaxation ?

- Mott, electronic correlations…

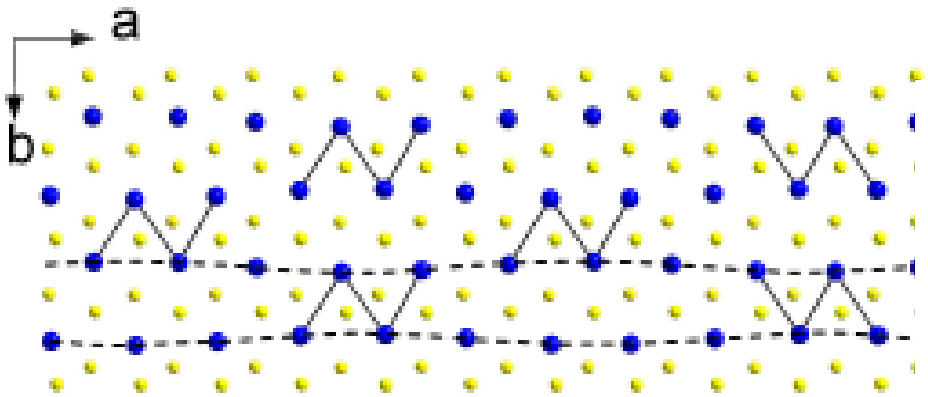
  Not close to half-filling, no strong renormalization => *unlikely*

- Disorder

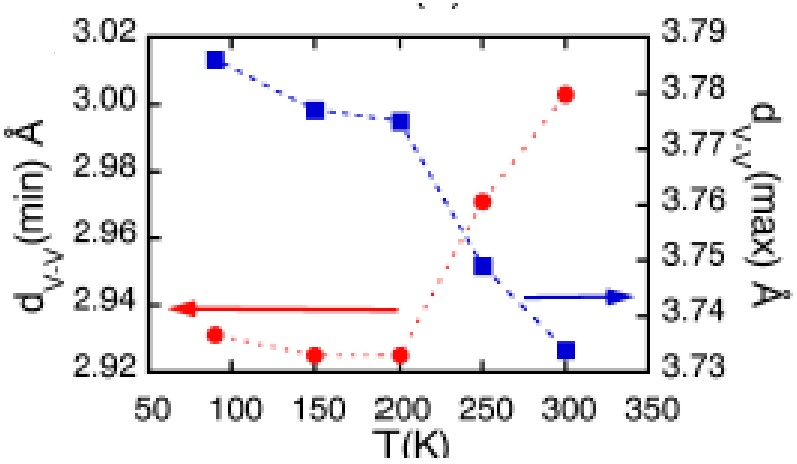
  Clear dispersion in ARPES, clear modulation in x-ray => *unlikely*

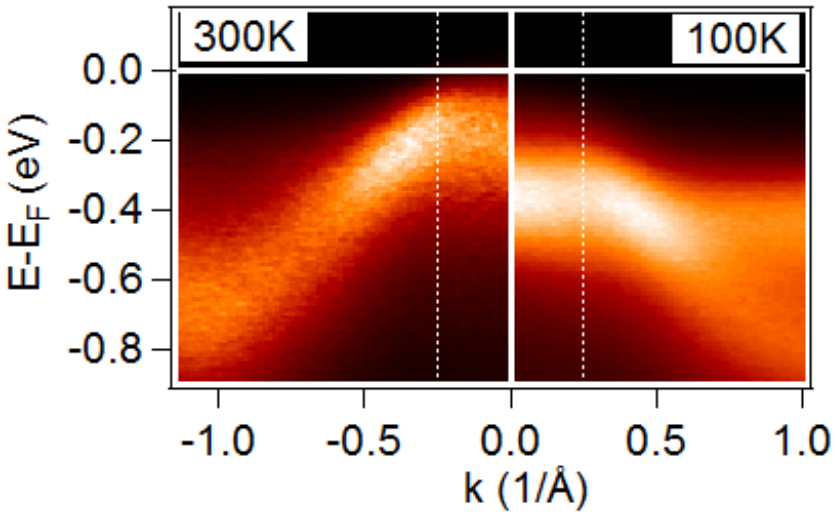
Change of electronic structure with T

There is no gap opening at low T but a complete change of electronic structure



Modulation amplitude vs T





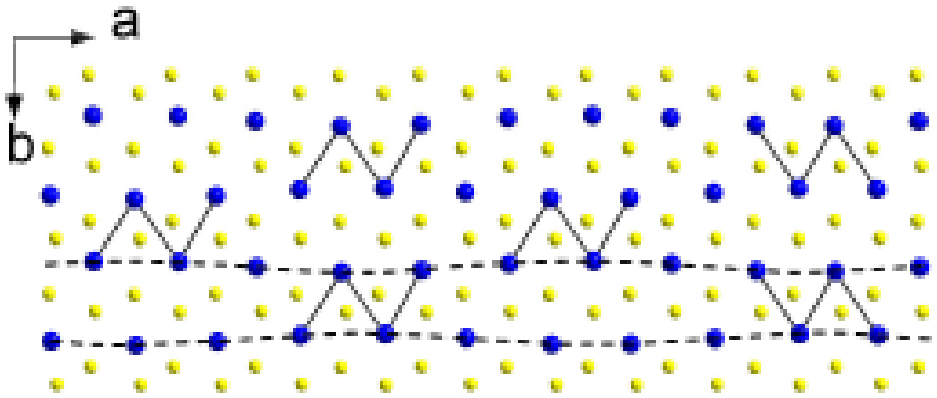
⇒ The distorsion brings atoms close to chemical bonding in V clusters

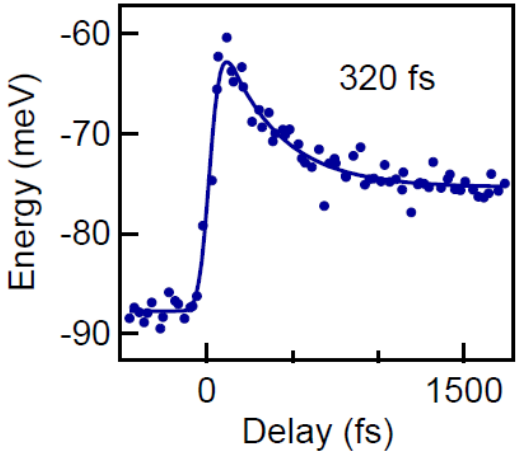
⇒ Very strong electron-phonon coupling !

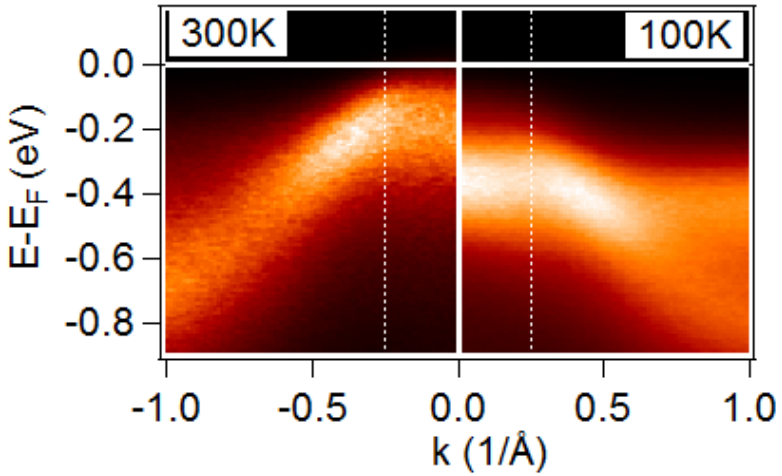
Strong coupling between distorsion and electronic properties

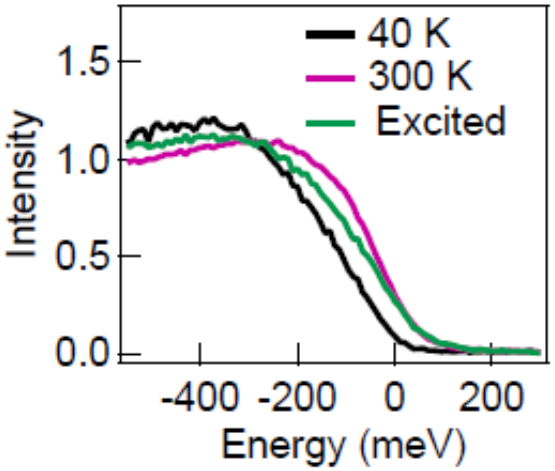
⇒ No collective excitation

⇒ Filling of the gap by local and incoherent excitations









Conclusion :

nature of the instability in $[LaS]_{1.2}VS_2$?

« purely electronic »

coupled

« purely structural »

Origin of the instability

Mott insulator

Charge Density Waves

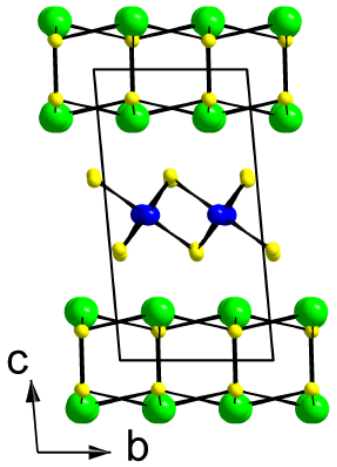
Band insulator

Excitonic insulator

$[LaS]_{1.2}VS_2$?

weak — strong

el-ph coupling



On the origin of charge-density waves in select layered transition-metal dichalcogenides

Table 1. Qualitative comparison of weak-coupling and strong-coupling CDWs.

	Weak-coupling CDW	Strong-coupling CDW
PLD/CDW amplitude	Small	Large
Energy gap	Small ($\Delta / E_F \ll 1$)	Large ($\Delta / E_F \lesssim 1$)
Coherence length	Large ($\xi / a \gg 1$)	Small ($\xi / a \gtrsim 1$)
Electronic energy gain	Arising mostly near k_F ($\propto \Delta^2 \ln \Delta$)	Spread over Brillouin zone ($\propto \Delta$)
CDW periodicity w.r.t. original lattice	Incommensurate ($\lambda_0 = \pi / k_F$)	Tends to be commensurate
Thermal disordering	Due to electronic entropy	Due to lattice entropy
Electron–hole pairing above T_0	No	Yes, but pairs are incoherent
Qualitative picture	Fermi surface instability	Local chemical bonding

K. Rossnagel et al., J. Phys Cond Mat 2011

Conclusion :

nature of the instability in $[LaS]_{1.2}VS_2$?

« purely electronic »

coupled

« purely structural »

Origin of the instability

Mott insulator

Excitonic insulator

Charge Density Waves

Band insulator

$[LaS]_{1.2}VS_2$?

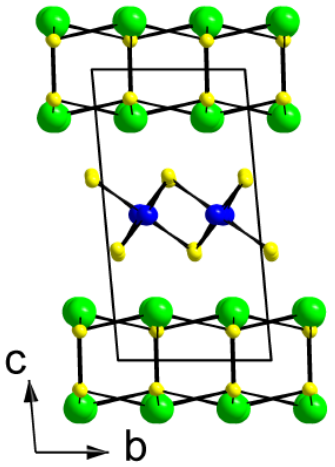
weak

strong

el-ph coupling

collective

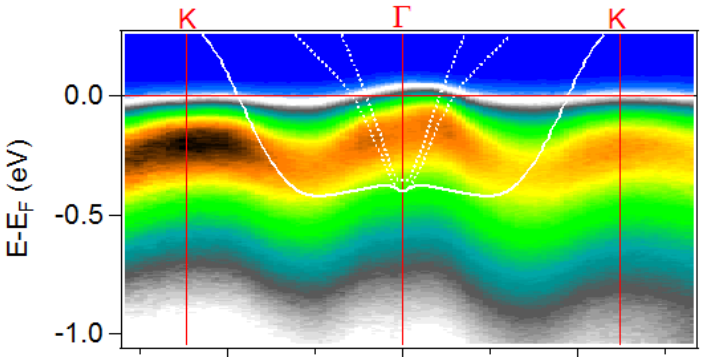
molecular



⇒ Such « hidden » chemical bonding could play a role in many « CDW » systems or valence bond solids ($LiVS_2$)

⇒ High sensitivity to perturbations : resistive switching

GK dipsersion



Band structure for a=3.4Å and 3.5Å