Probing exotic superconductors by high sensitivity microcalorimetry

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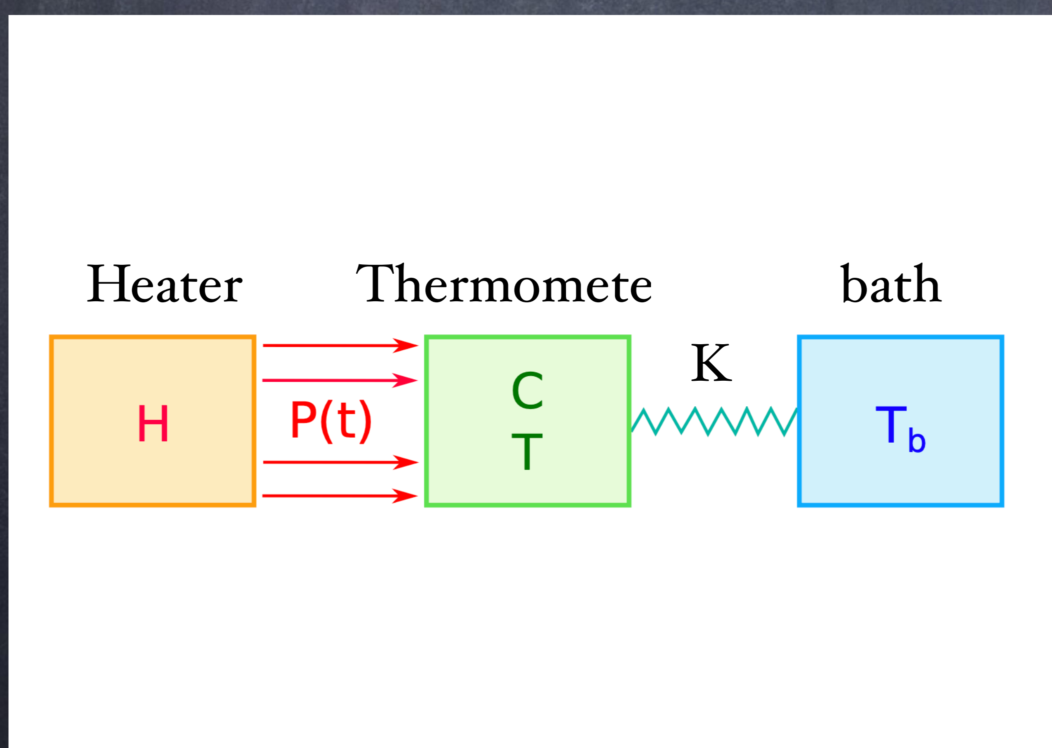
Alternative calorimetry

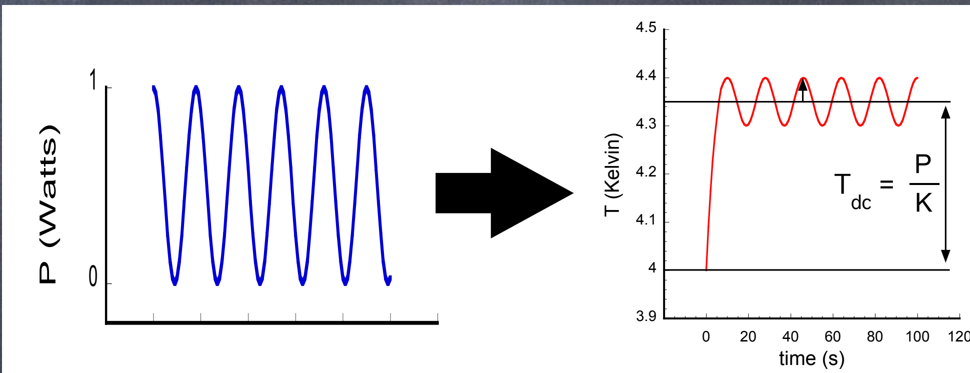
applying an alternative **heating** power P_{ac} (at ω)

to a sample of heat capacity **C**

linked to the thermal bath through K

and recording corresponding **temperature oscillations** T_{ac}





$$T_{ac} = \frac{P_{ac}}{K + jC\omega}$$

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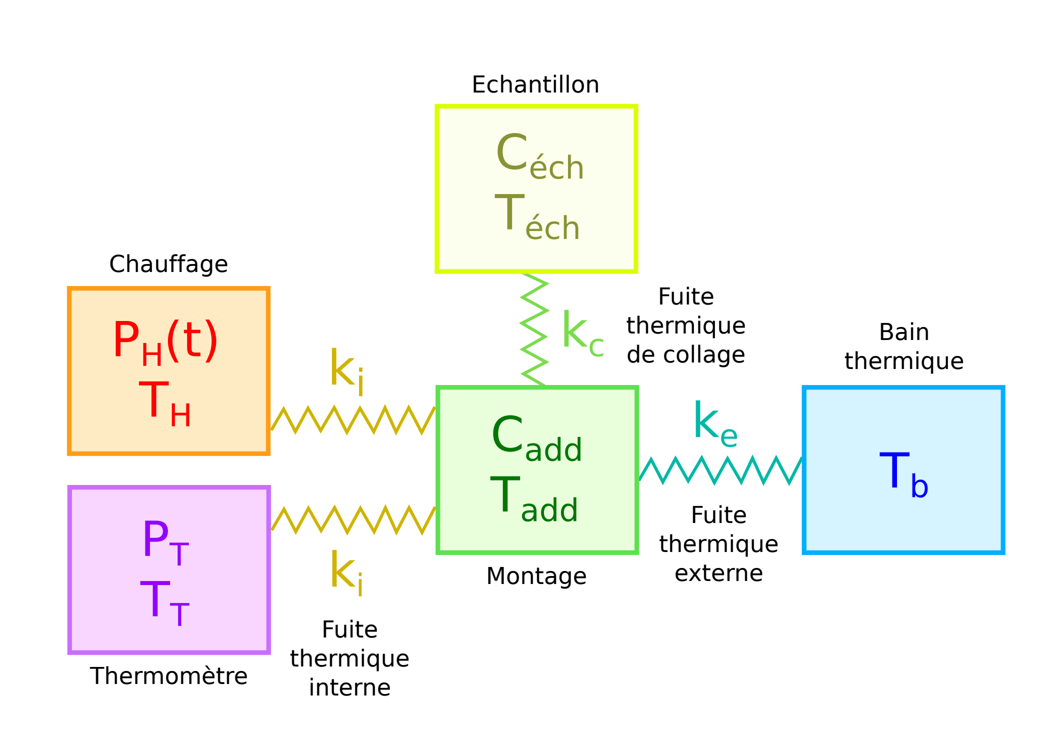
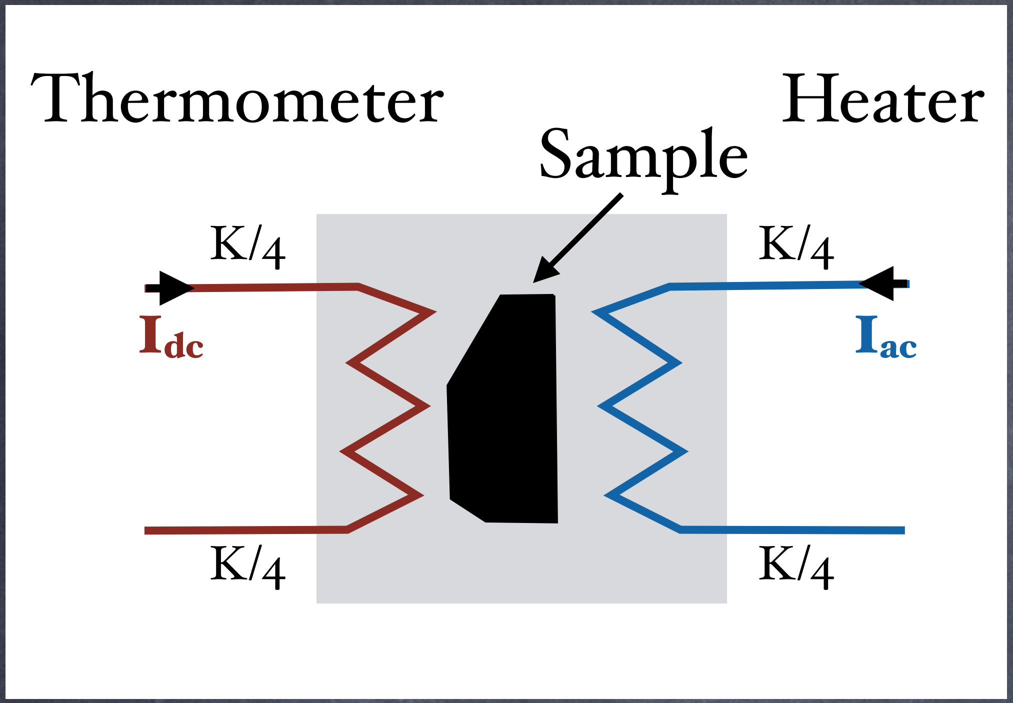


Unfortunately life is a little bit more complicated (internal couplings)

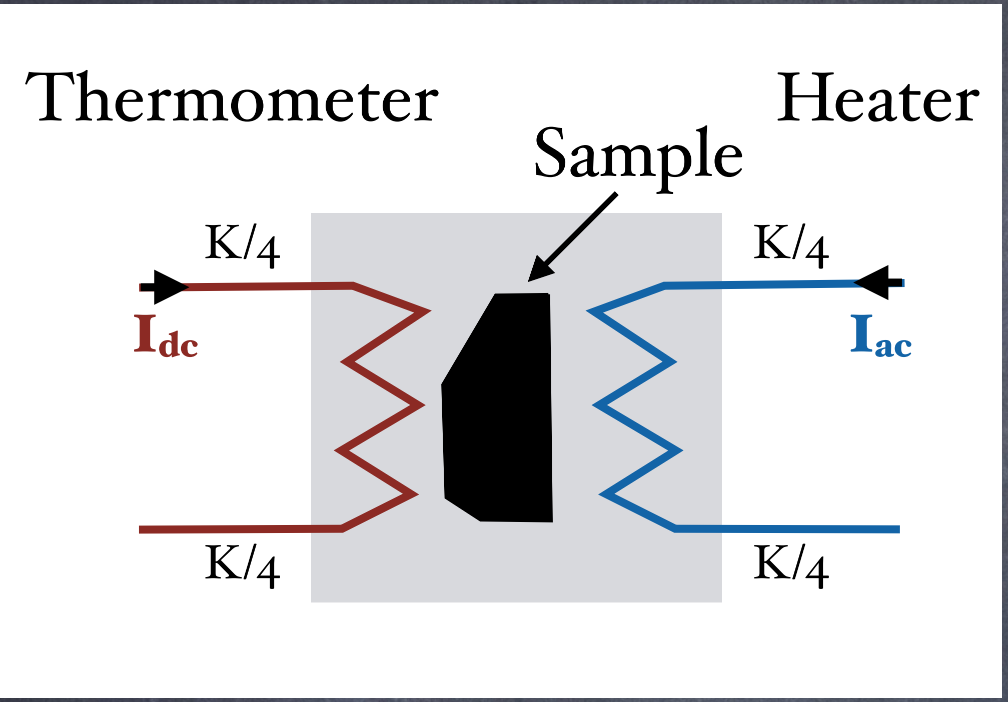
$$T_{\mathrm{ac}} = \frac{P_{\mathrm{ac}}}{K_{\mathrm{eff}} + jC_{\mathrm{eff}}\omega}$$

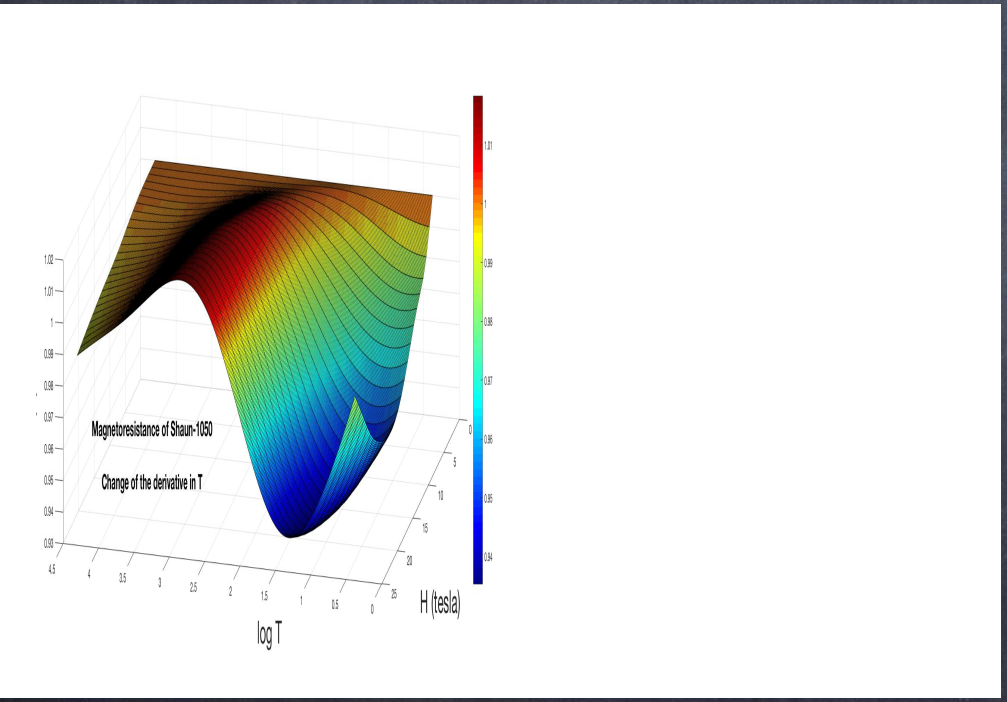
but $C_{\mathrm{eff}} \rightarrow C$ in the *good* conditions...



HEATER : **resistance R_H** (or optical fiber,...)

THERMOMETER : **R_T** (or thermocouple,...)





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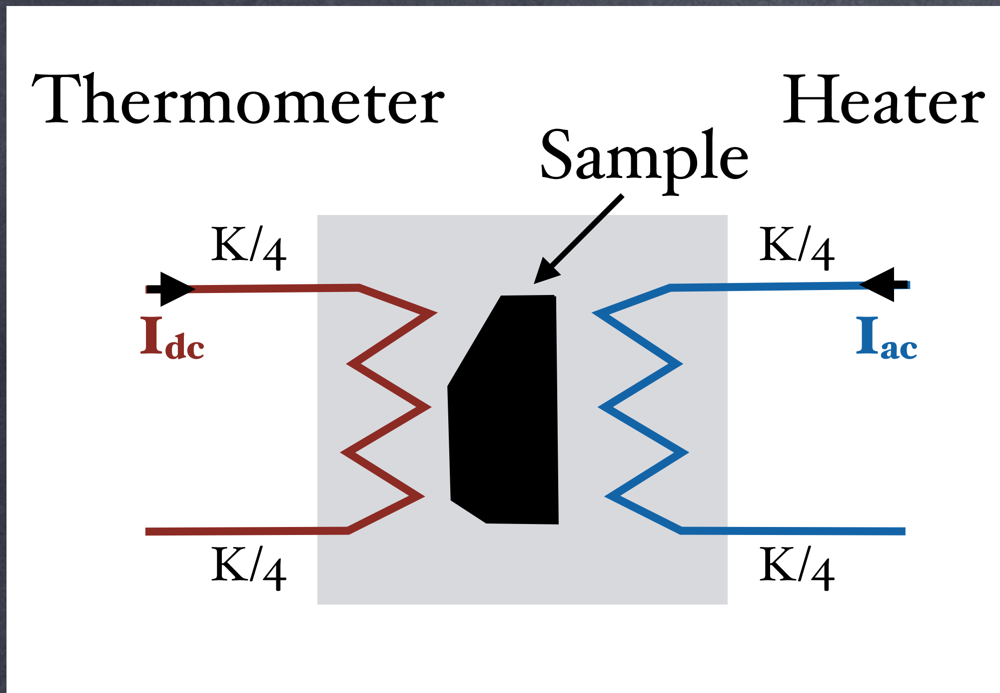
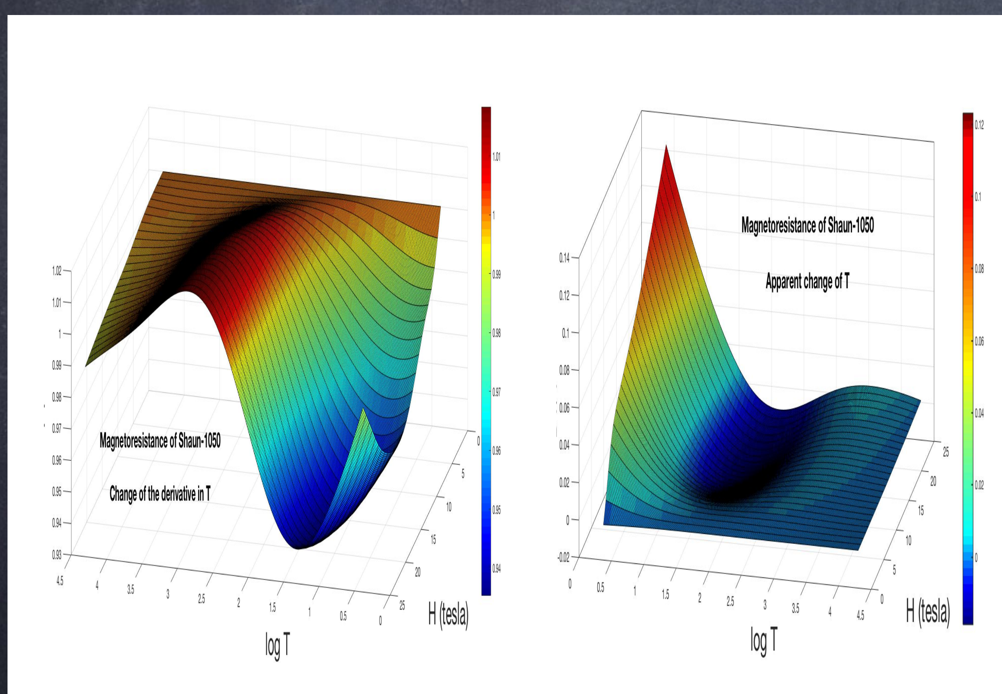
and this technique then requires

very demanding calibrations

for R(T,H)

$$V_{dc} = R_T(T, H)\, I_{dc} \rightarrow T$$





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$$V_{dc} = R_T(\mathrm{T, H})\, I_{dc} \rightarrow T$$

<u>AND dR/dT</u>

$$V_{ac} = \frac{dR_T}{dT}(\mathrm{T, H})\, T_{ac} I_{dc} \rightarrow C$$

- **Small samples**
  a few milligrams down to ~ 10 micrograms (or less)

- **Continuous H and/or T sweeps**
  in **extreme conditions** : 36T-0.1K (even under pressure)

- **Good absolute accuracy (> 95%)**
  as deduced from measurements in ultra pure Cu (1mg)

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  in **extreme conditions** : 36T-0.1K (even under pressure)

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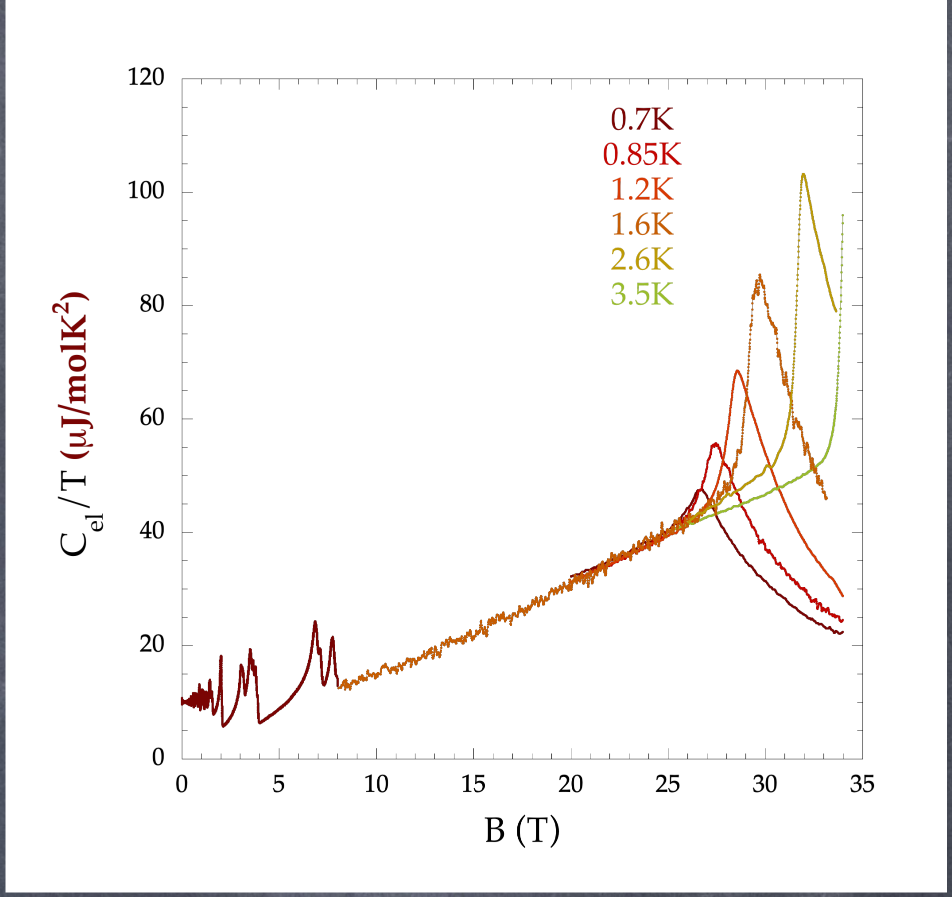
- **excellent resolution (down to a few 10^{-5}** of the total signal)

  Lock-in detection, filters ➜ $\Delta C \sim 10^{-13}$ J/K

⇒ detection of **small *features*** in C/T

as for instance quantum oscillations

in semimetals (Graphite)



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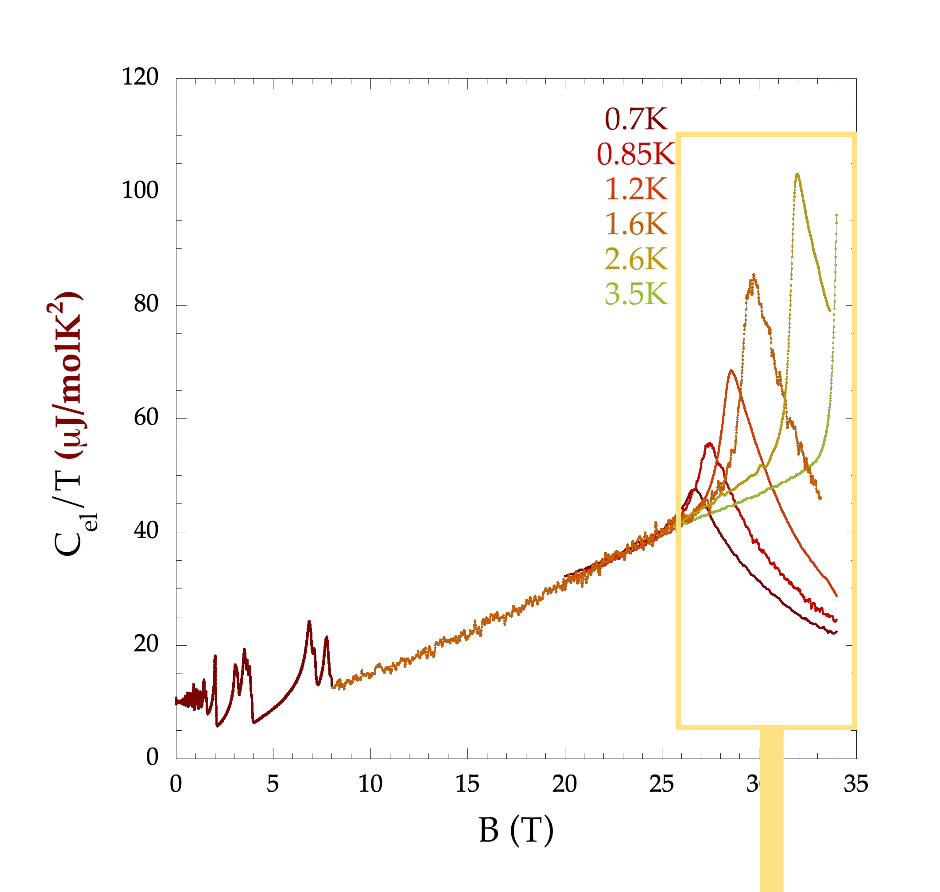

Electronic phase transition in the quantum limit (1D)

Marcenat *et al.* PRL 2021

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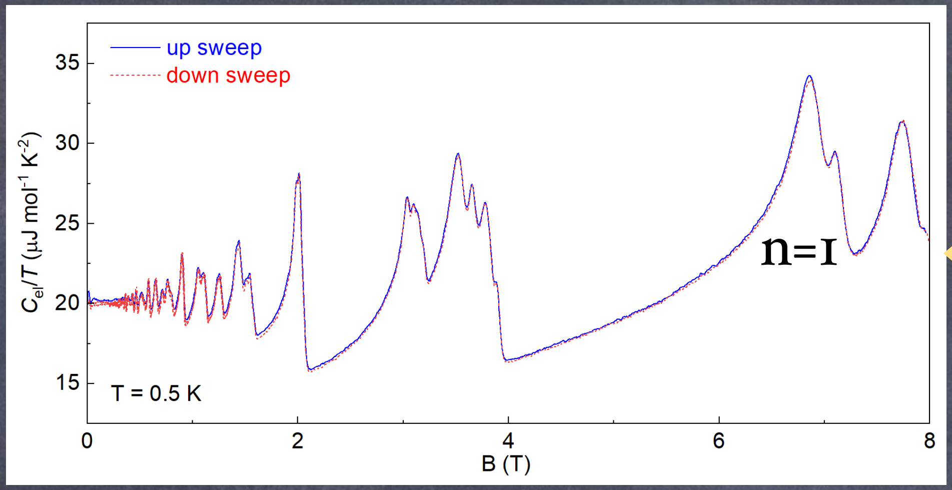
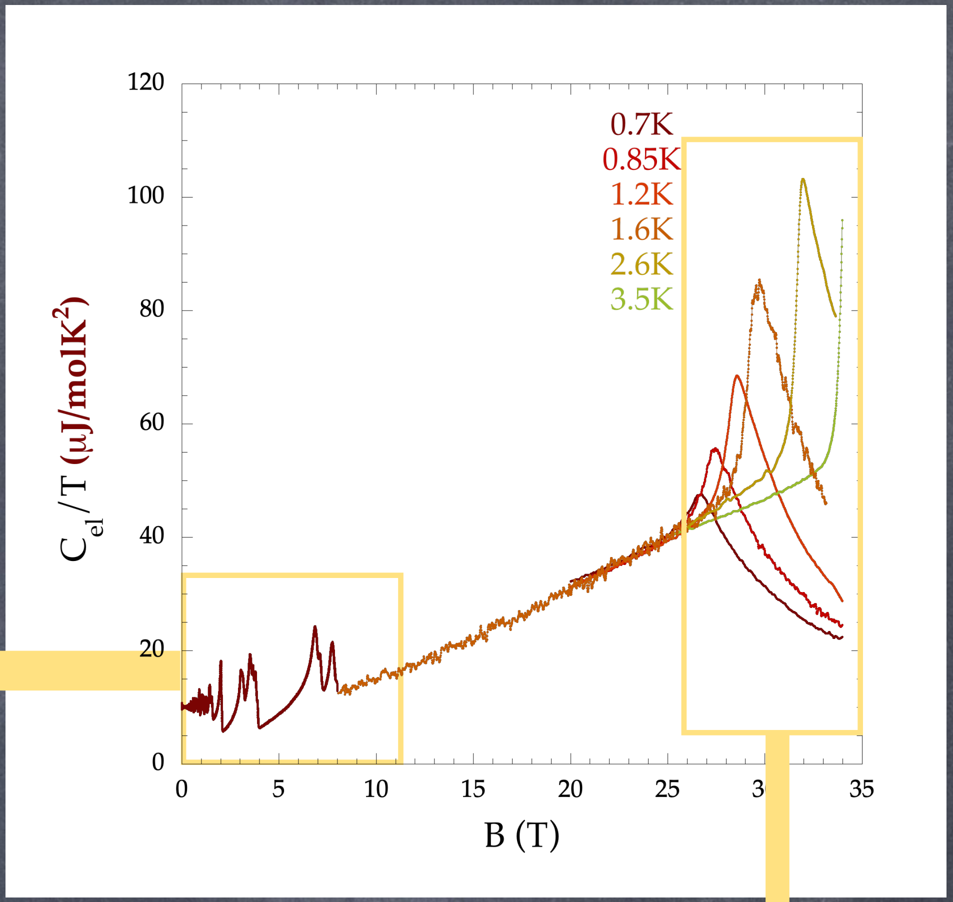
1nJ/K^2





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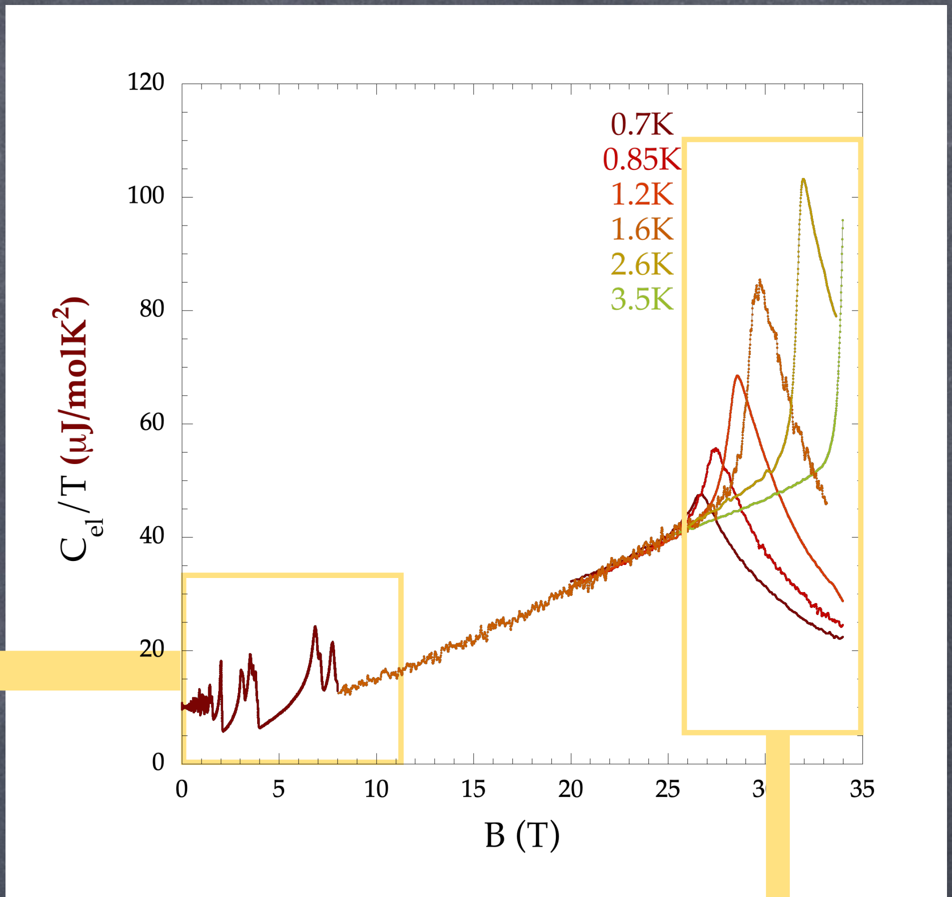
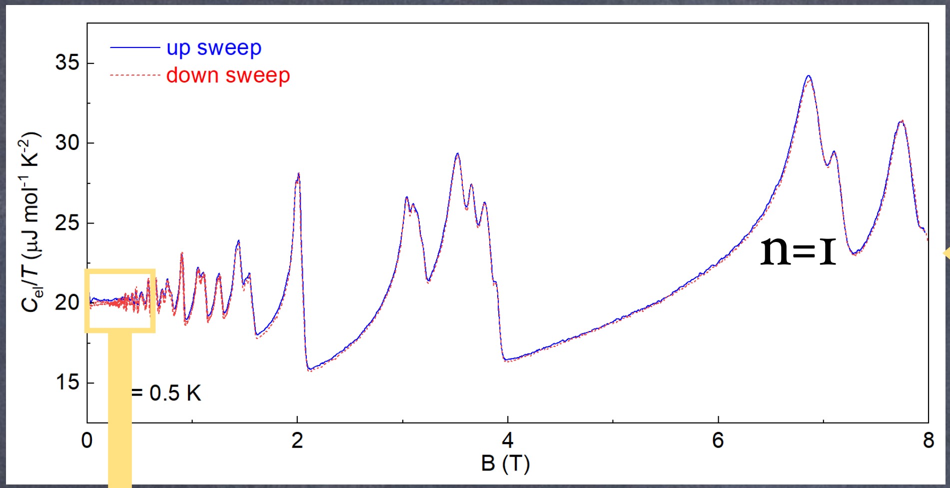
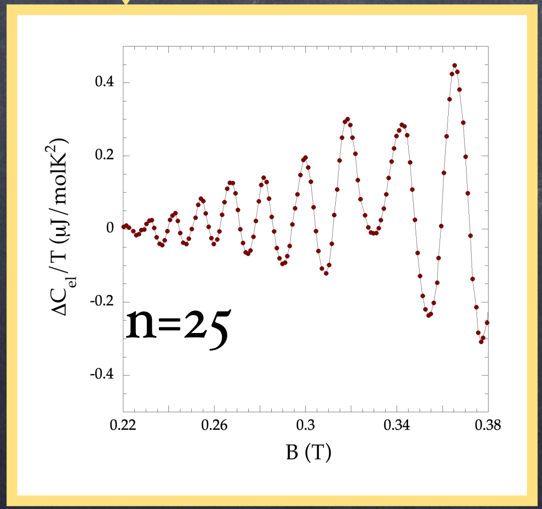




1nJ/K²



0.05nJ/K²

$\Delta C/C \sim 5\,\%$

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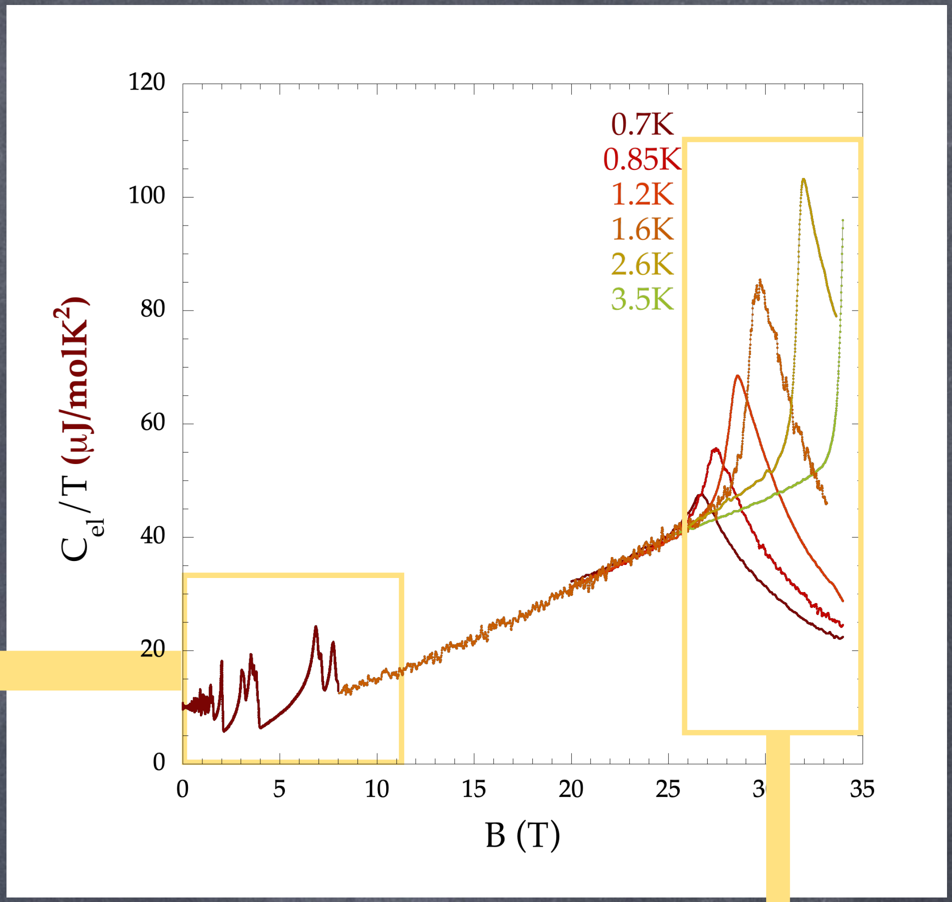

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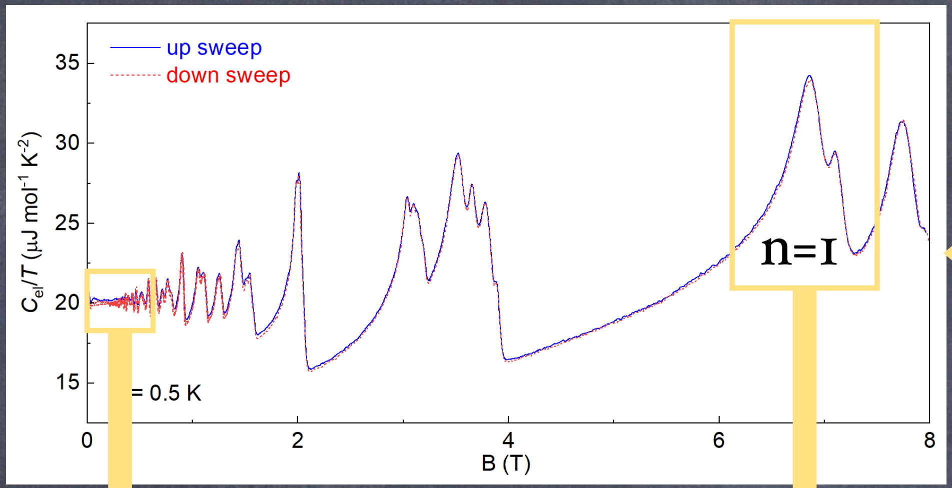

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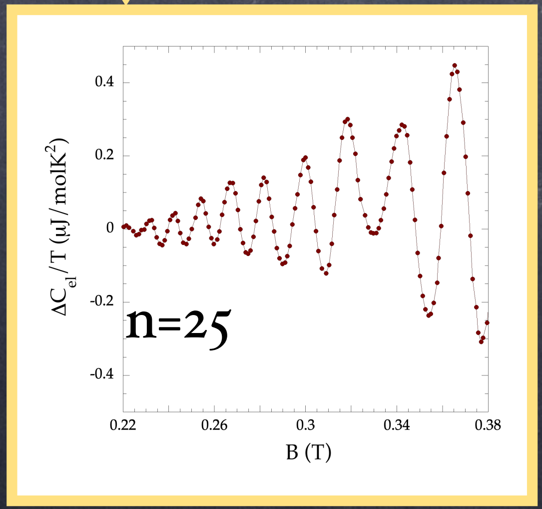

Note the *splitting* of the peaks characteristic of C/T measurements

Zhuo *et al.* Nature Com 2023

Electronic phase transition in the quantum limit (1D)

Marcenat *et al.* PRL 2021

Outline

Things that we do understand and that we do **NOT** understand on :

- **The FeSe nematic superconductor**

  H-T phase diagram and T & H dependence of the gap structure

- **Thermodynamic properties of the normal state in cuprates**

  Quantum criticality at the onset of the pseudo-gap and charge order

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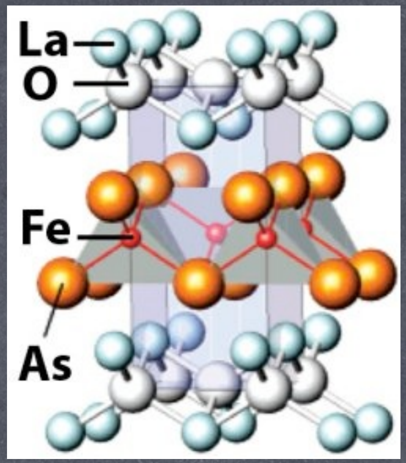
FeSe = **iron based** superconductor



attracted considerable interest due to the interplay between superconductivity and magnetism

and T_c rising up to ~ 50K in Gd(O,F)**Fe**As (so called 1111-phase)

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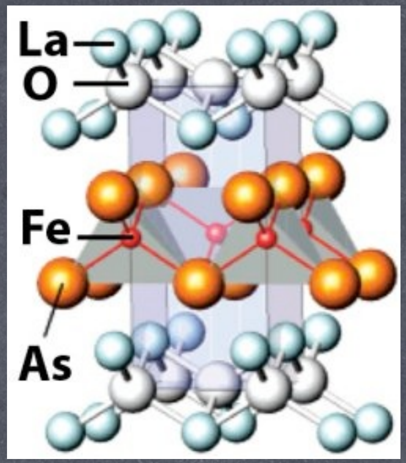
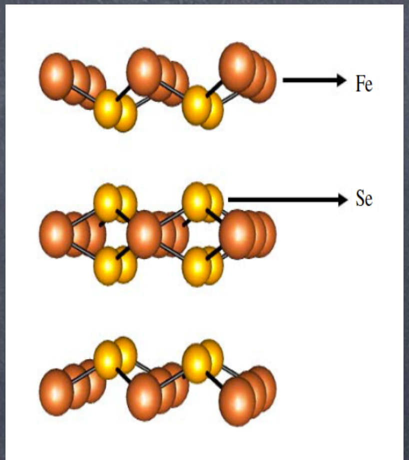


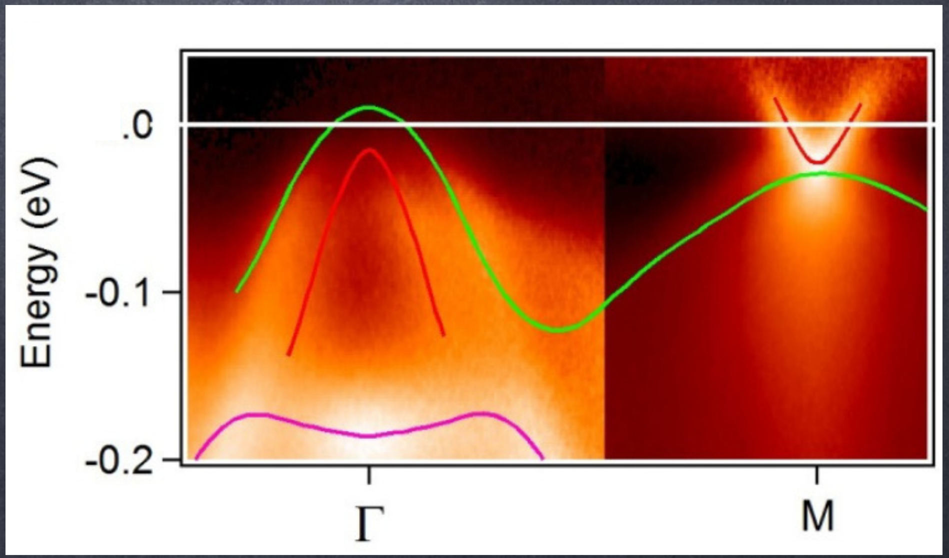
- **FeSe** = c-axis stacking of SC layers **without** any charge reservoir.
- Moderate T_c~9K, but very low carrier concentration



semi-metal : small *e+hole pockets*

$E_F \sim \Delta$: *high* T_c (two gap-) superconductor

at the verge of a Bose Einstein Condensation



L.Fanfarillo *et al.*, PRB 2016

- interesting **vortex physics**

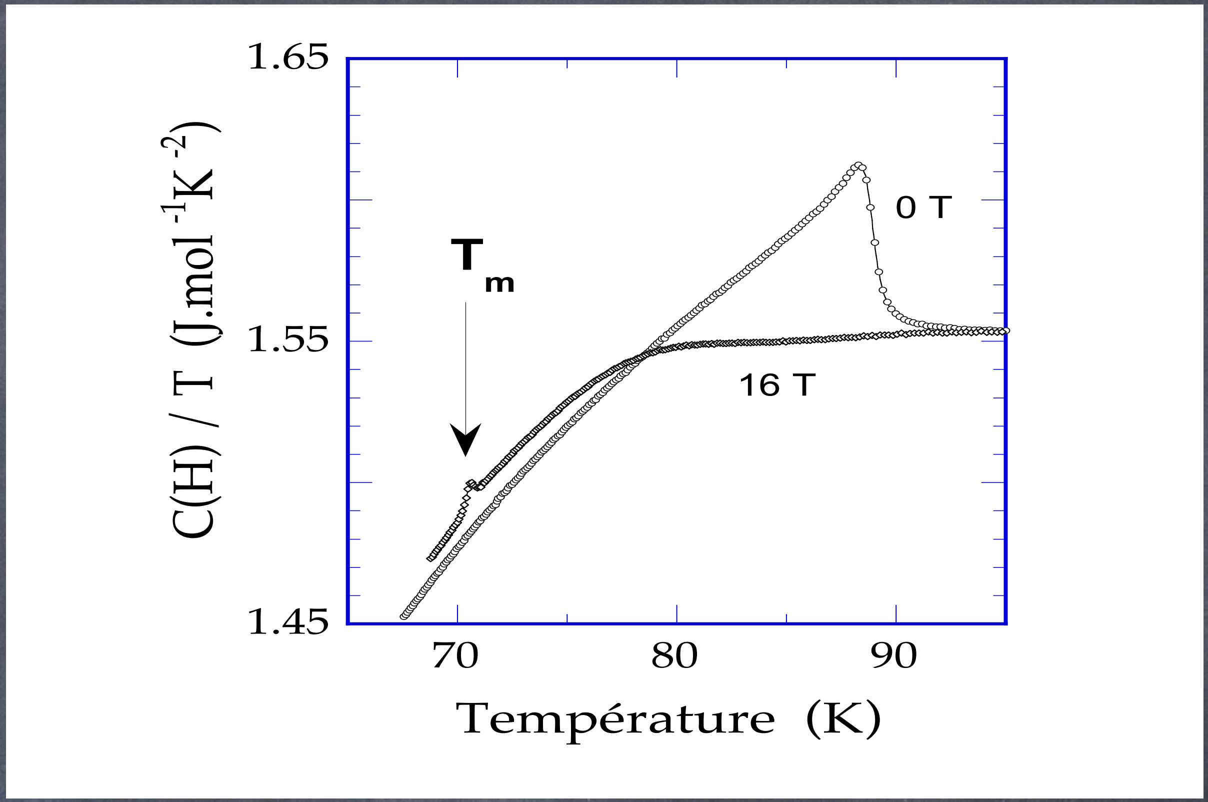
It all started in the 90s in high T_c cuprates

$\Rightarrow$ Strong thermal fluctuations

$$G_i = (1/8)(k_B T_c/\epsilon_{cond})^2 \sim 10^{-2}$$

where $\epsilon_{cond} = \epsilon_0 \xi$ with $\epsilon_0 = \Phi_0^2/4\pi\lambda^2$

$\Rightarrow$ **melting** of the vortex solid



Sharp (1st order) peak in C/T

... but observed only in (a few)

high quality optimally doped

YBaCuO single crystals

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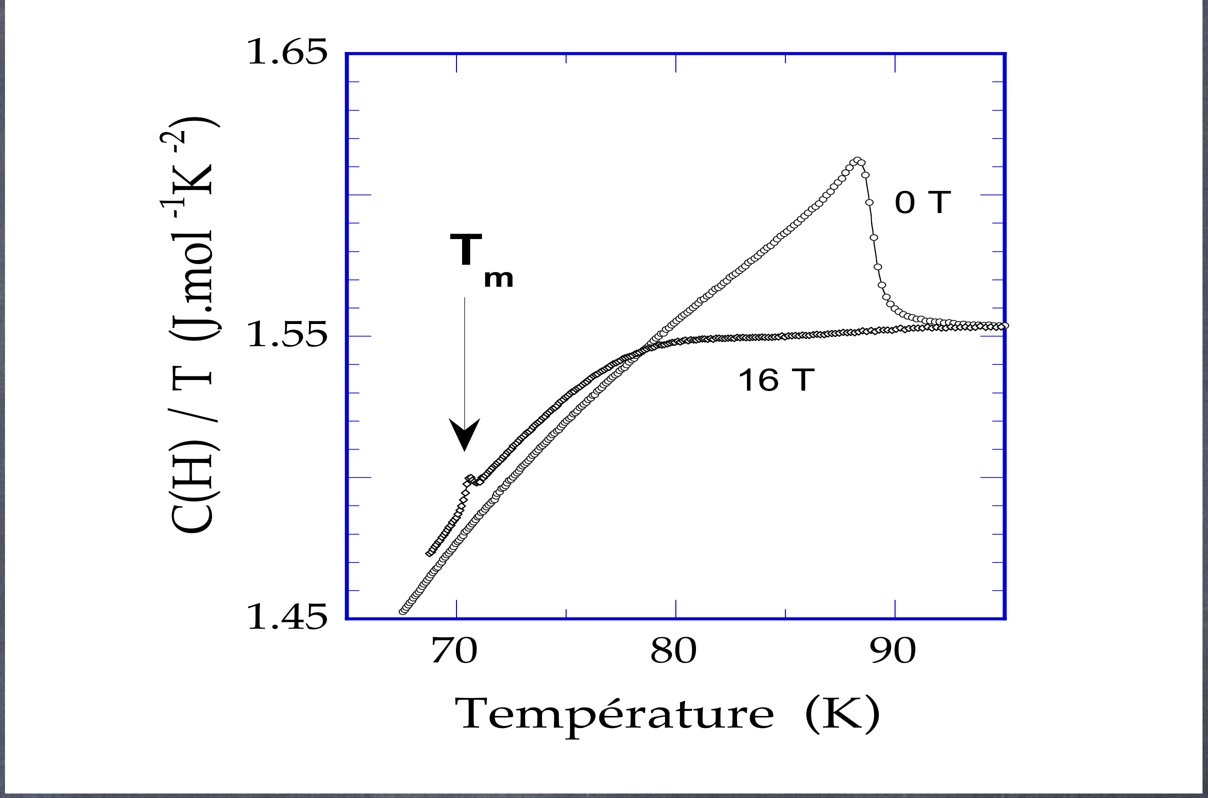

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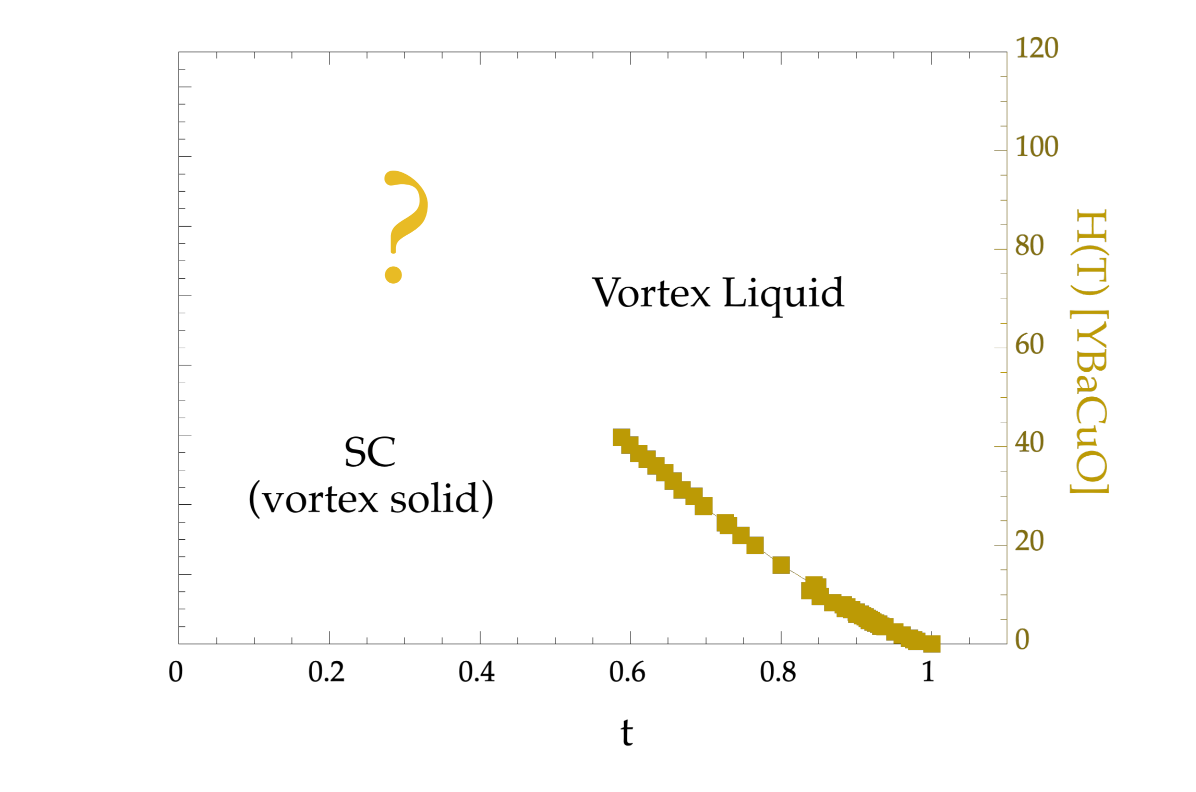


The melting line can only be

tracked down to $\sim T_c/2$

FeSe : Small superfluid density

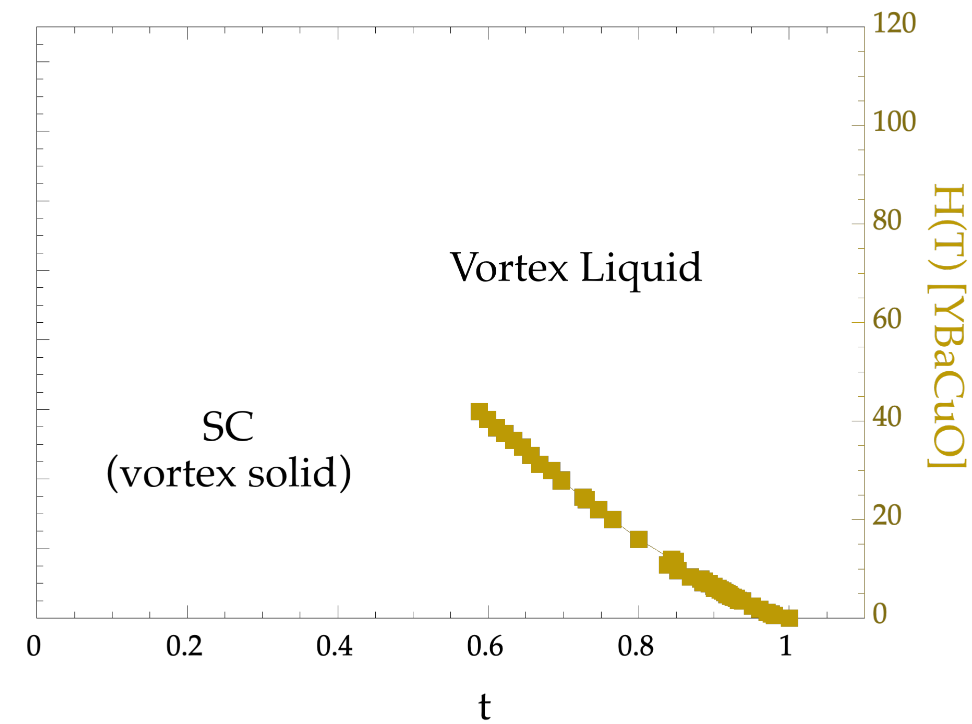
$\Rightarrow$ large λ value

$\Rightarrow$ **Very** small condensation energy

$\epsilon_{cond} = \epsilon_0 \xi_c$ ~ 160K

$\Rightarrow$ (still a) **Large Gi value**

	FeSe	Nb	YBaCuO
T_c	9	9	92
λ (A)	5000	400	1200
ε_o (K/A)	8	1200	140
ξ_c (A)	20	400	4
ε_{cond} (K)	**160 !**	5.10^5	560
Gi	10^{-3}	3.10^{-9}	5.10^{-3}



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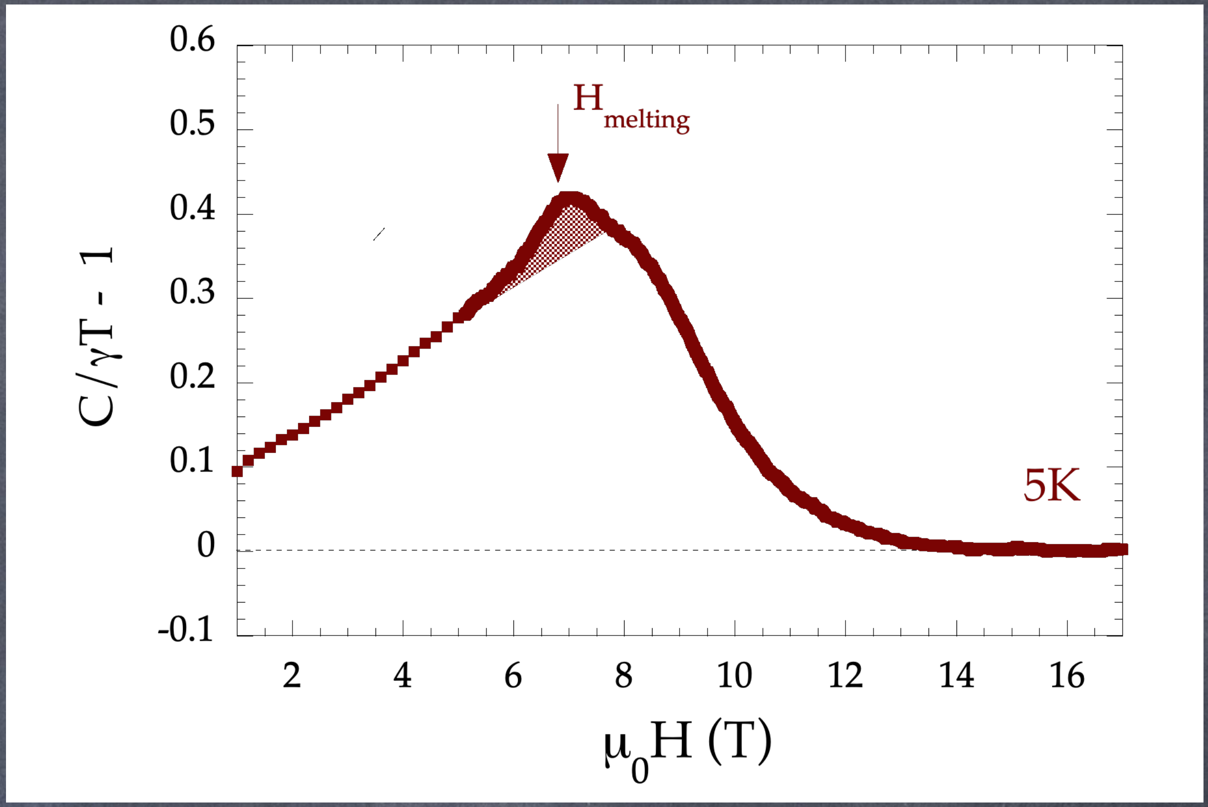

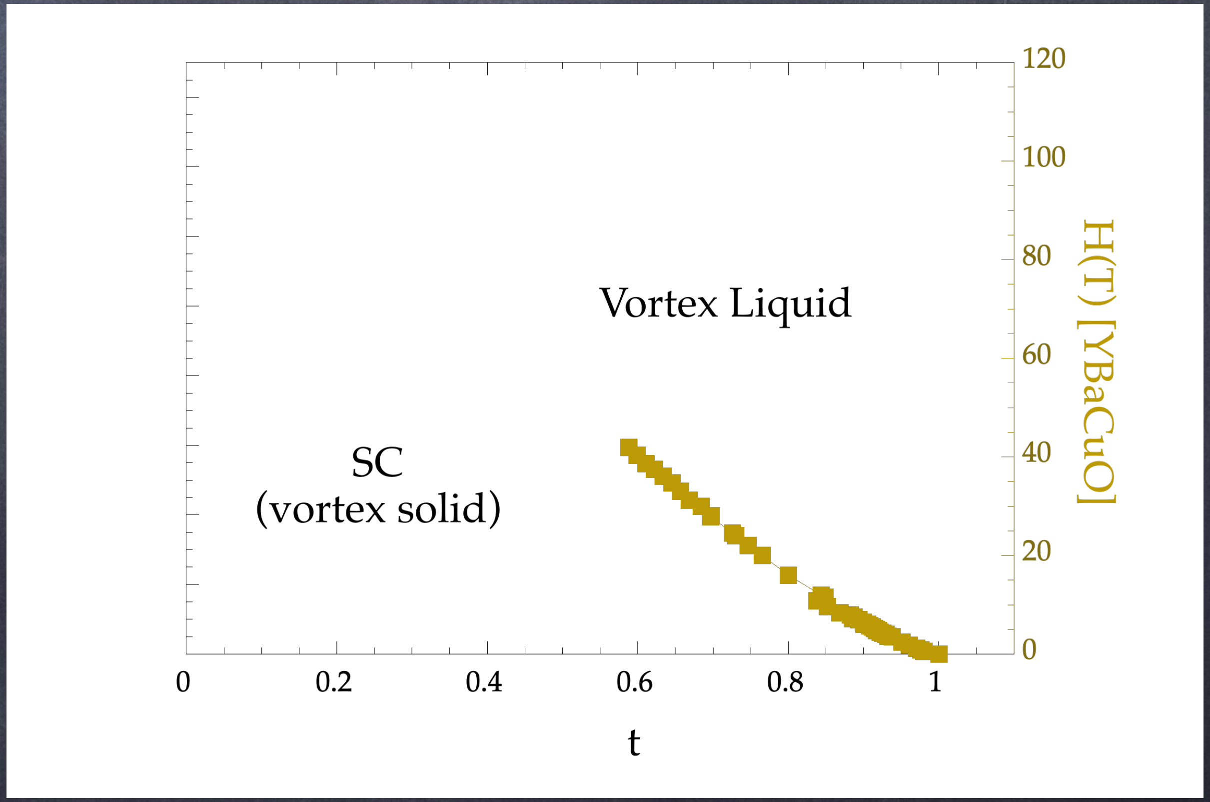

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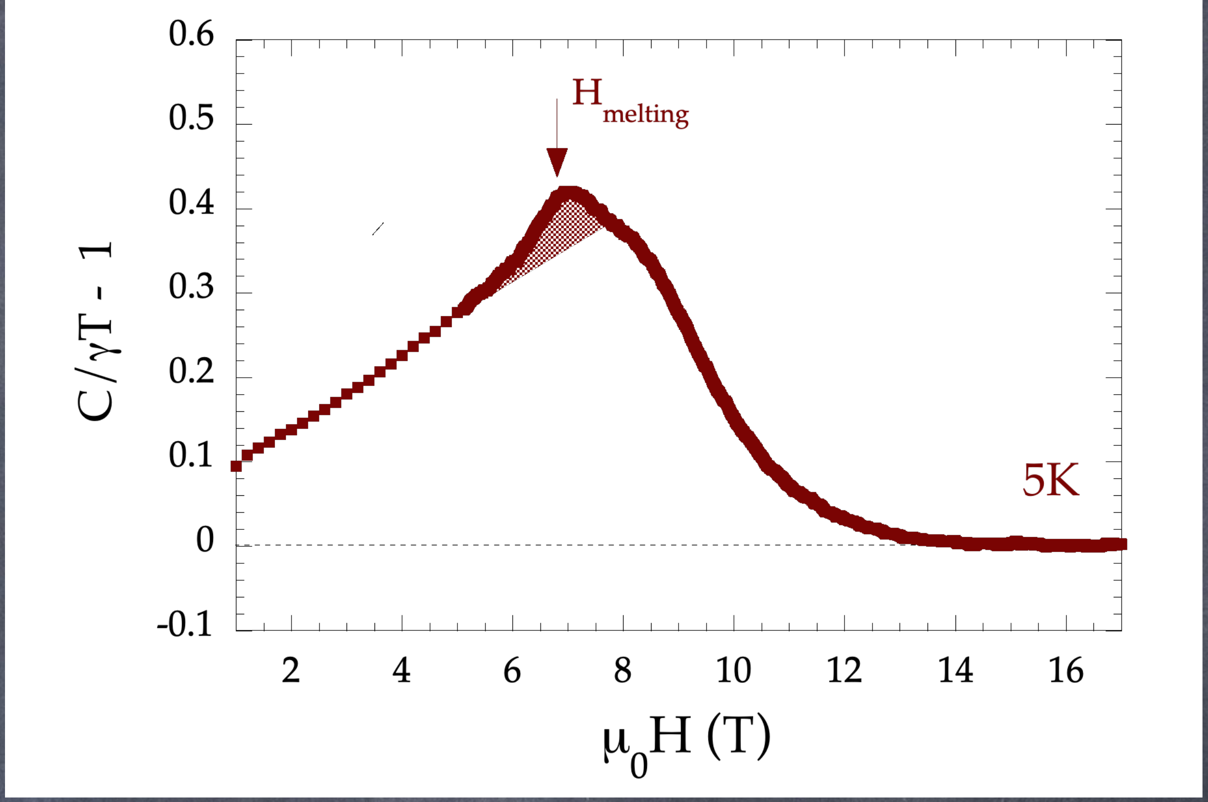
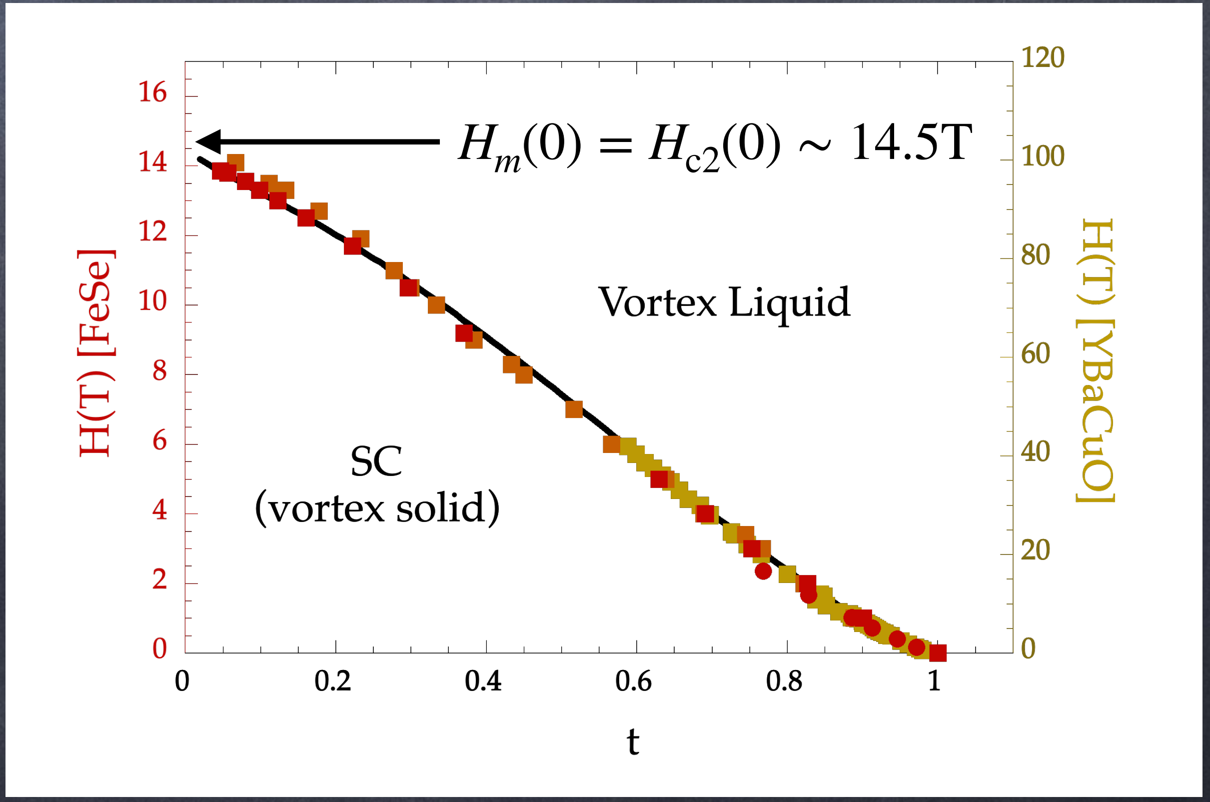
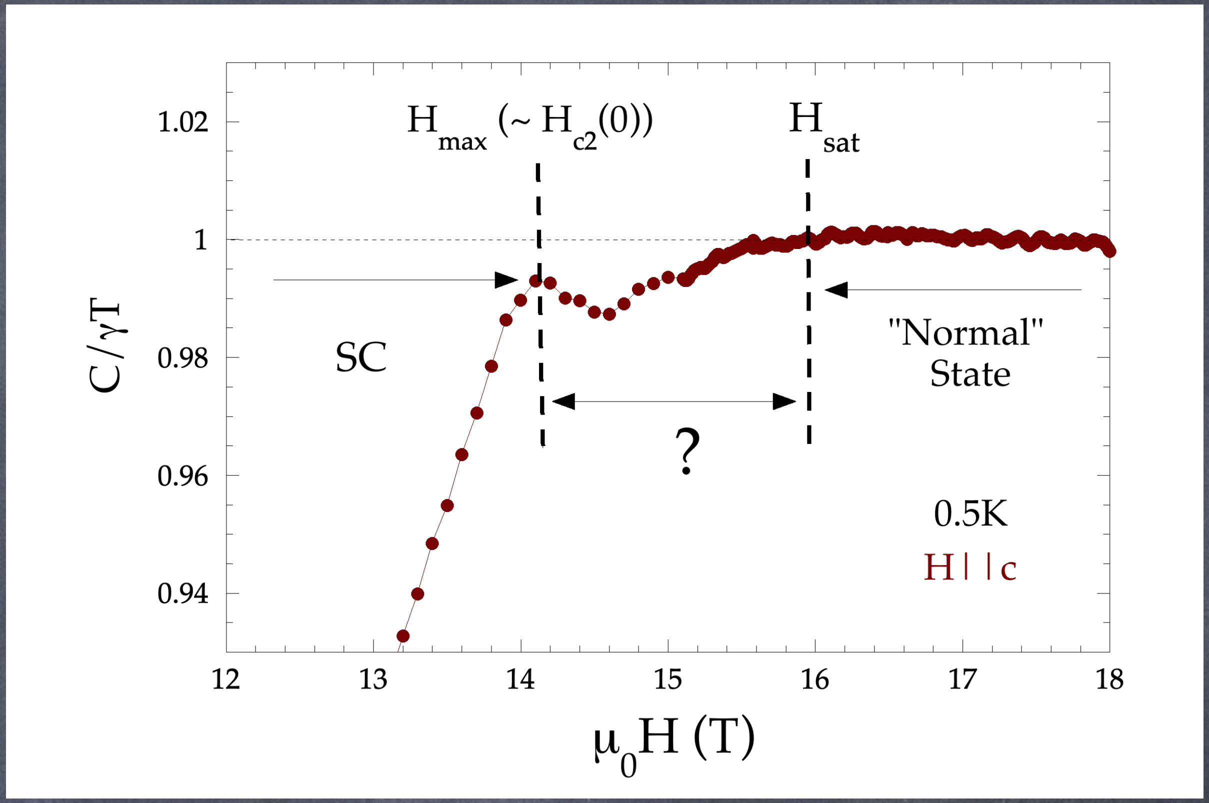
...which can be studied (and well fitted) **down to the lowest T**
($H_{c2}(0) \sim 14.5$T)
(interplay with paramagnetic limit for H||ab see F.Hardy *et al.* PRR 2020)





but C/T still increases with H
well above $H_{c2}(0)$???
and only saturates above H_{sat}



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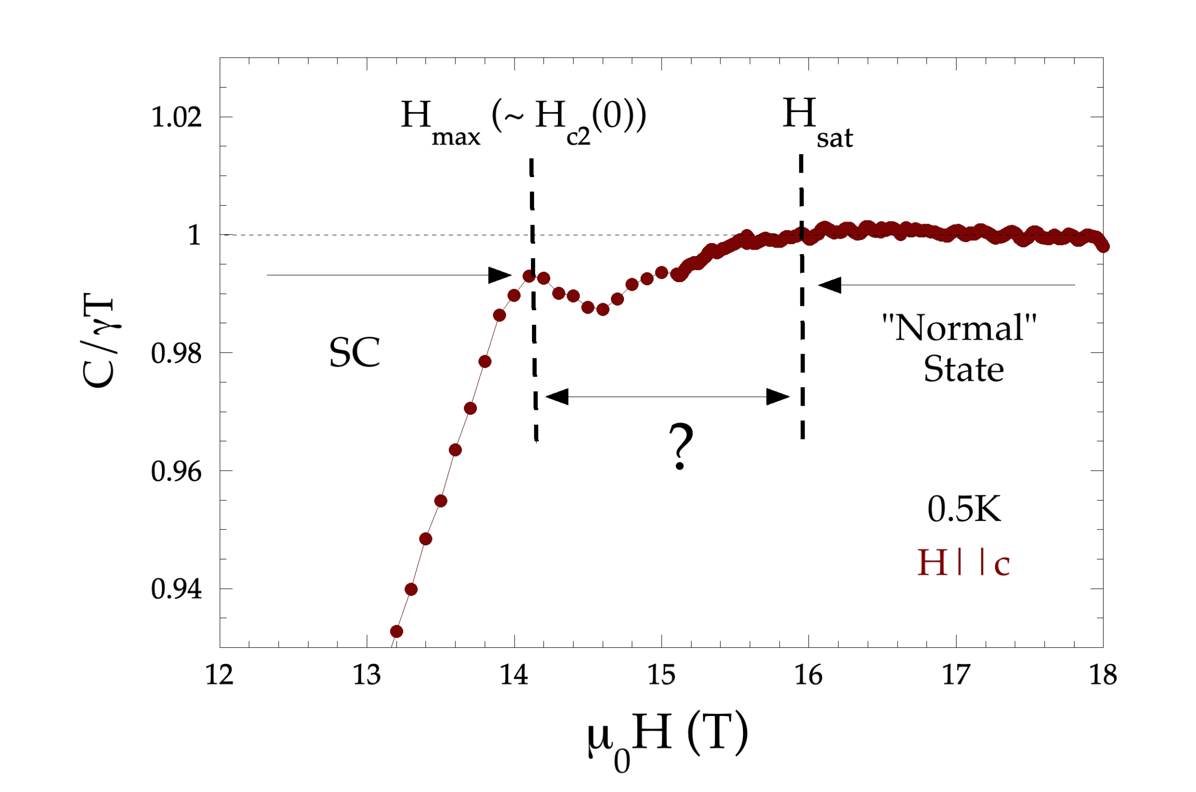
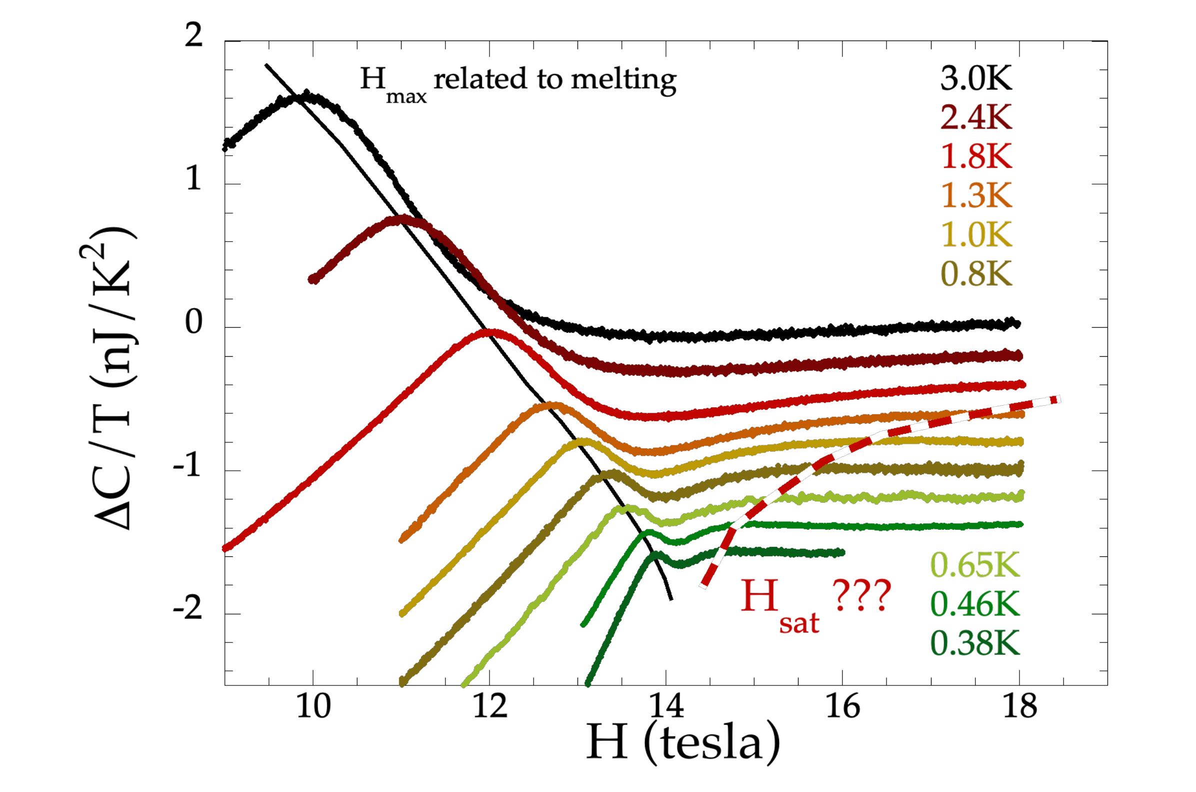


Nature of the H_{sat} field ?

increases with T !

(→ 22 T @ 1.8K)



Signature of an electronic transition ?

- No magnetic order (still a structural phase transition)

  but orbitally ordered nematic state (breaking of the C_4 symmetry)

Mukherjee *et al.* PRL 2015, Aichhorn *et al.* PRB 2010, Yamakawa *et al.* PRX 2016, Watson *et al.* PRB 2017

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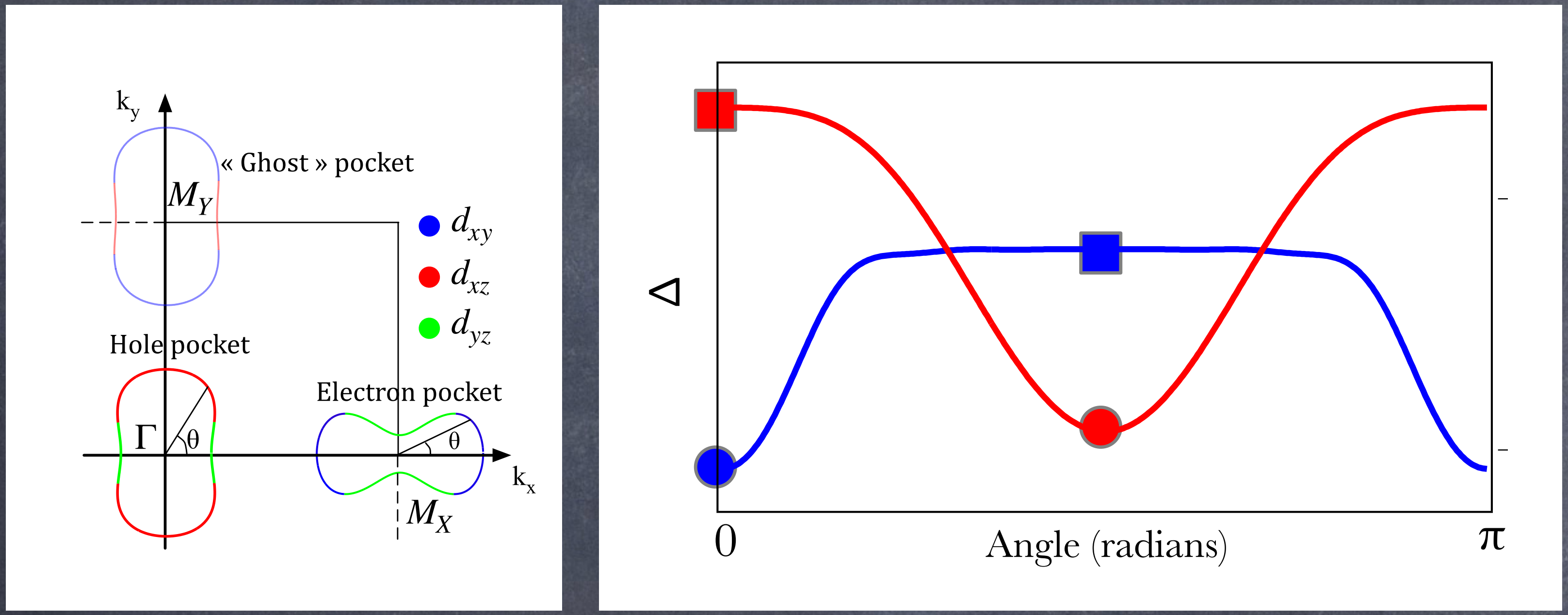
C2 anisotropy of (the spectral weight and) superconducting **gaps**



$\Delta_{\text{hole}}^{\max} \sim 2 - 3\text{meV}$

$\Delta_{\text{elec}}^{\max} \sim 1 - 2\text{meV}$

H.Cercellier *et al.*, PRB 2019 and references therein

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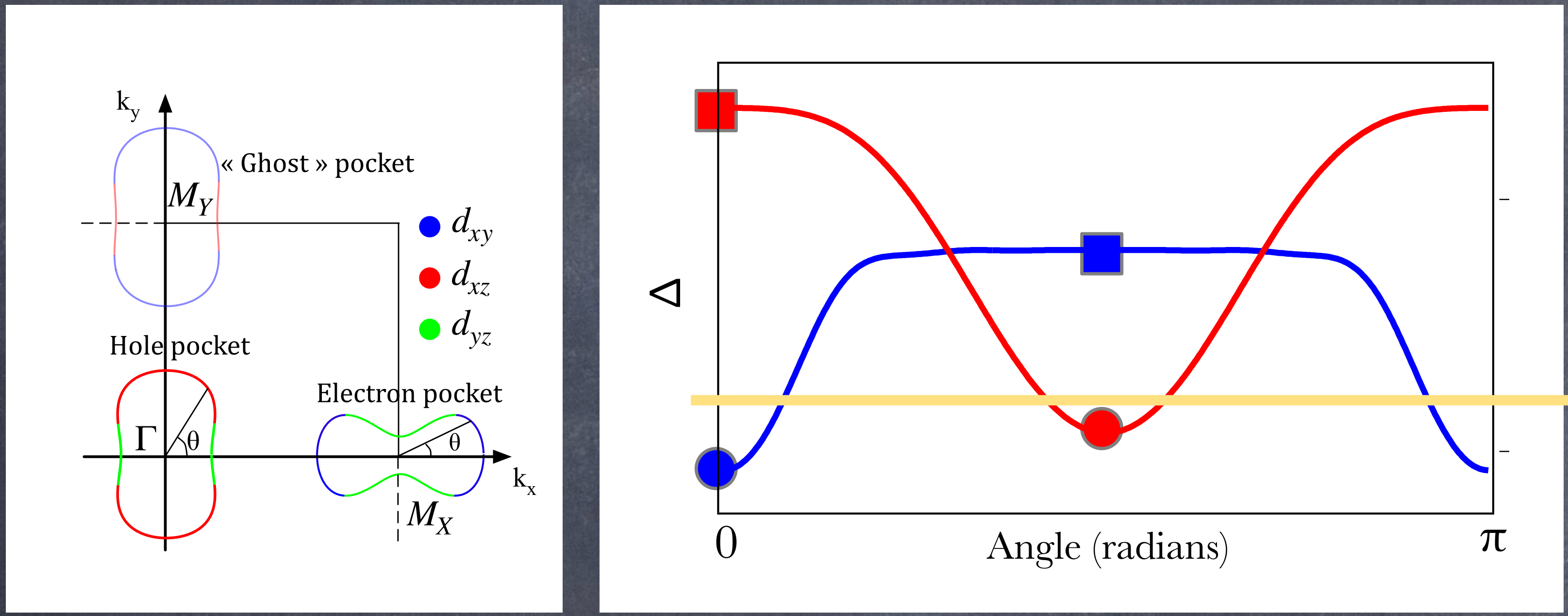


H.Cercellier *et al.*, PRB 2019 and references therein

$\Delta_{\rm hole}^{\rm max} \sim 2 - 3{\rm meV}$

$\Delta_{\rm elec}^{\rm max} \sim 1 - 2{\rm meV}$

0 ?

$\Delta_{\rm hole}^{\rm min}, \Delta_{\rm elec}^{\rm min} < 0$?

accidental nodes ?

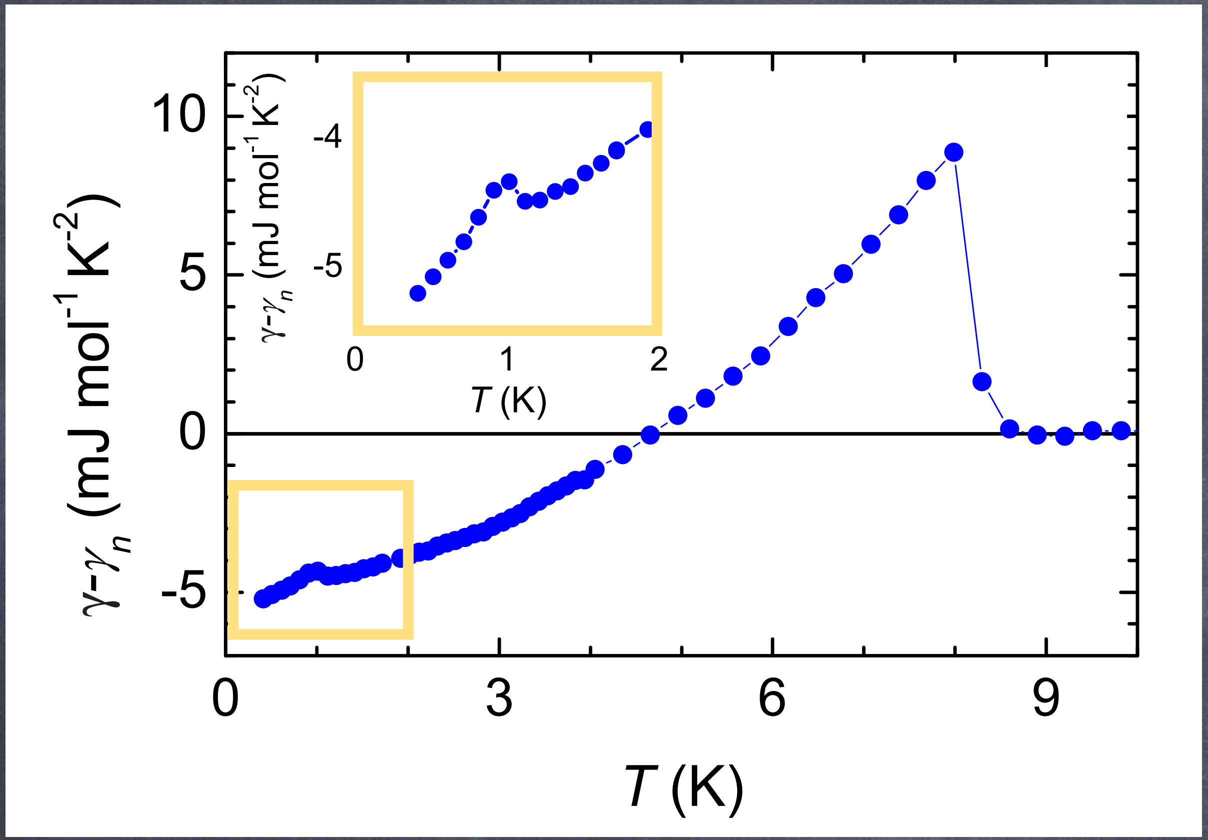
Nematic order couples the s and d wave harmonics of the gap (s+d symmetry)

Δ_s and Δ_d sensitive to **nematicity and intra/interband couplings**.

➡ *Accidental* gap nodes can show off if $\Delta_d > \Delta_s$

Moreover : *anomalous* specific heat anomaly **around 1K**

(but not observed in all samples ?)



G.Y.Chen *at al.* PRB 2017

possible $s + d \rightarrow s + e^{i\theta}d$

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suggested by

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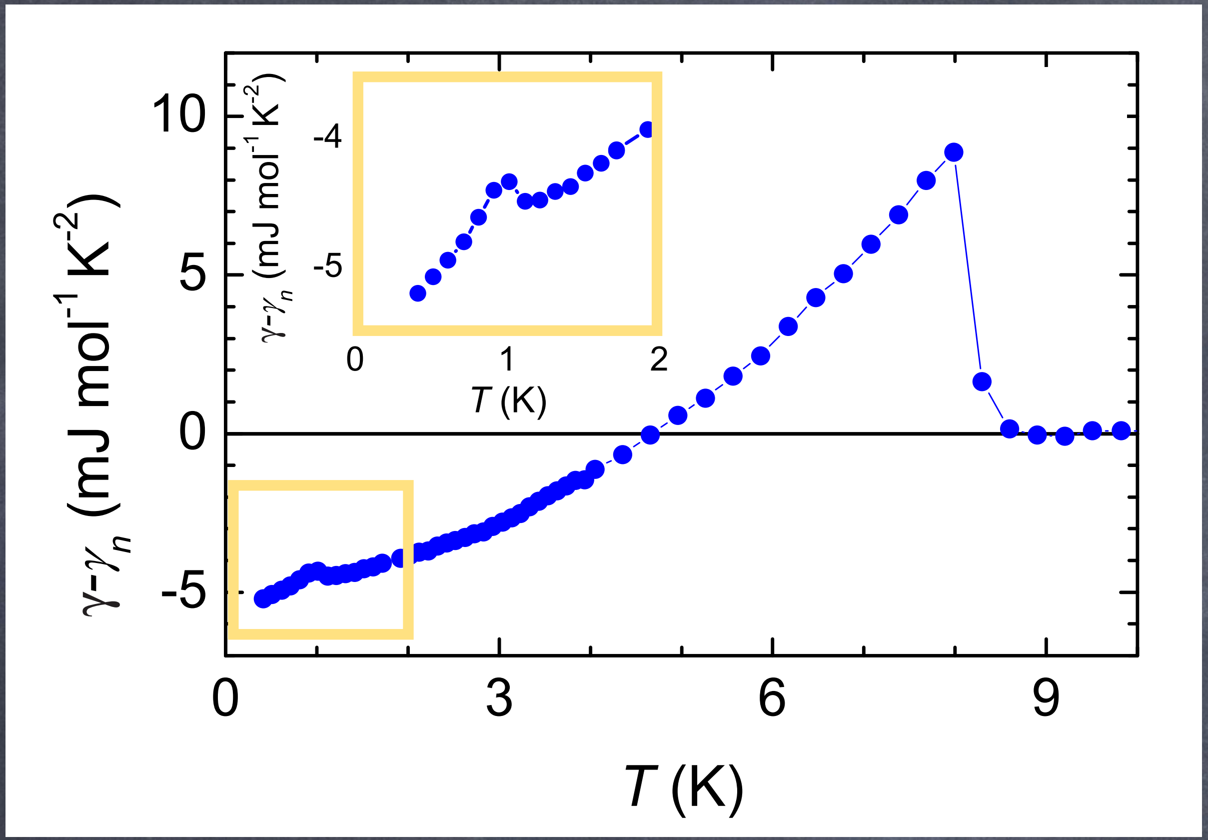

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The symmetry of the gap is still an open question

which can be probed by specific heat measurement

in **rotating** H||ab (Oxy plane)

DOS (=C) maxima when Doppler shift : $\overrightarrow{v_s} \,.\, \overrightarrow{k_F} \sim \Delta$

velocity of vortex screening current = rotating field

(anisotropy of) electronic structure

anisotropy of the superconducting gap

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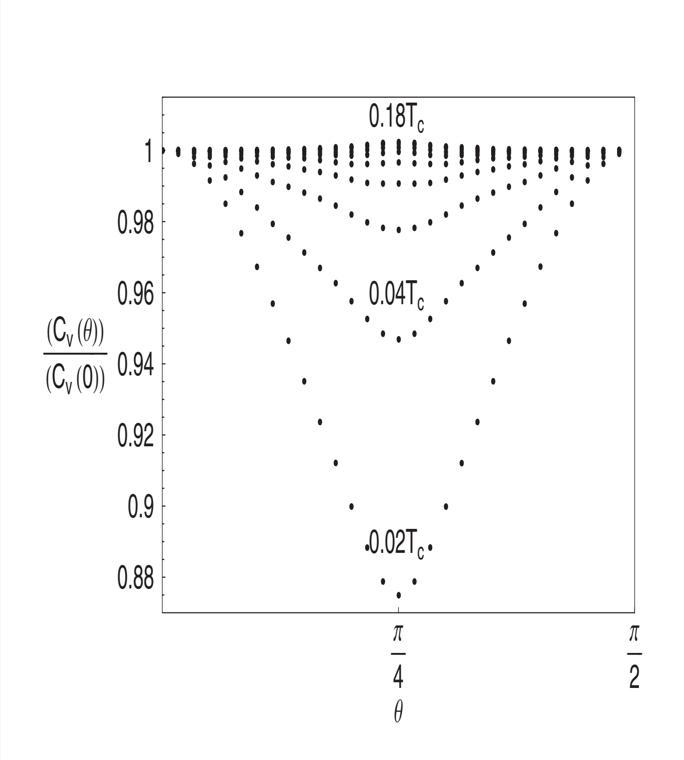

Numerical calculations
d-wave gap
G.R.Boyd *et al.* PRB 2009
(see also Vorontsov *et al.* PRB 2007)

However
$\Delta C/\gamma T < 1\,\%$
above $T_c/10$

and $\pi/4-$shift
(min $\Longleftrightarrow$ max)
for $T \sim T_c/10$

DOS (=C) maxima when Doppler shift : $\vec{v_s} \cdot \vec{k_F} \sim \Delta$

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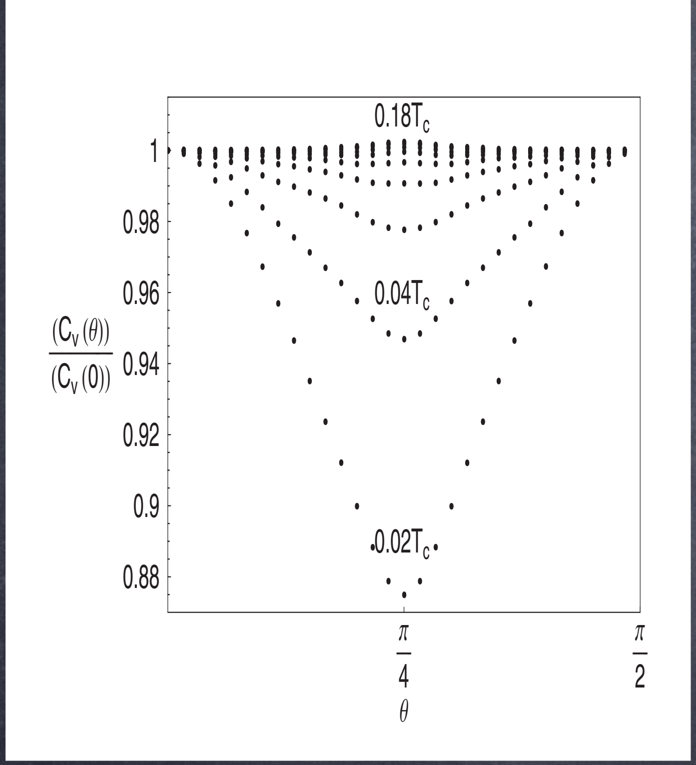

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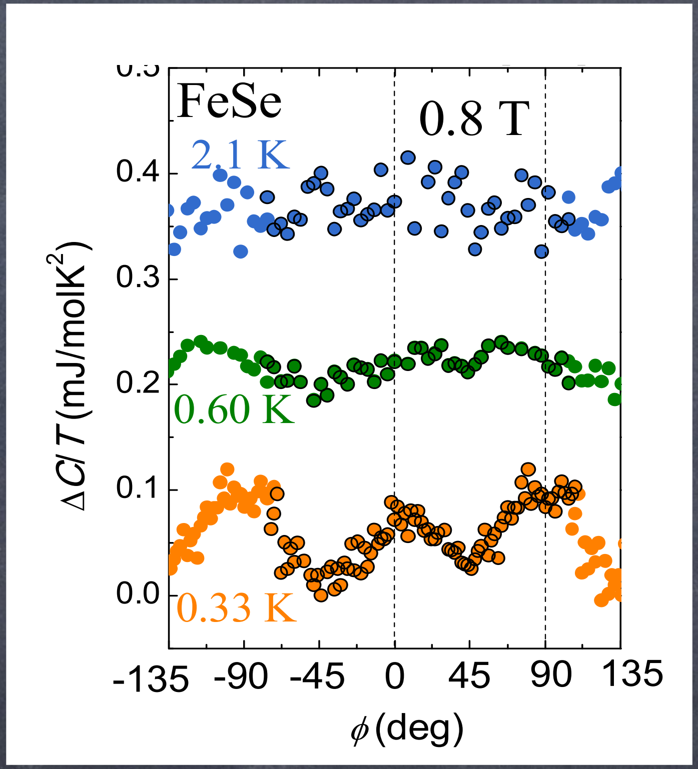

Observed in FeSe

but only low T/H

Sun *et al.* PRB 2017

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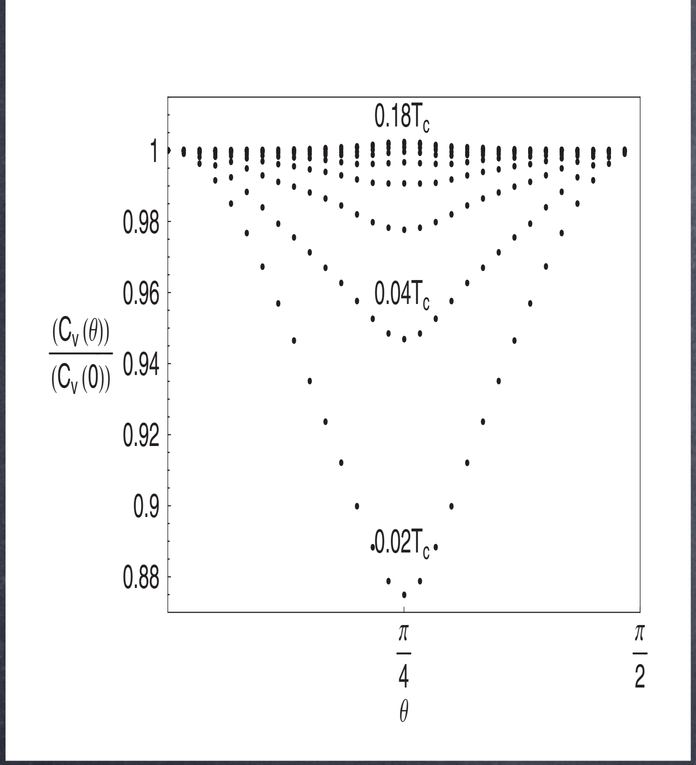

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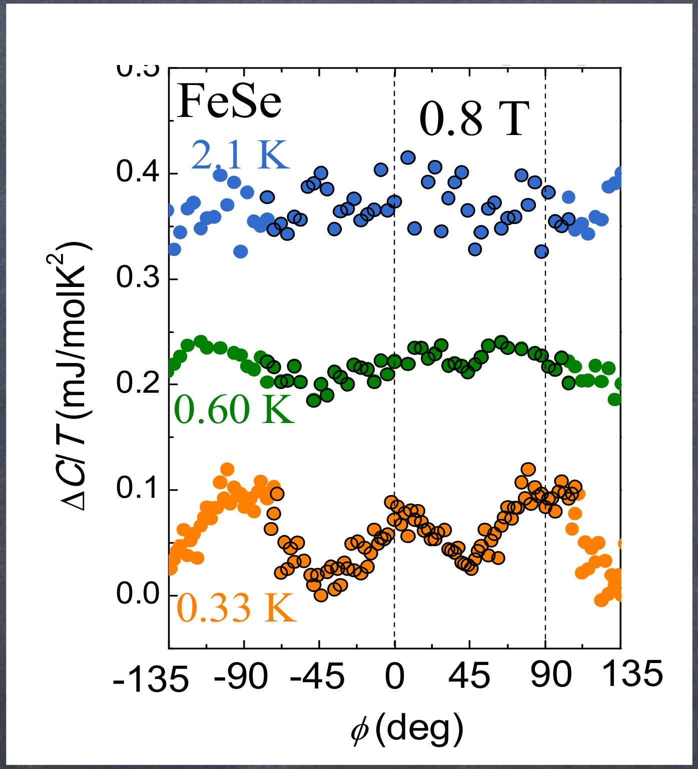

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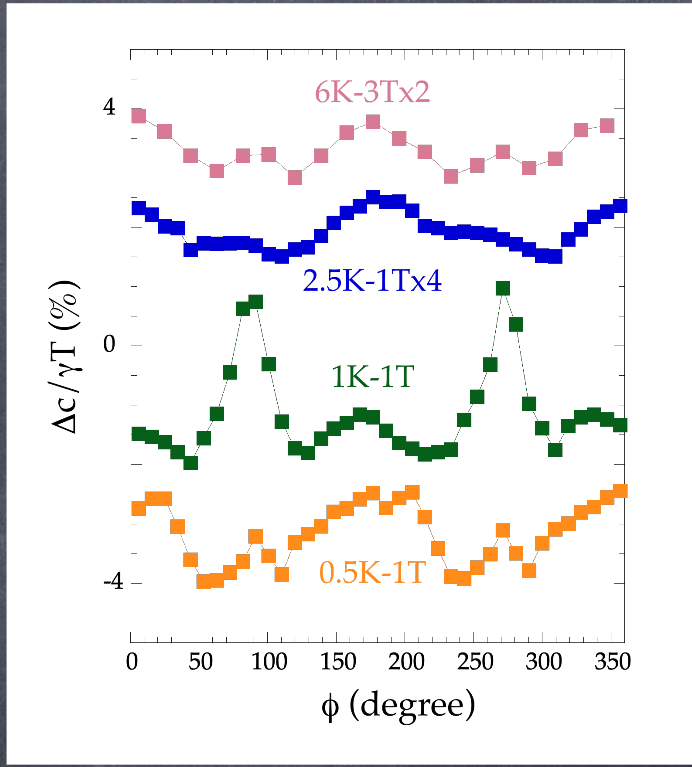


$\Delta C/C$

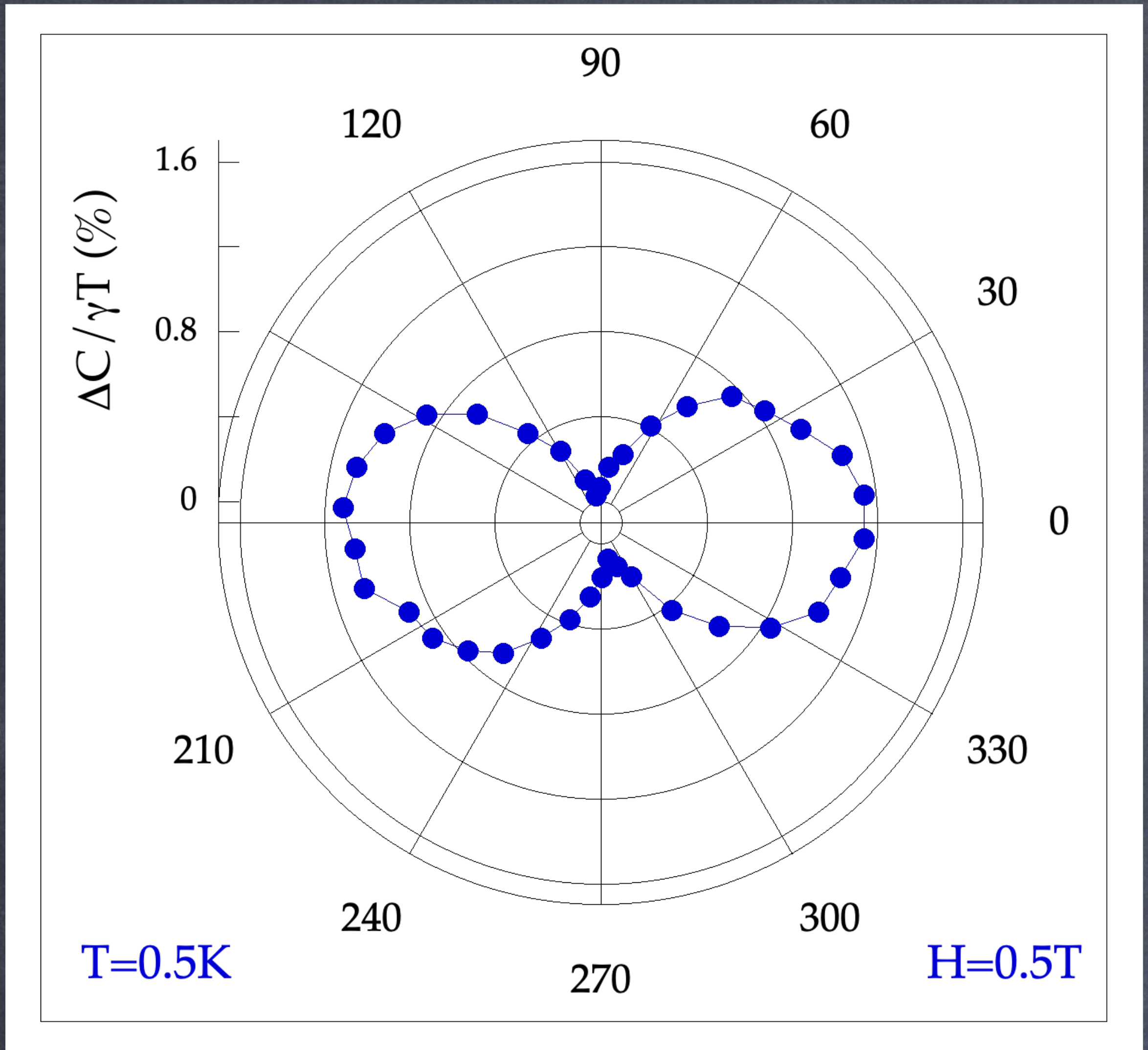
0.06 %

3 %

Observed in FeSe

up to 6K and/or 6T

AC measurements

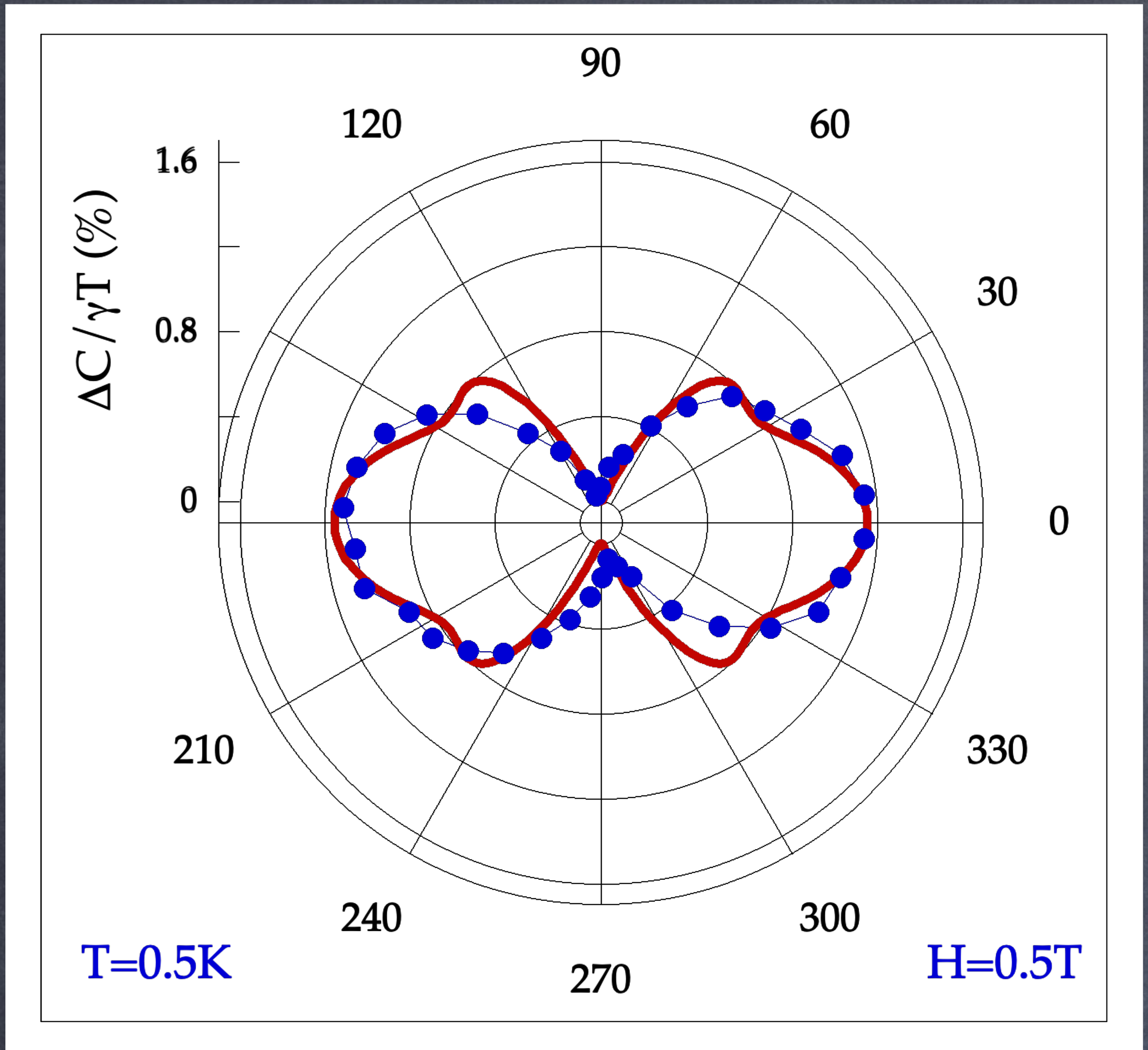


twofold (C2) symmetry

at low T/low H

= nematic character of FeSe

from thermodynamic data



twofold (C2) symmetry

at low T/low H

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Good **quantitative** agreement

with numerical calculations

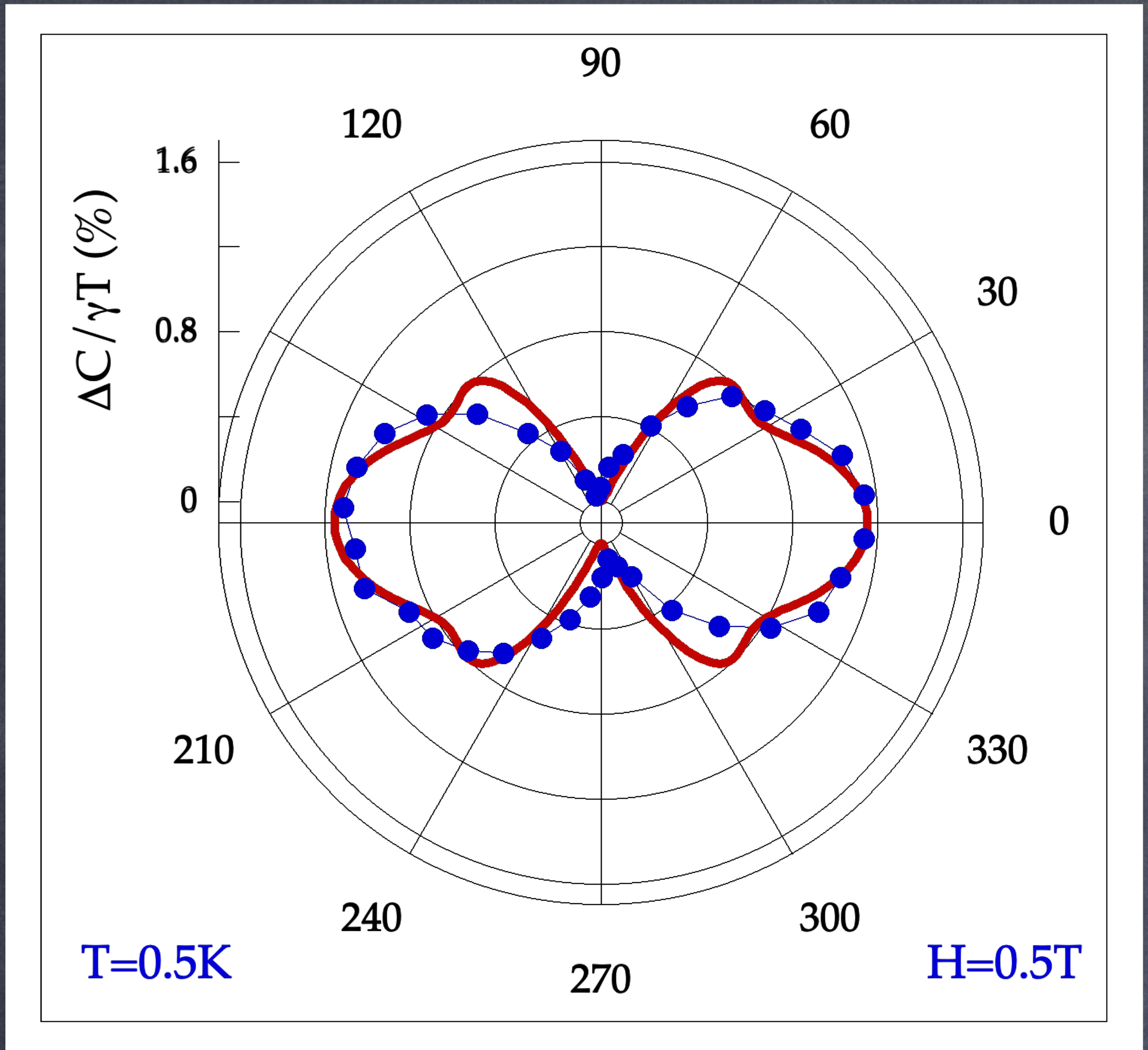
(H.Cercellier)

$\Delta_{\mathrm{hole}}^{\mathrm{max}} \sim 2\mathrm{meV}$

$\Delta_{\mathrm{elec}}^{\mathrm{max}} \sim 1\mathrm{meV}$

in very reasonable agreement with other measurements

H.Cercellier *et al.*, PRB 2019



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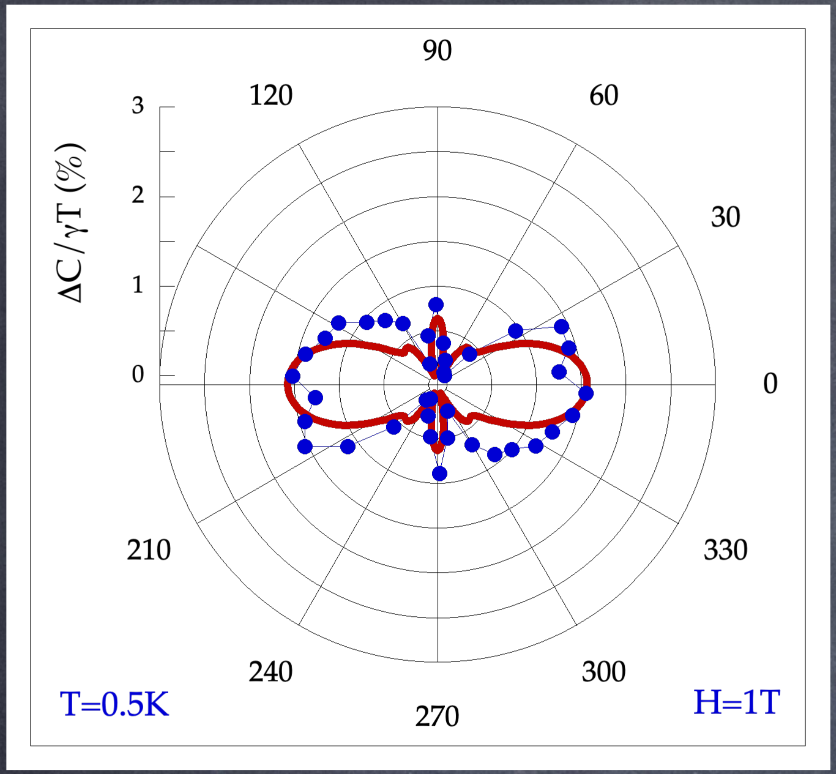
but *exotic* temperature and field dependence of

the gap structure !...

increasing temperature



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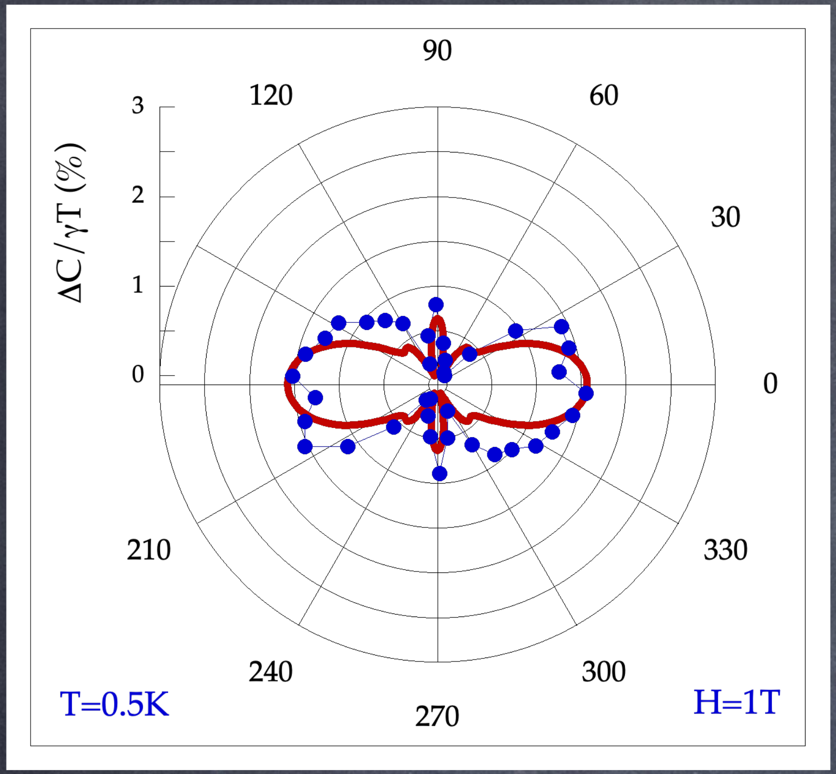

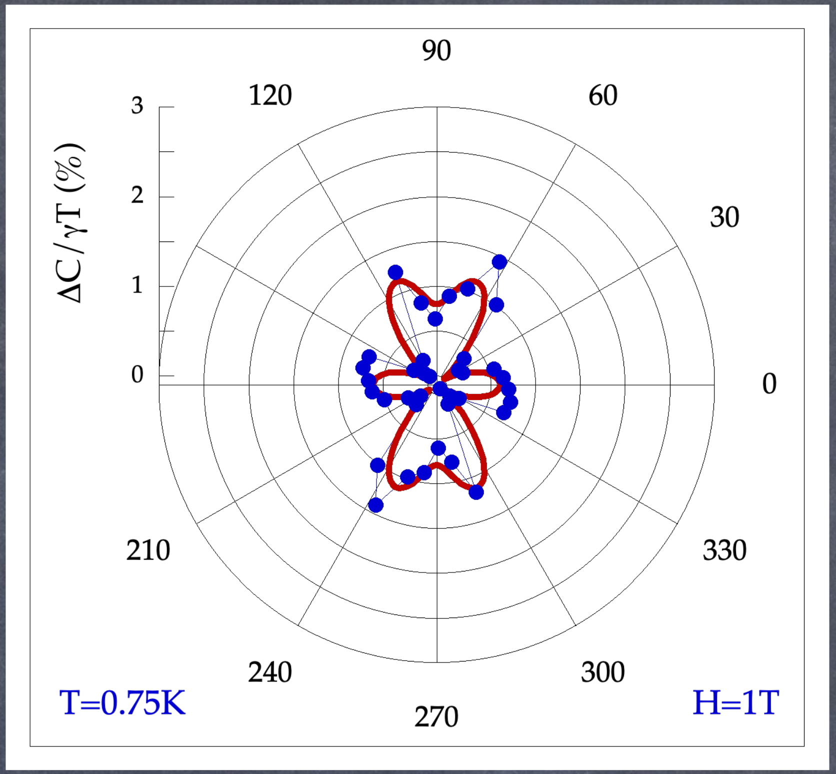

intriguing «butterfly» structure

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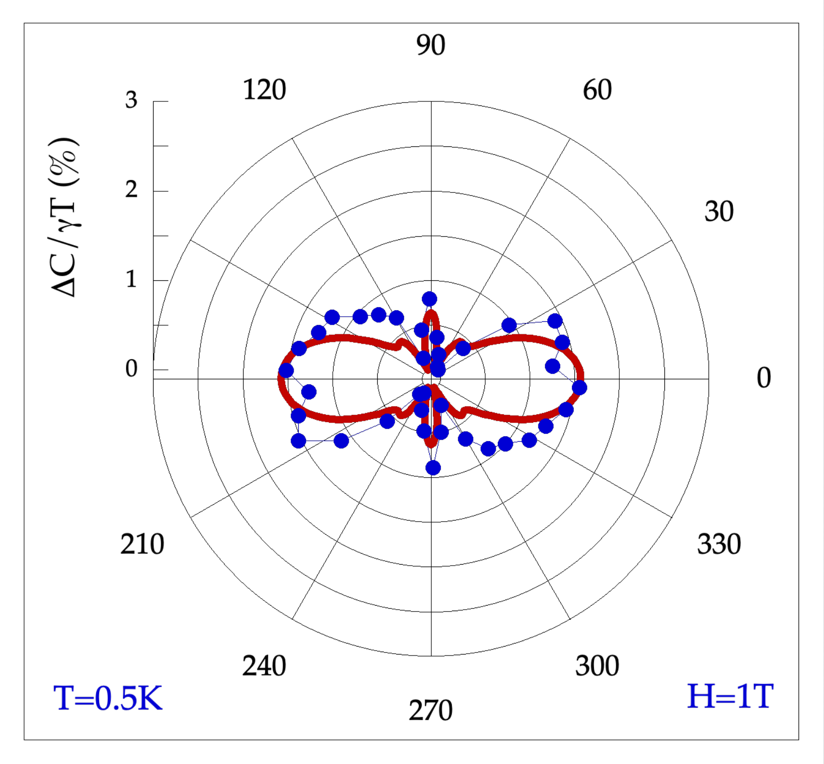

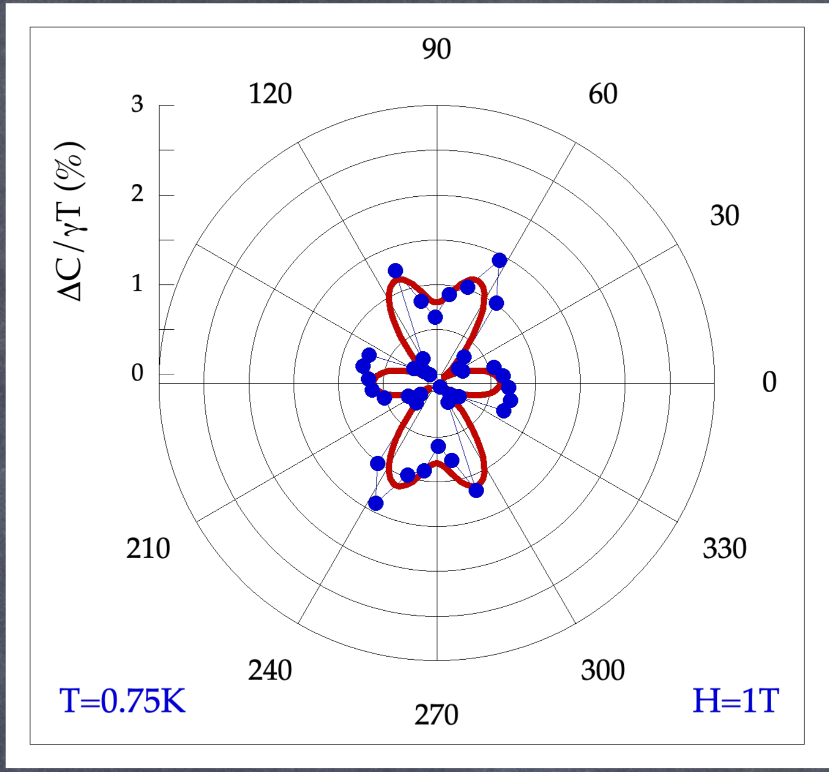

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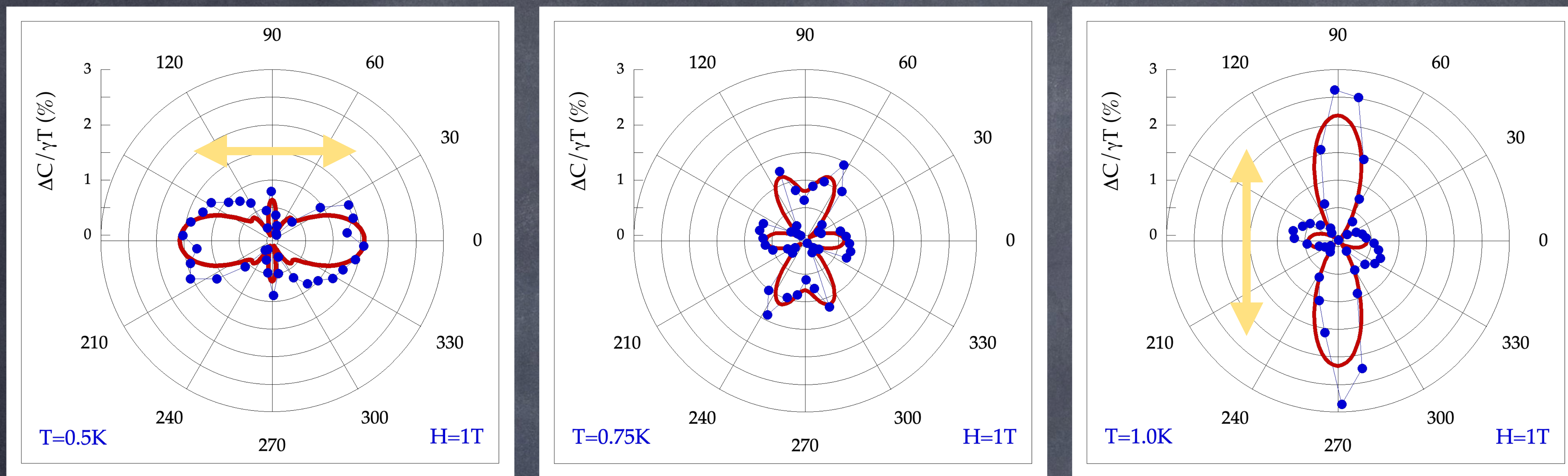
characteristic of accidental nodes

$\Delta_{hole}^{min} \sim \Delta_{elec}^{min} \sim -0.2meV$

increasing temperature



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intriguing «butterfly» structure

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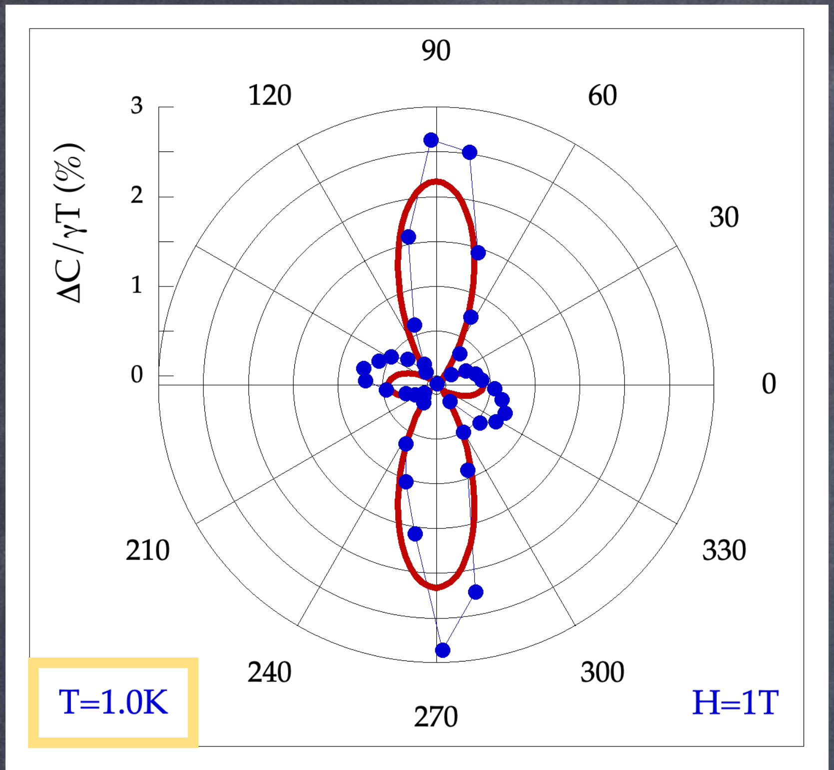
$\Delta^{min}_{hole} \sim \Delta^{min}_{elec} \sim -0.2meV$

$\pi/2-$shift of the symmetry axis (for $T \sim T_c/10$) also well reproduced

increasing temperature



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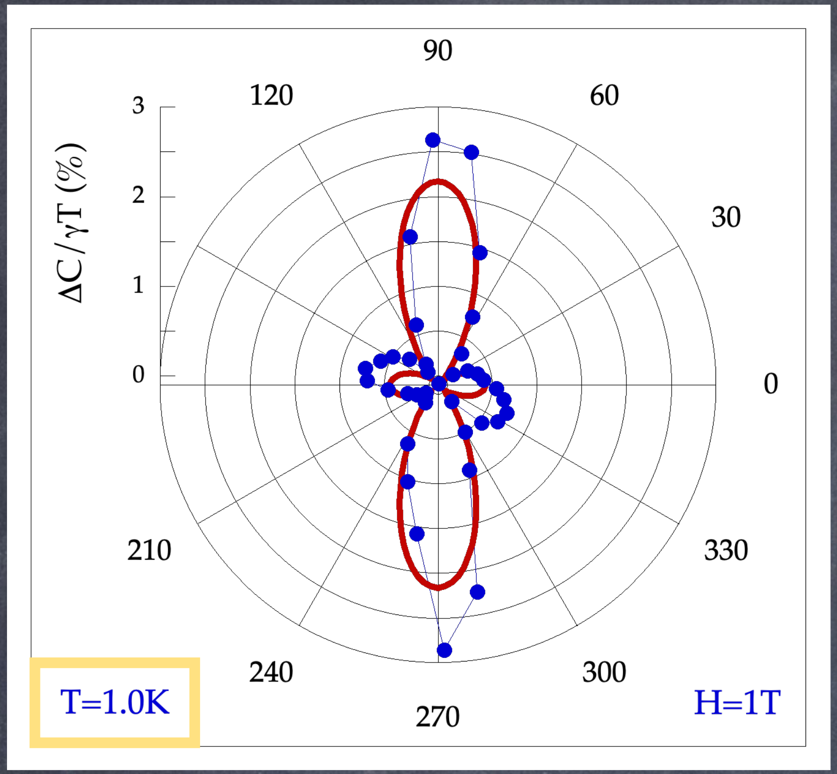


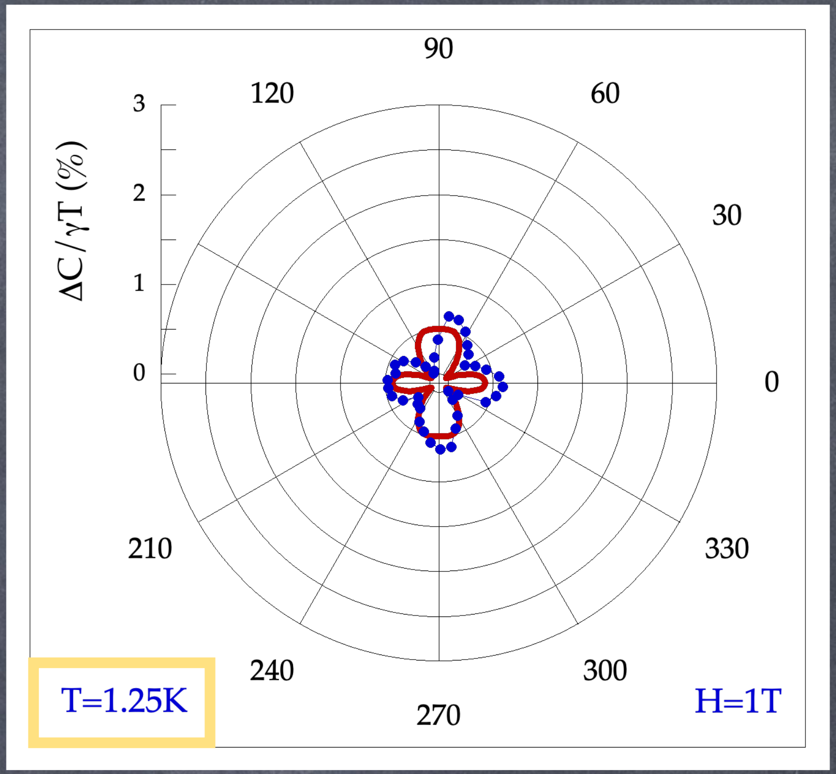
but

drastic drop

of the

amplitude

above 1K ?



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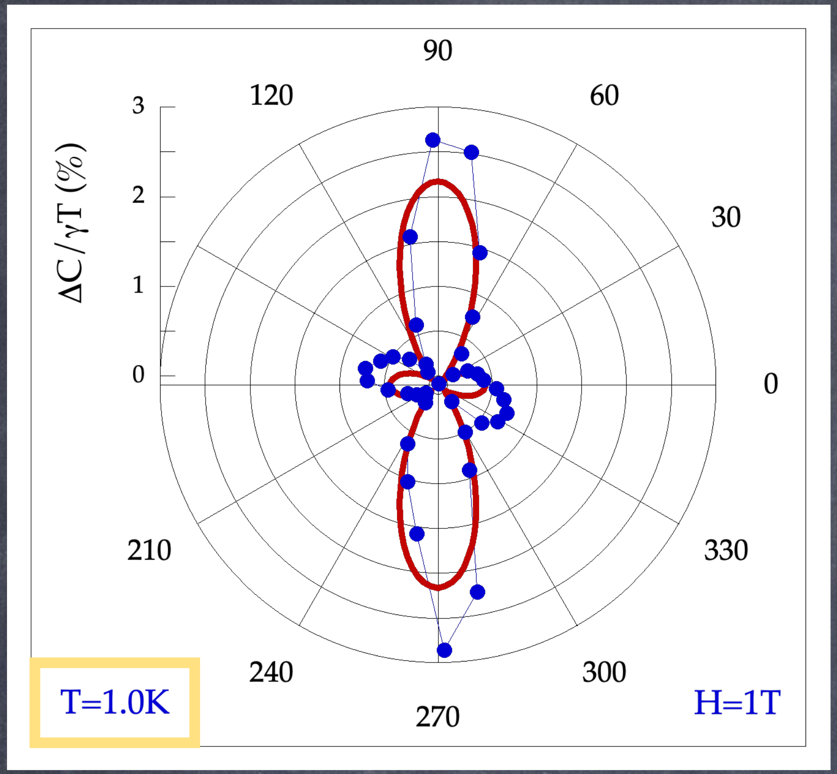

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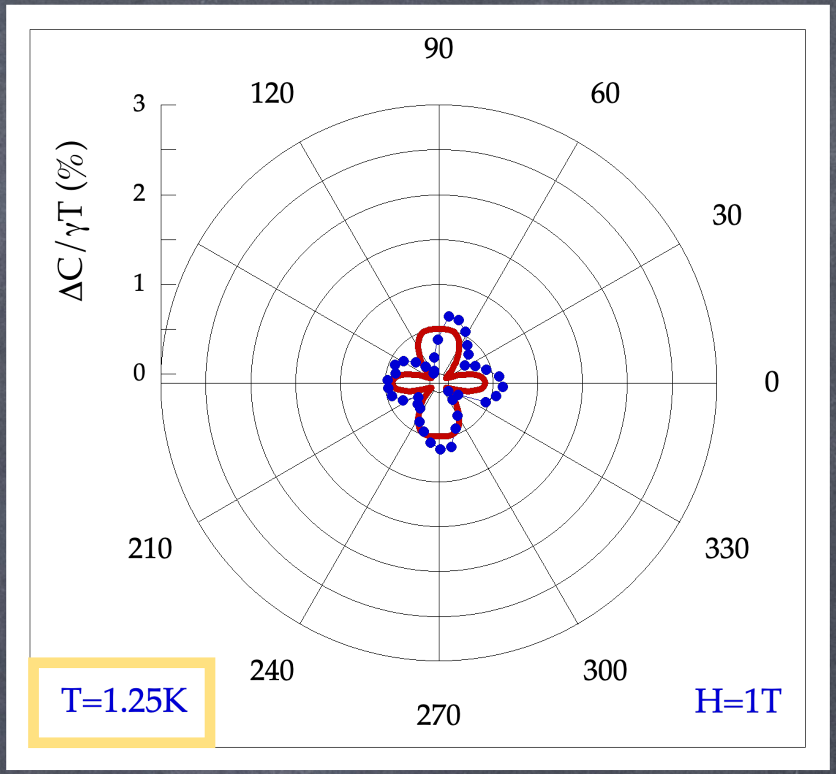

Sudden change in the gap structure

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→

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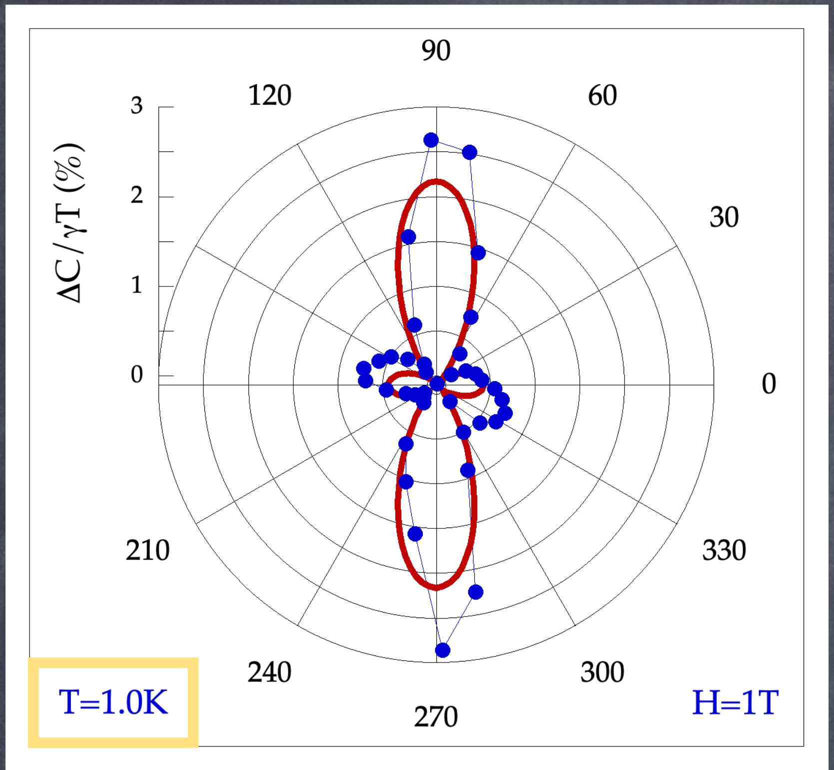

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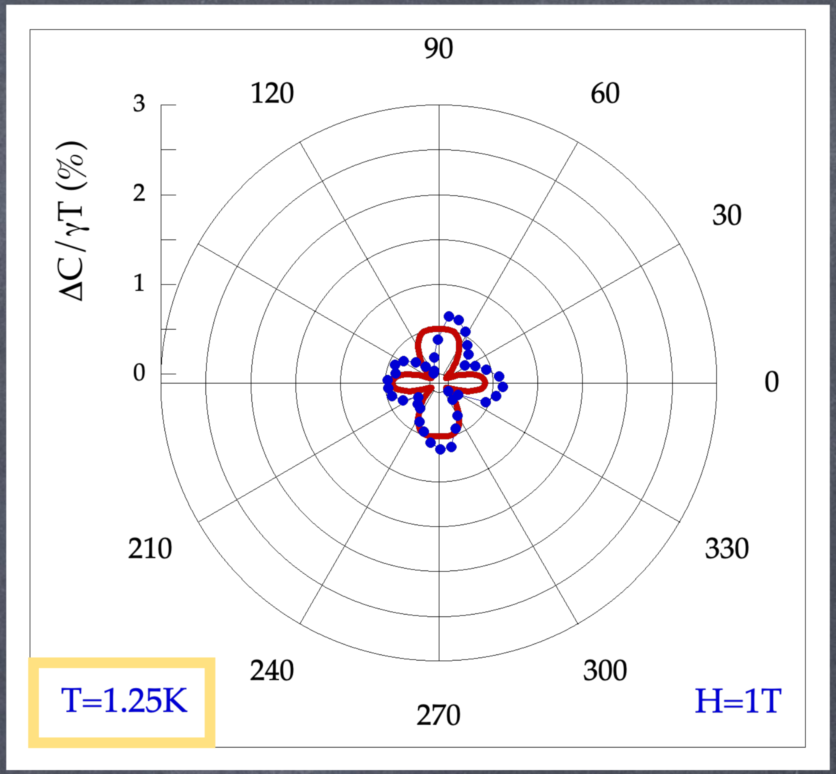

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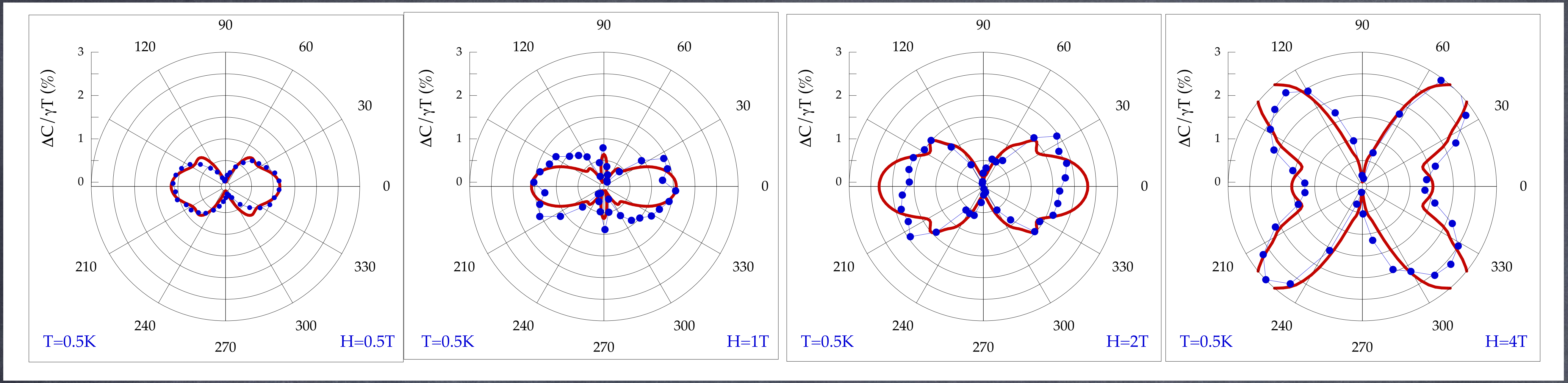
... leading to the observed specific anomaly around 1K increasing field

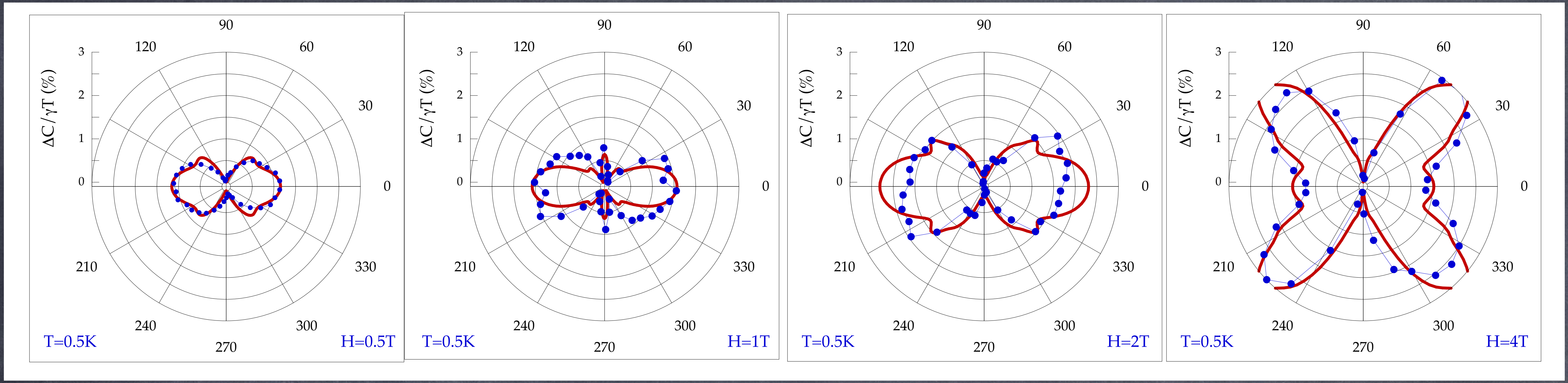


and lobes develop
around 45°
above ~4T

field dependent gap amplitude

$\Delta^{max}_{hole} \rightarrow \Delta^{max}_{elec} \rightarrow 1.5\mathrm{meV}$

increasing field



and lobes develop
around 45°
above ~4T

field dependent gap amplitude

$\Delta^{max}_{hole} \rightarrow \Delta^{max}_{elec} \rightarrow 1.5meV$

This T and H dependence of the gap structure still has to be understood

(not observed in all samples...)

H.Cercellier *et al.* submitted to PRL (2024)

Outline

- **The FeSe nematic superconductor**
  H-T phase diagram and T & H dependence of the gap structure

- **Thermodynamic properties of the normal state in cuprates**
  Quantum criticality at the onset of the pseudo-gap and charge order

part of the PhD work of **B.Michon & C.Girod**

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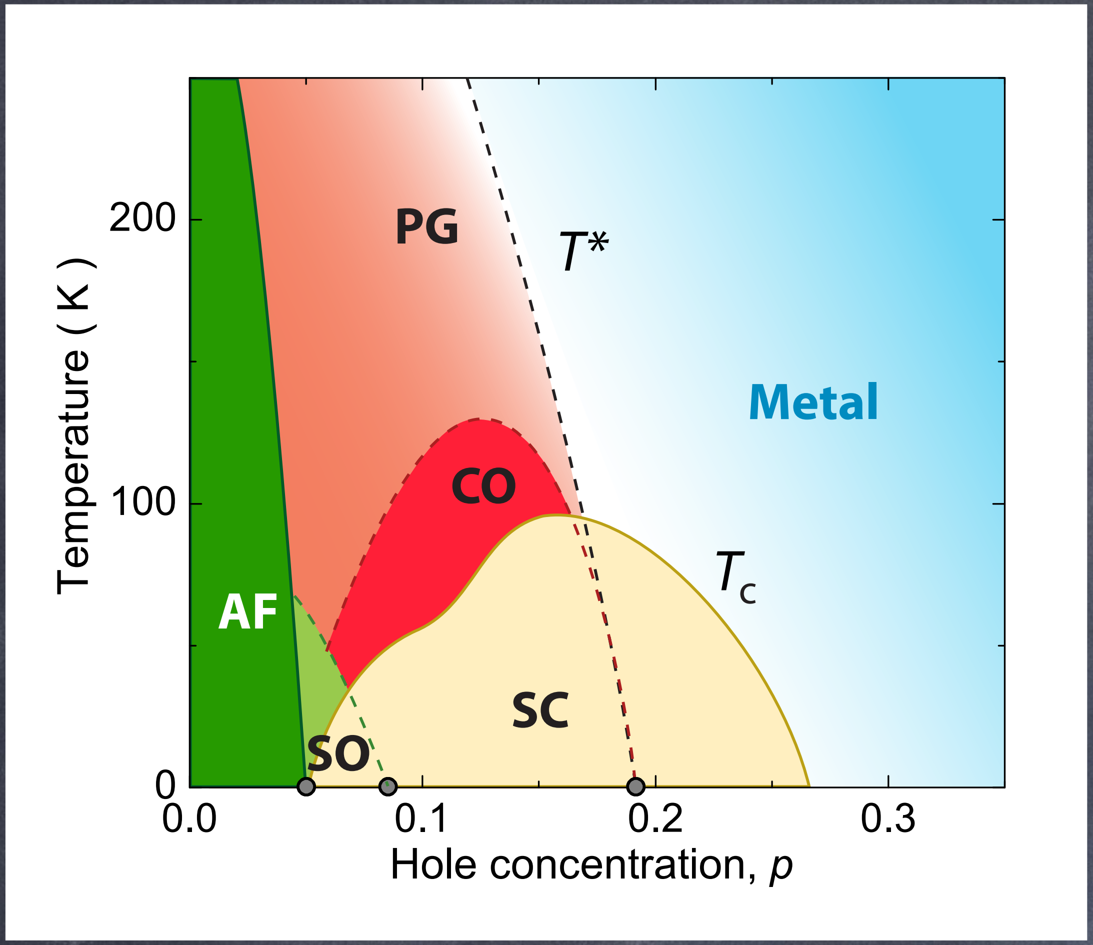

YBaCuO, Courtesy N.Doiron-Leyraud, Univ. of Sherbrooke

Cuprates

very complexe phase diagram

CO=charge order
SO=spin order
PG= *mysterious* pseudo-gap phase
SC=superconductor

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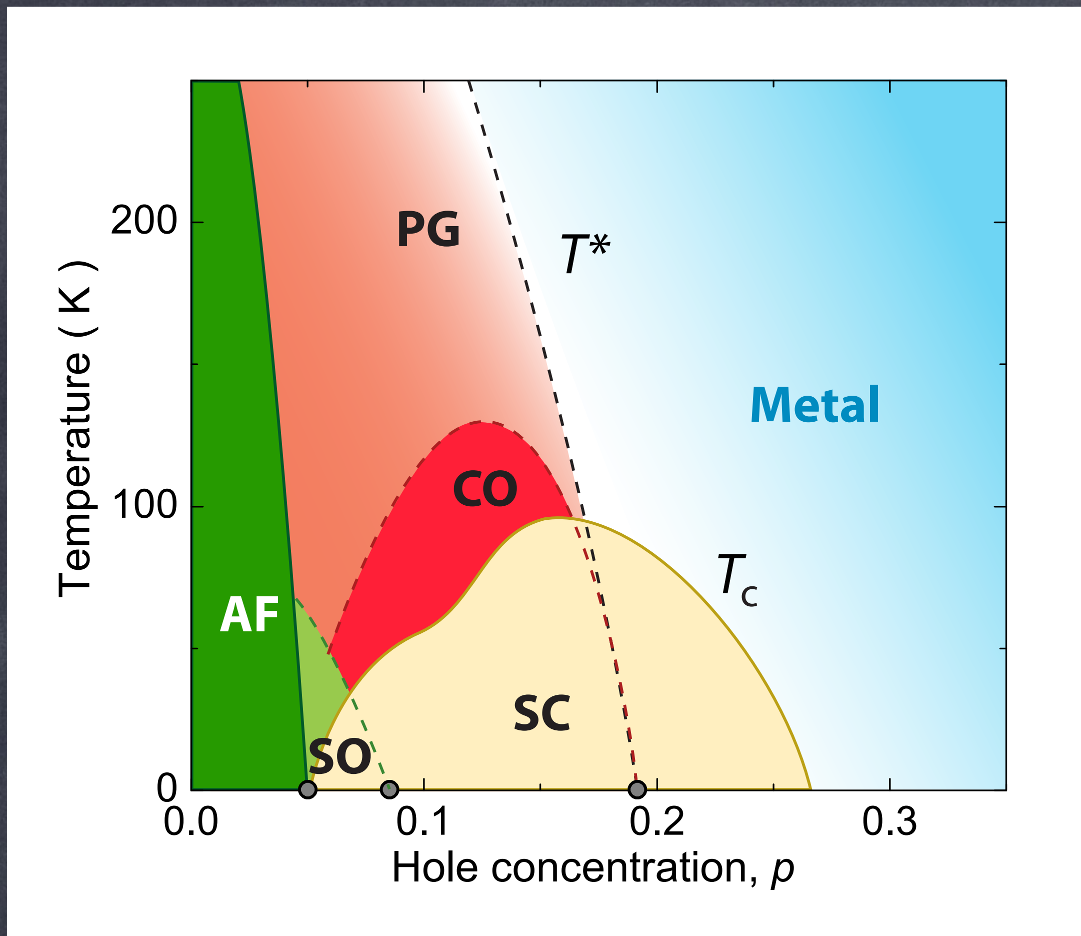
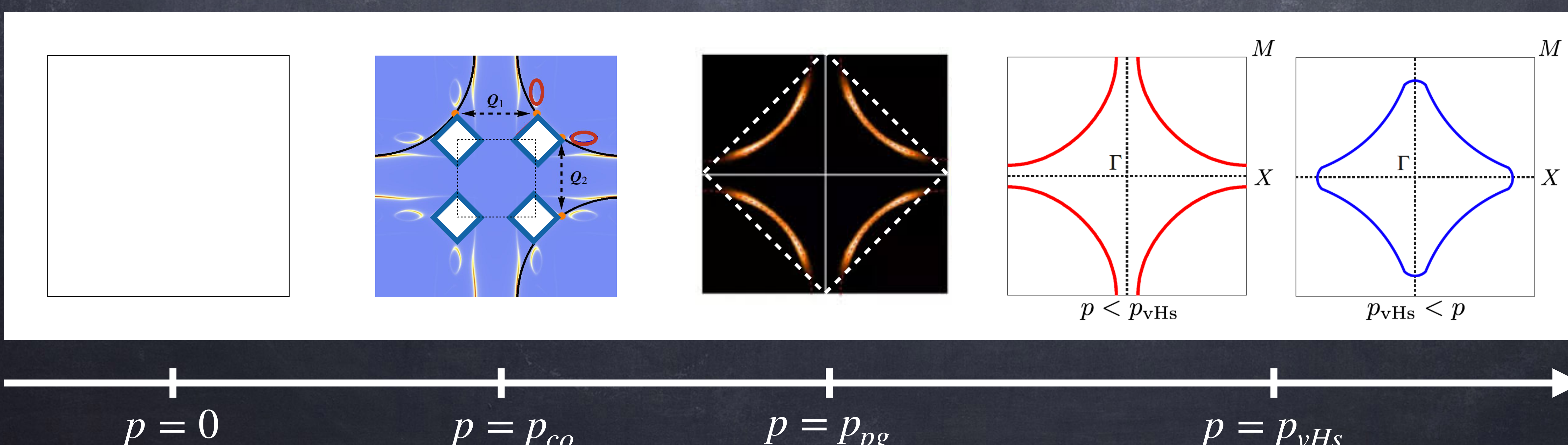


YBaCuO, Courtesy N.Doiron-Leyraud, Univ. of Sherbrooke

CO=charge order
SO=spin order
PG= *mysterious* pseudo-gap phase
SC=superconductor

associated with major changes in the topology of the Fermi surface

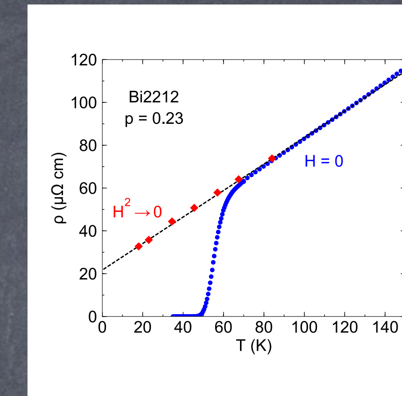


Let's start with the pseudo-gap phase…

Linear temperature dependence

of the resistivity : so-called *Planckian* diffusion

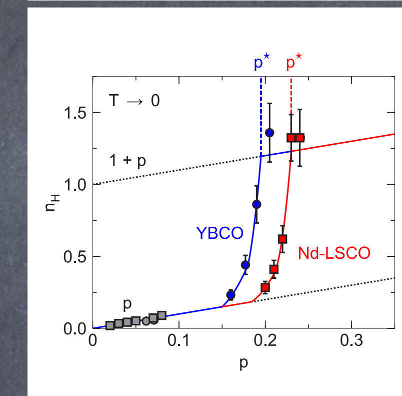
C. Collignon et al., PRB 2017, A. Legros et al., Nature 2019



Drop in the carrier density from n=1+p to n=p

consistent with (AF) reconstruction of the FS

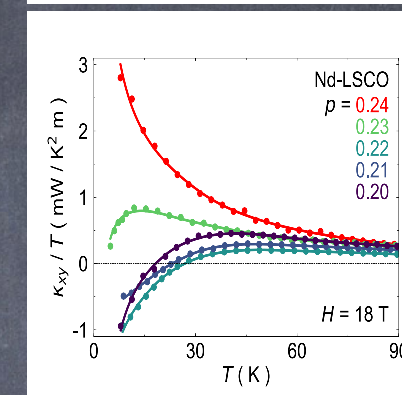
C. Collignon et al., PRB 2017, S. Badoux et al., Nature 2016

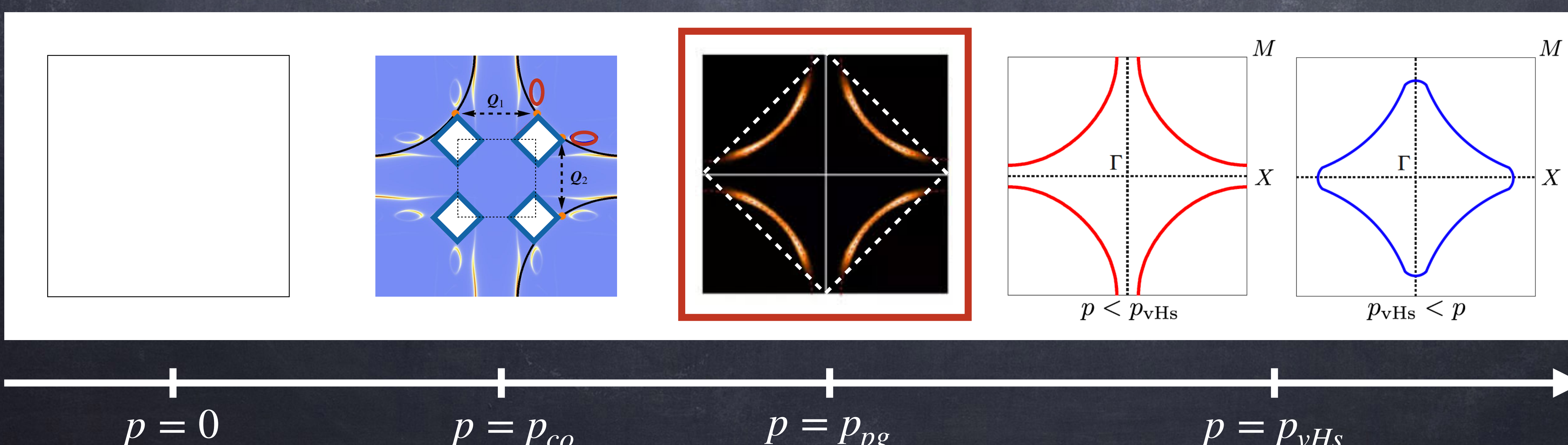


Giant negative thermal Hall effect

from p_{pg} down the Mott insulator (p=0)

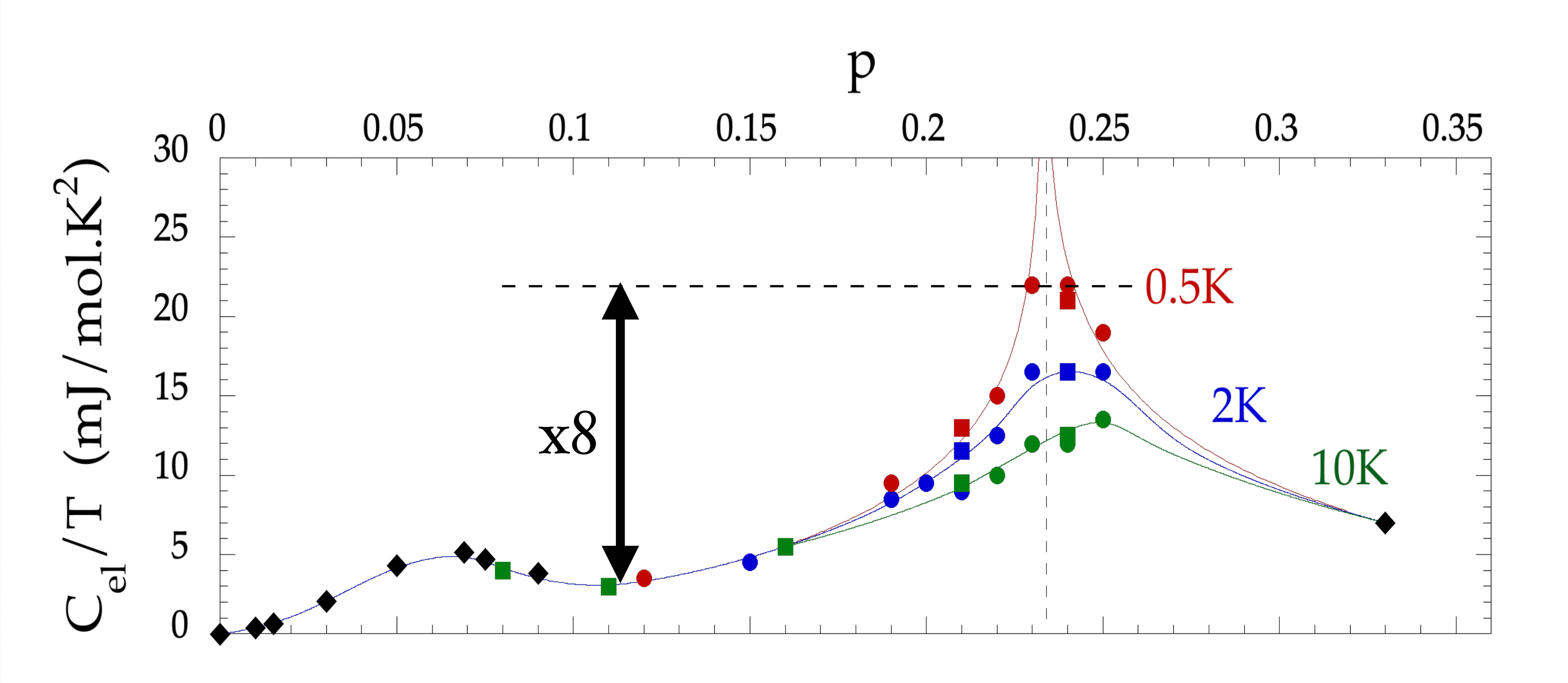
G. Grissonanche et al., Nature 2019 & 2020, M-E. Boulanger et al., Nat. Com 2020





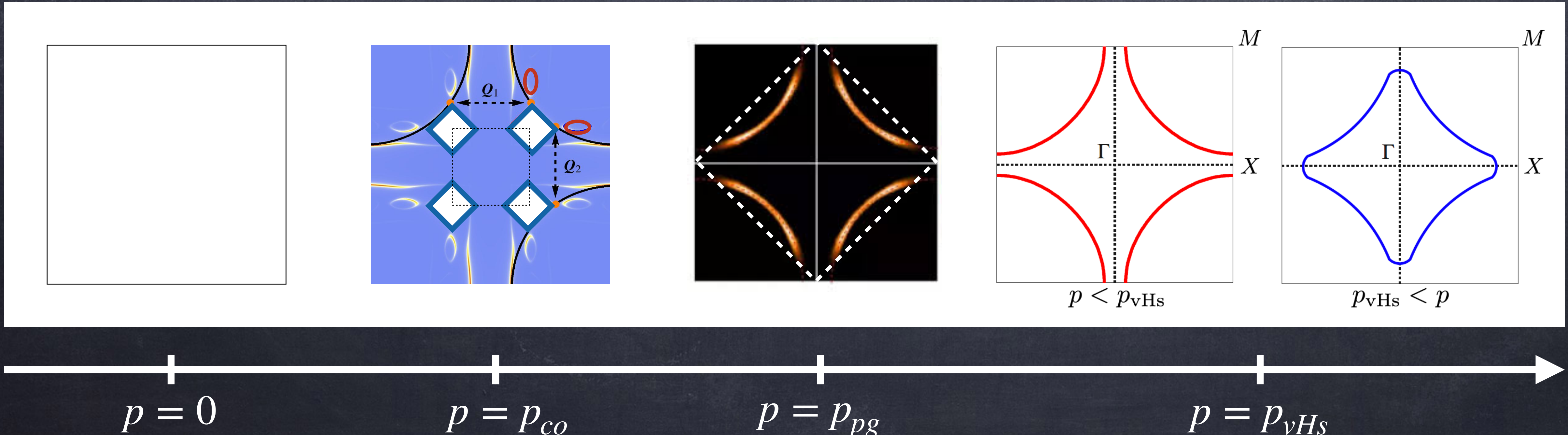
We have shown that the onset of the PG is also associated with **pronounced peak** of the Normal state specific heat for $T \to 0$

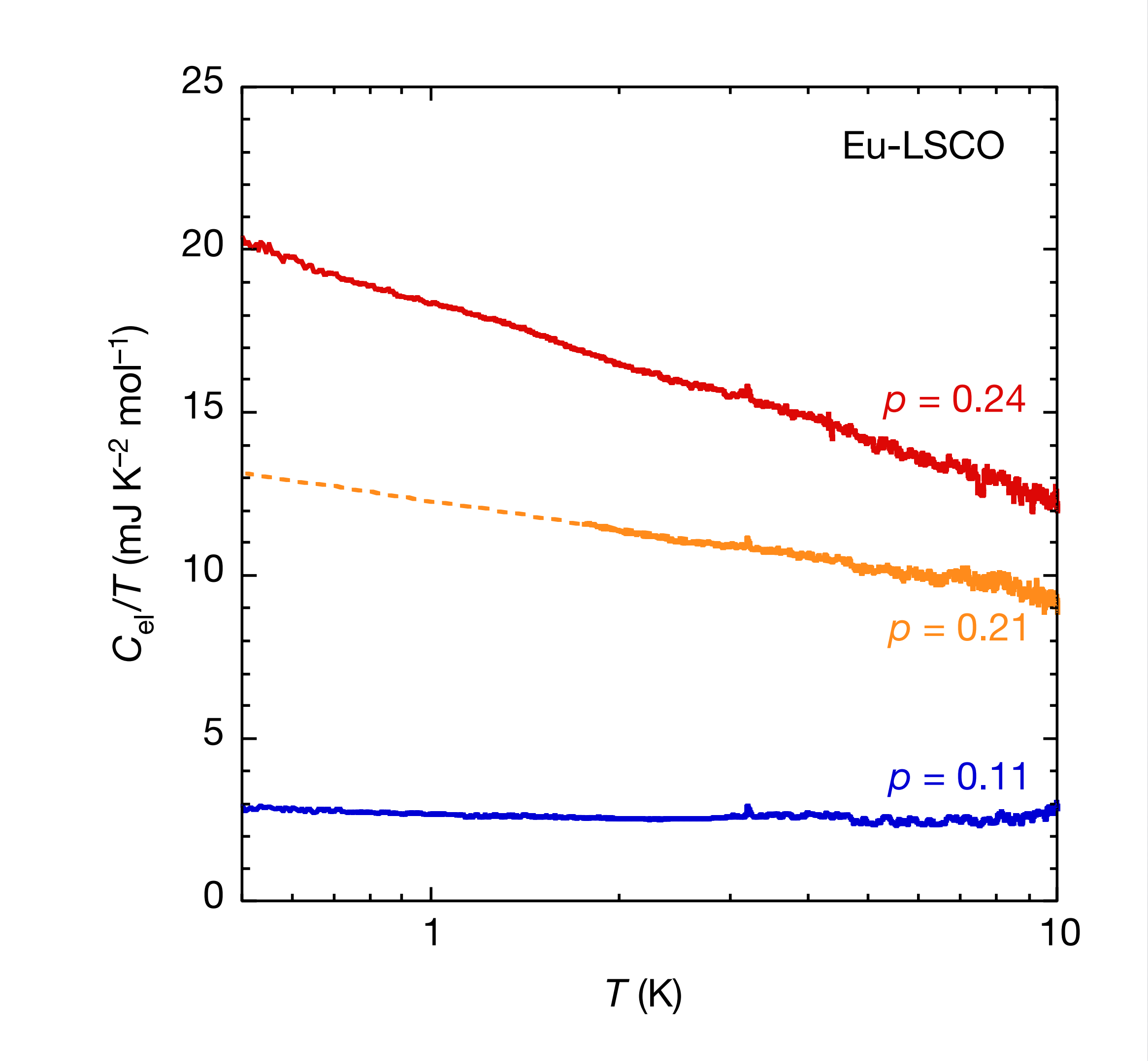
(aas mentioned yesterday by Chandra Warma)



Nd/Eu-LSCO :
$T_c < 15K$
$H_{c2}(0) < 18T$

B.Michon *et al.* Nature 2019

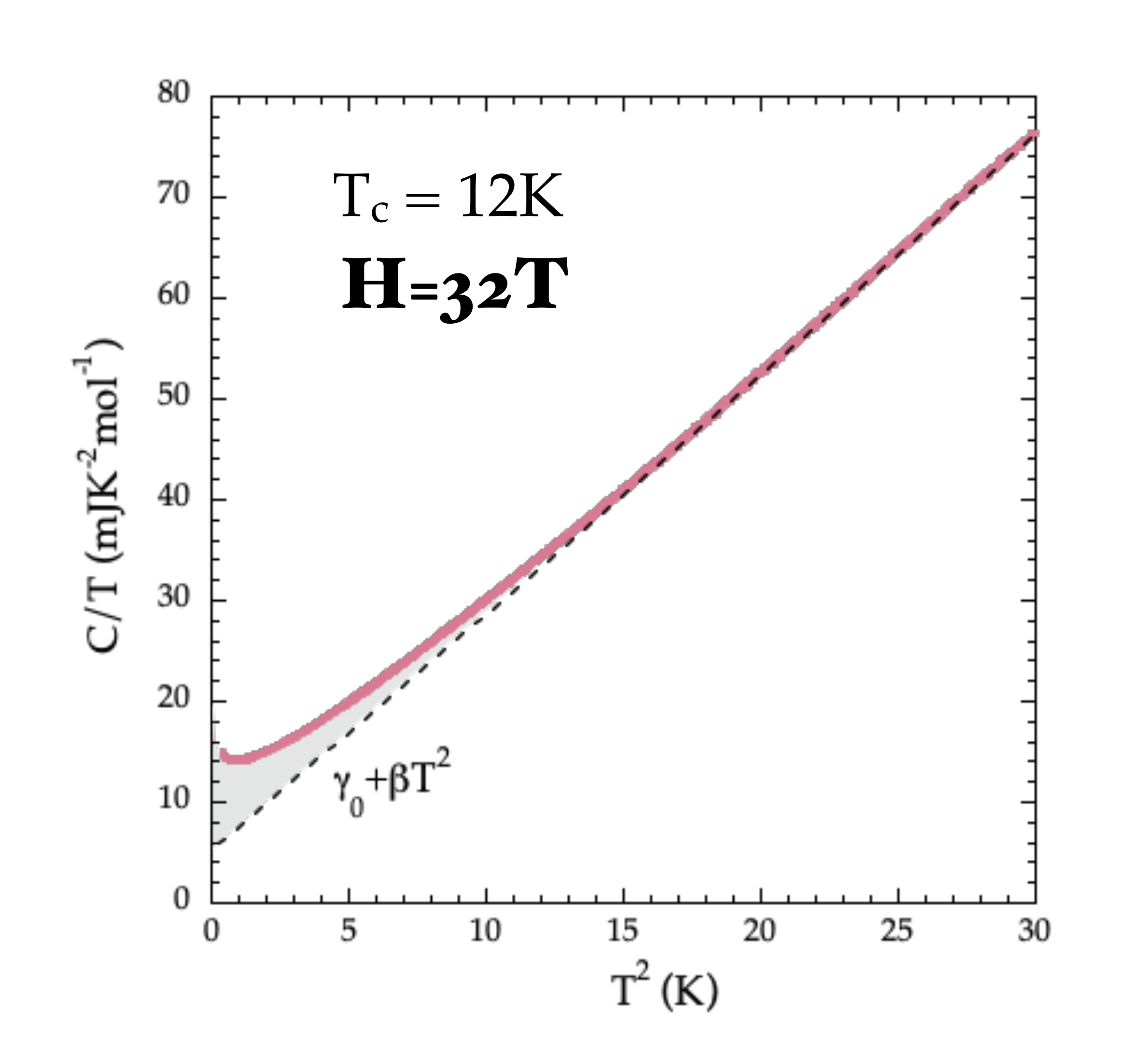




associated with a $LogT$ dependence of C_{el}/T for $p \rightarrow p_{pg} \sim 0.24$

(here $C_{el}/T = C/T - \beta T^2$)

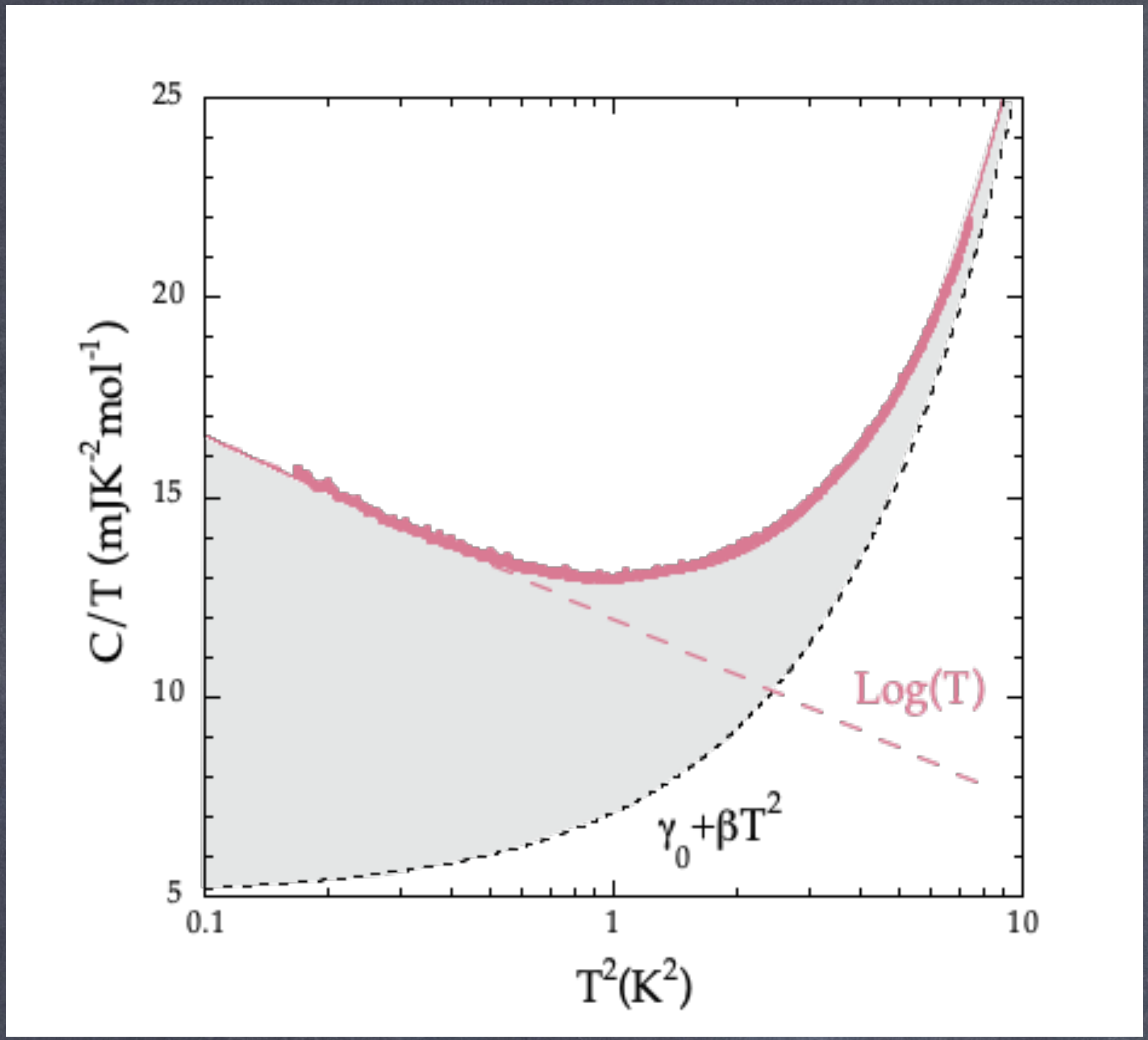
Michon *et al.* Nature 2019



clear deviation from the standard temperature dependence also observed in

$Bi_{2+y}Sr_{2-x-y}La_xCuO_{6+\delta}$

C.Girod *et al.* PRB 2021



(again) very good fit
to the data with
$C/T = \beta T^2 + BLog(T/T_0)$

clear deviation from the standard temperature dependence also observed in

$Bi_{2+y}Sr_{2-x-y}La_xCuO_{6+\delta}$

C.Girod *et al.* PRB 2021

LnT contribution to the specific heat pour $p \sim p_c$	strong C/T peak for $p \to p_c$ at low T

= *classic* thermodynamic signature of a $z = d$ **Quantum Critical Point**

BUT…**what is the diverging correlation length ?**

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strong C/T peak for $p \to p_c$ at low T

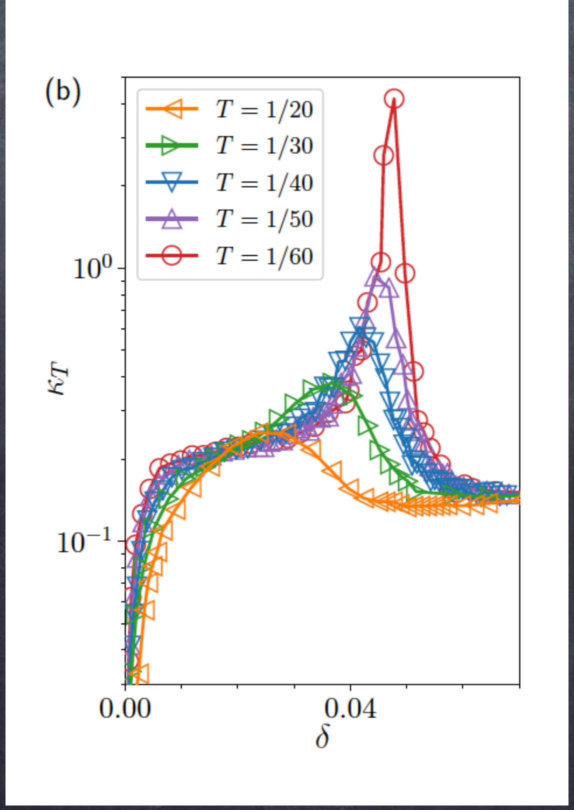
= *classic* thermodynamic signature of a $z = d$ **Quantum Critical Point**

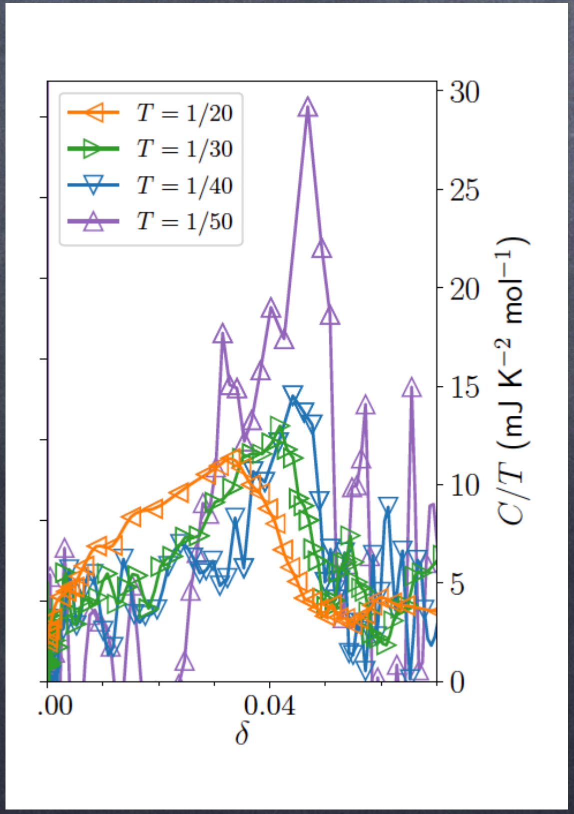
BUT…**what is the diverging correlation length ?**

cluster-DMFT calculations in the t-U model (correlated systems)

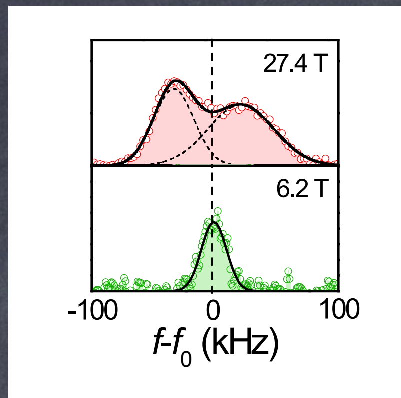
G.Sordi *et al.* Sc. Reports 2012, 2010

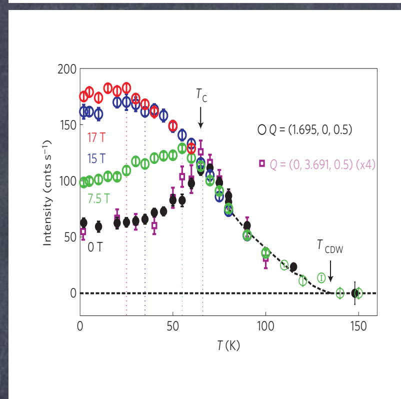
T dependent peak in the electronic compressibility $K_T = 1/n^2(dn/d\mu)_T$ (diverging at the critical point)

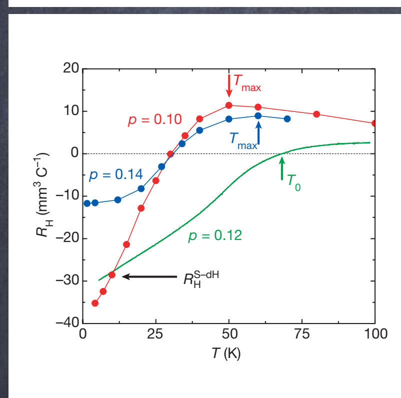




AND in the specific heat ! **(as observed)**







and a few words on the Charge Order...

Splitting of the NMR lines

modulation of the charge density

T.Wu et al. Nature 2011

X-ray diffraction peaks

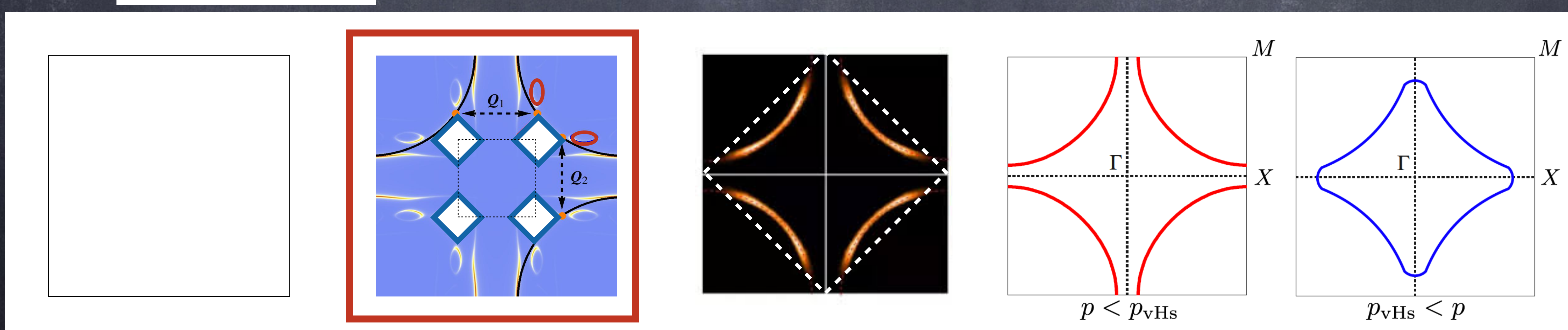
⇒ Long range CDW at high field/low T

J.Chang et al. Nature Physics 2012, D.Leboeuf et al. Nature Physics 2013 (and more)

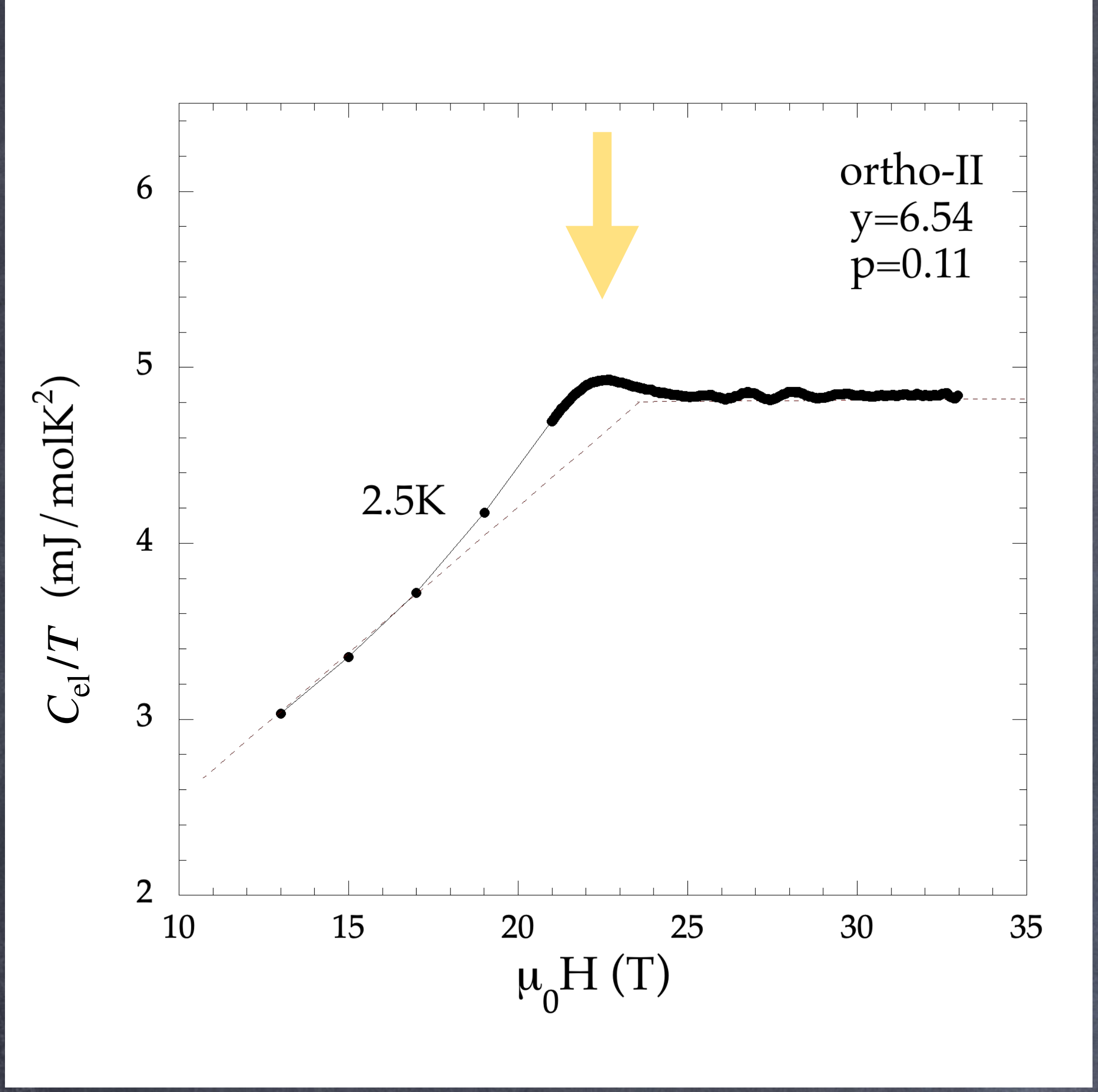
Change of sign of Hall effect

+ low frequency quantum oscillations (FS reconstruction) & reduced H_{c2}

D.Leboeuf et al. Nature 2007, Nicolas Doiron-Leyraud et al. Nature 2007,...

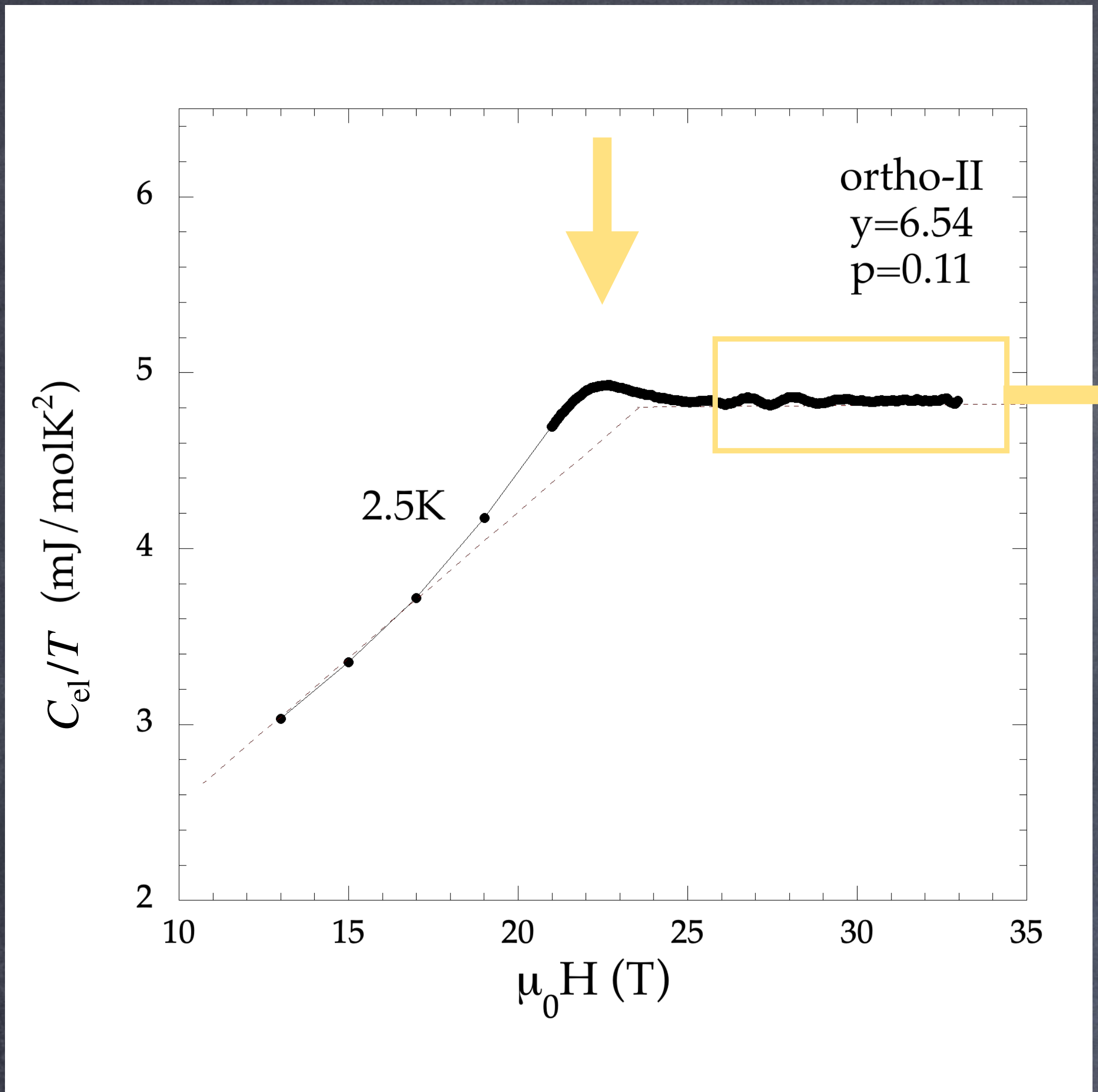


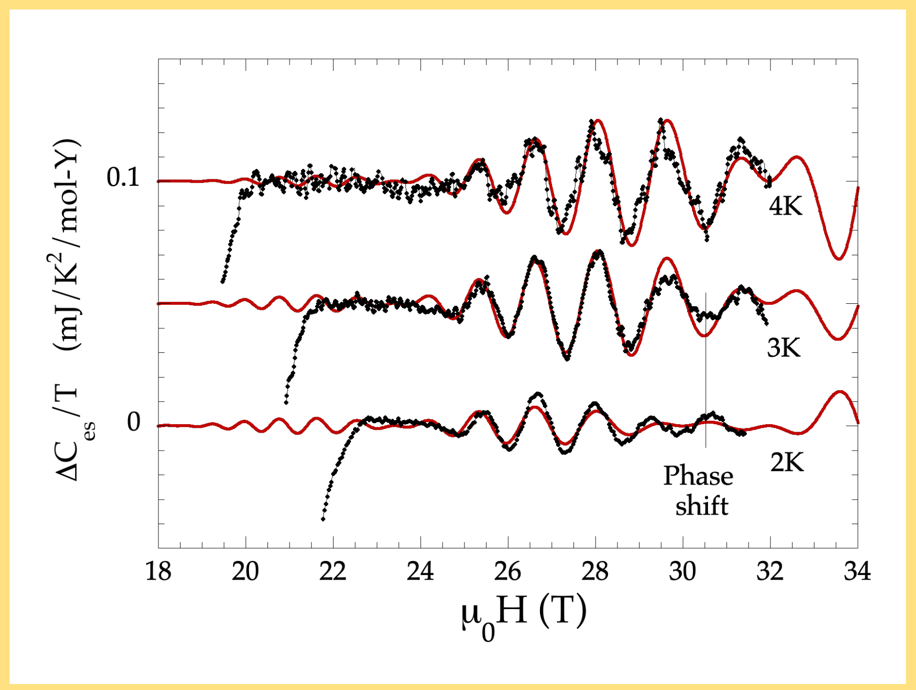
$p = 0$ $p = p_{co}$ $p = p_{pg}$ $p = p_{vHs}$



C/T saturates for $H > H_{DOS}$

(not H_{c2} due to thermal fluctuations)





Note the presence of well resolved quantum oscillations in good agreement with **small electron pockets**

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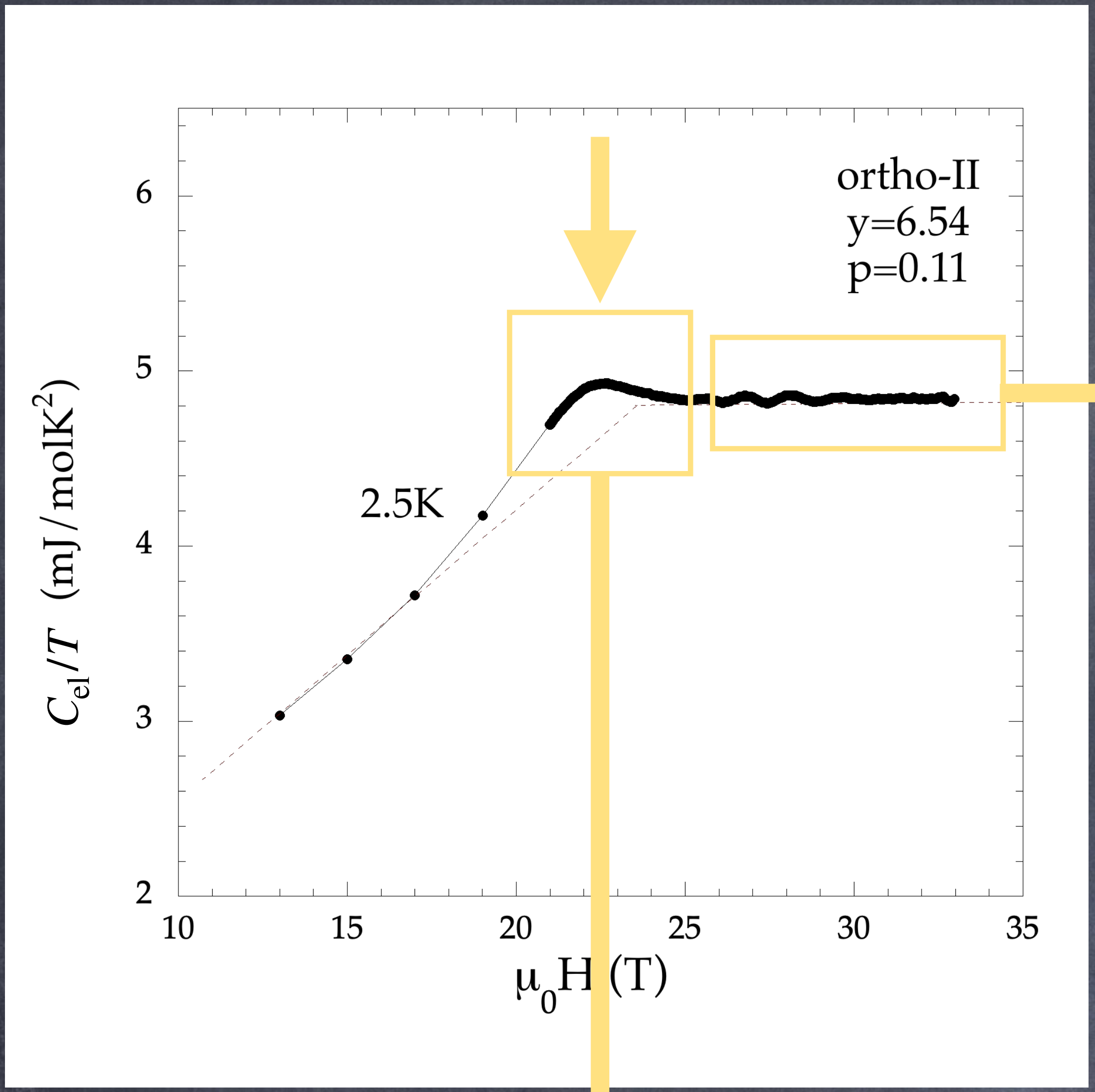

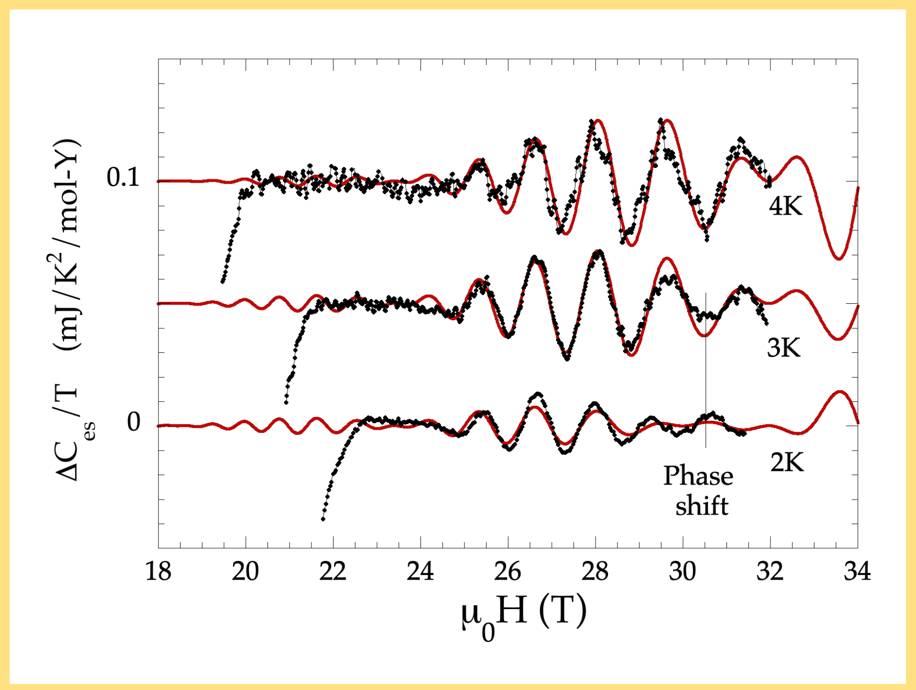

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C/T saturates for $H > H_{DOS}$

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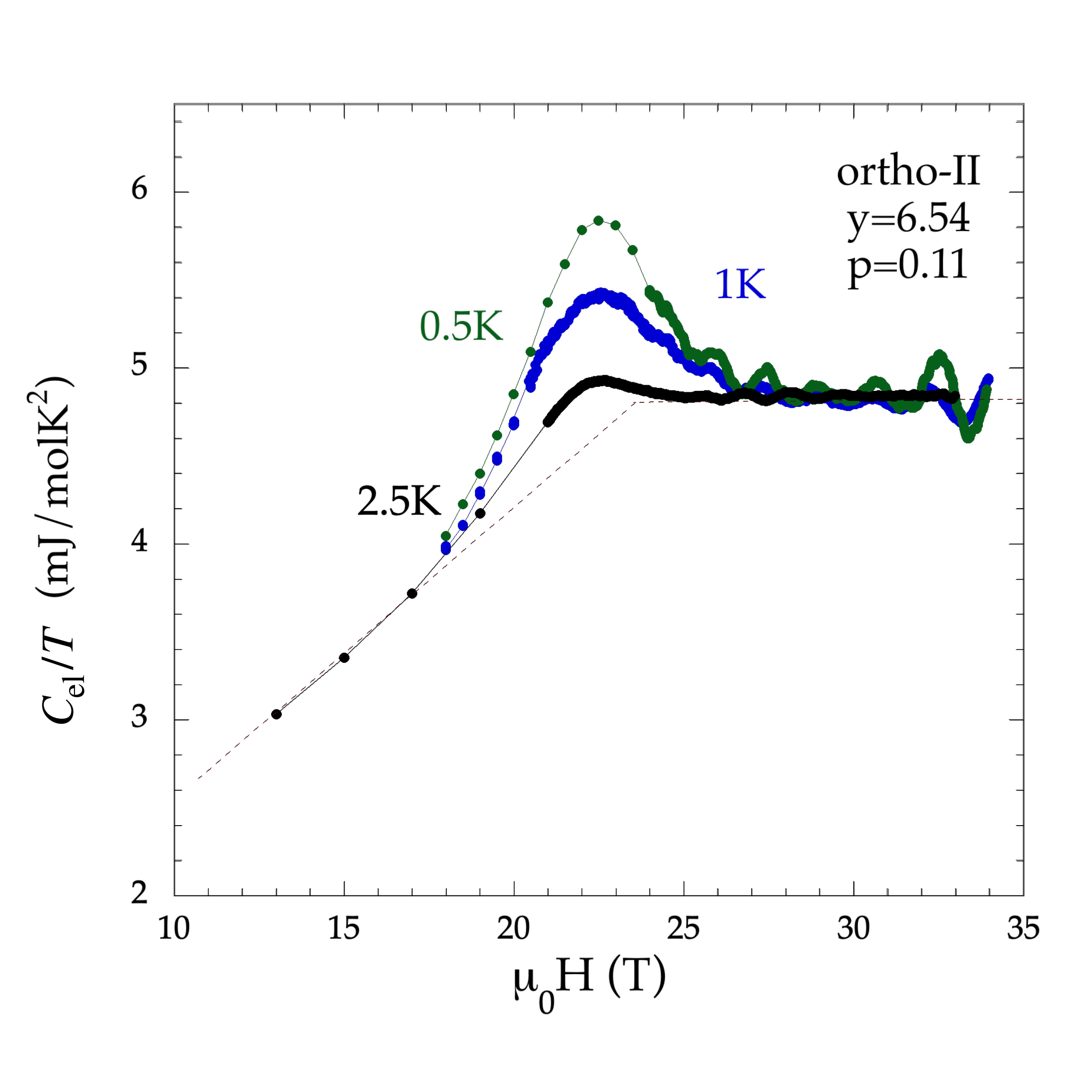
note also the small

Overshoot at 22T ?



which **increases**

for decreasing temperature !

(unpublished)

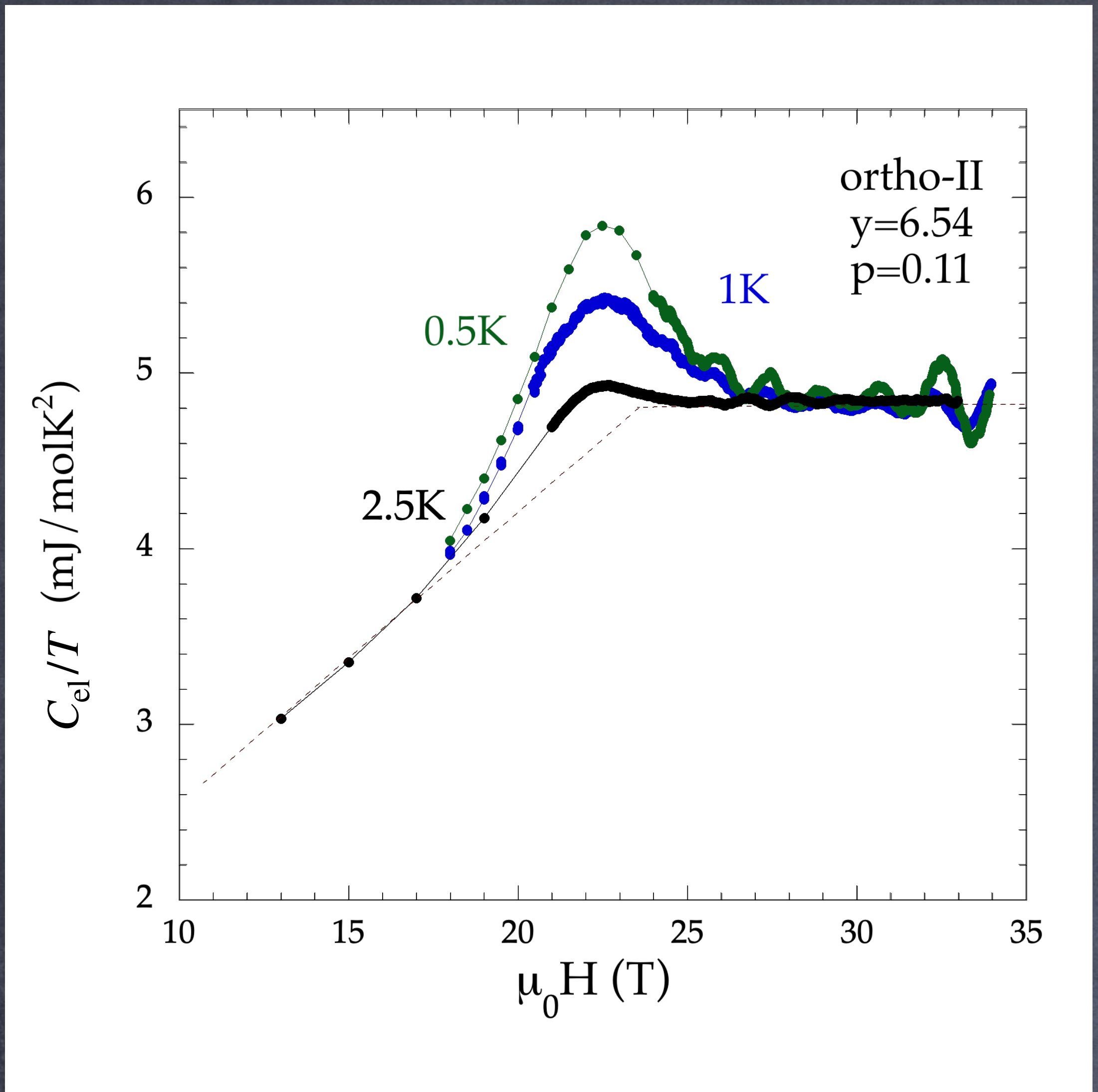
Can **not** be the standard overshoot

observed at the SC transition

Its origin still has to be clarified…

(another field induced)

quantum critical point ???



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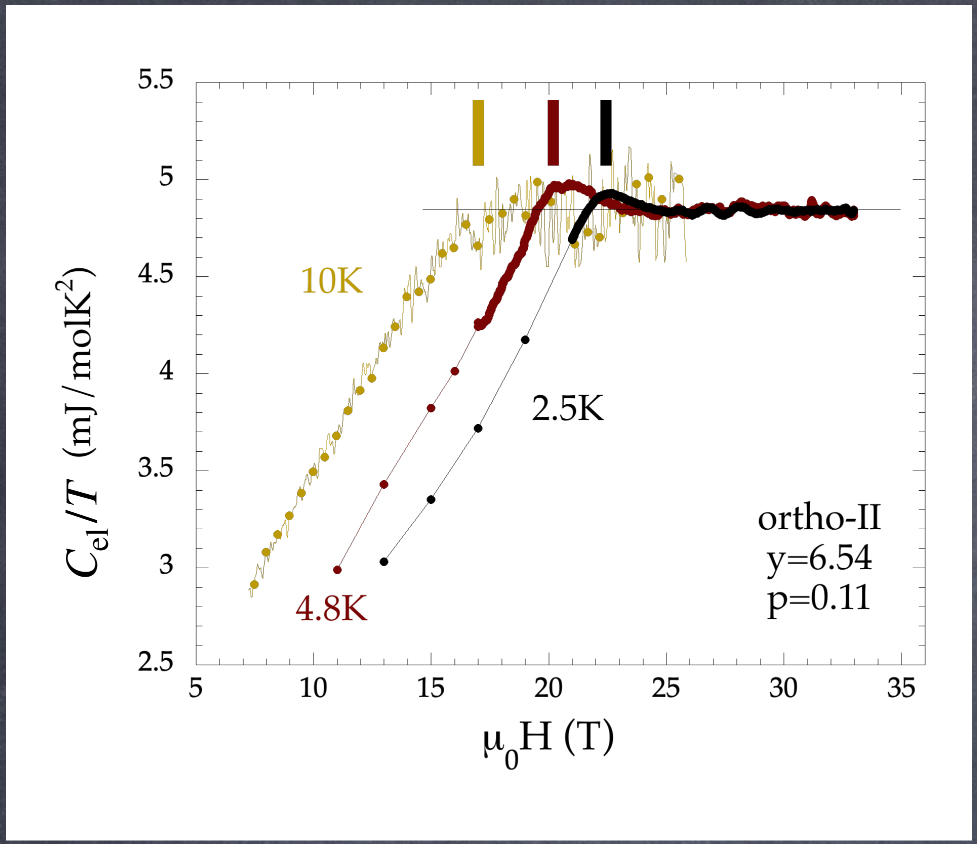
but let's follow the temperature dependence of H_{DOS}

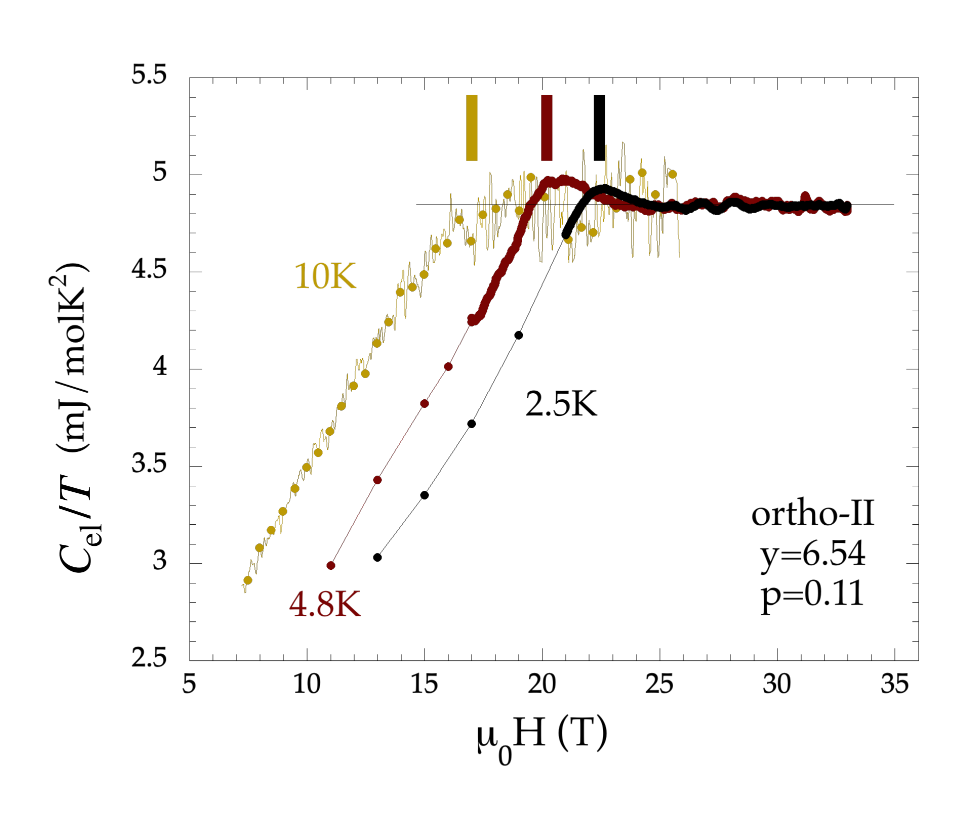


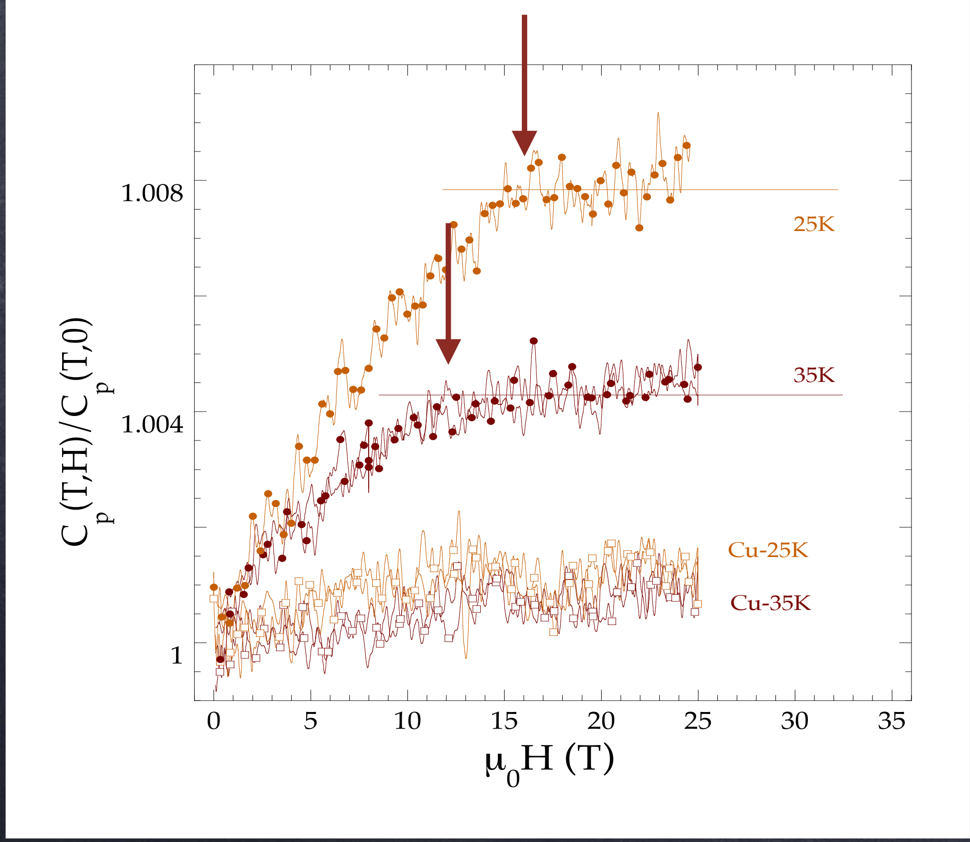
As expected H_{DOS} decreases with T

but... $$\frac{C_{el}}{T} = \frac{C}{T} - \underline{\beta T^2}$$

and the phonons contribution rapidly increases

⇒ increasing signal/noise ratio





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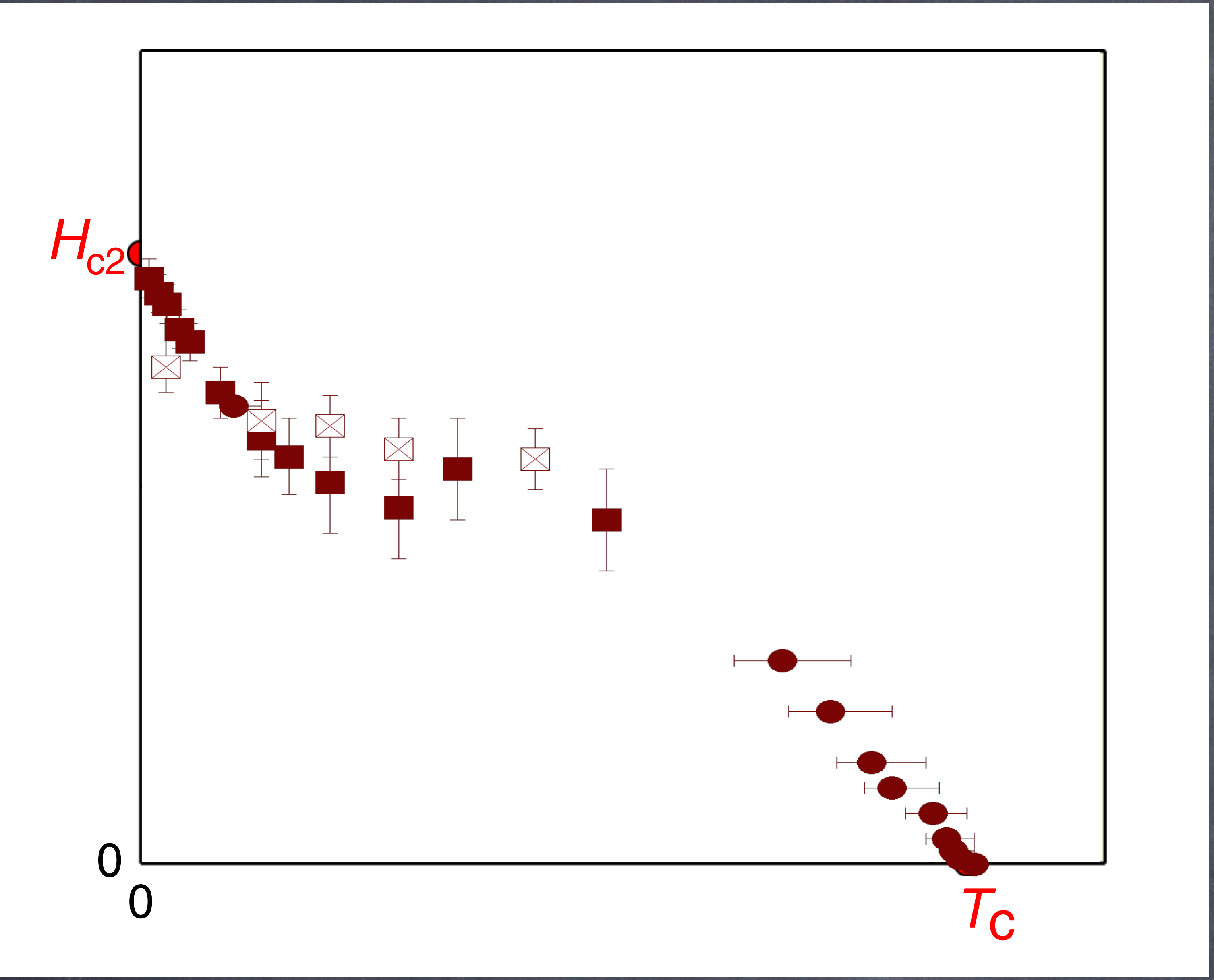
⇒ **the variation in C_{el}/T becomes smaller that 1% above 20K !**

but can still be determined

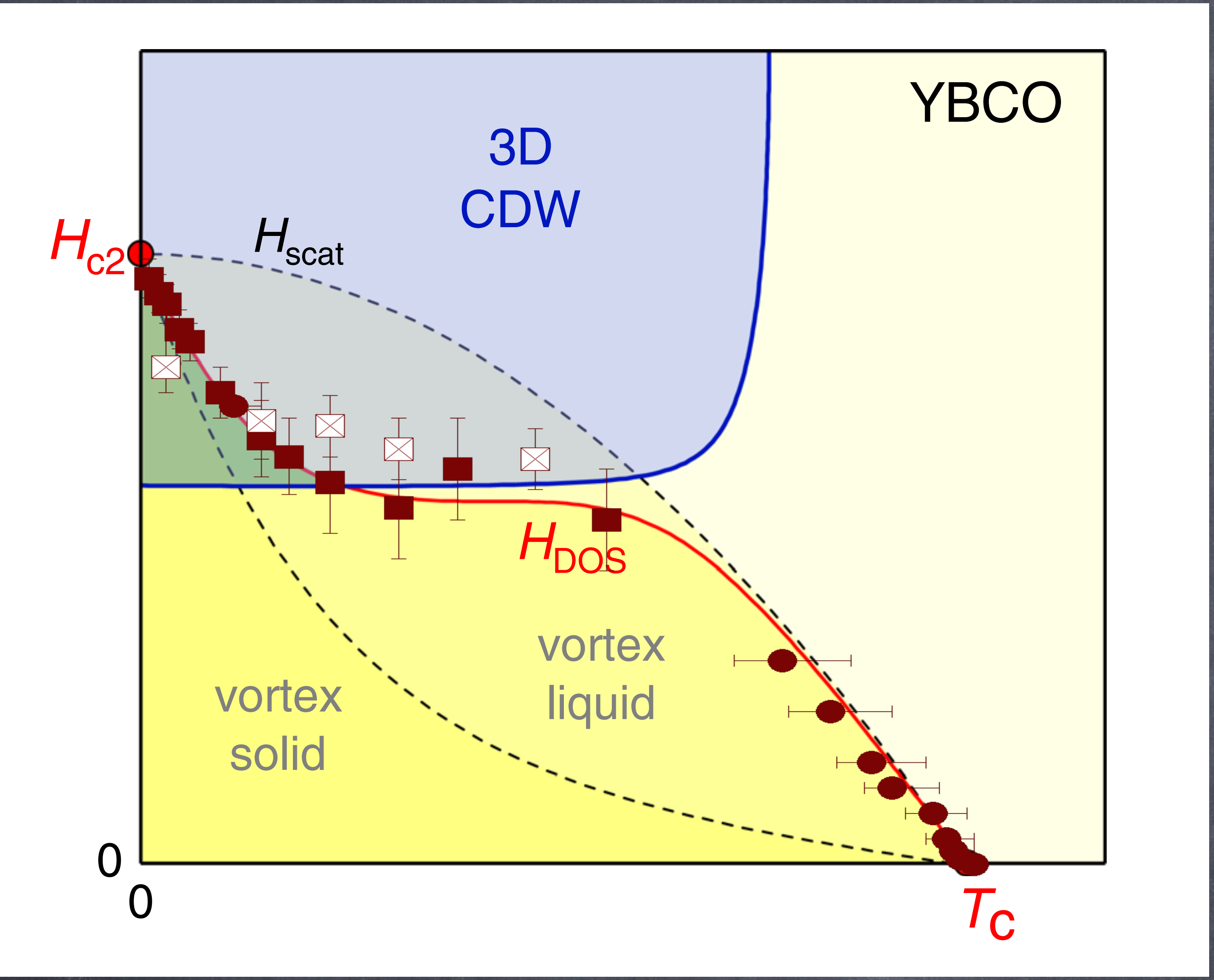
Confirmed by NMR (Knight shift) measurements

Group of M-H. Julien

see J.Kacmarcik *et al.* PRL 2019

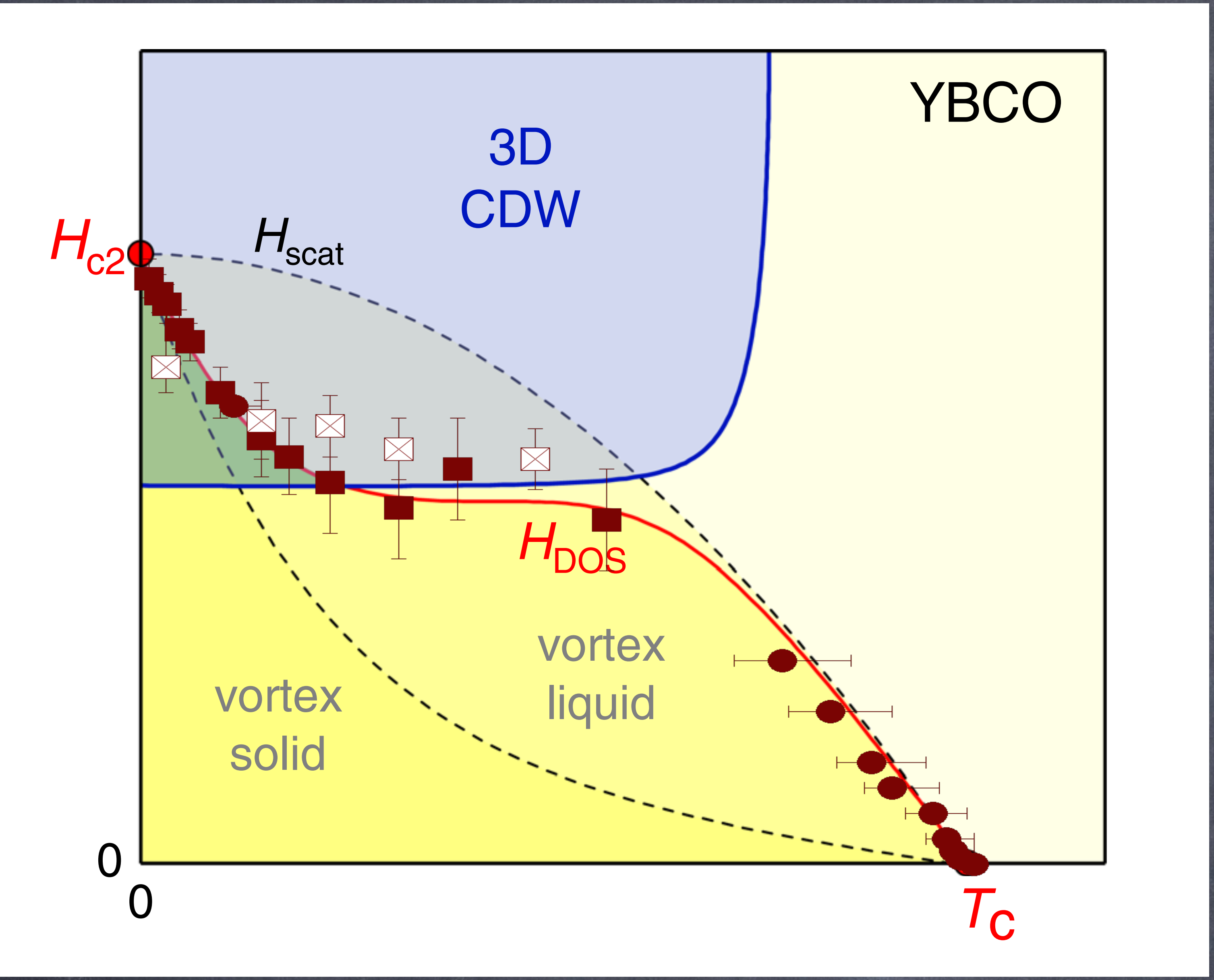


Upturn of $H_{\mathrm{DOS}}(T)$ at low T ?



Upturn of $H_{DOS}(T)$ at low T ?

SC & CDW eventually establish a form of **cooperation** at low T



Upturn of $H_{DOS}(T)$ at low T ?

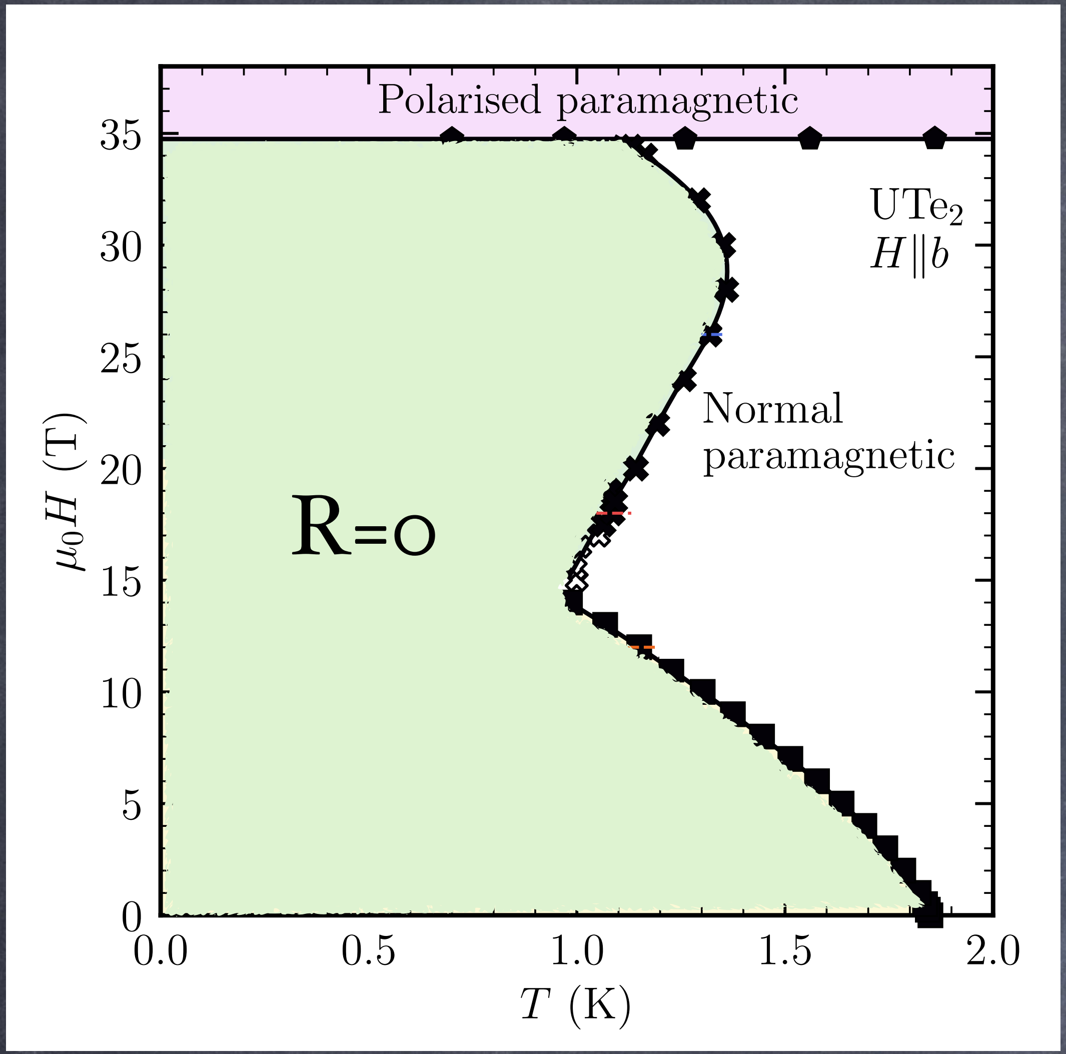
SC & CDW eventually establish a form of **cooperation** at low T

giving rise to some kind of **filamentary SC** at the interface of CDW domains ? (or *fragile* SC at CDW dislocations). Yu & Kivelson PRB 2019, Leridon et al. New J.Phys. 2020

or «FFLO-like» upturn, related to the **pair-density wave order ?**

Agterberg et al. PRB 2015, Berg et al. PRL 2007, Dai et al. PRB 2018

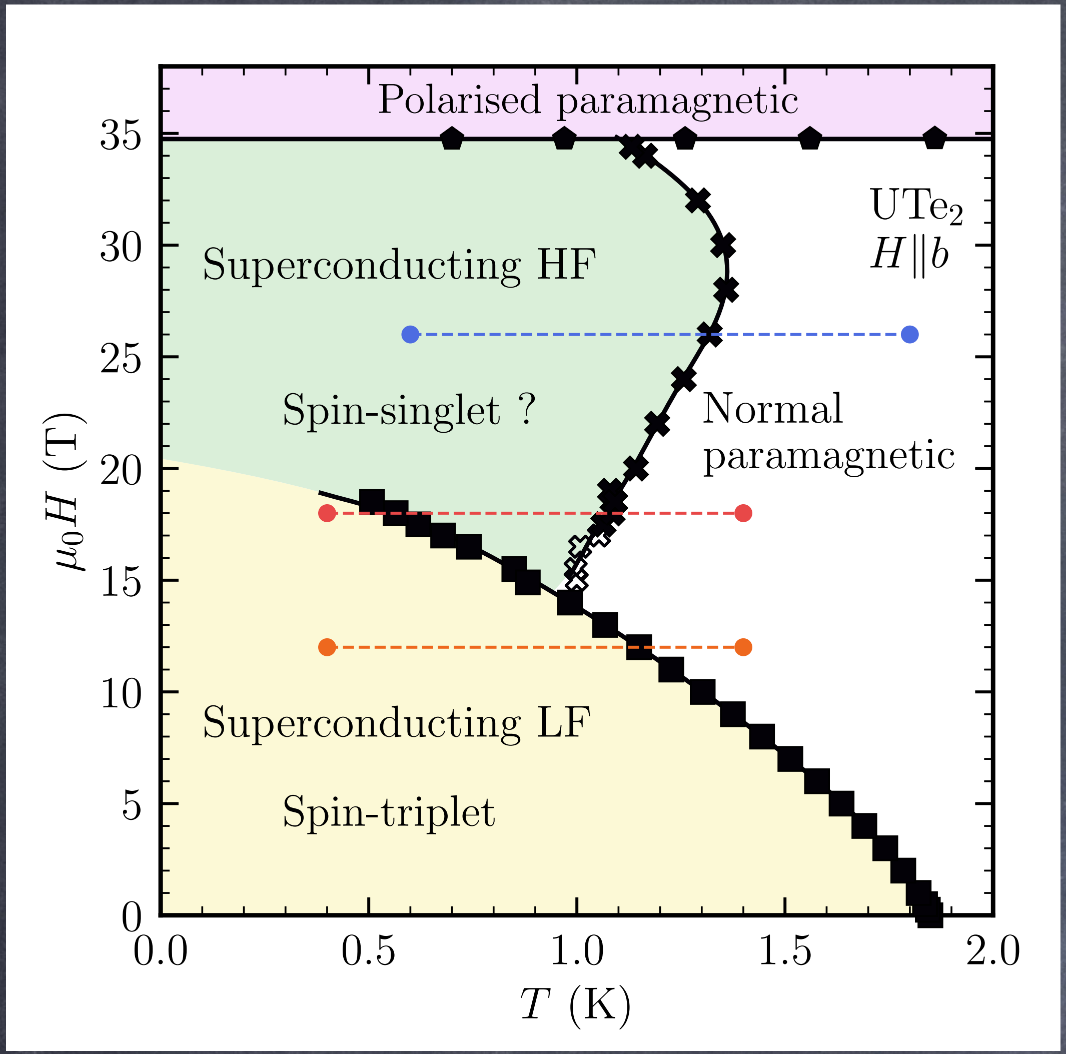
A last exemple to flash....

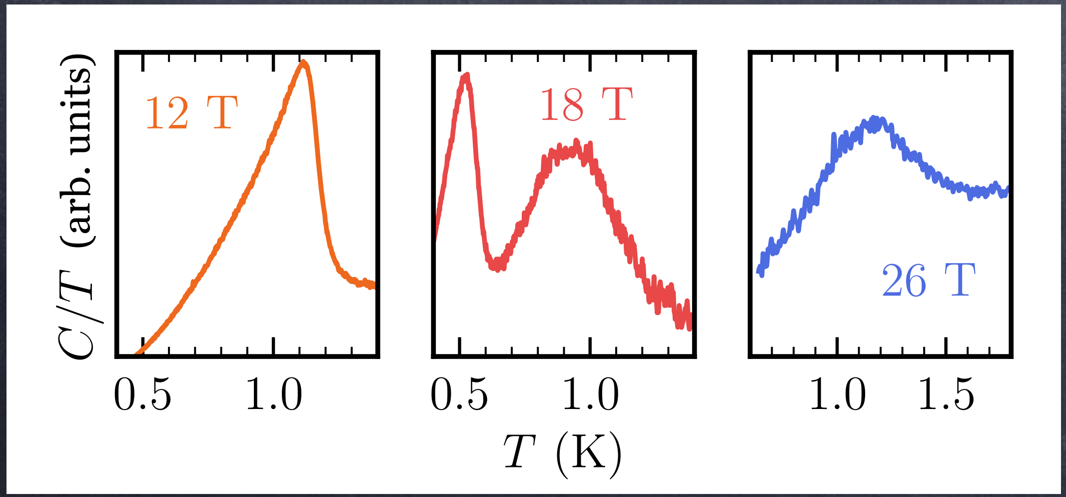


Re-entrant superconductivity in UTe_2

(possible spin-triplet and topological SC)

A last exemple to flash....





Re-entrant superconductivity in UTe_2

(possible spin-triplet and topological SC)

C/T measurement

⇒ phase transition **within** the superconducting state

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Thank you for your attention