Condensed Matter Physics

Charge Density Waves and Superconductivity in High-T_c materials

Ted Forgan

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EuCAS, Geneva

September 2017

Georg Bednorz and Alex Muller's discovery

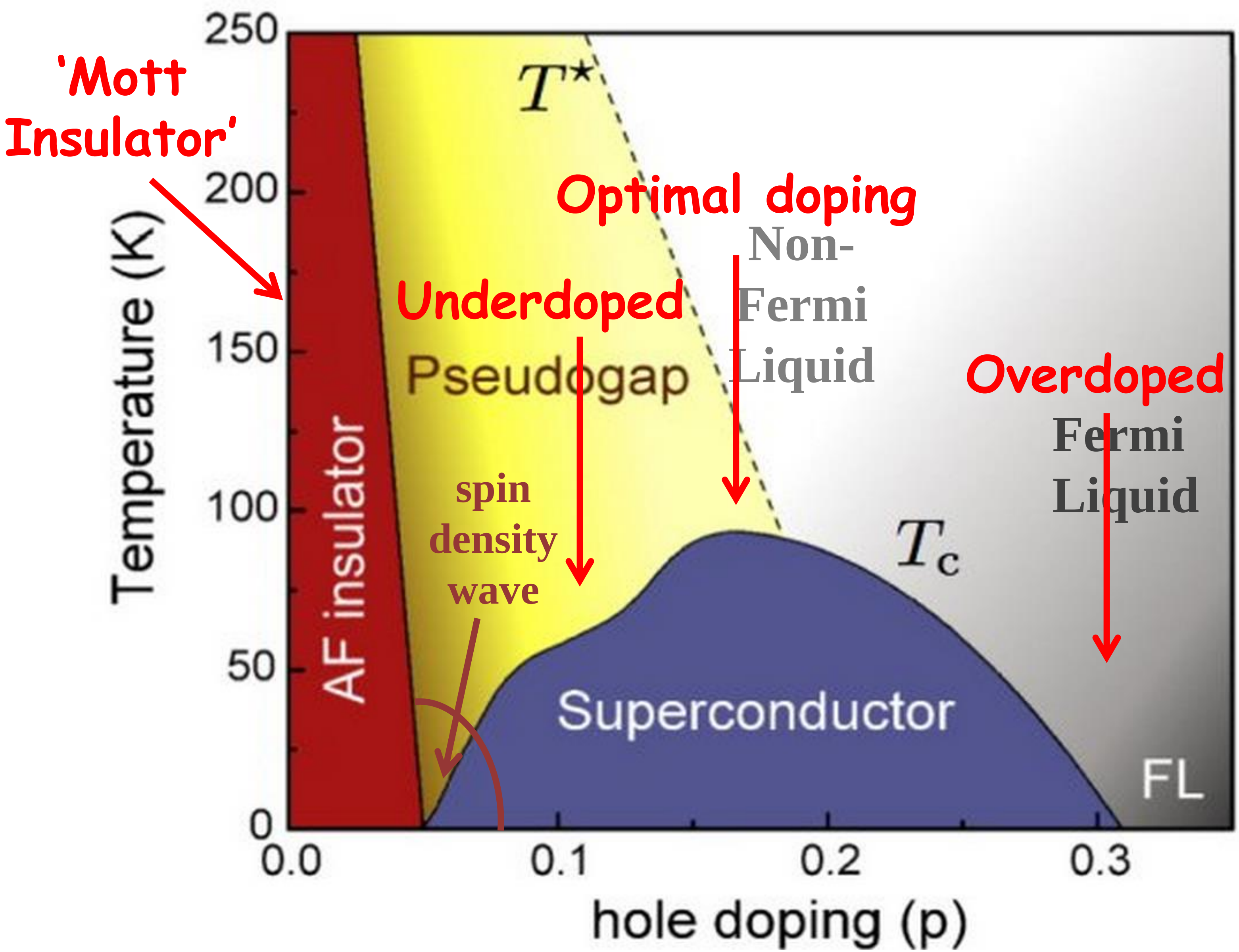
http://www.phys.ntnu.no/brukdef/prosjekter/super/Profiles/bednmull.jpg

received the Nobel Prize 1987 for discovery of the first of the copper-oxide superconductors

> 30 years & ~ 10^5 papers later and we still don't understand these materials!

- applications are nothing like as widespread as hoped in those heady early days...

High-T_c properties versus hole doping



modified from LCMI website

Some simple "chemistry"

La_2CuO_4

Lanthanum: 3+ ; La_2 => 6+

Oxygen: 2- ; O_4 => 8-

∴ Copper: 2+

But La_2CuO_4 is a "Mott insulator": electron repulsion keeps them localised on Cu++ and their spins line up antiferromagnetically

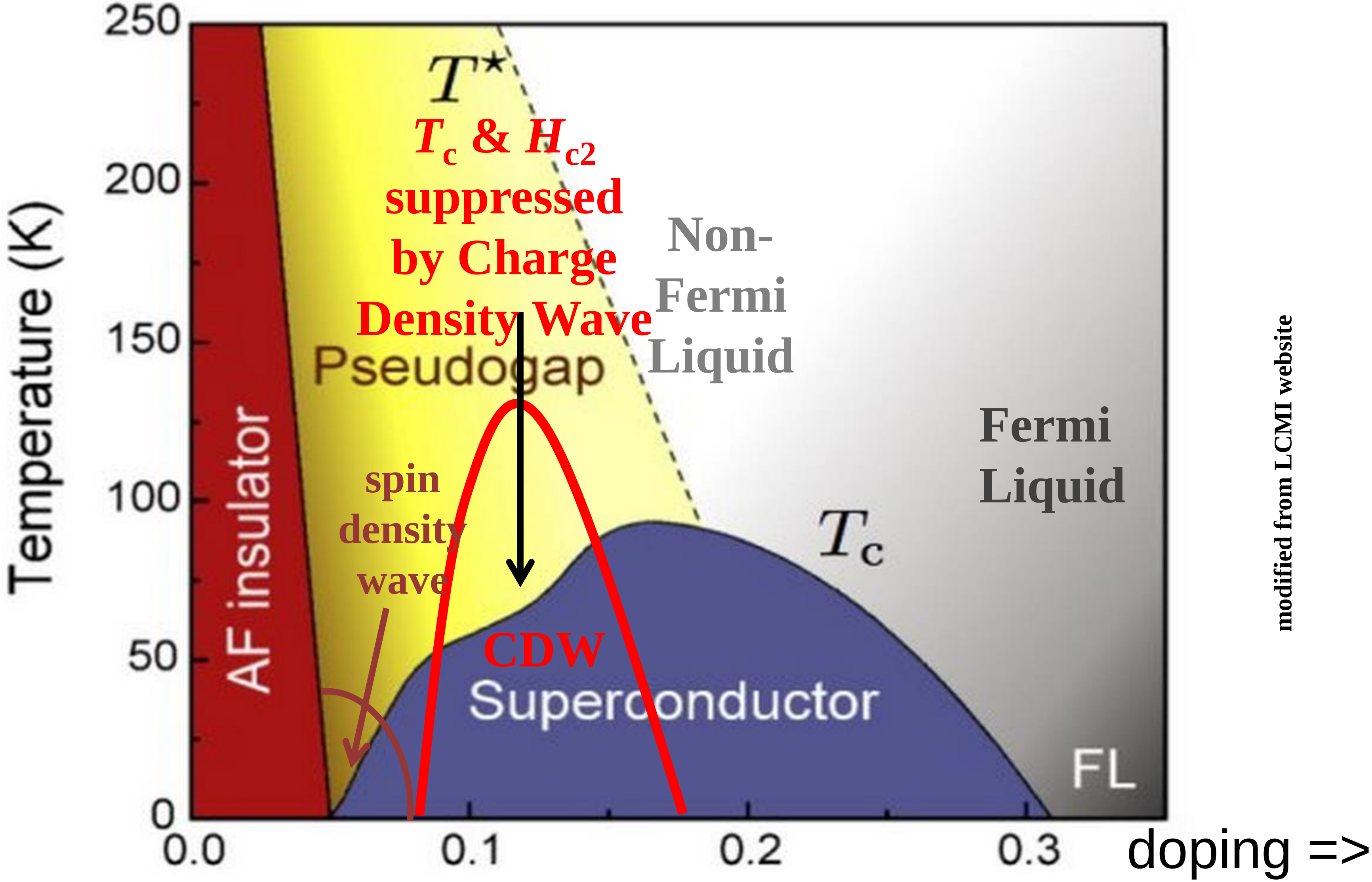
Cu has $3d^{10}$, $4s^1$

∴ Cu^{++} has $3d^9$, i.e. 1 hole (unpaired electron) in the d-shell..

Consider $YBa_2Cu_3O_7$: (slightly overdoped superconductor) Doing the same calculations, we get an average of 1.33 holes per $Cu^{2.33+}$

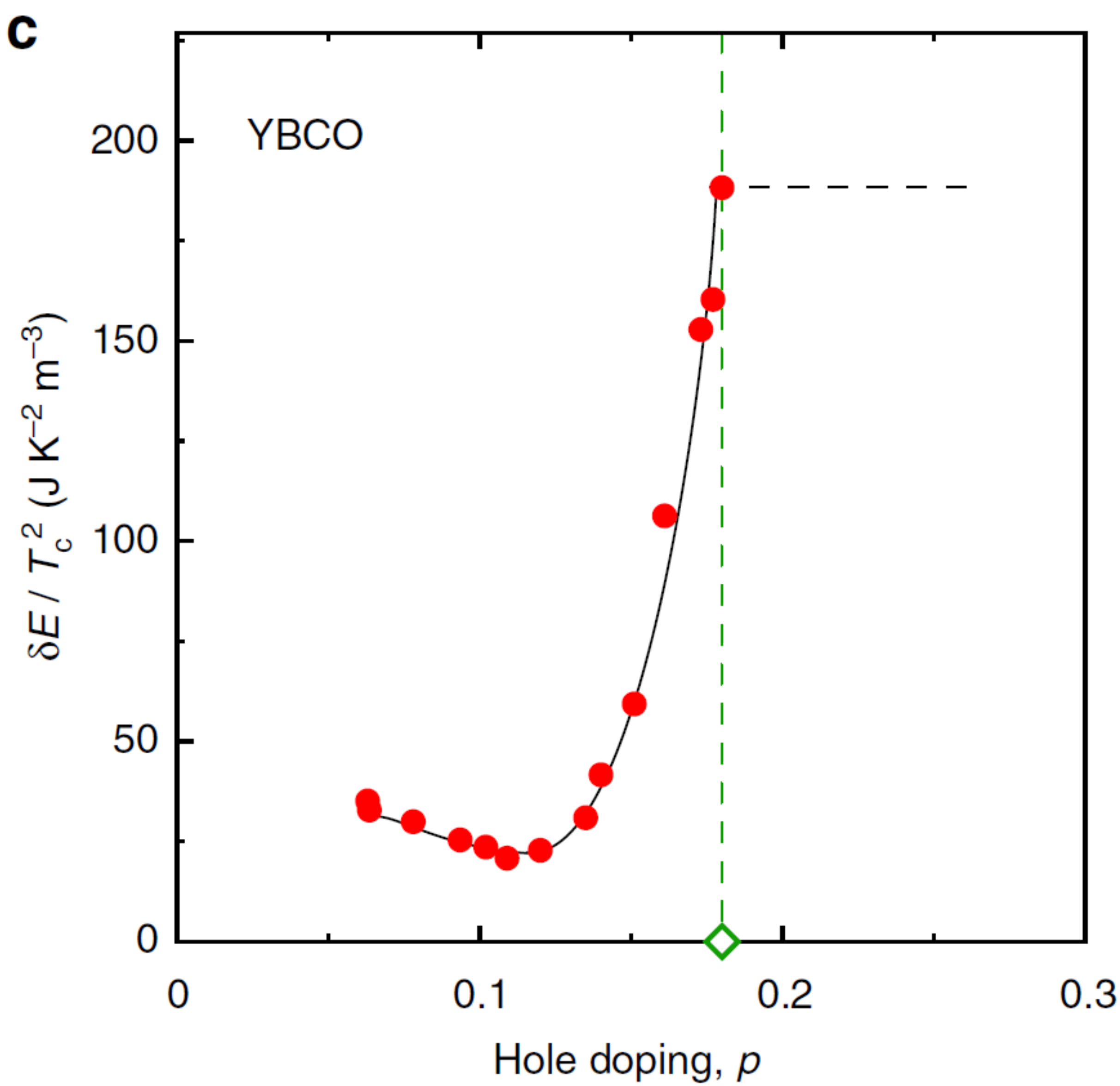
What happens as we go from an insulator to a metal?

High-T_c properties versus hole doping - current ideas



modified from LCMI website

Superconductivity *vs.* doping in YBCO



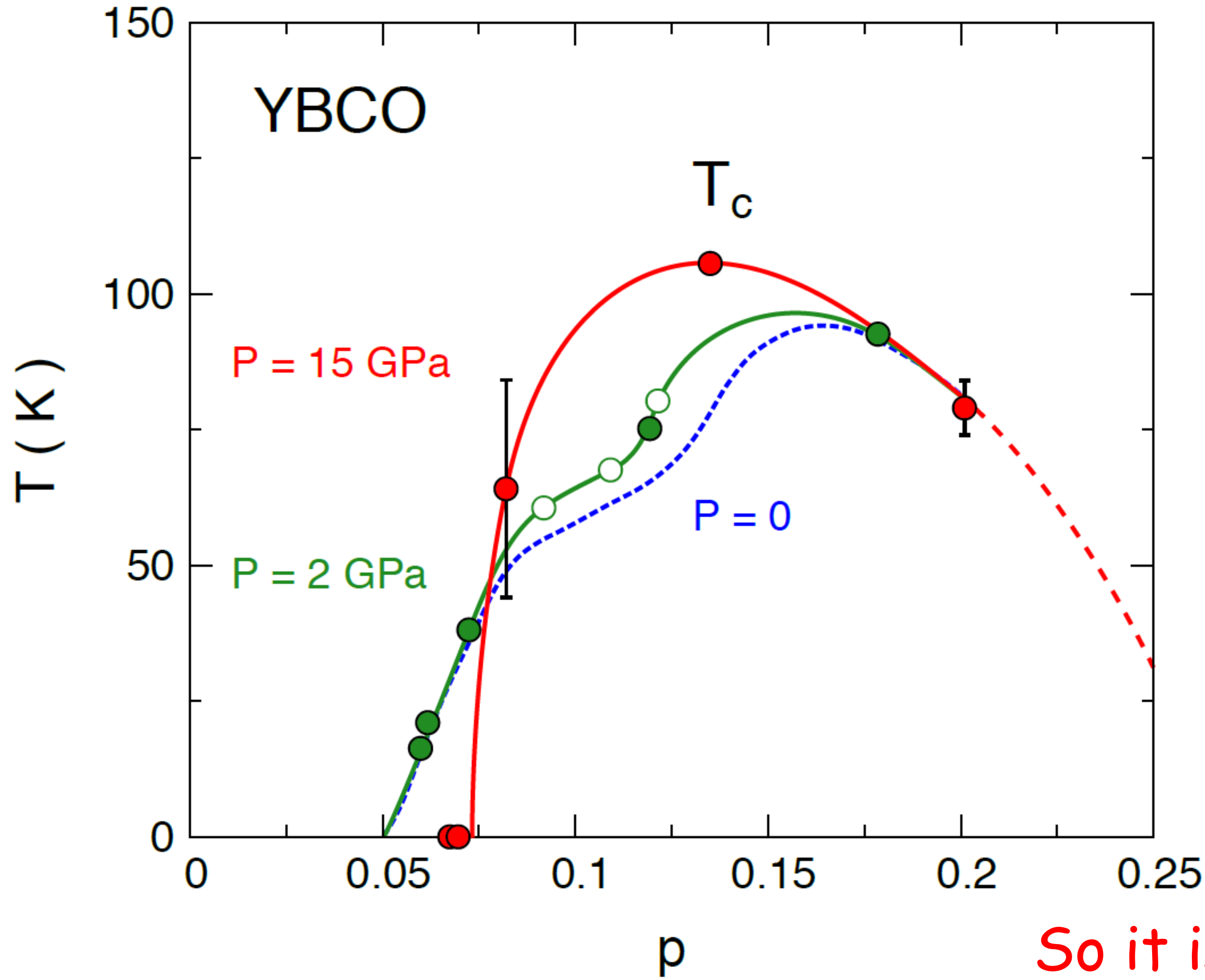
Grissonnanche *et al.* Nature Comms. 5, 3280 (2014)

B_{c2} drops down to ~22 T at the maximum CDW

T for ρ = 0 as a function of doping for various fields.

Superconducting condensation energy versus doping.

Can the Charge Density Wave be avoided?



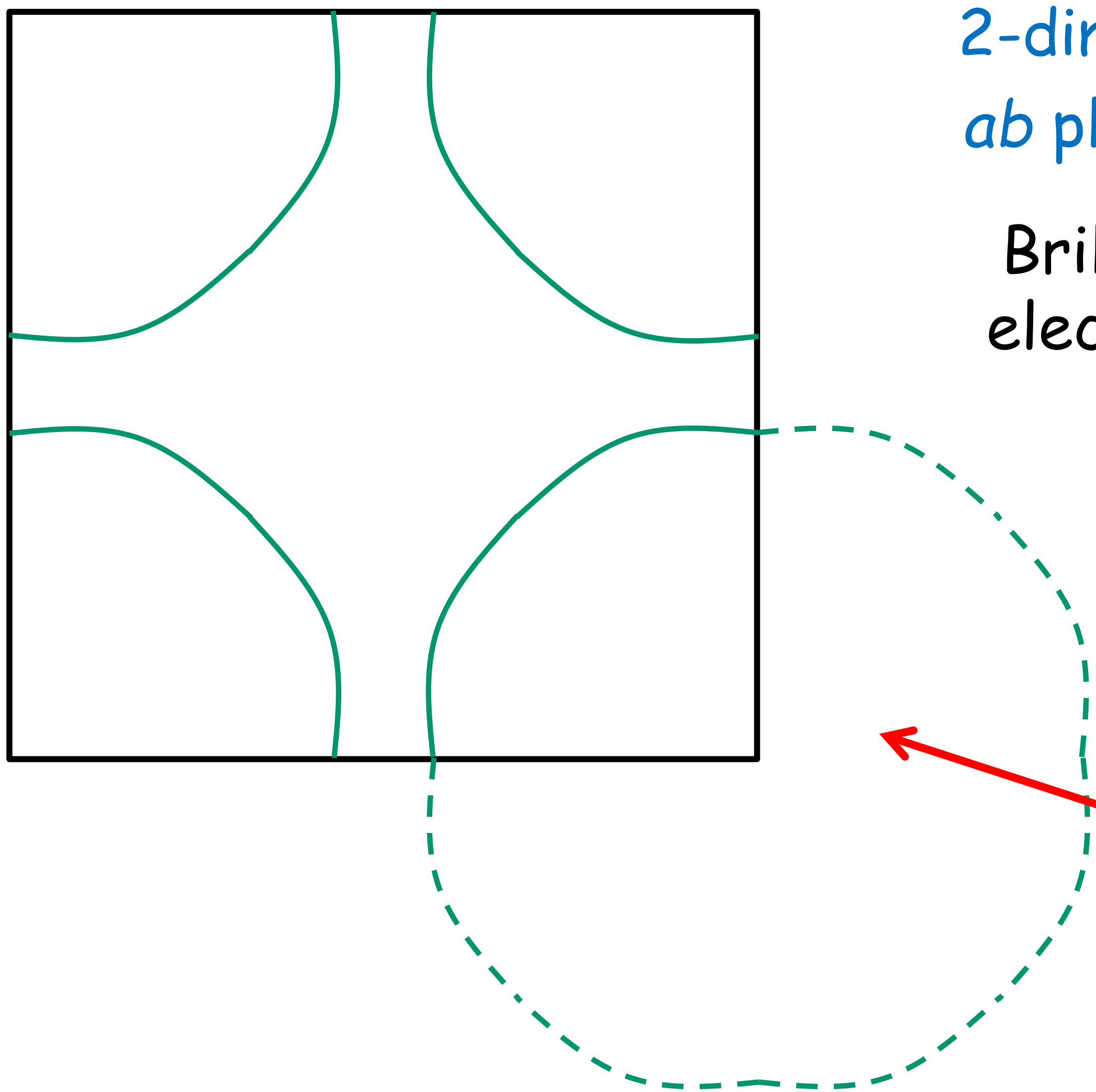
Not very good error bars, but it appears that pressure suppresses the CDW

The max. value of T_c rises and moves closer to the AFM region

So it is important to understand the CDW

Cyr-Choiniere *et al.* arXiv:1503.02033 (2015)

What "should" a High-T_c Fermi Surface look like?



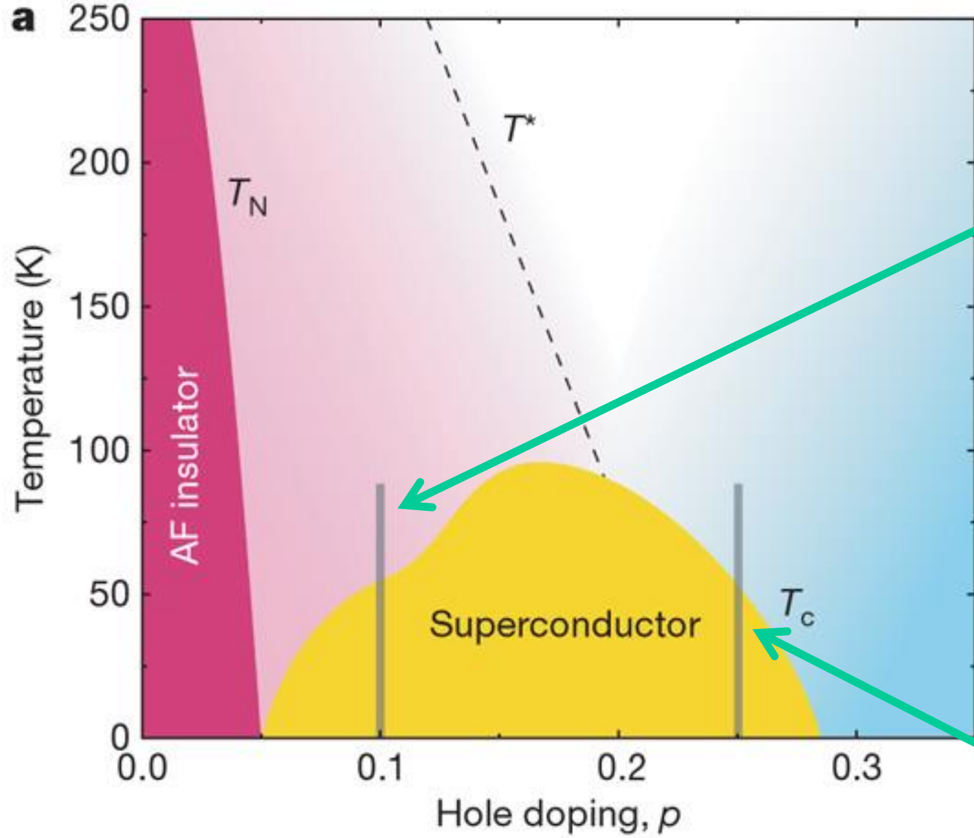
2-dimensional: -look at *ab* plane cross-section

Brillouin Zone holds 2 electrons or holes/cell

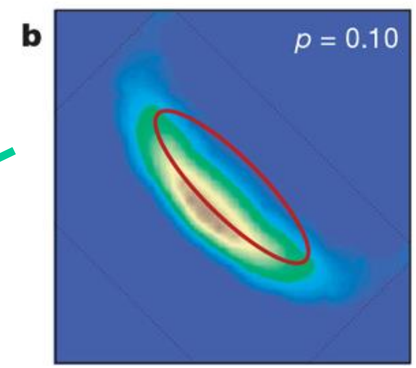
1 hole/Cu at zero doping: $p = 0$

This area would be $\propto (1+p)$ if all the electrons are free to move

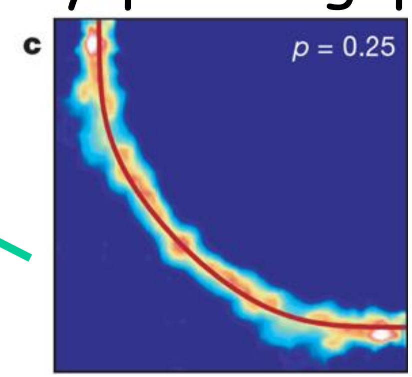
ARPES (photo-electron spectroscopy) shows changes in Fermi Surface with doping



"Fermi arc"



FS ends removed by"pseudogap"



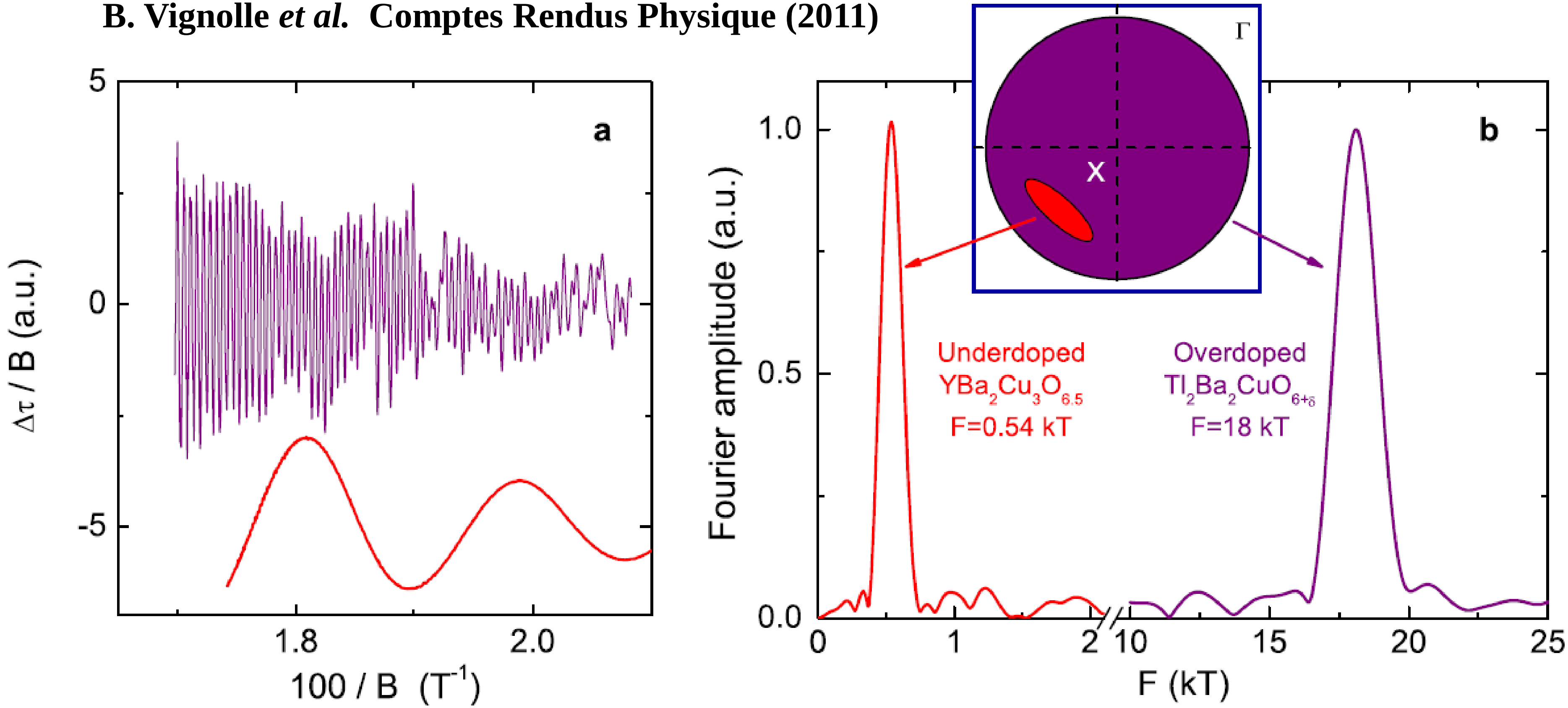
Full Fermi surface

Doiron-Leyraud *et al.* Nature (2007)
Platé, *et al.* Phys. Rev. Lett. (2005)
Shen, *et al.* Science (2005)

Some Quantum Oscillation Data

Overdoped – all holes visible - obeys Luttinger theorem

B. Vignolle *et al.* Comptes Rendus Physique (2011)

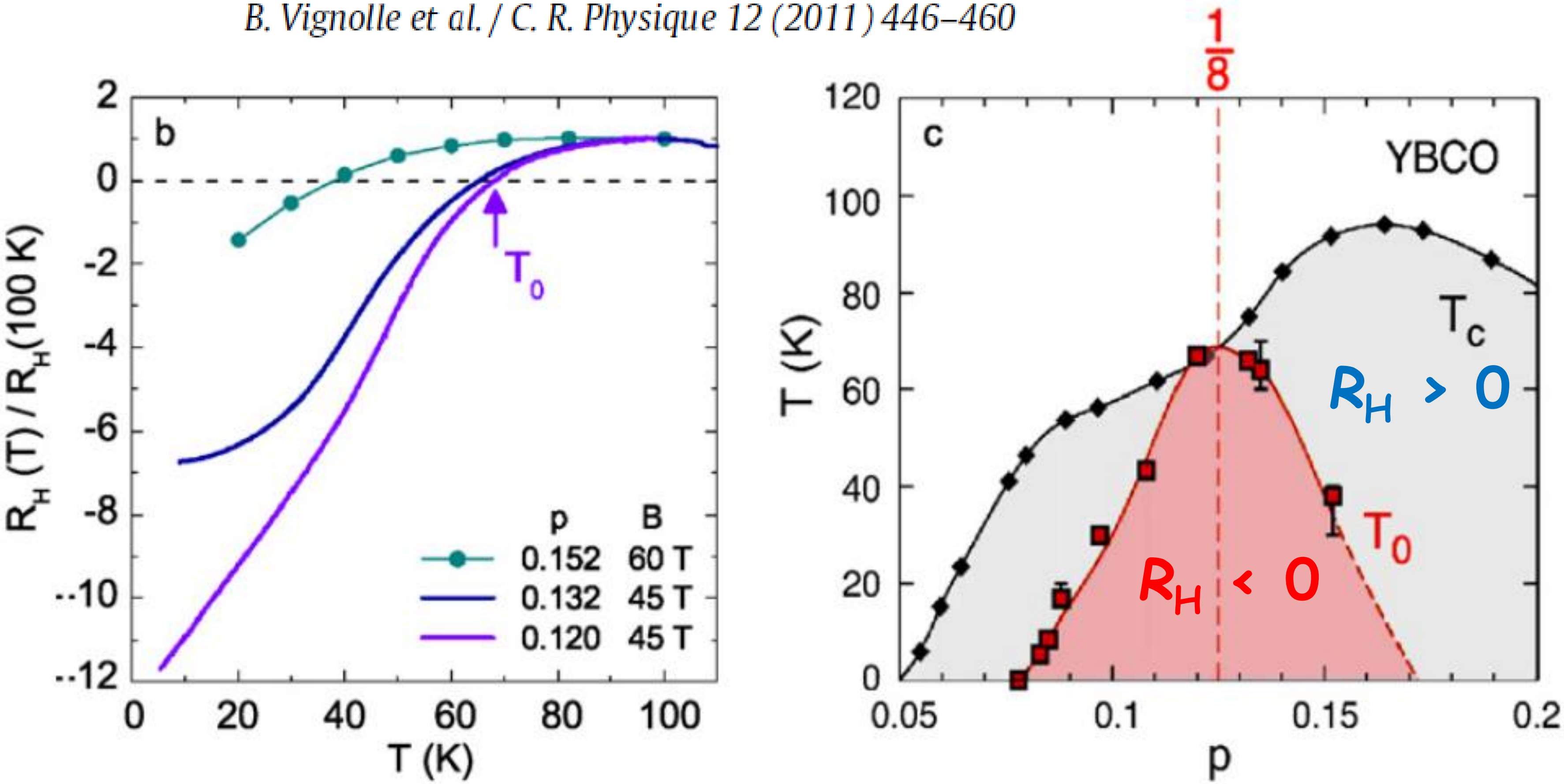


Underdoped – tiny number of *electrons* not holes

N.B. QOs give the *area* of the electron pocket, not shape

At low T, the Hall effect changes sign in underdoped $YBCO_y$

B. Vignolle et al. / C. R. Physique 12 (2011) 446–460

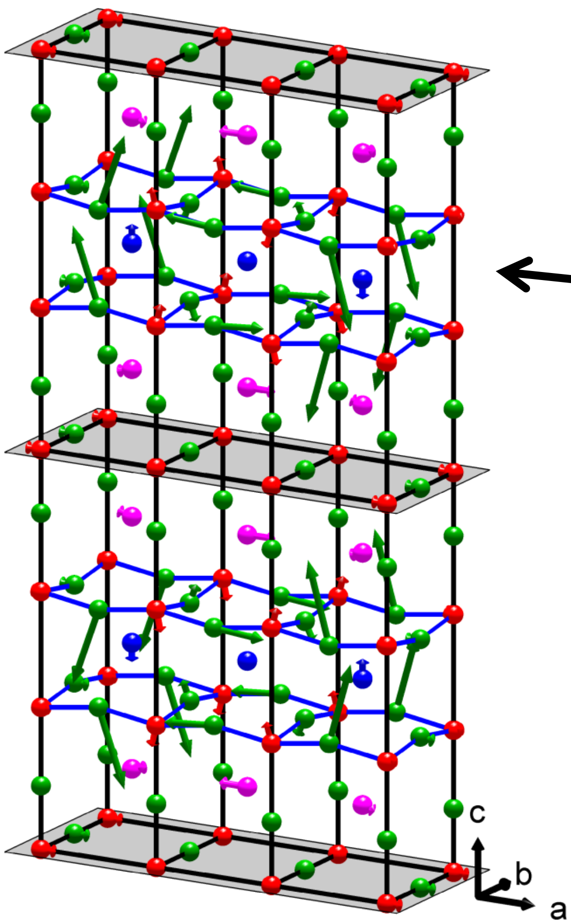


This suggests that for doping levels around $p \sim 1/8$ the Fermi surface changes topology below $>\sim T_0$ …

… from big hole FS (R_H small, +ve) to tiny electron FS (R_H large, -ve)

What do we think is causing this?

Charge Density Wave (CDW) order - a tiny modulated charge density – and associated lattice distortion, - which forms in a *wide range* of slightly under-doped cuprate high-T_c materials.



It is centred on the CuO_2 layers and *competes* with superconductivity

Exaggerated view of a CuO_2 plane displacements (oxygen, copper)

This CDW order has an *incommensurate* period ~3 unit cells along *both* ***a*** and ***b***. (***a*** shown)

It disappears as doping is increased to about optimum for superconductivity

Observing the CDW by diffraction - 100 keV X-rays, 17 T

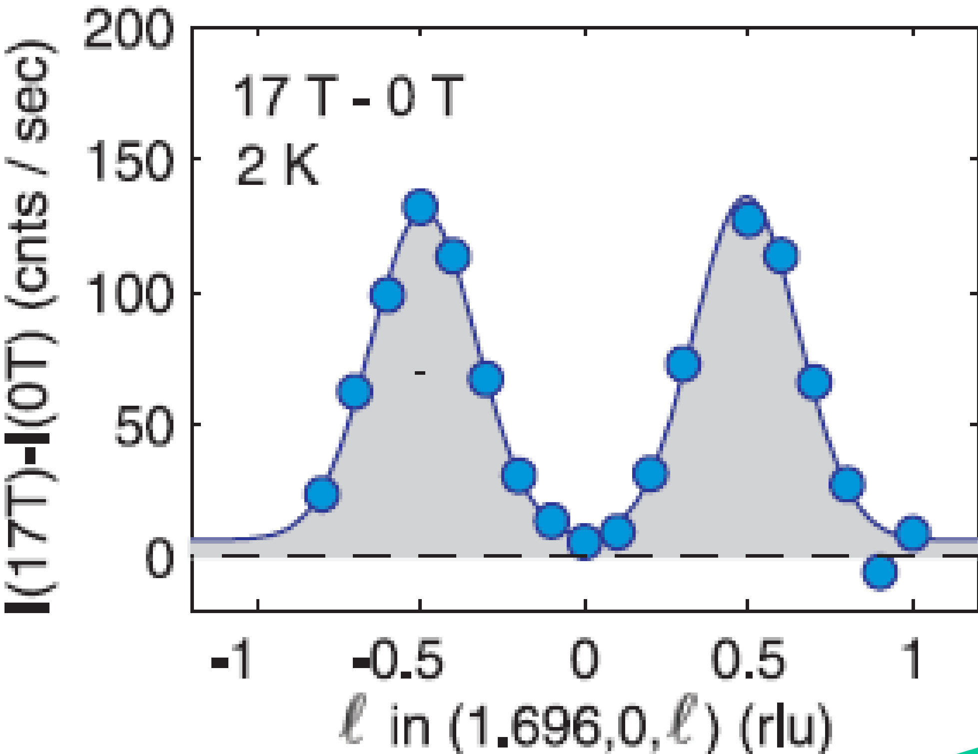
Our first experiment:
was on $YBCO_{6.67}$
$3.1 \times 1.7 \times 0.6$ mm^3
99% detwinned
$T_C = 67$ K

Others measured at zero field using Cu-L-edge resonant X-rays*

BW5 – on DORIS (RIP),
HASYLAB, DESY, Hamburg
- using the Birmingham beamline cryomagnet –taken there by truck

*Ghiringhelli, G. *et al.* Science **337**, 821 (2012)

Our results: a Field- & Temperature-dependent diffracted peak



Intensity: few x 10^{-6} of the (200) (strongest charge peak)

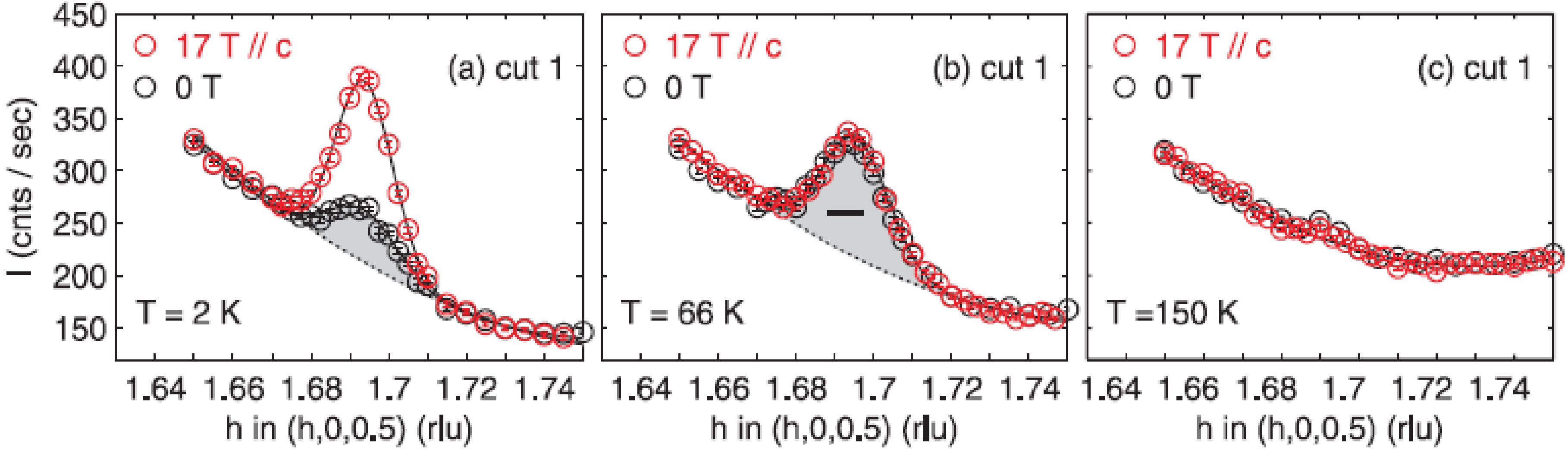
Incommensurate

At zero field, adjacent cells along the *c*-direction in antiphase

$$\boldsymbol{q}_1 = (0.305, 0, 0.5)$$

Accompanied by a similar modulation along ***b***

What happens as we change temperature?



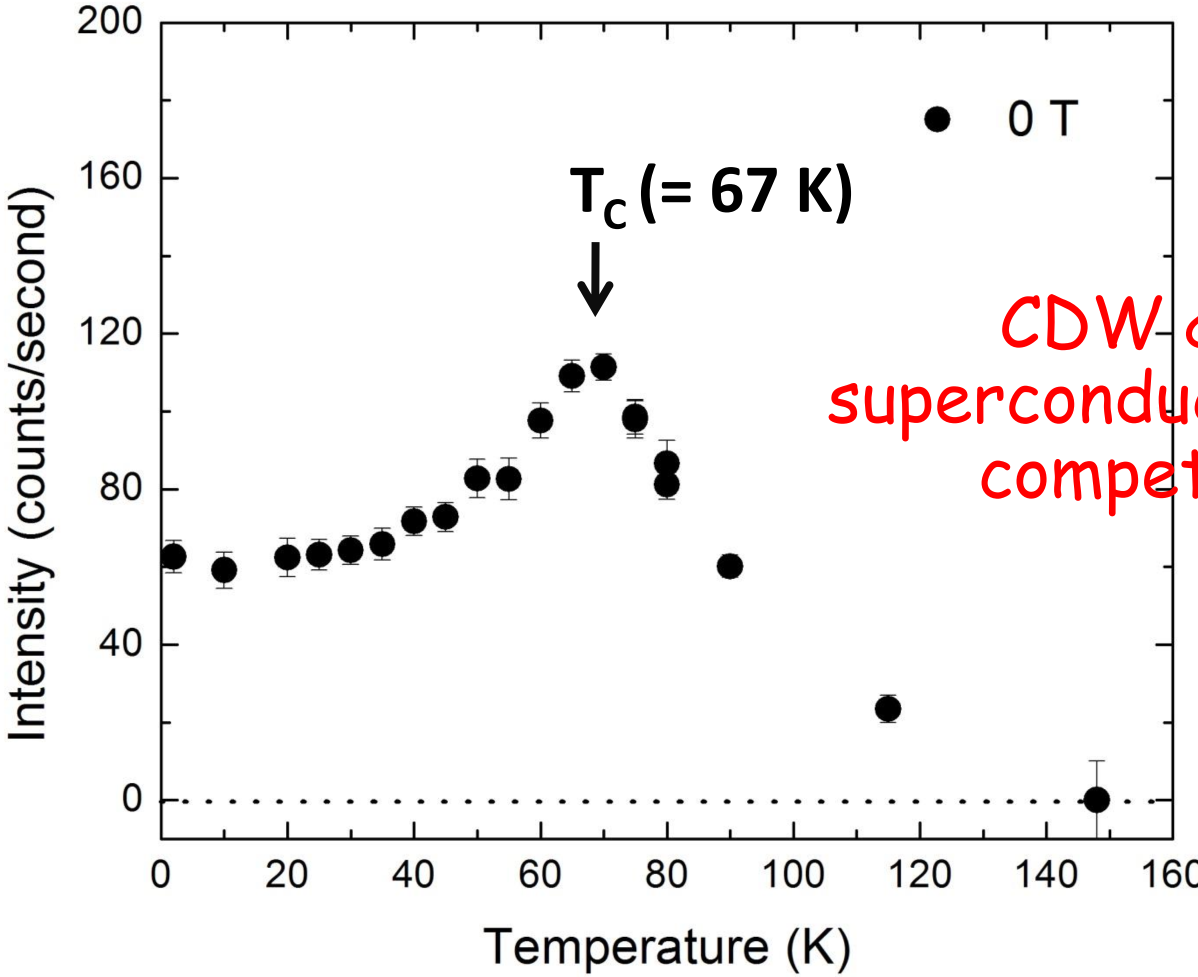
CDW Peak is always finite width – order is finite range

CDW Peak disappears at high T

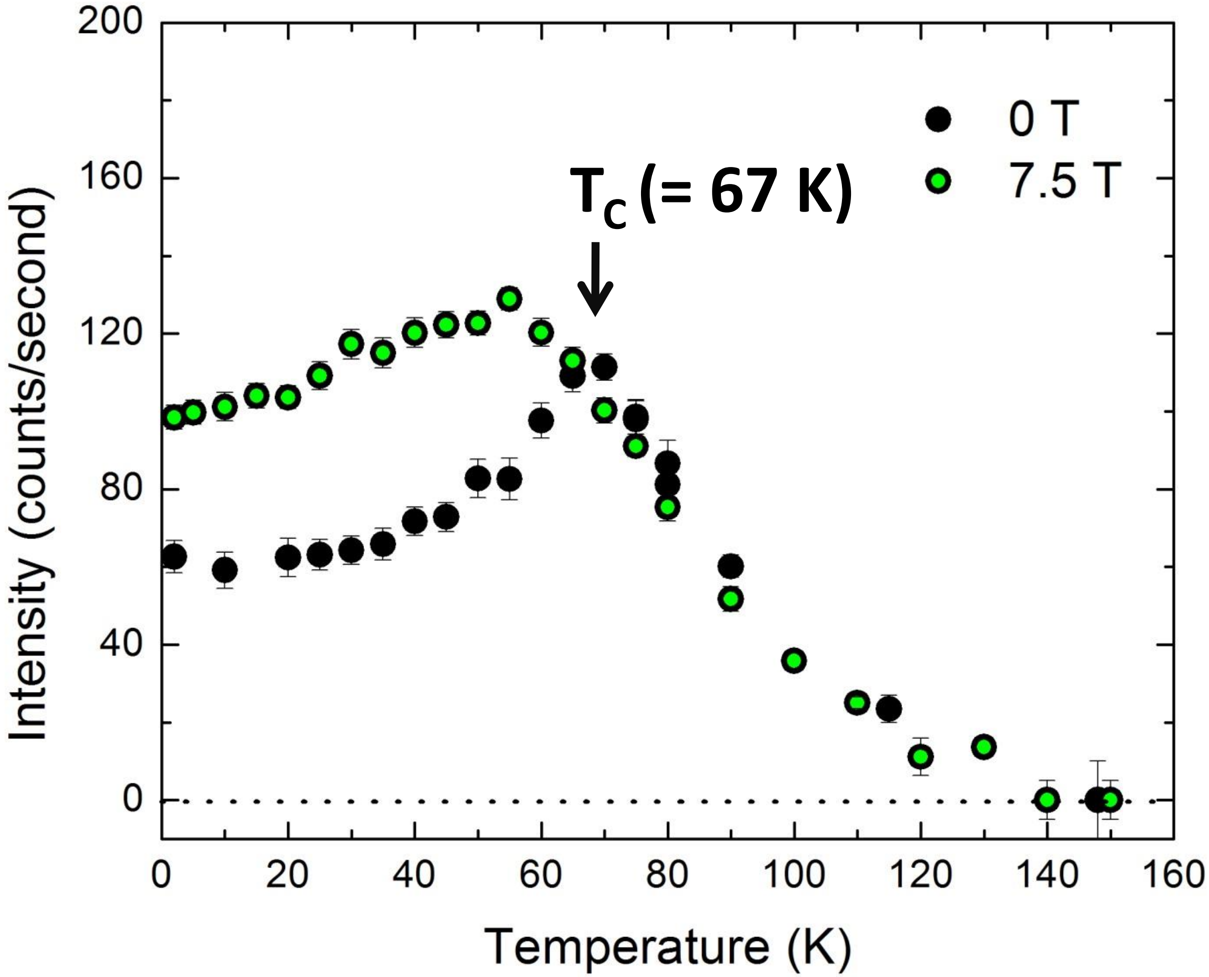
No field-dependence above superconducting T_c

However, at low T, superconductivity is suppressed by the B-field, and the CDW intensity increases

What happens as we change temperature?



What happens as we change temperature?



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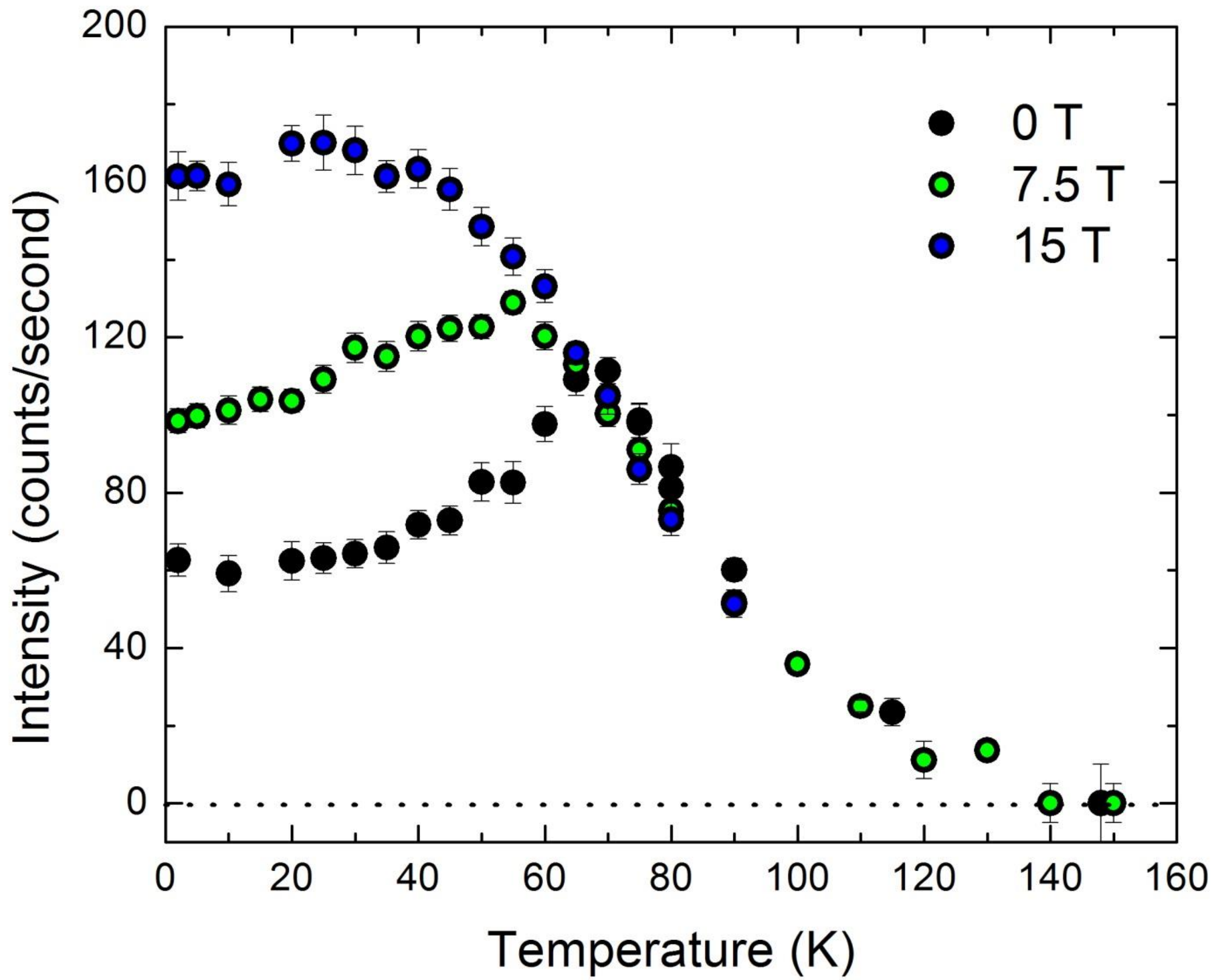

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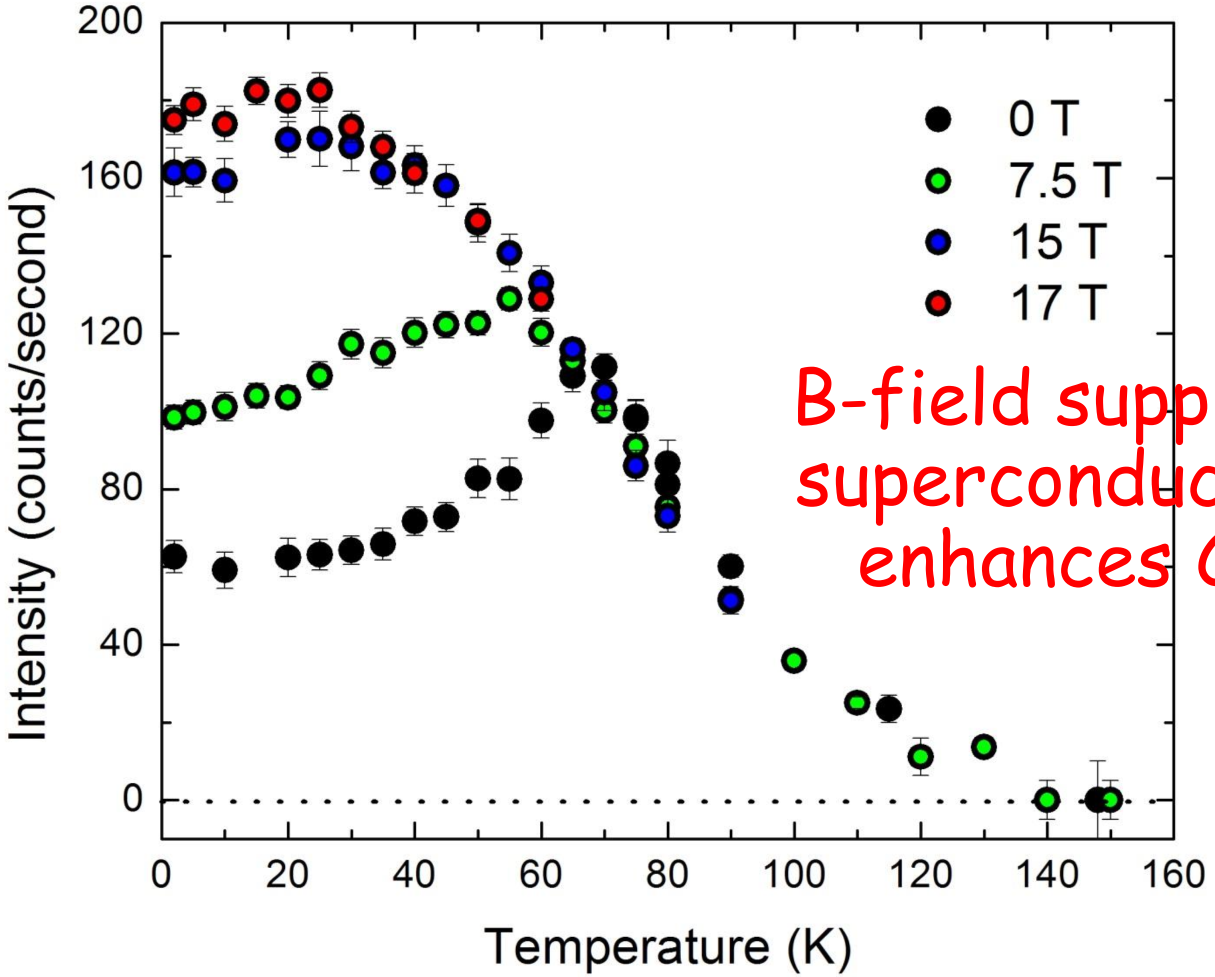
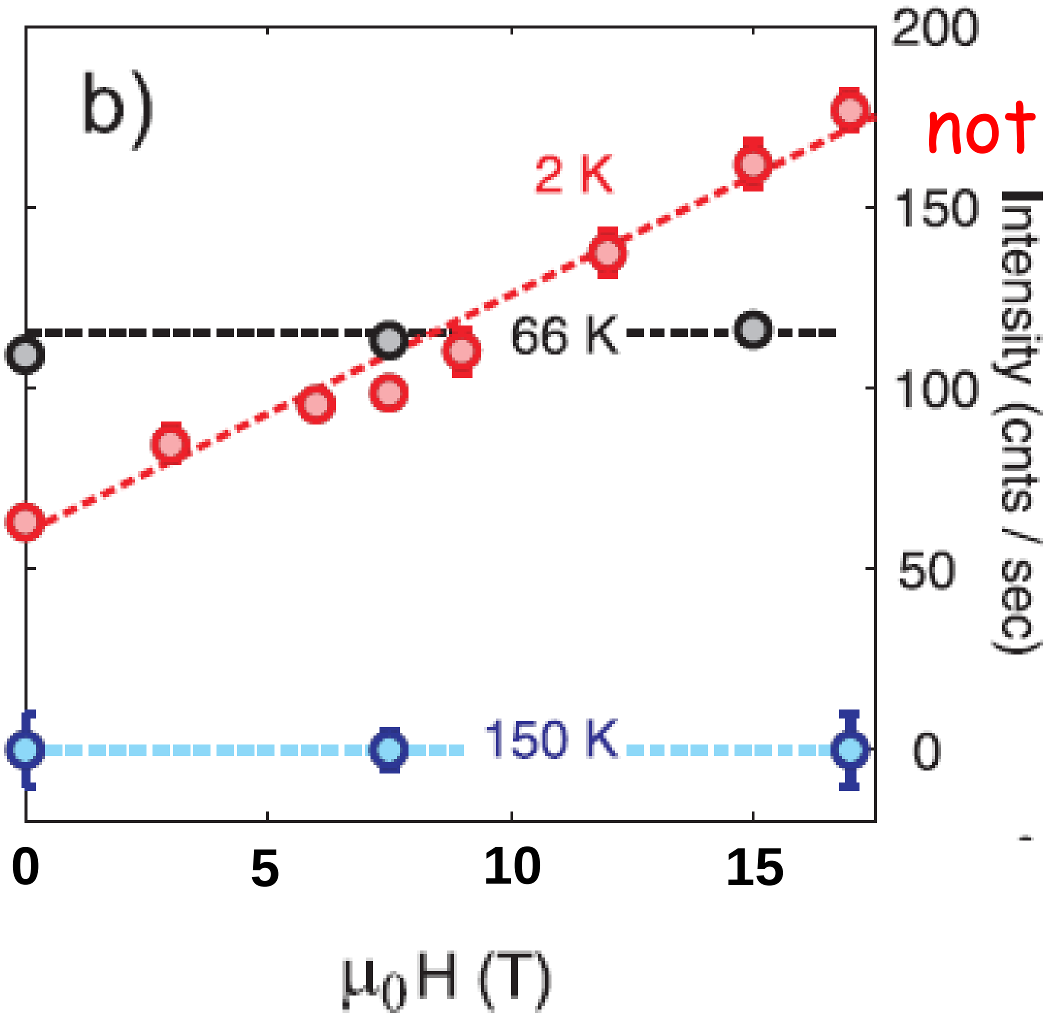


B-field suppresses superconductivity, enhances CDW

Field dependence of CDW Intensity

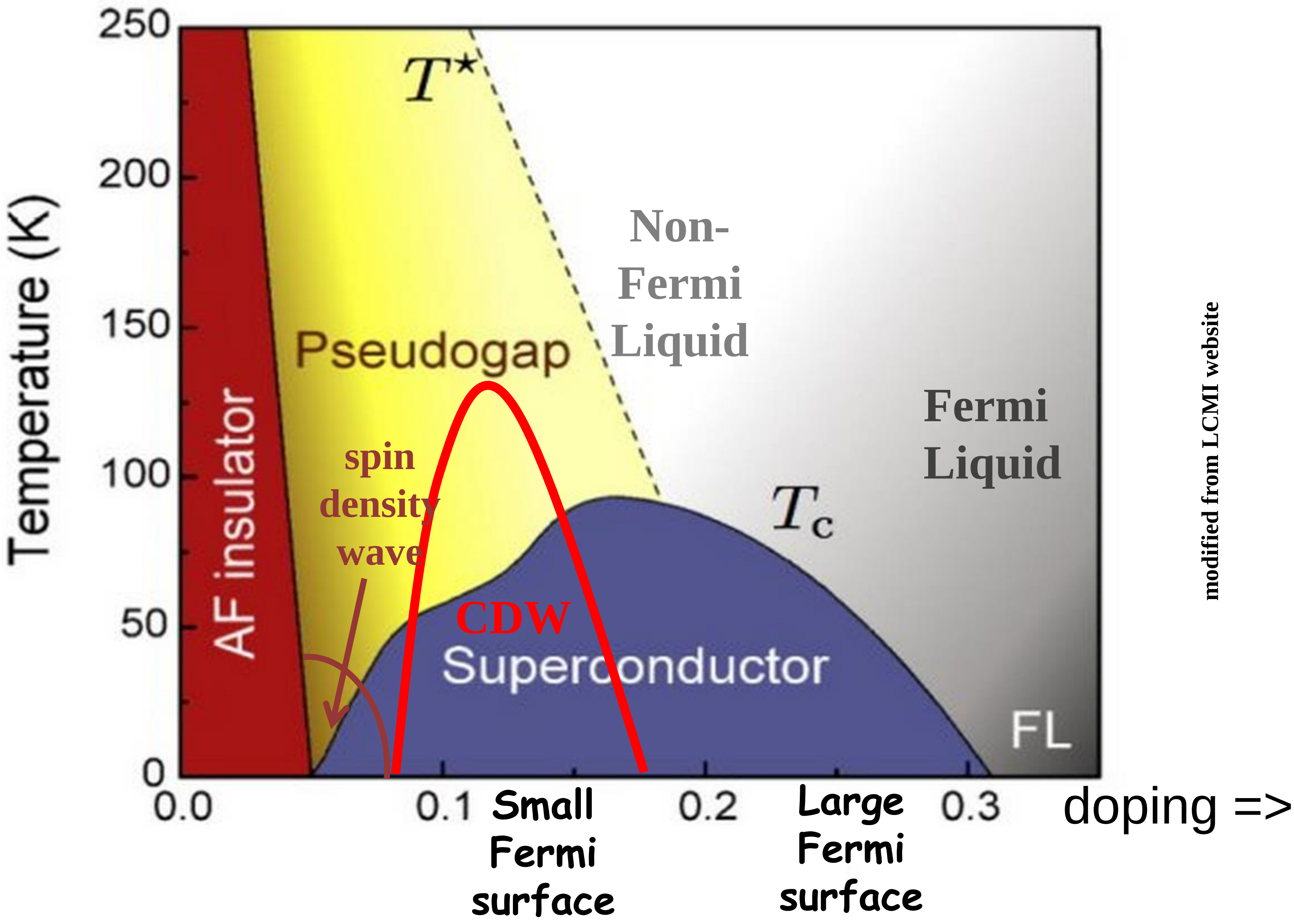


not quite saturated

Above T_c:
no effect of
B-field on CDW

Chang, J. *et al.* Nature Phys. **8**, 871 (2012)

High-T_c properties versus hole doping including CDW



What is the *structure* of Charge Density Waves?

Measure sufficiently many (>200) different X-ray diffraction satellites due to the CDWs to derive the atomic displacements that fit the data. Needs zero *B*-field for flexibility

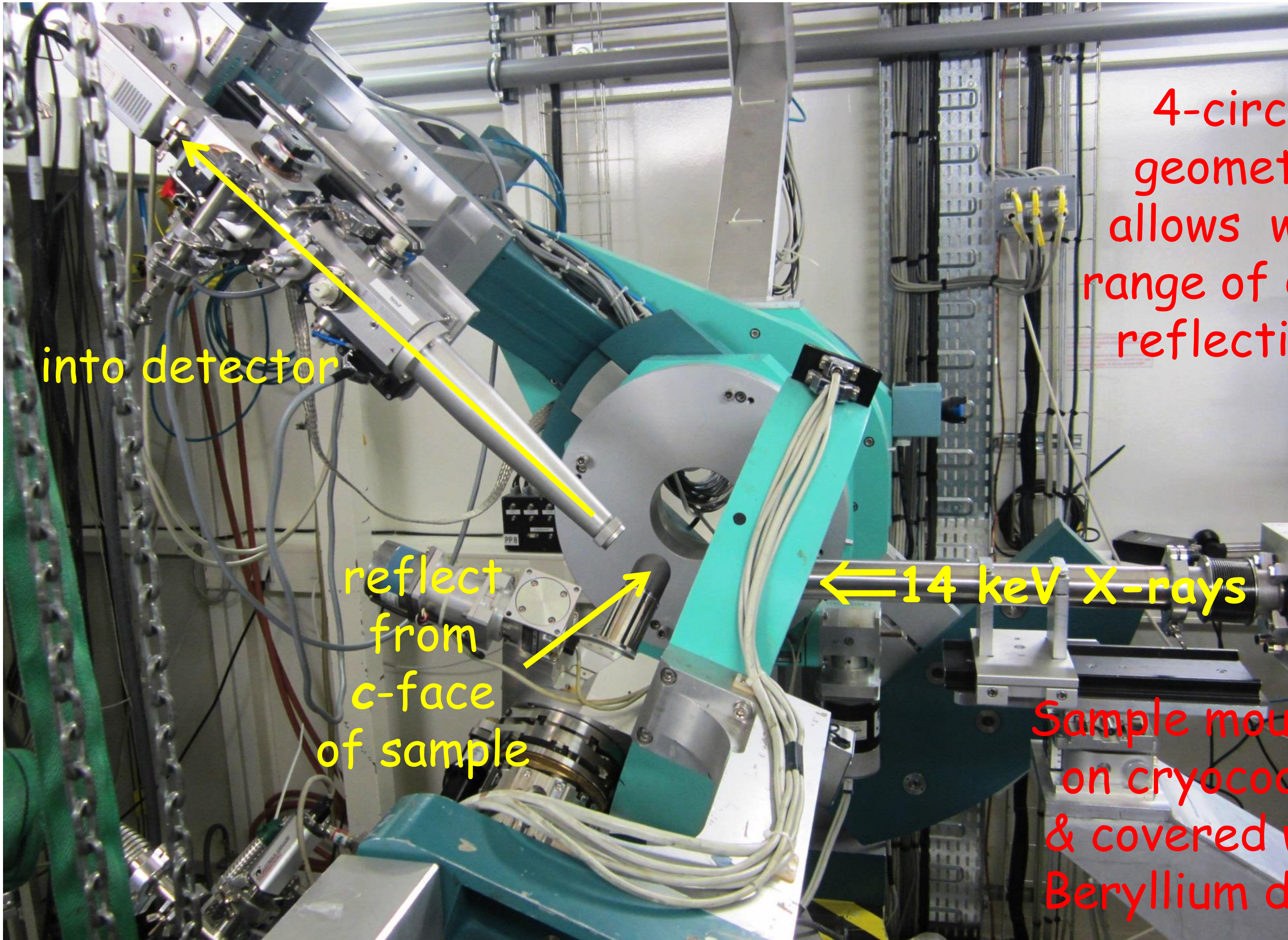
If possible, deduce something about the physics of the CDW from these atomic displacements

But non-resonant X-rays see *ALL* the 13 atoms in the YBCO unit cell, so the results are difficult to analyse!

Group theory allows us to solve this problem; the symmetry of the derived displacements is quite surprising

We then use the properties of the CDW to propose how the Fermi Surface reconstruction occurs
- and learn something about High-T_c

Apparatus: the XMaS (UK) beamline at ESRF



Considerations used in analysis of results

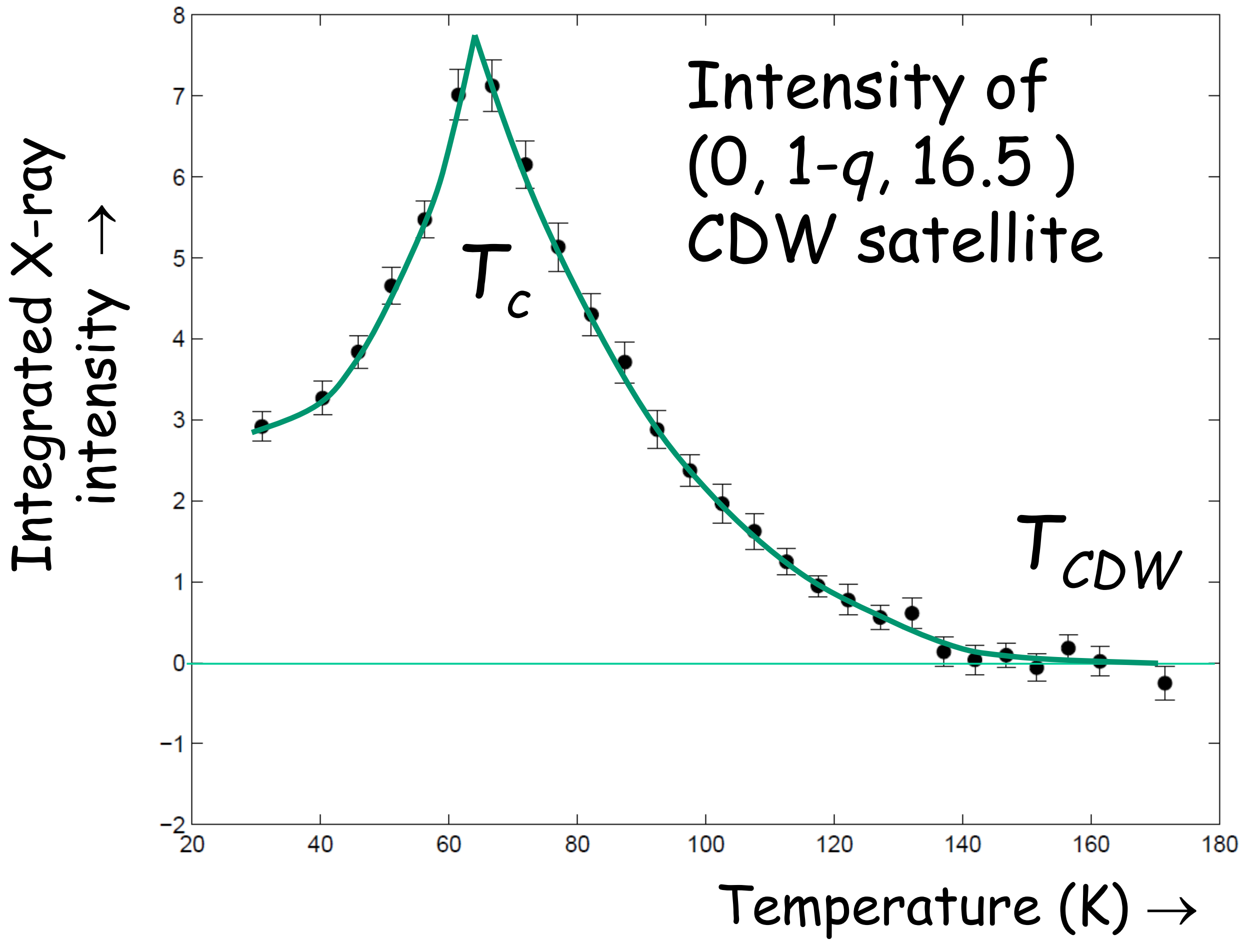
Non-resonant X-rays are insensitive to small charge density changes.
Instead they respond to the associated/resultant atomic *displacements* from their usual positions.
(because *ALL* the electrons in a displaced atom scatter X-rays)

A single CDW can be described by an incommensurate ***q***-vector along either the ***x*** or ***y*** (***a*** or ***b***) crystal directions.

Adjacent unit cells in the ***c***-direction are in antiphase
(Doubled cell indicated by CDW satellites at half-integral ℓ)

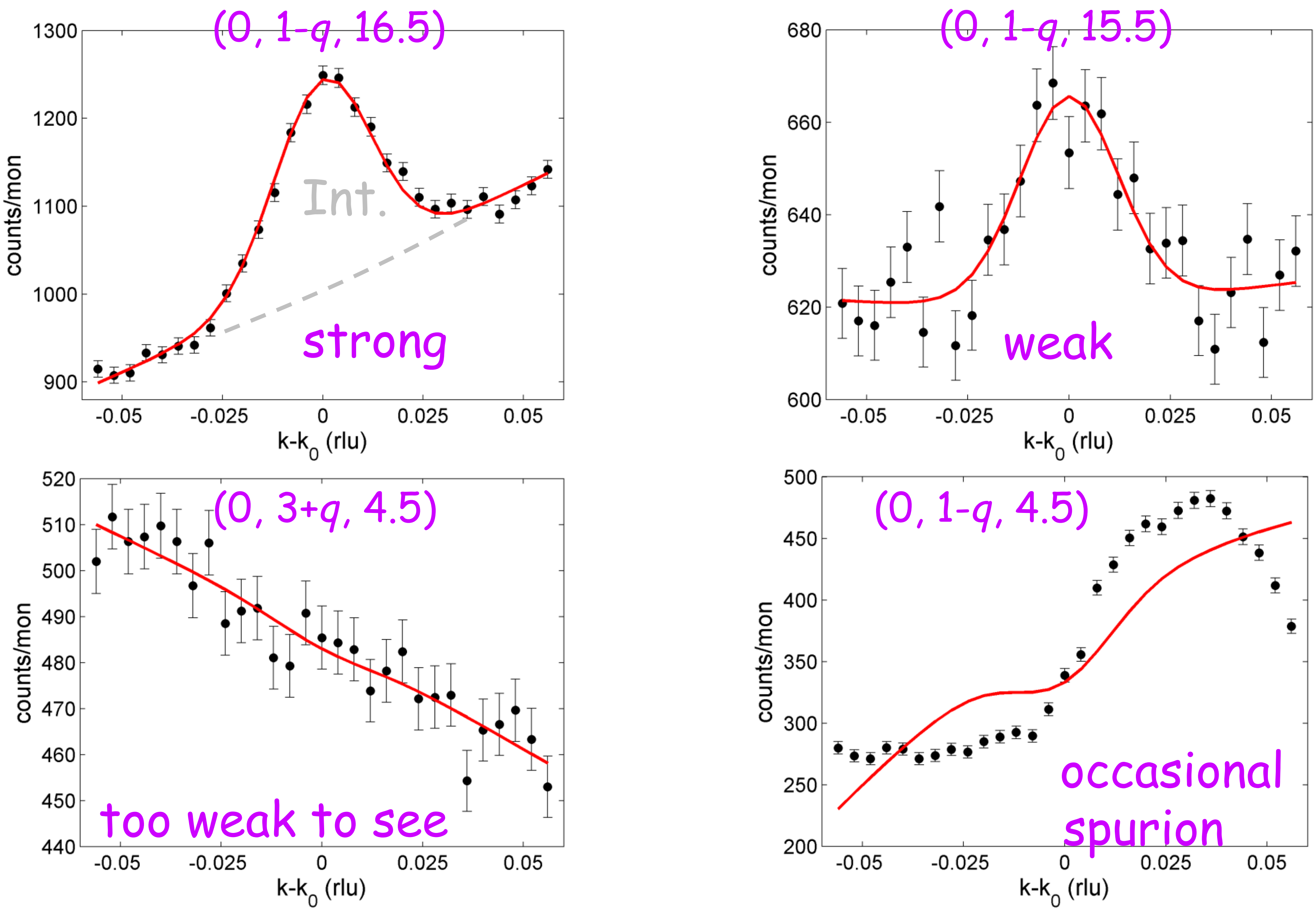
CDWs are longitudinal, with atomic displacements
(e.g. for ***q*** // ***y***) along *both* ***y*** & ***z*** directions.

Temp-dependence of CDW order in $YBCO_{6.54}$

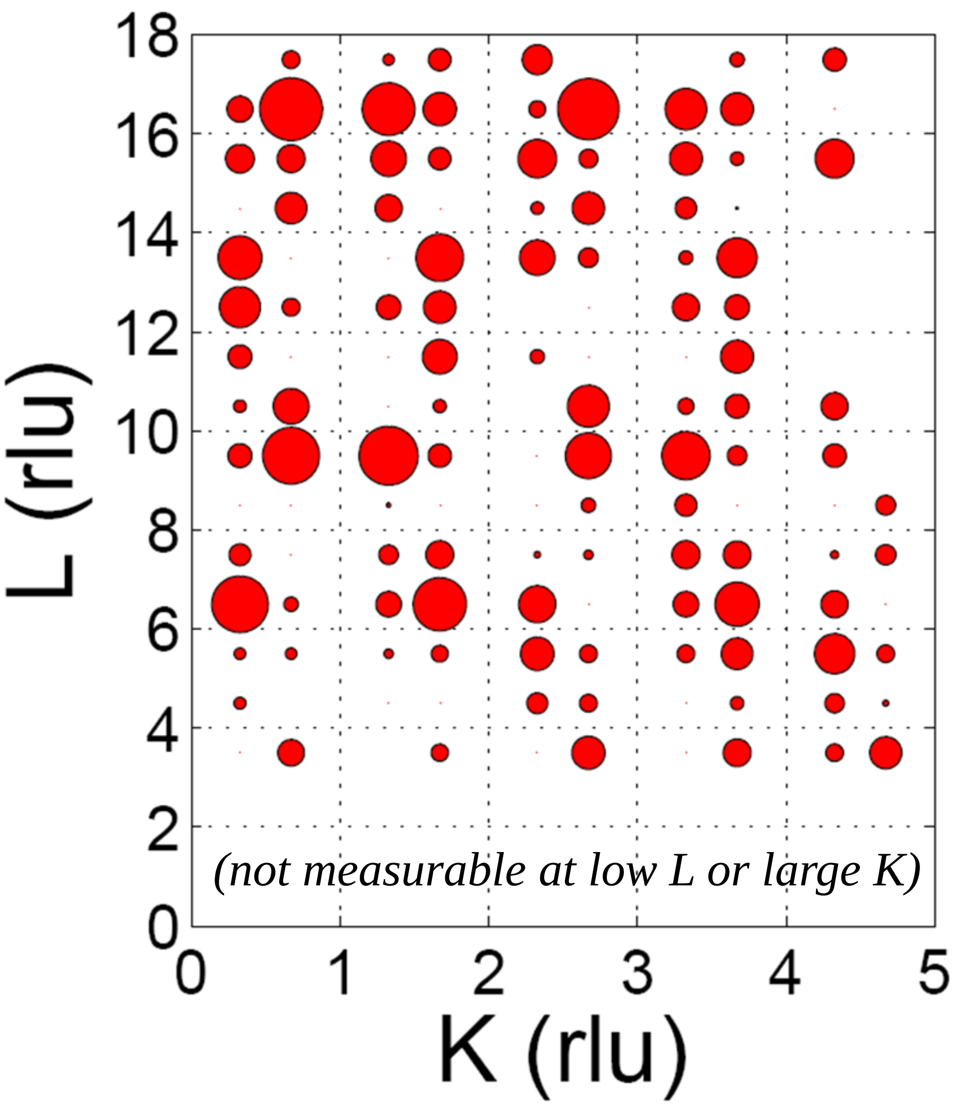


Make all observations of CDW intensities at
T_c (superconductivity) = 60 K

Typical observations of CDW satellites at 60 K



A typical CDW satellite intensity pattern



You can always get from a model to the diffraction pattern - but not vice versa

not a simple pattern so the displacements do not involve just one or two atoms

A total of 269 satellite positions observed for $\boldsymbol{q}_b$ and 193 for $\boldsymbol{q}_a$

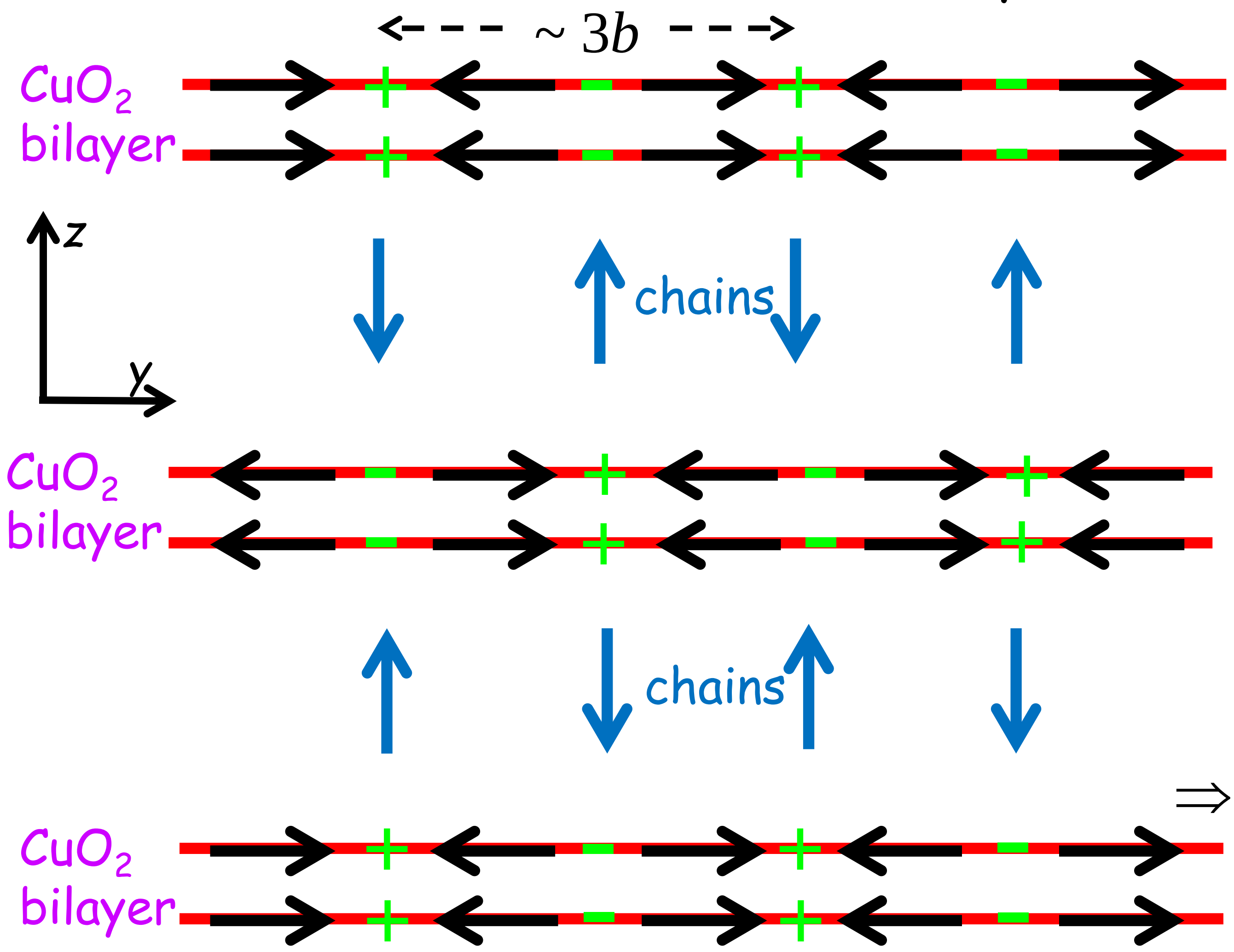
Area of circle $\propto$ Intensity

blank = not measured

Expected structure of the CDW order

We expect atomic displacements with this *symmetry*

$\leftrightarrow$ motion is even in *z* about bilayer , and $\uparrow\downarrow$ is odd in z



CDW $\Leftrightarrow$ atomic displacements

Next unit cell in antiphase ($\equiv \ell = 0.5$)

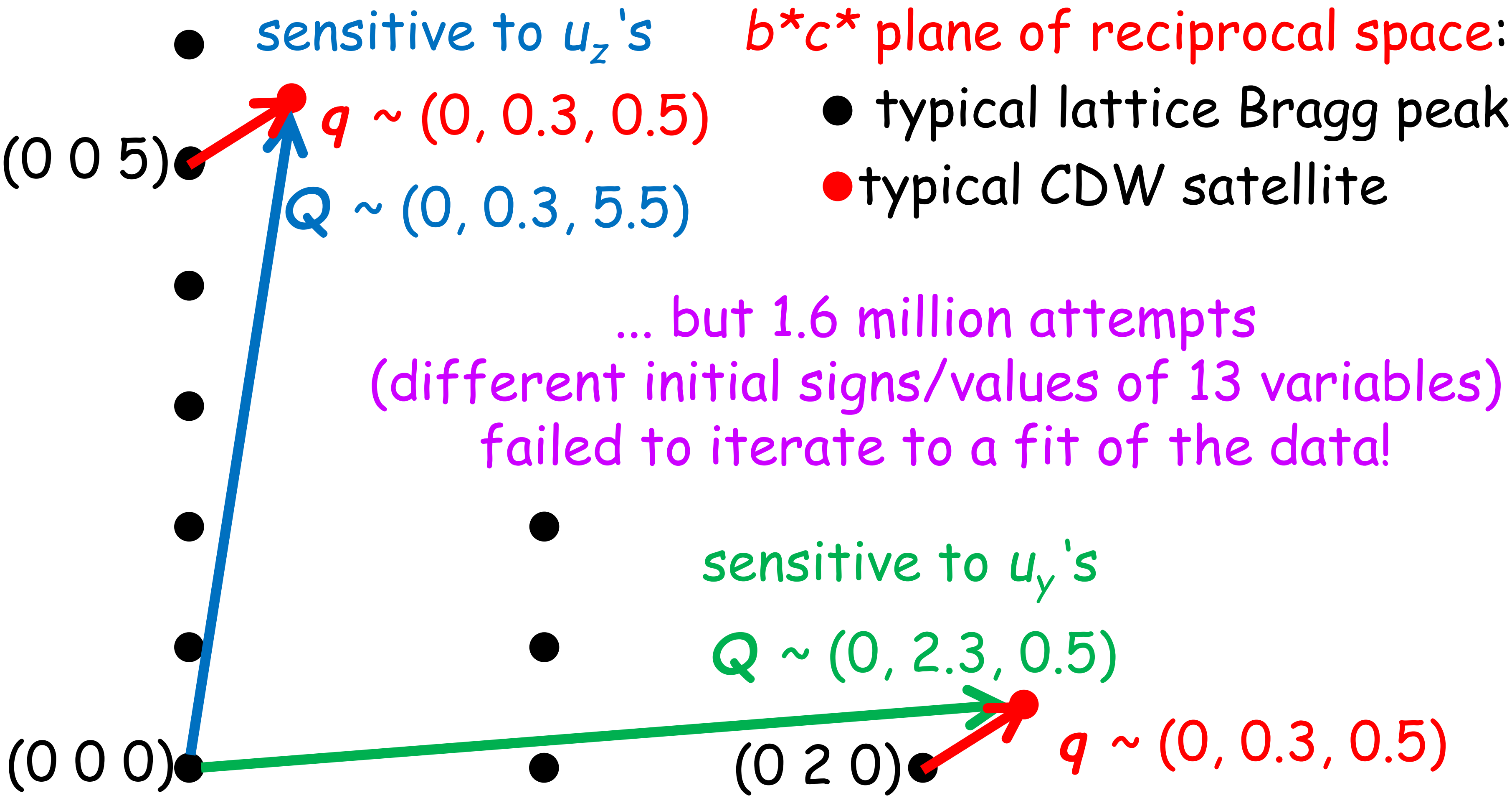
also $\exists$ *c*-axis displacements

$\Rightarrow$ total of 13 atomic motion variables to fit the data

How to deduce atomic displacements *u* in the CDW

CDW satellite intensities are proportional to $(\mathbf{Q}.\mathbf{u})^2$

So we can detect basal and ***c***-axis displacements u_y u_z.

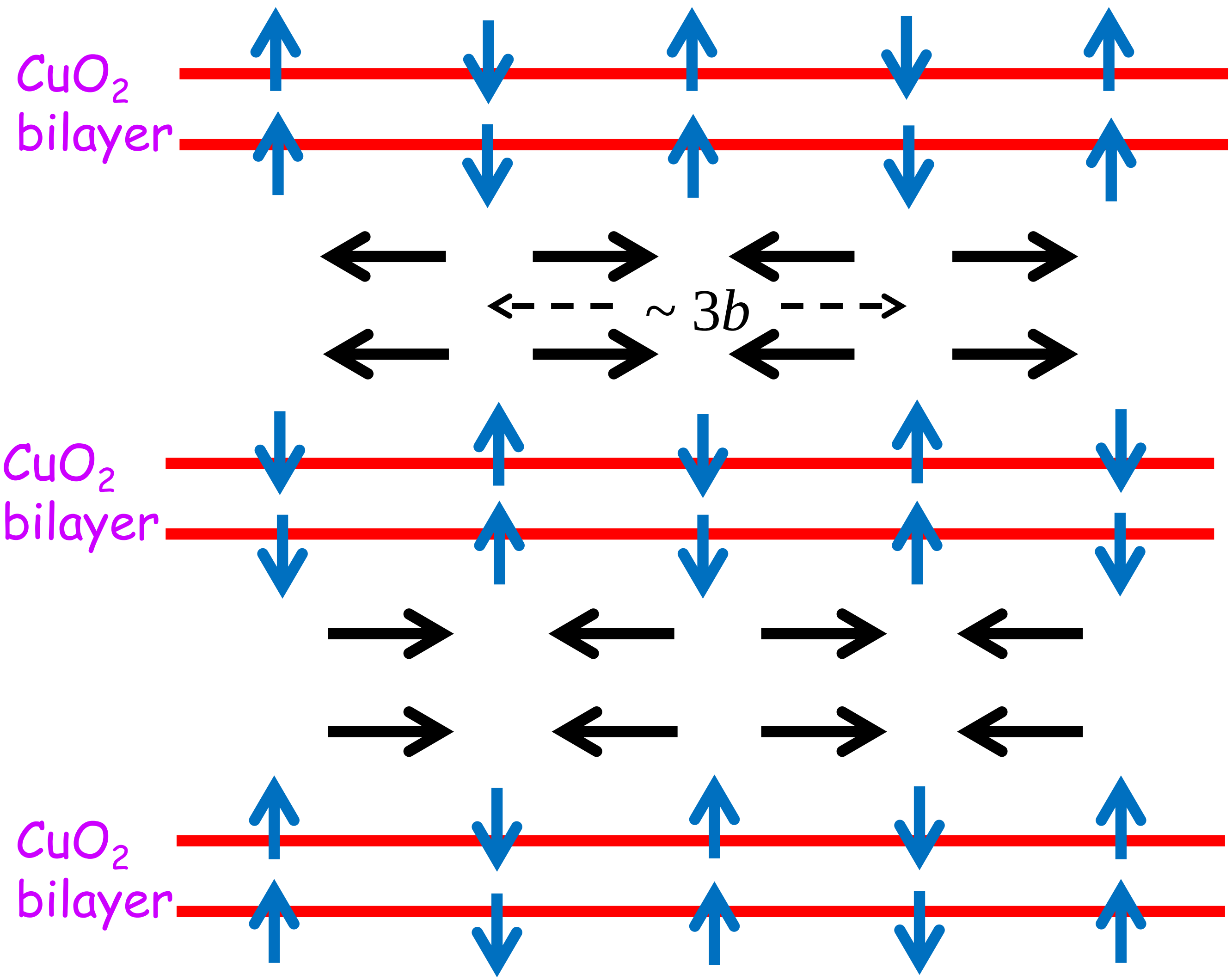


... but 1.6 million attempts
(different initial signs/values of 13 variables)
failed to iterate to a fit of the data!

Only other possible model for the atomic movements

We are forced to consider displacements of this *symmetry*

↑↓ motion even in *z* about bilayer, and ↔ odd in z



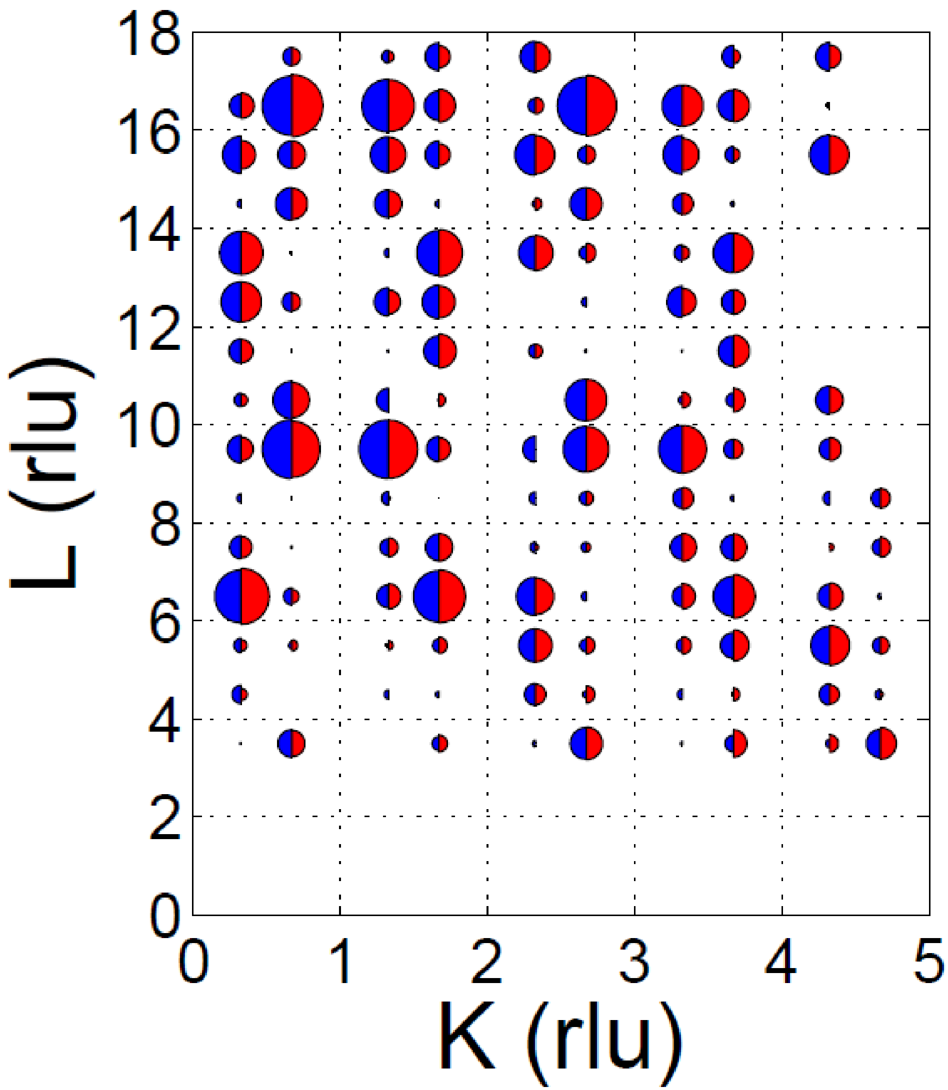
CuO_2 bilayers *sheared* – not compressed

Next unit cell in antiphase (≡ ℓ = 0.5)

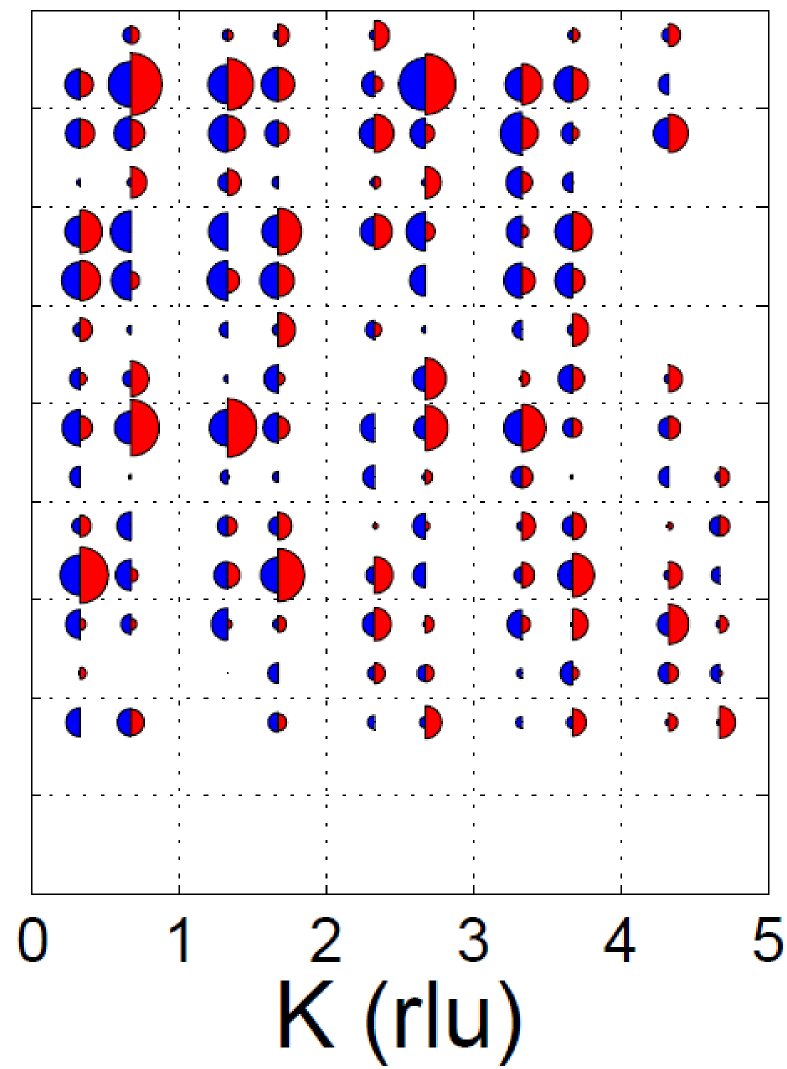
y & ***z*** atomic displacements => total of 13 variables

fits the data!

Good fit - bad fit…



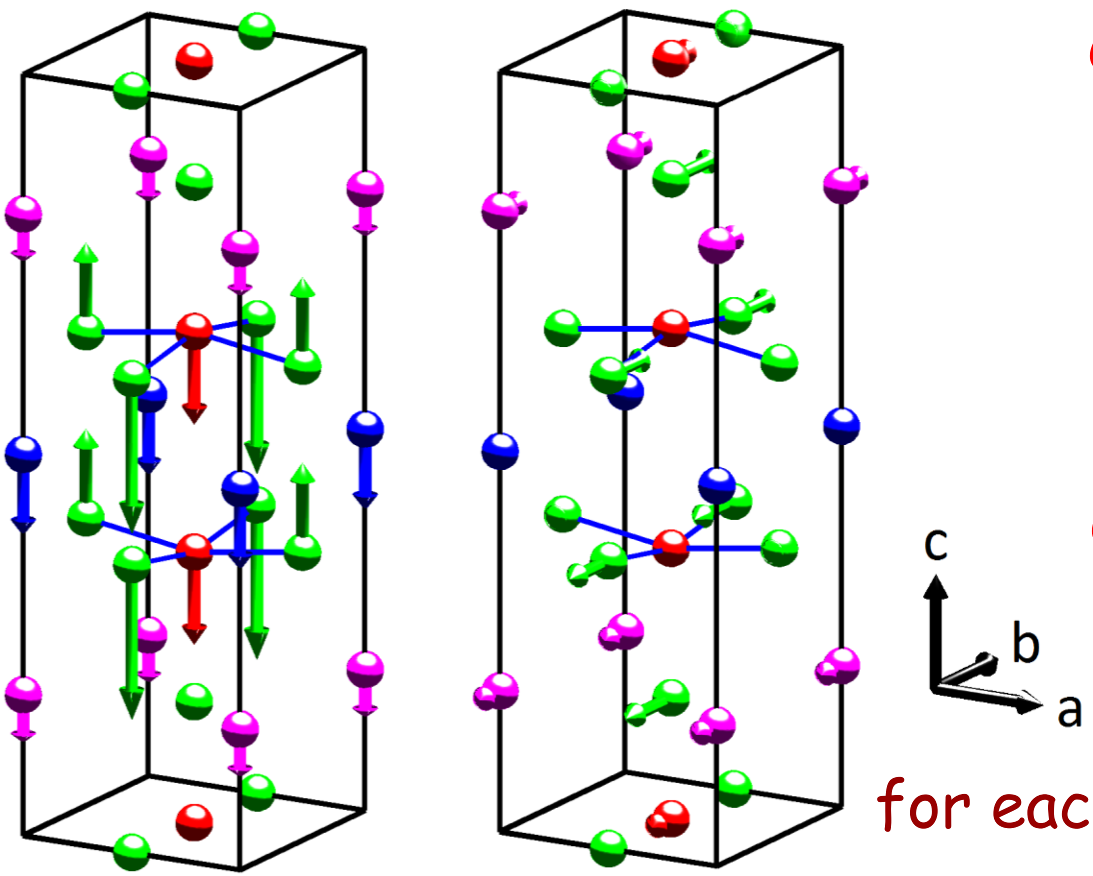
CuO$_2$ bilayers sheared



CuO$_2$ bilayers compressed

The motif which is modulated to form the CDW

- from the results of the good fit to the $\boldsymbol{q}_b$ mode



$\boldsymbol{u}_c$ $\boldsymbol{u}_b$

CuO chains

BaO layer

CuO_2 plane

Y layer

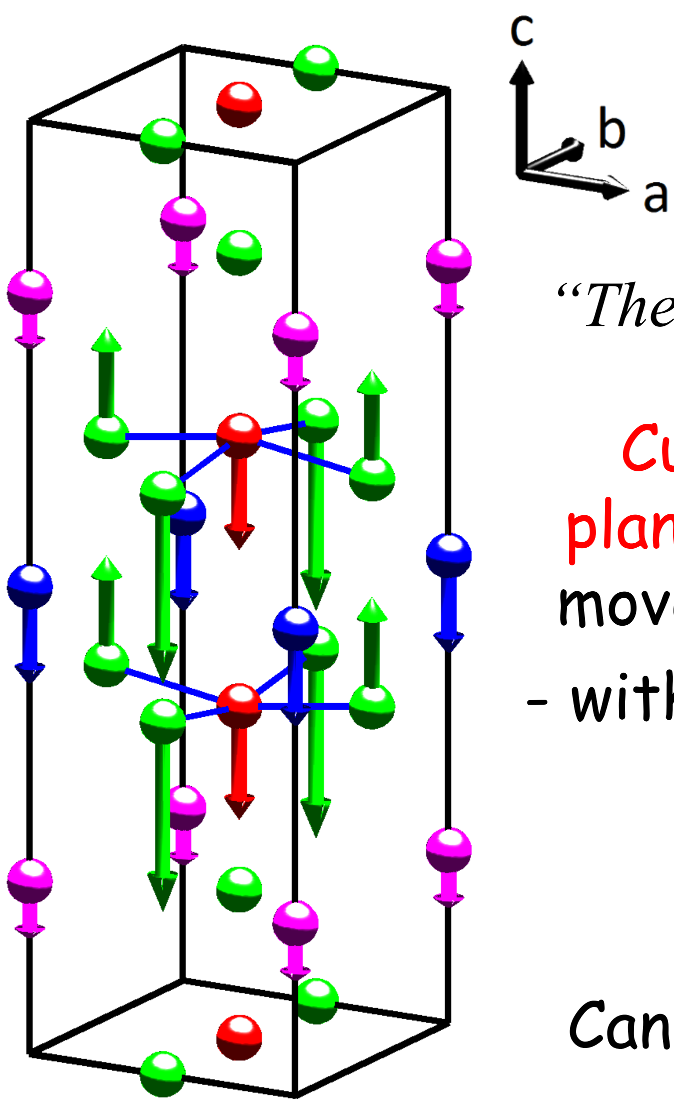
CuO_2 plane

for each atom, $\boldsymbol{r}_0 \Rightarrow \boldsymbol{r}_0 + \boldsymbol{u}(\boldsymbol{r}_0)$

$\boldsymbol{u}(\boldsymbol{r}_0) = \boldsymbol{u}_c \cos(\boldsymbol{q}.\boldsymbol{r}_0) + \boldsymbol{u}_b \sin(\boldsymbol{q}.\boldsymbol{r}_0)$

In zero field, next unit cell along $\boldsymbol{c}$ is in antiphase

The motif which is modulated to form the CDW



- concentrating on the ***c***-axis displacements which dominate

"The change in strain is mainly out of plane"

Cu's in the planar bilayers move together

- with the Y's

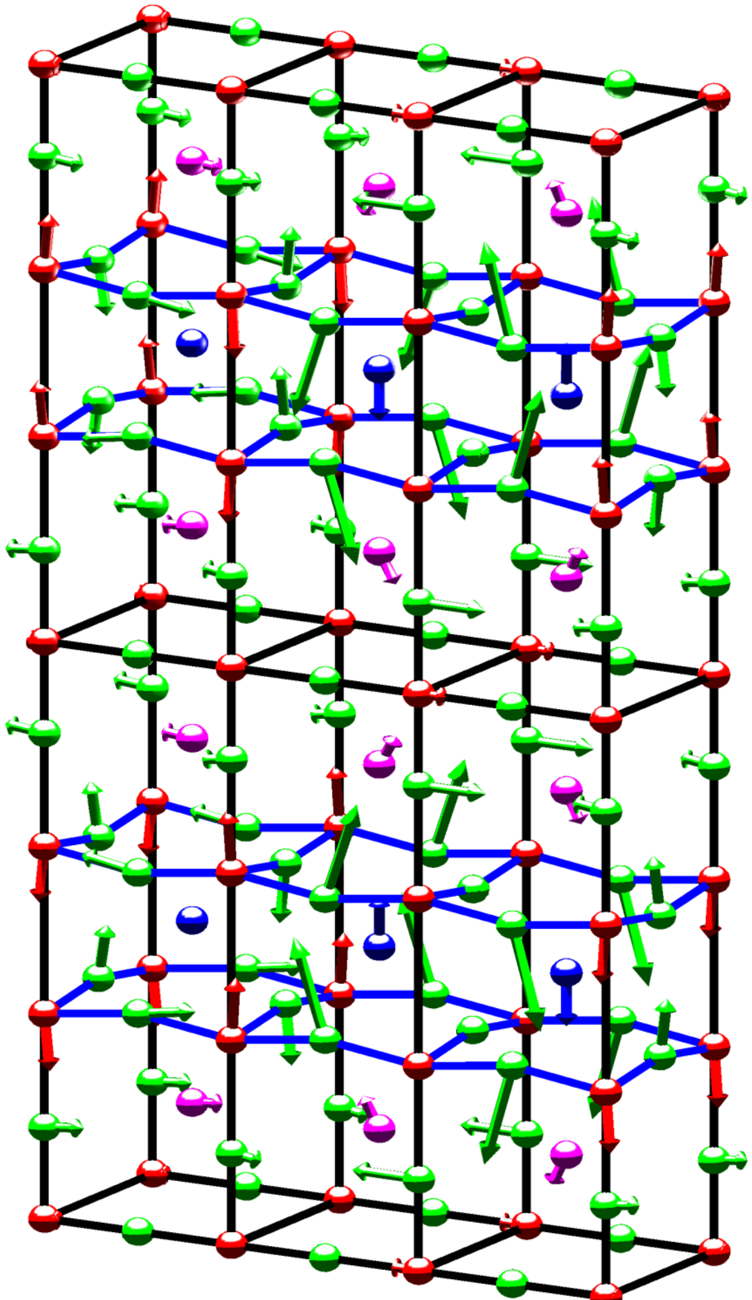
O_x & O_y move *oppositely* to each other

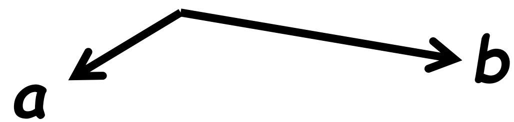
Actual amplitude ~ 10^{-3} of an atomic spacing!

Can this tiny effect be important? Yes!

CuO chains don't move (symmetry)

Resulting modulated ionic displacements





period only ~ 3 unit cells
so π phase change in only $1\frac{1}{2}$ cells

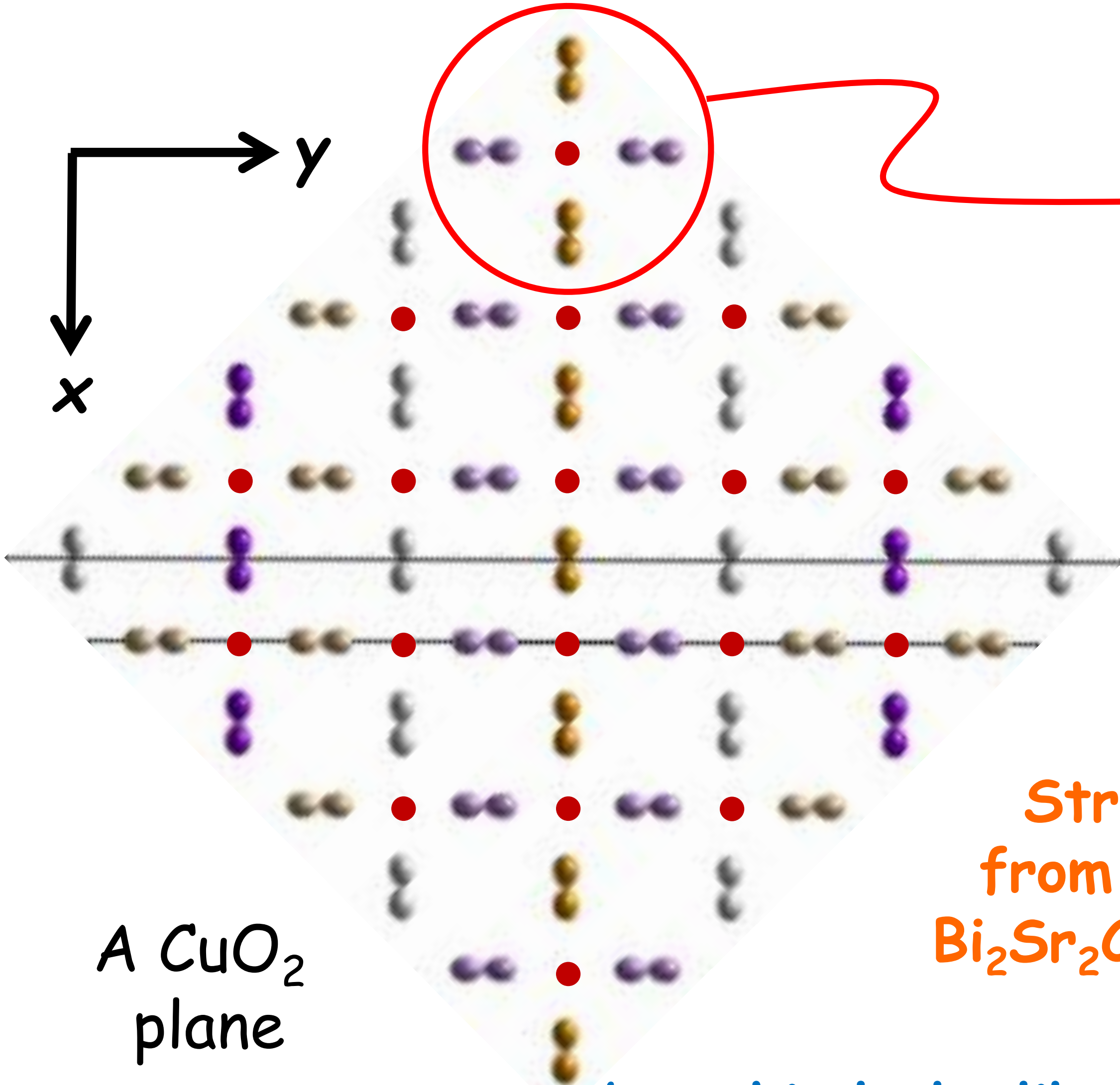
not tilted CuO_5 half-octahedra

Plus a similar modulation in the perpendicular direction

Almost certainly in the same region of space:

"double-***q***" or "biaxial" order => Fermi surface reconstruction

STM suggests "*d*-density wave" on planar oxygens



A CuO_2 plane

a small charge transfer from one pair of oxygen orbitals to the other

● Cu
O_x
O_y

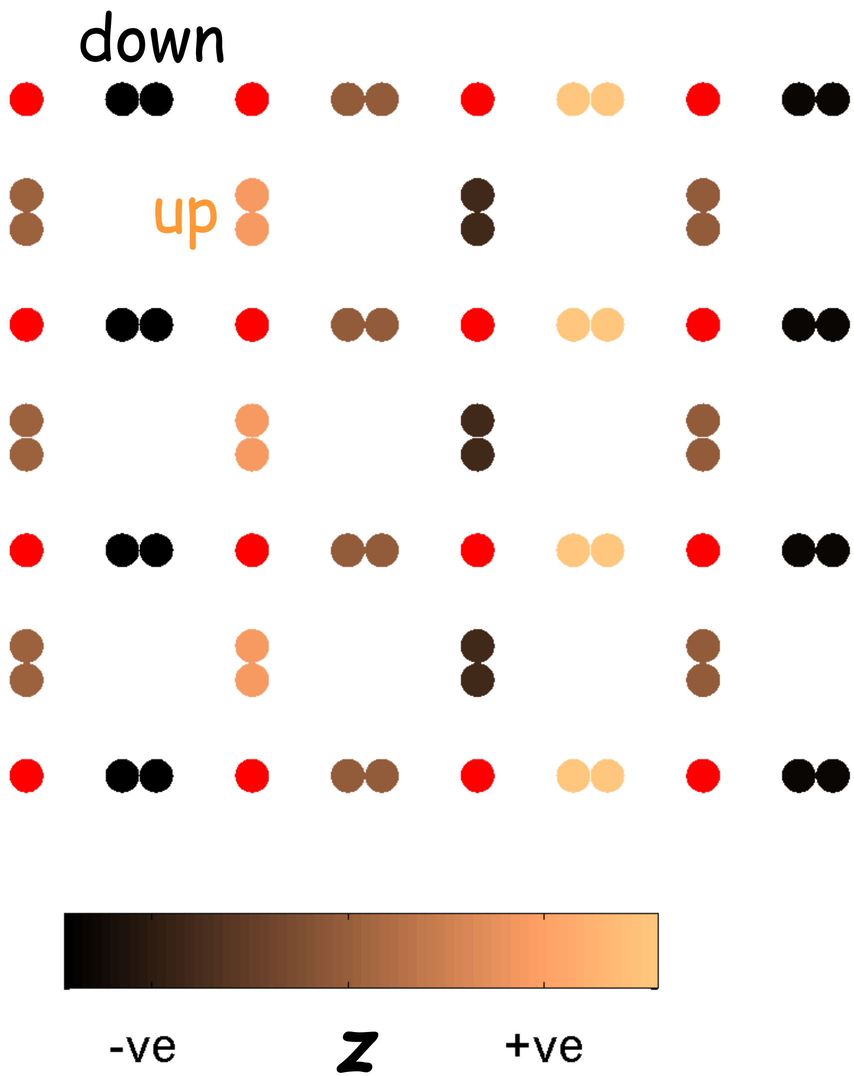
Spatially modulated along y

Strong evidence for this from STM measurements in $Bi_2Sr_2CaCu_2O_8$ & $Ca/NaCuO_2Cl_2$

... but this looks like our ***unsuccessful*** model !

S. Sachdev & J.C. Seamus Davis *group*, PNAS 2014

A plot of the modulated oxygen *z*-displacements



for a single CDW mode

modulation direction ➔

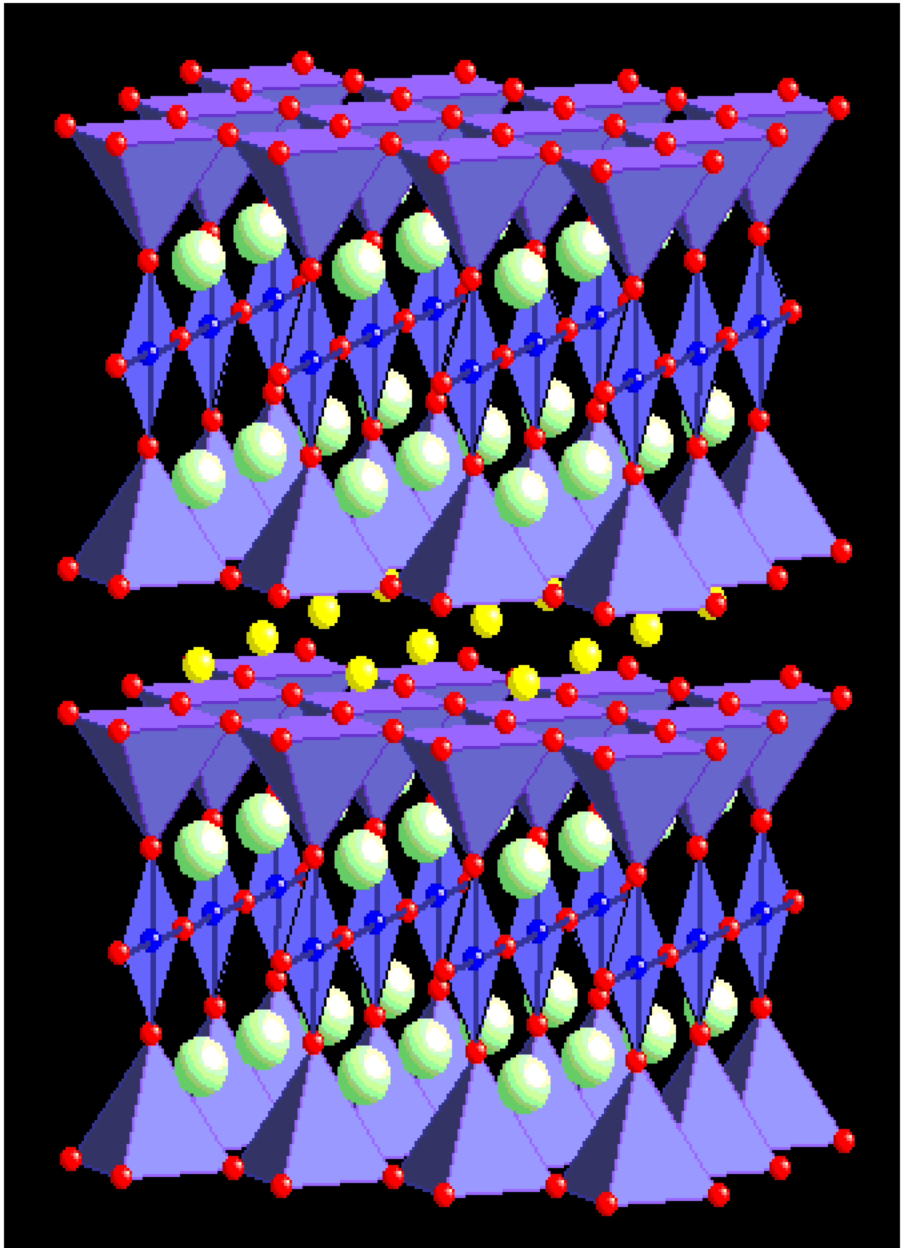
You have seen this pattern before…

Motion of an ion in the *z*-direction can alter the local doping

So our CDW shear *is* a "bond *d*-density wave"

CDW Structure determination: Nature Comms. 2015

Electron states in a CuO_2 bilayer in $YBCO_{6.5}$



YBCO structure from: www.ncl.ox.ac.uk/icl/heyes/structure_of_solids/lecture4/lec4.html

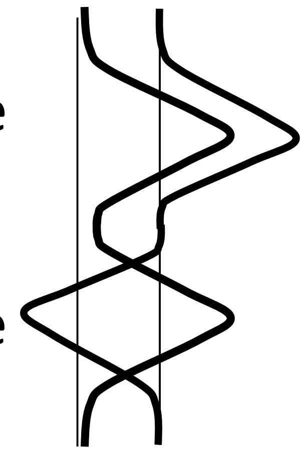
Superconductivity resides mainly in the CuO_2 planes

Cu O chains: O ½ occupied – electrically inactive

Cu O_2 plane

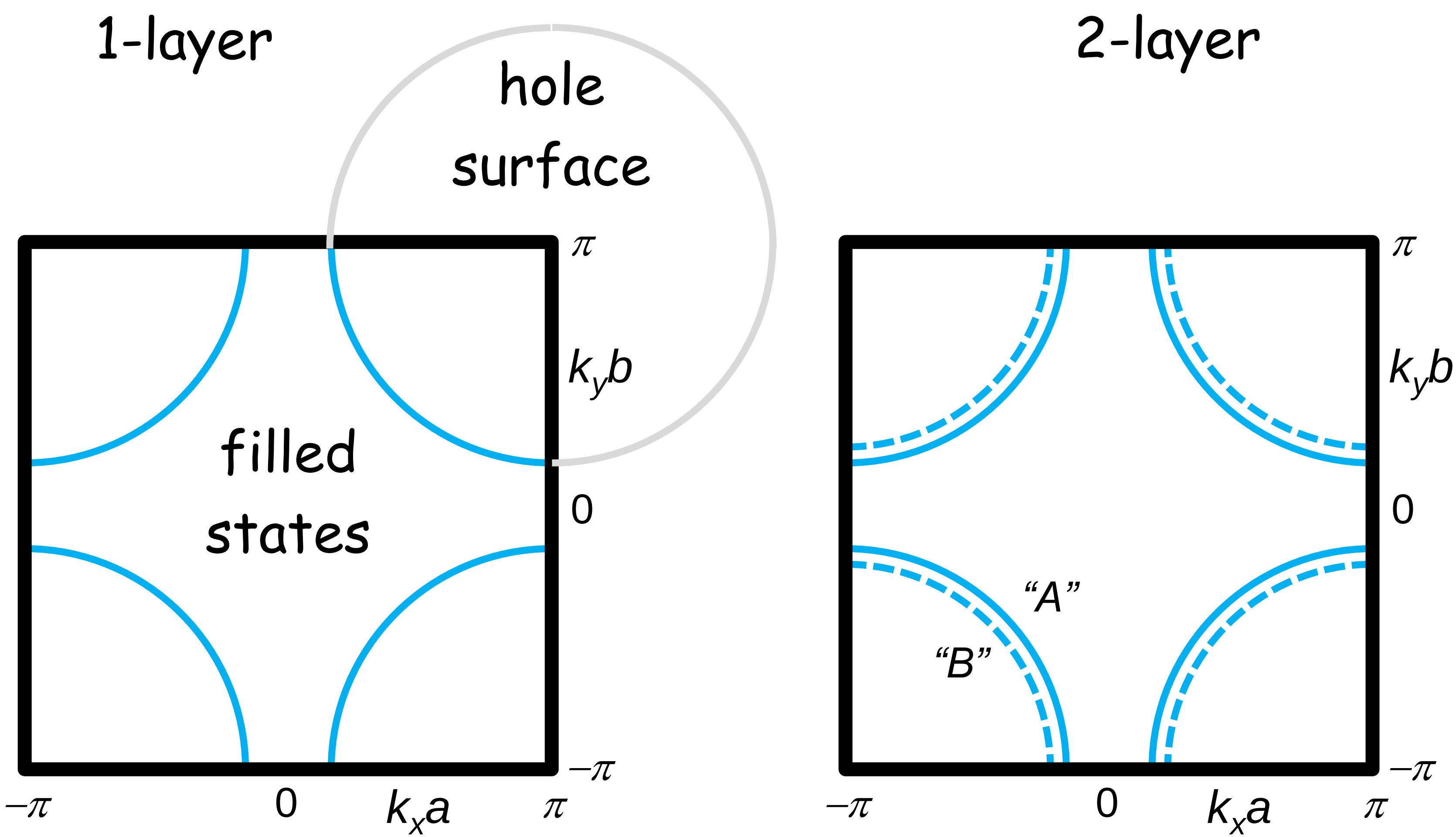
Y layer

Cu O_2 plane



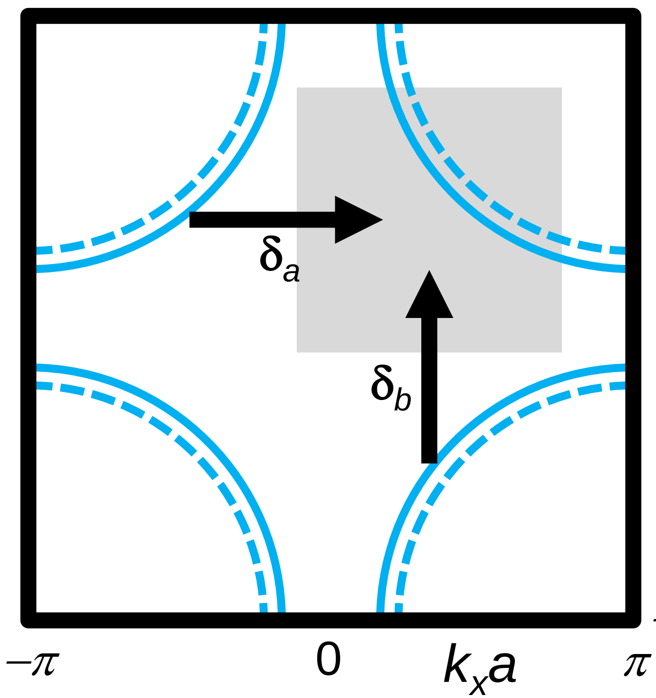
There are two ways of combining the wavefunctions of the states in the two halves of a bilayer

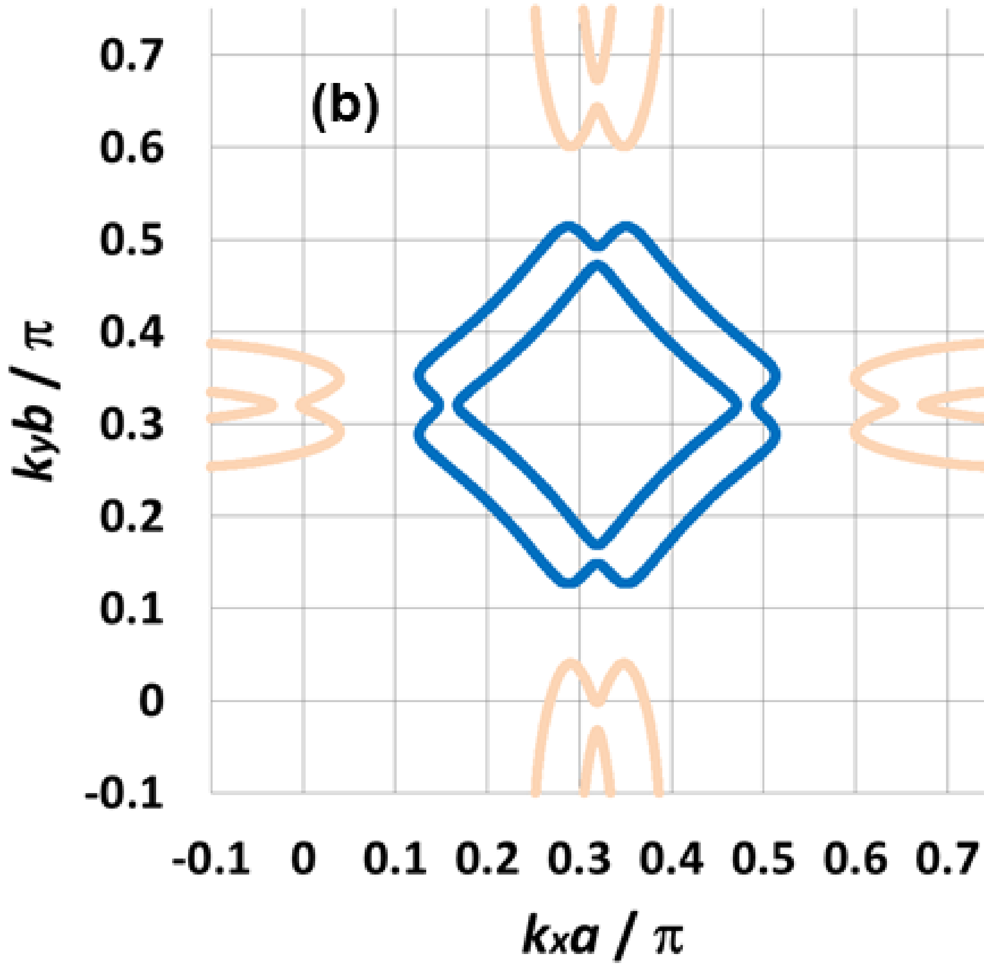
Single-layer & Bilayer Fermi Surfaces – no reconstruction



Reconstruction by CDW with basal wavevectors δ_a & δ_b

states can pick up wavevector of CDW



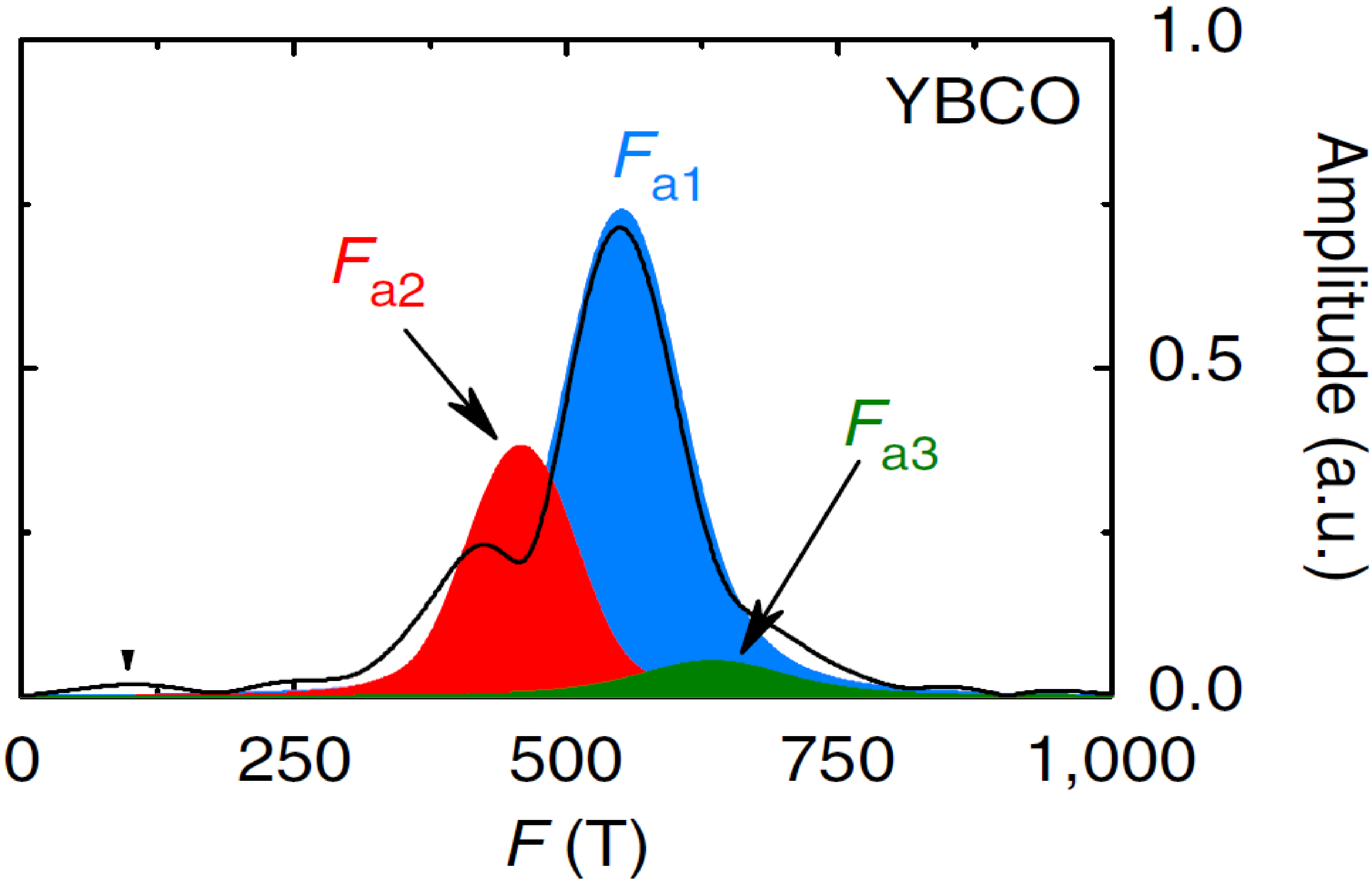


and may hybridise where degenerate

○ A – B degeneracy

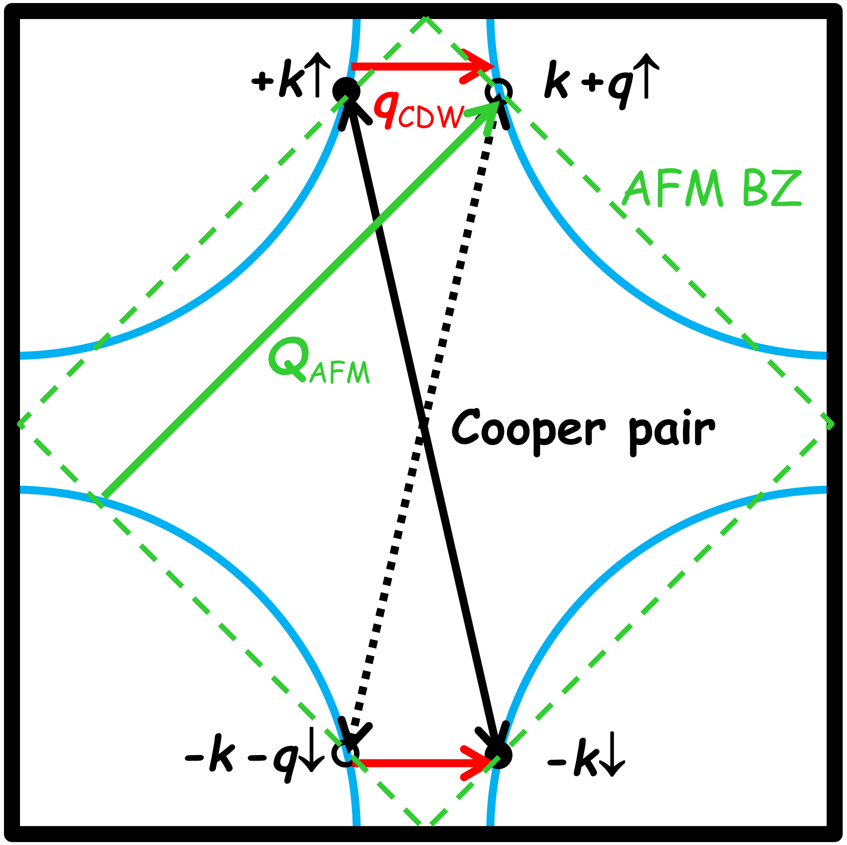
● A – A & B - B degeneracy

Due to bilayer-split FS, QO results in YBCO show multiple Fermi Surface areas



Fermi Surface Reconstruction: Phys Rev B 2016

How does this all hold together? "SU(2) theory"



A CDW can be regarded as the Bose condensation of electron-hole pairs

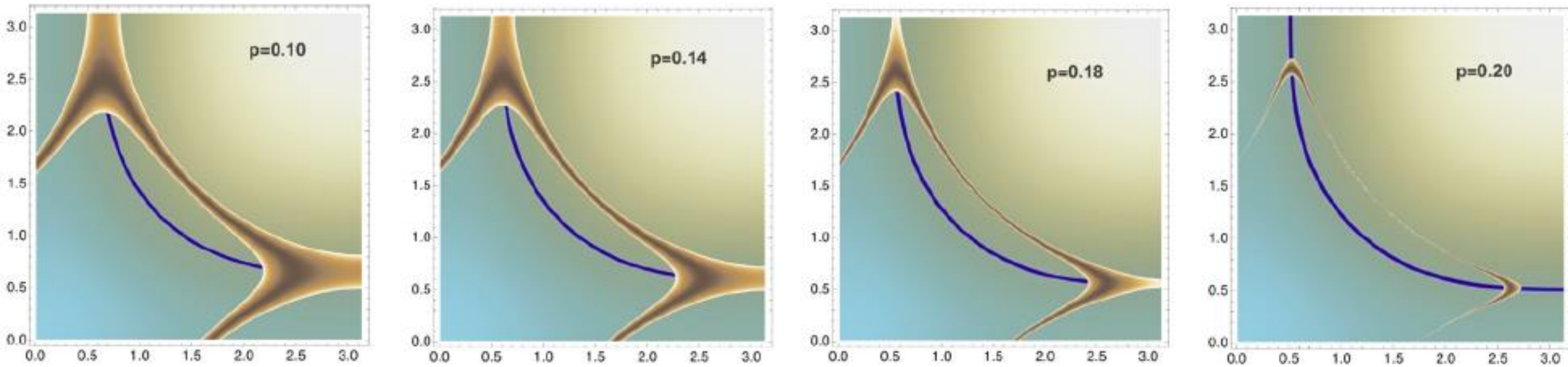
A superconductor can be regarded as the Bose condensation of electron-electron (Cooper) pairs

An underdoped cuprate has a superposition of both orders related by an SU(2) symmetry

How does antiferromagnetism come in?
The CDW occurs near the AFM "hot spots"
where the SU(2) symmetry is exact
and AFM fluctuations cause pairing

How does this all hold together? “SU(2) theory”

calculation* of SU(2) fluctuations *vs.* doping =>



It is proposed that these fluctuations create the pseudogap

- which removes the ends of the “Fermi arcs”

-and creates the conditions for the CDW
and Fermi Surface reconstruction to occur

*C. Pepin group, Phys Rev. B **95** 104510 (2017)

How High-T_c theory appears to me in 2017

CDW appears in fairly flat parallel regions of Fermi surface

Antiferromagnetic fluctuations link CDW & Superconductivity

The CDW and the superconductivity share the same *d*-wave symmetry (though they don't need to by theory), and they compete for the same electrons

Highest T_c where SU(2) fluctuations/pseudogap are reduced

Workers on LBCO or LSCO who see antiferromagnetic + CDW **stripes** as important would not agree!

Some numerology

John Bardeen, Leon Cooper & Bob Schrieffer 1957

http://superconductors.org/history.htm#resist
http://www.nobel.se/physics/laureates/1913/onnes-bio.html

Kamerlingh Onnes 1911 - explained 46 years later

- or 32 years after Quantum Mechanics came along in 1925

- 1986 + 32 = 2018 – are we approaching the explanation of HiTc?

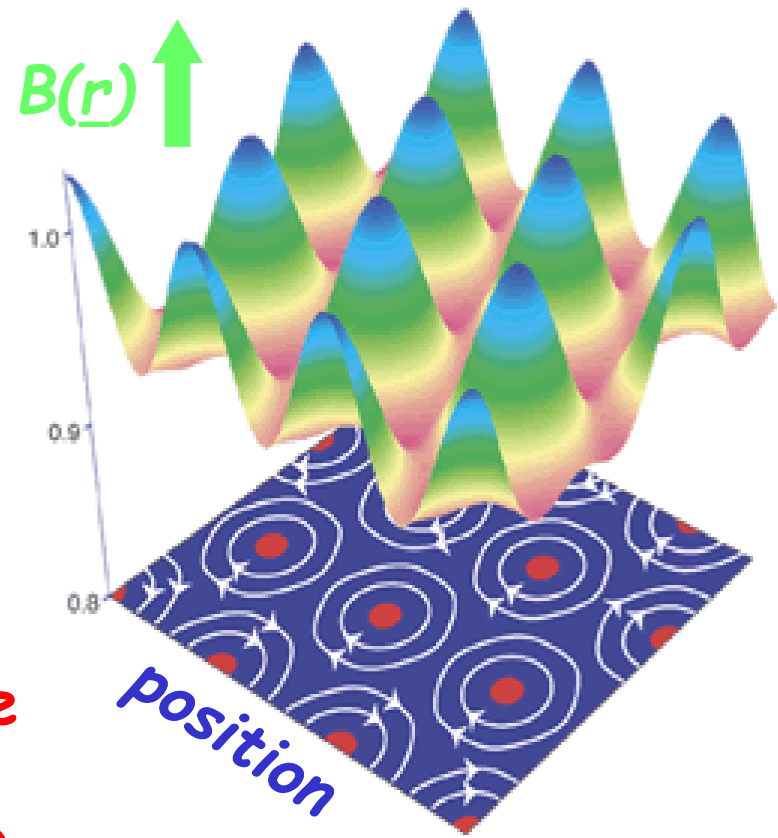
That's all Folks!

Research on YBCO at high magnetic fields

Neutrons are slightly magnetic so can be diffracted by the field in the mixed state

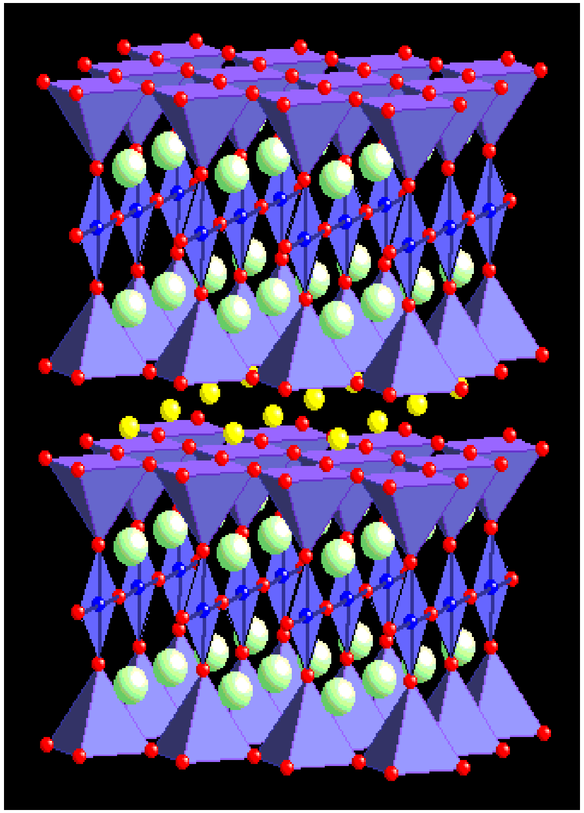
The pattern of first-order diffraction spots is identical to the arrangement of flux line nearest neighbours

- rotated by 90° about the field direction
- (property of 2-*d* lattices)



- we first used this technique at 0.2 Tesla in 1990 and have been going to higher fields ever since

Reminder of $YBa_2Cu_3O_y$ crystal structure

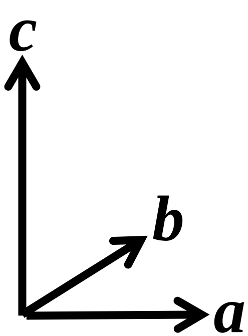


Cu O chains
Ba O layer
Cu O_2 plane
Y layer
Cu O_2 plane
Ba O layer
Cu O chains

Superconductivity mainly resides in the CuO_2 bi-layered planes

Zero doping corresponds to 1 hole per Cu in the planes (=> AFM ordered insulator)

Doping of YBCO is lowered by reducing oxygen content by removing O from the chains running along ***b***



Flux lines observed in slightly overdoped O_7; CDWs observed in $O_{<7}$

YBCO structure from: www.ncl.ox.ac.uk/icl/heyes/structure_of_solids/lecture4/lec4.html

d-wave predictions for flux lattice structure

Calculations of flux lattice structure and energy by Machida *et al.*

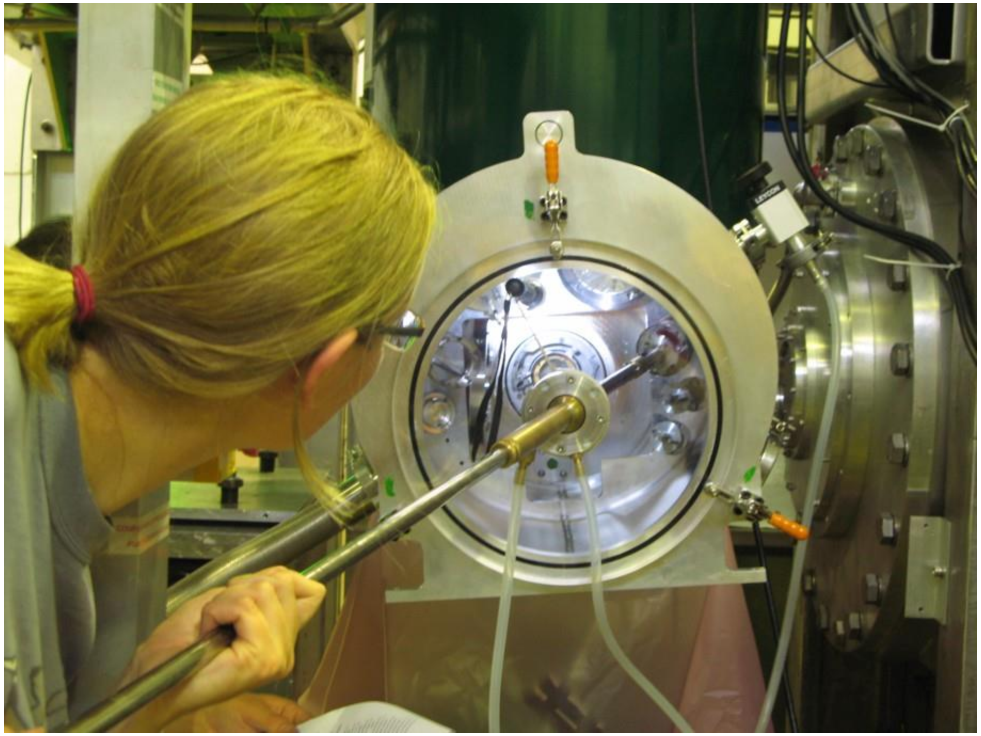
Hexagonal lattice predicted for low fields

Square lattice orientation predicted for high fields in a *d*-wave superconductor

Apparently observed in YBCO at ~ 12 T

Going to high field ~ 17 T

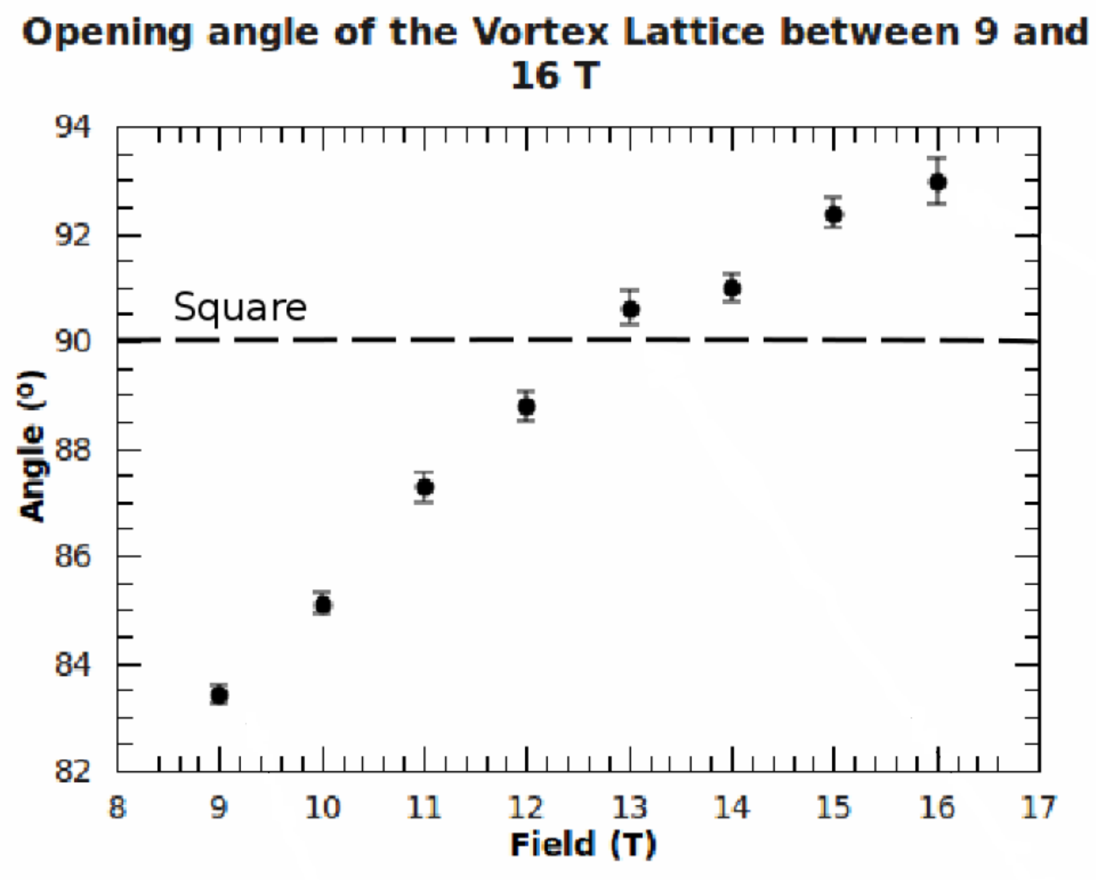
17 T, 60 mK to 300 K
B'ham cryomagnet



"side-loading" of samples into
cryostat vacuum

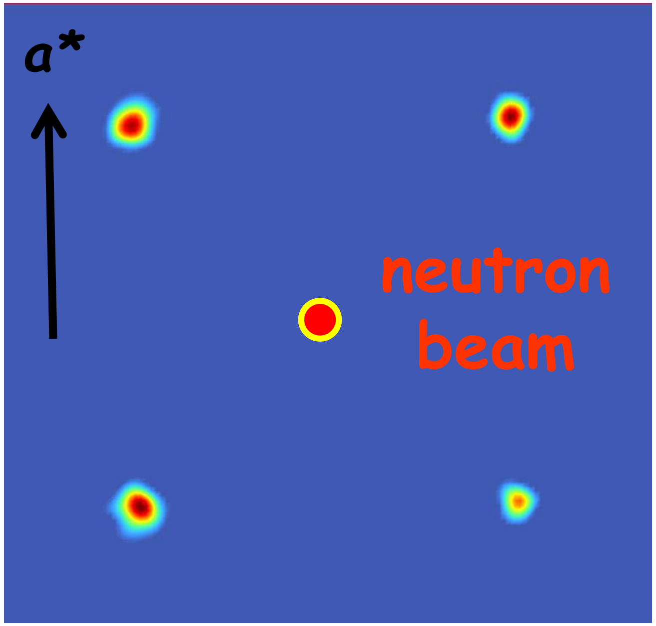
FLL in YBCO up to 16 T

Our cryomagnet used to observe FLL in YBCO up to 16 T



A.S. Cameron *et al.* *PRB* **90**, 054502 (2014)

B = 16 T diffraction pattern



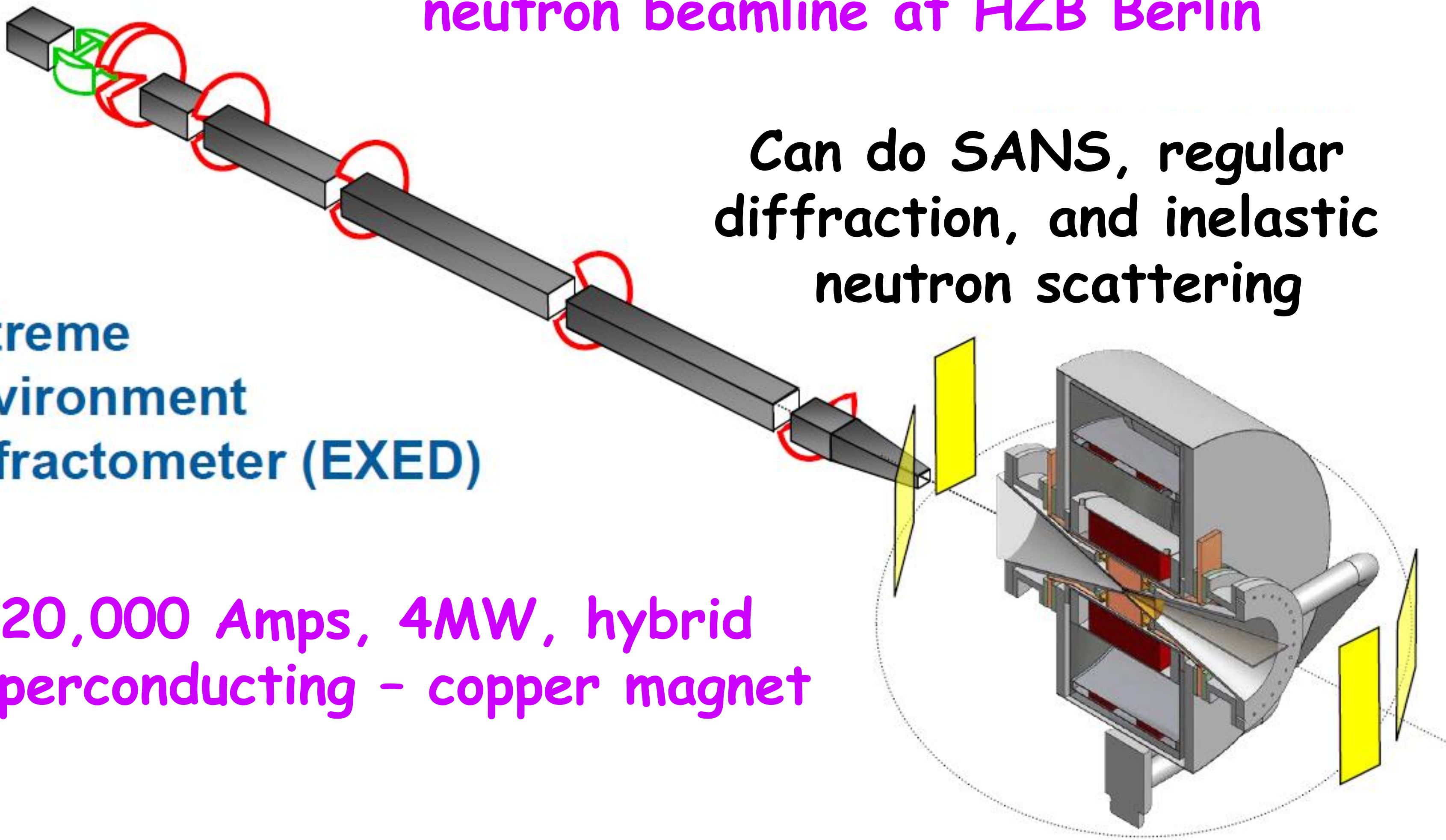
Angle goes straight through 90 degrees !

What happens at higher field?

Up to 26.2 T steady *B* field on a neutron beamline at HZB Berlin

Can do SANS, regular diffraction, and inelastic neutron scattering

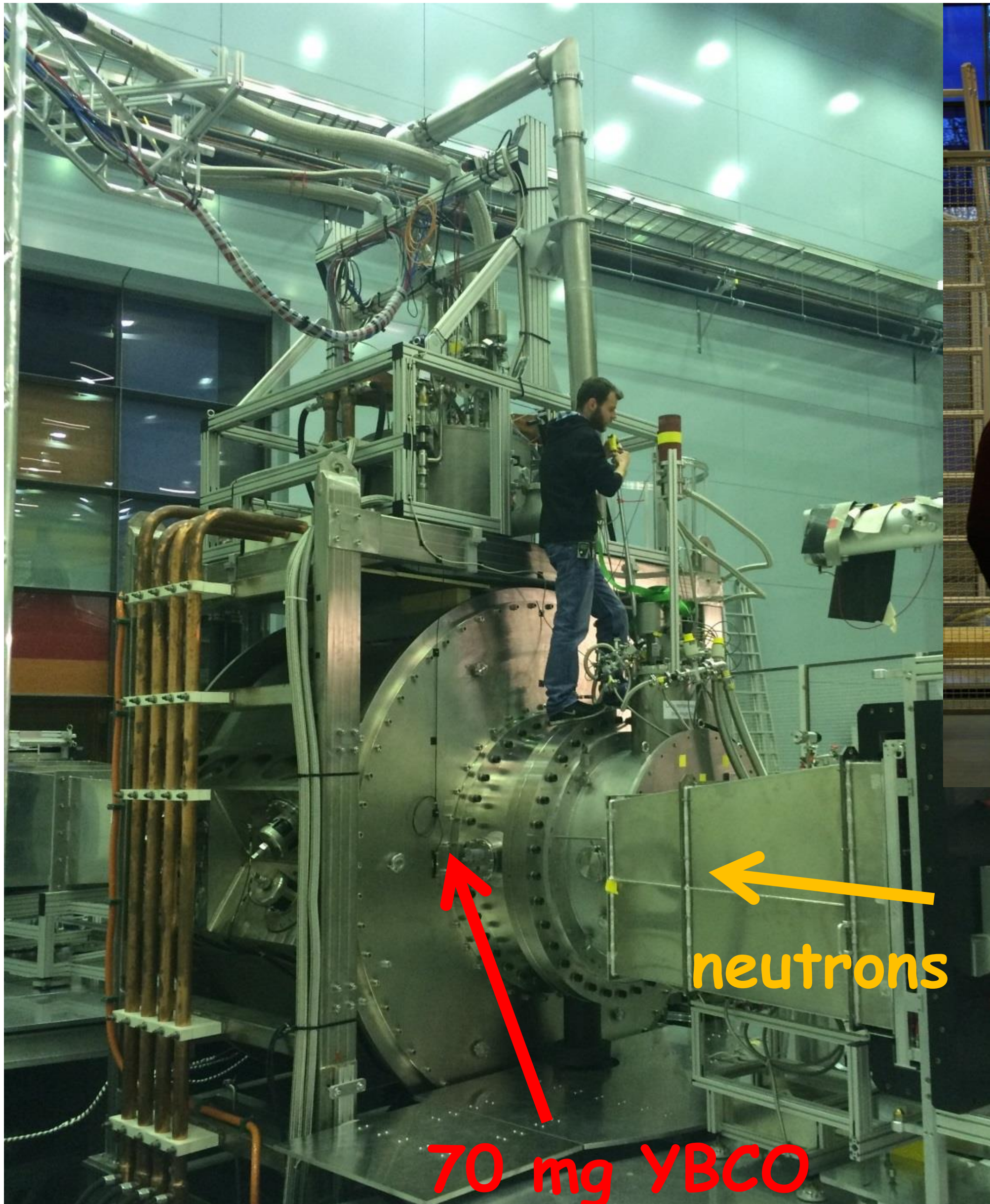
Extreme Environment Diffractometer (EXED)

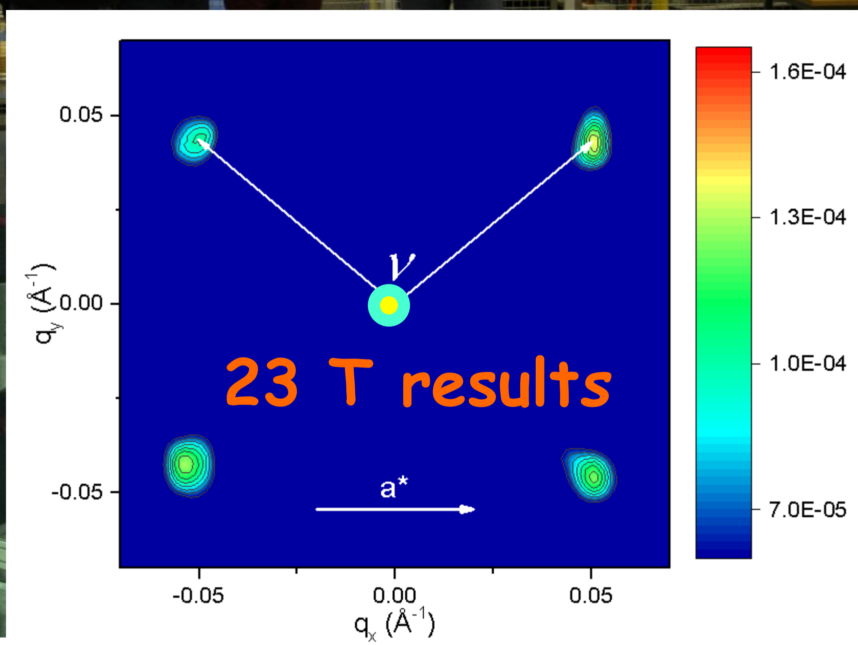


20,000 Amps, 4MW, hybrid superconducting – copper magnet

Hopefully in a few years HiTc insert cooled with liquid He

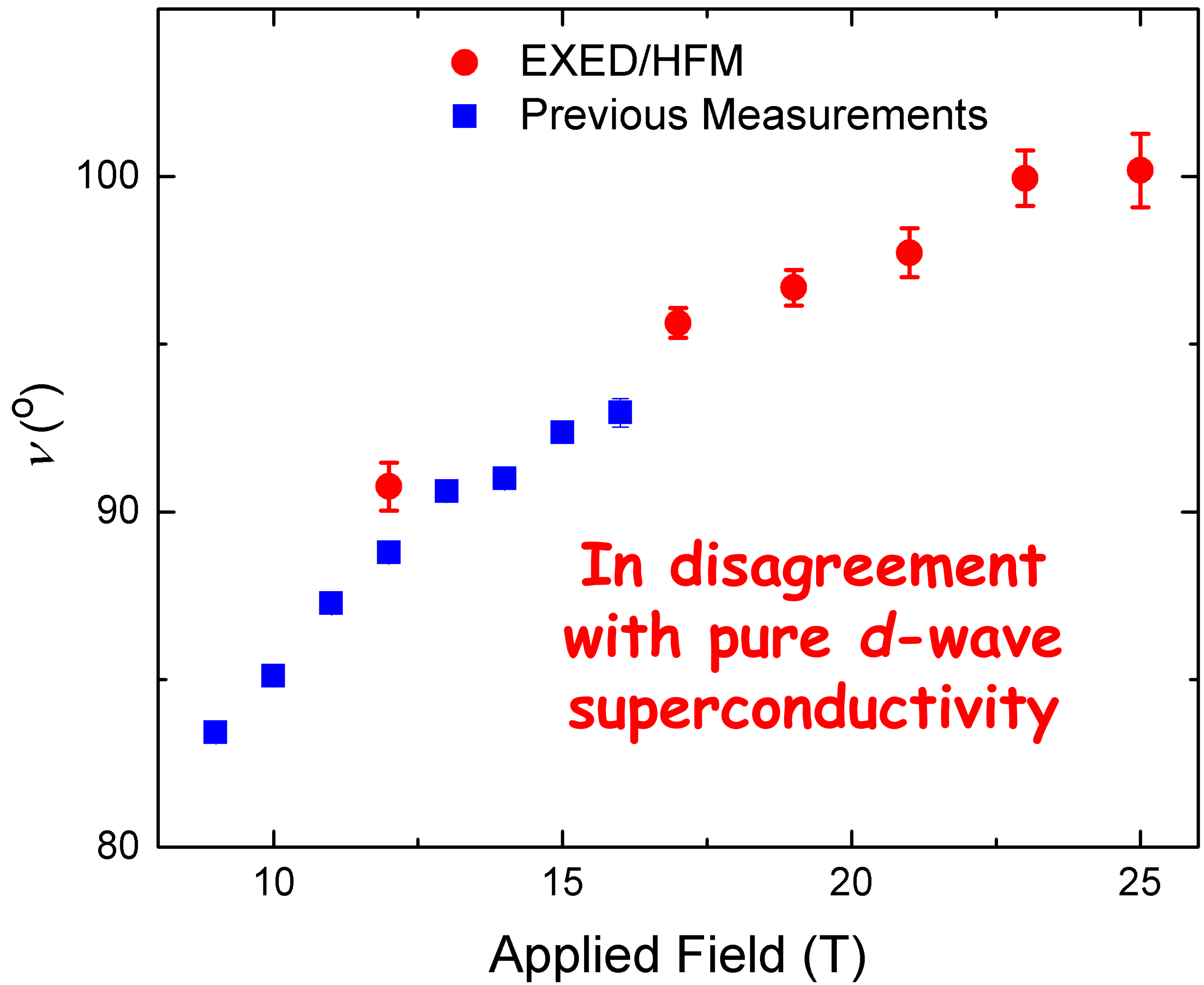
The beamline at HZB Berlin





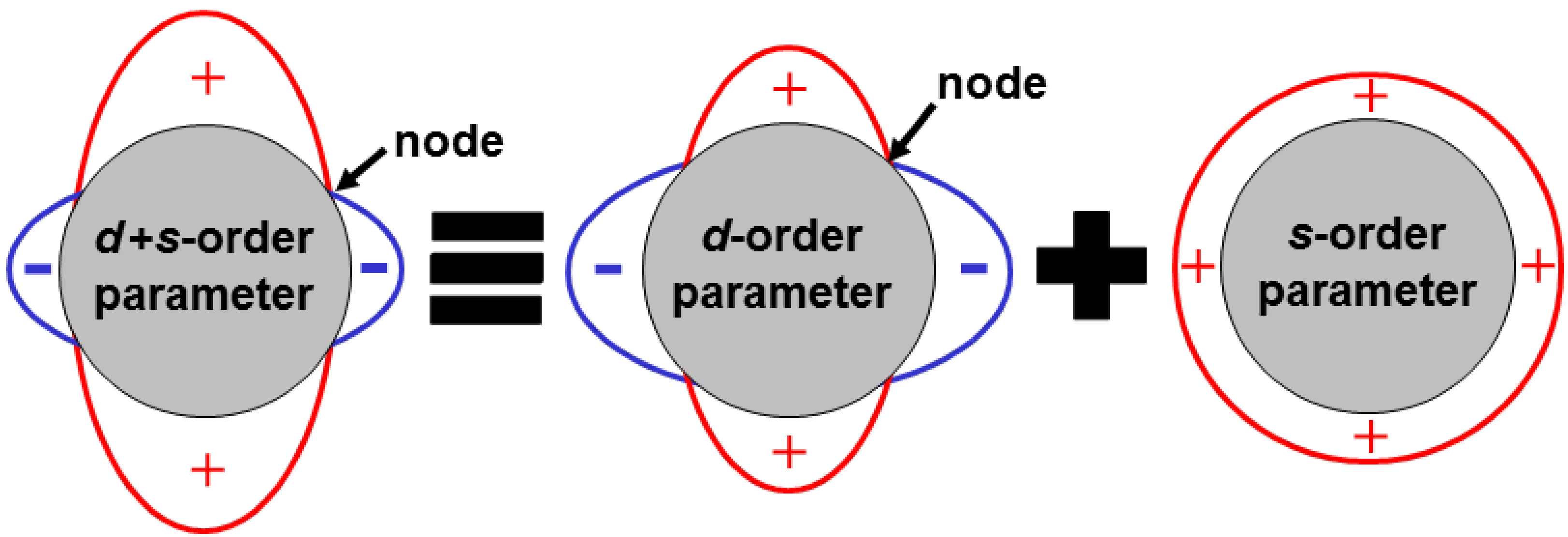
Results for Flux lattice structure

The flux lattice structure is *even less* square at higher fields!



(d + s)-wave superconductivity

d + s combination has orthorhombic symmetry
– so is expected in (orthorhombic) YBCO, which has
CuO_2 planes *plus* CuO chains along the crystal *b*-direction



The nodes are not at 45° : they are nearer the direction with *weaker* superconductivity

(d + s)-wave superconductivity

Our structure results are explained if:

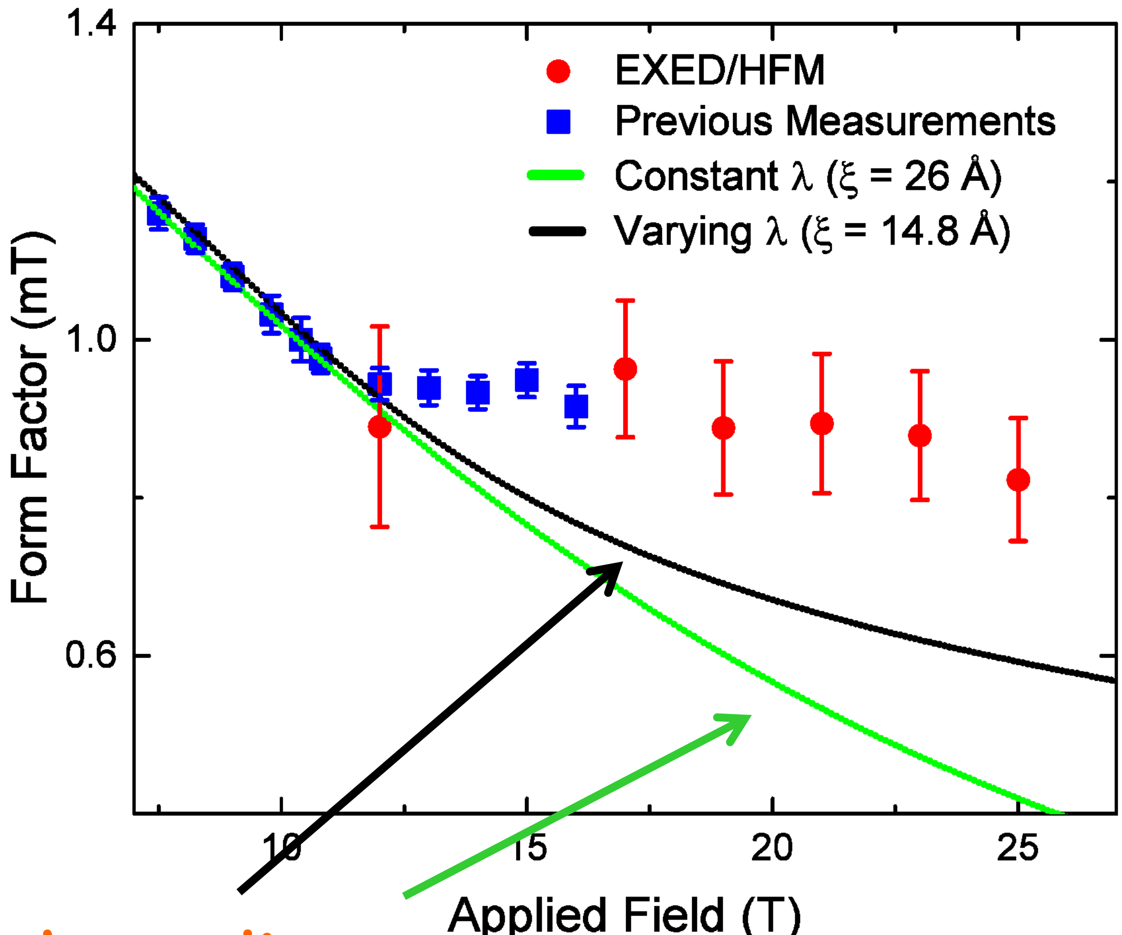
- The flux line nearest neighbour directions tend to lie along the nodal directions

- at *low* fields, the CuO chains along *b* are super-conducting, so superconductivity is *stronger along b*

- at *high* fields, the CuO chains along *b* have turned normal, so superconductivity is *weaker along b*

We also have evidence from our results (not shown) that at high fields superconductivity in YBCO is being weakened by *B* lining up the antiparallel spins

Magnetic contrast at high fields



The flux lattice signal is *hardly suppressed* by high fields!

- We could measure still higher in field - if available!

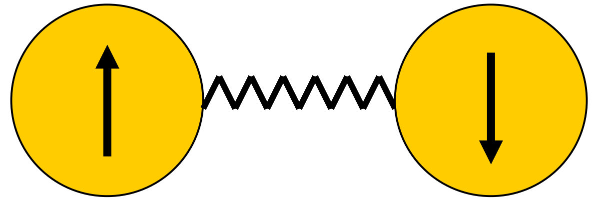
... but - WHY?

Two theory lines for vortex core overlap

"Pauli-limited" superconductivity

s- or *d*-wave'

Cooper pairs



... have *antiparallel* spin electrons - which can't line up with a field

- but in the vortex core, the electrons become *unpaired*

so the vortex cores become magnetised and increase the signal,
- but also start to destroy the superconductivity!

This is important in any superconductor for which:
B_{c2} in Tesla is larger than T_c in Kelvin

Pauli paramagnetism may be the effect limiting the upper critical field in cuprate and pnictide superconductors