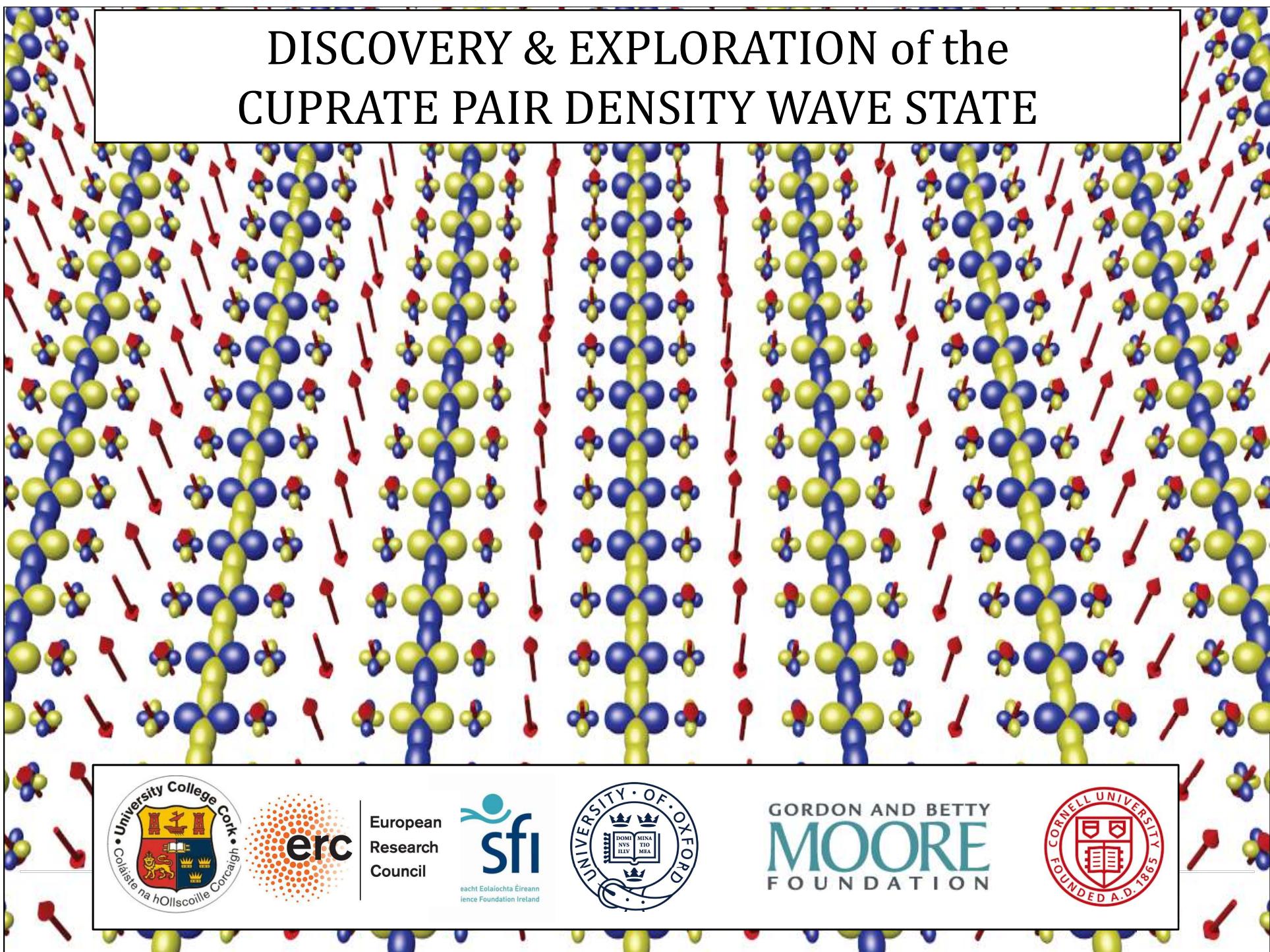


# DISCOVERY & EXPLORATION of the CUPRATE PAIR DENSITY WAVE STATE



European  
Research  
Council



each Eolaíochta Éireann  
ience Foundation Ireland

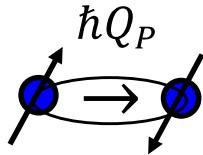


GORDON AND BETTY  
**MOORE**  
FOUNDATION



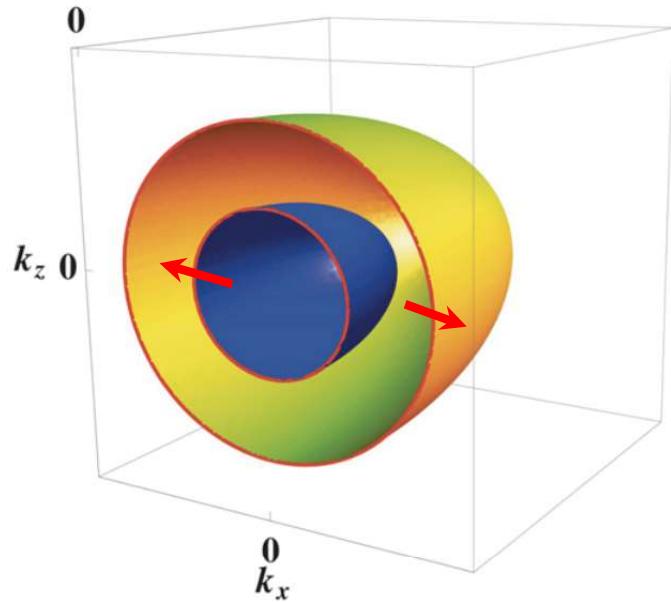
# COOPER-PAIR DENSITY WAVE (FFLO) THEORY

# COOPER PAIR DENSITY WAVE STATE (PDW)

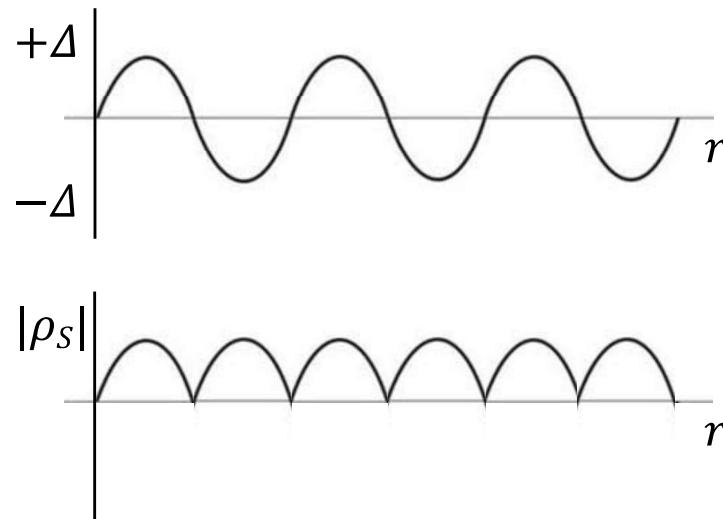


$$\langle c_{k\uparrow}^\dagger, c_{-k+Q_P\downarrow}^\dagger \rangle$$

*Phys. Rev.* 135, A550 (1964) ; *Zh. Eksp. Teor. Fis.* 37 , 1146 (1964).

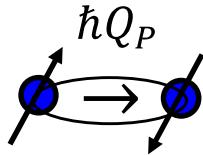


$$\Delta(\mathbf{r}) = \Delta_P [e^{i\mathbf{Q}_P \cdot \mathbf{r}} + e^{-i\mathbf{Q}_P \cdot \mathbf{r}}]$$



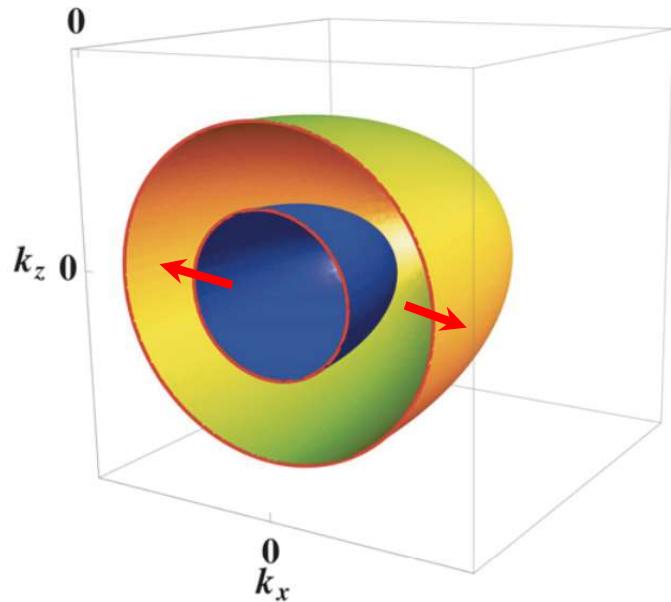
Cooper-pairs non-zero COM momentum: Modulating Cooper-pair Density

# COOPER PAIR DENSITY WAVE STATE (PDW)

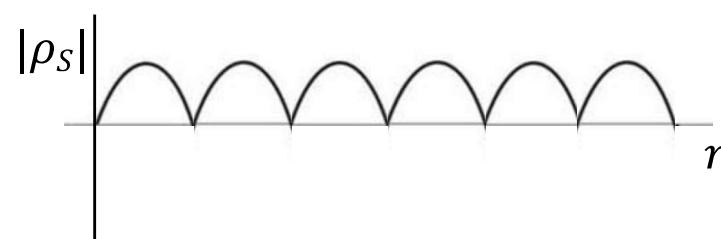
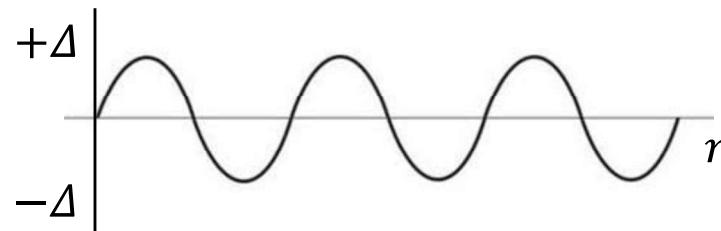


$$\langle c_{k\uparrow}^\dagger, c_{-k+Q_P\downarrow}^\dagger \rangle$$

*Phys. Rev.* 135, A550 (1964) ; *Zh. Eksp. Teor. Fis.* 37 , 1146 (1964).



$$\Delta(\mathbf{r}) = \Delta_P [e^{i\mathbf{Q}_P \cdot \mathbf{r}} + e^{-i\mathbf{Q}_P \cdot \mathbf{r}}]$$



NEVER OBSERVED DIRECTLY

# EXCITON PAIR DENSITY WAVE (CDW)

**PDW**

$$H = \sum_k \Psi_k^\dagger \hat{h}_k \Psi_k$$

$$\langle c_{k\uparrow}^\dagger, c_{-k+Q_P\downarrow}^\dagger \rangle$$

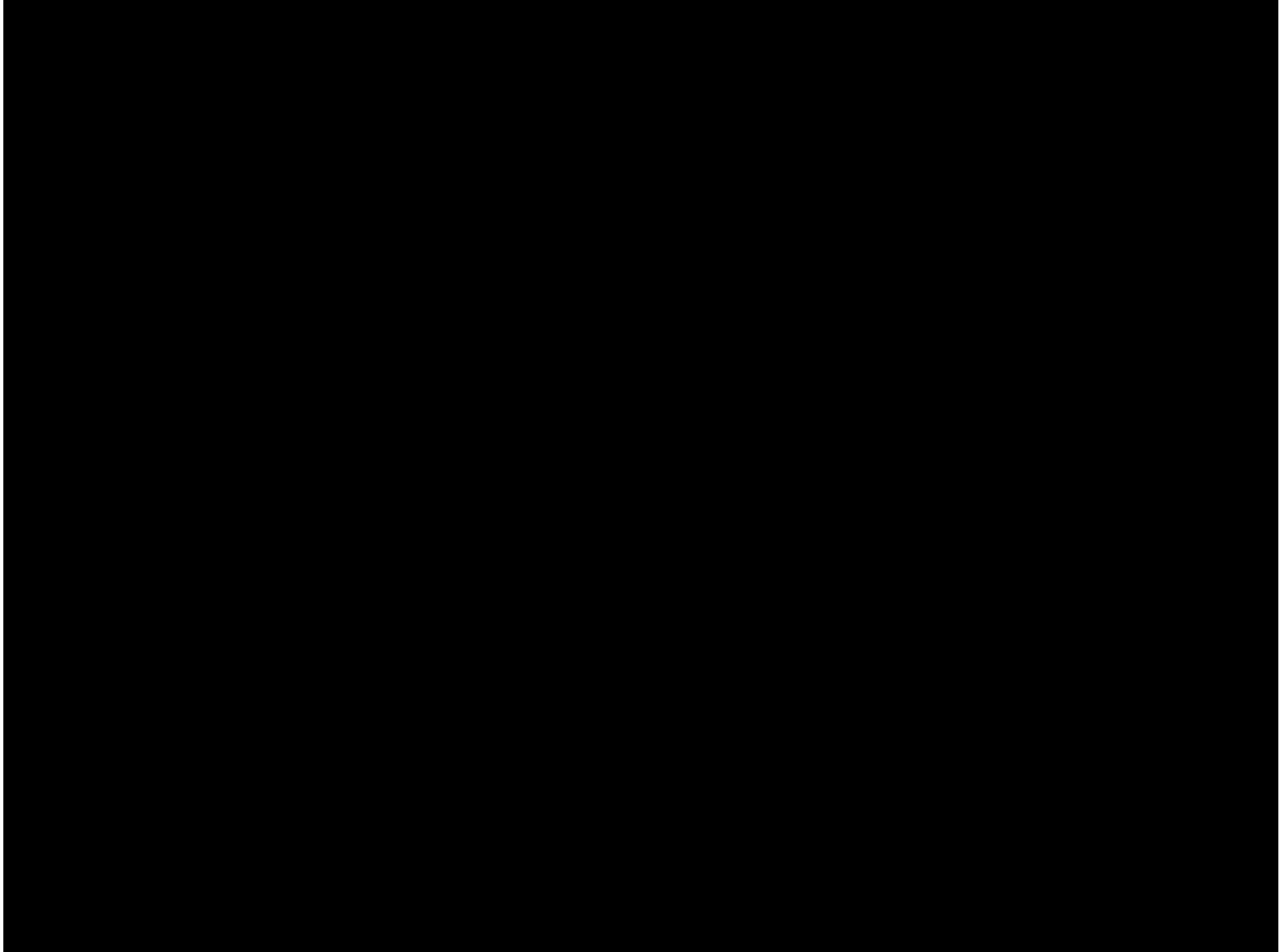
$$\hat{h}_k = \begin{pmatrix} \varepsilon_{\vec{k}} & \Delta_0 & \Delta_{\vec{k}+\vec{Q}_x/2} & \Delta_{\vec{k}-\vec{Q}_x/2} \\ \Delta_0 & -\varepsilon_{-\vec{k}} & 0 & 0 \\ \Delta_{\vec{k}+\vec{Q}_x/2} & 0 & -\varepsilon_{-(\vec{k}+\vec{Q}_x)} & 0 \\ \Delta_{\vec{k}-\vec{Q}_x/2} & 0 & 0 & -\varepsilon_{-(\vec{k}-\vec{Q}_x)} \end{pmatrix}, \Psi_k = \begin{pmatrix} c_{k,\uparrow} \\ c_{-k,\downarrow}^+ \\ c_{-(\vec{k}+\vec{Q}_x),\downarrow}^+ \\ c_{-(\vec{k}-\vec{Q}_x),\downarrow}^+ \end{pmatrix}$$

**CDW**

$$H = \sum_k \Psi_k^\dagger \hat{h}_k \Psi_k$$

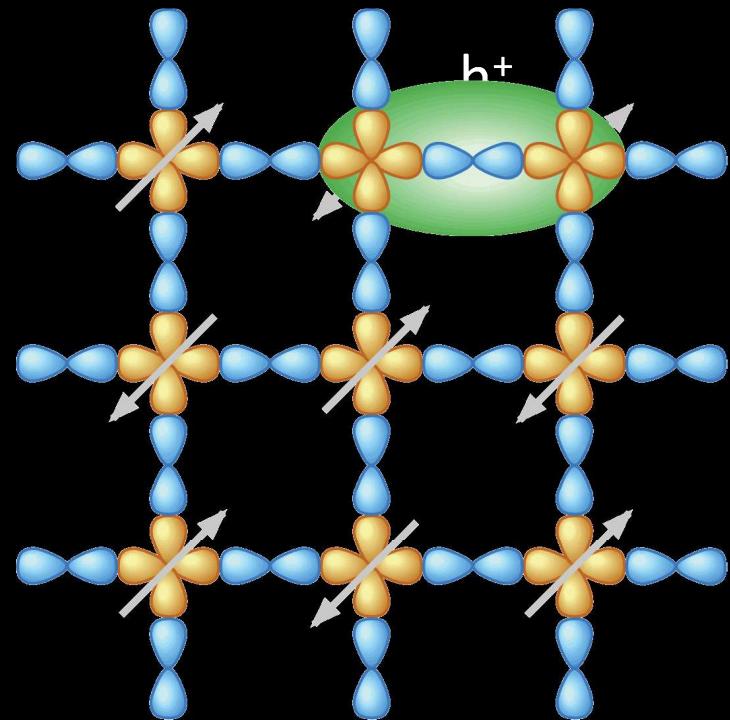
$$\langle c_{k\uparrow}^\dagger, c_{k+Q_C\downarrow} \rangle$$

$$\hat{h}_k = \begin{pmatrix} \varepsilon_{\vec{k}} & \Delta_0 & 0 & 0 \\ \Delta_0 & -\varepsilon_{-\vec{k}} & V_{\vec{k}+\vec{Q}_x/2} & V_{\vec{k}-\vec{Q}_x/2} \\ 0 & V_{\vec{k}+\vec{Q}_x/2} & -\varepsilon_{-(\vec{k}+\vec{Q}_x)} & 0 \\ 0 & V_{\vec{k}-\vec{Q}_x/2} & 0 & -\varepsilon_{-(\vec{k}-\vec{Q}_x)} \end{pmatrix}, \Psi_k = \begin{pmatrix} c_{k,\uparrow} \\ c_{-k,\downarrow}^+ \\ c_{-(\vec{k}+\vec{Q}_x),\downarrow}^+ \\ c_{-(\vec{k}-\vec{Q}_x),\downarrow}^+ \end{pmatrix}$$



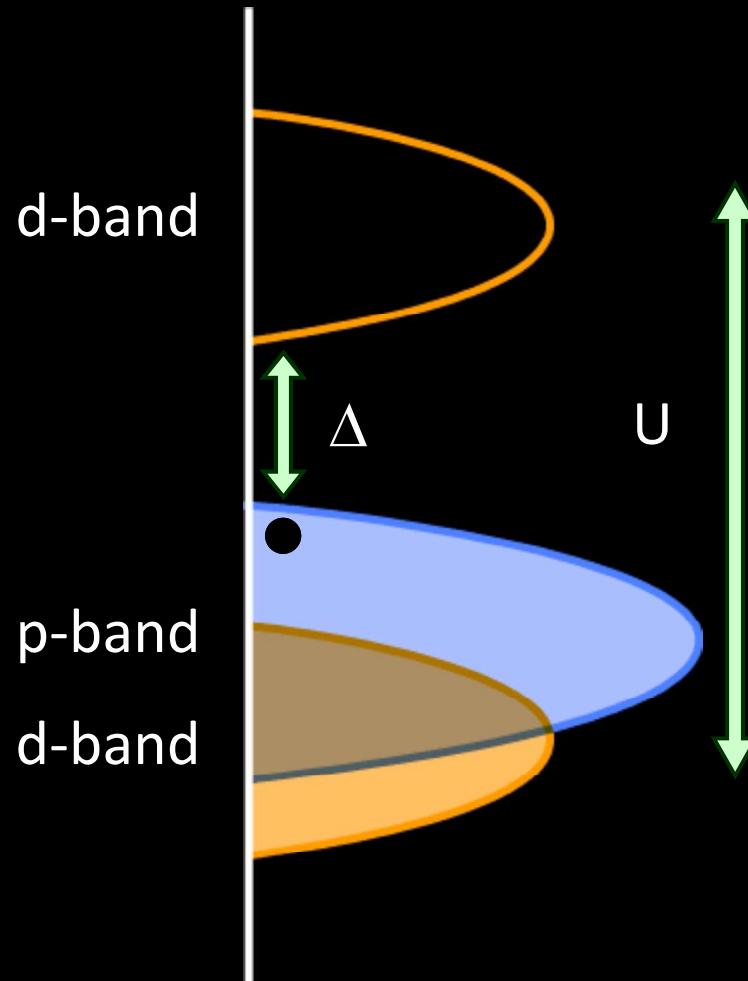
CUPRATE PAIR DENSITY WAVE

# HOLE-DOPED CuO<sub>2</sub> MOTT INSULATOR

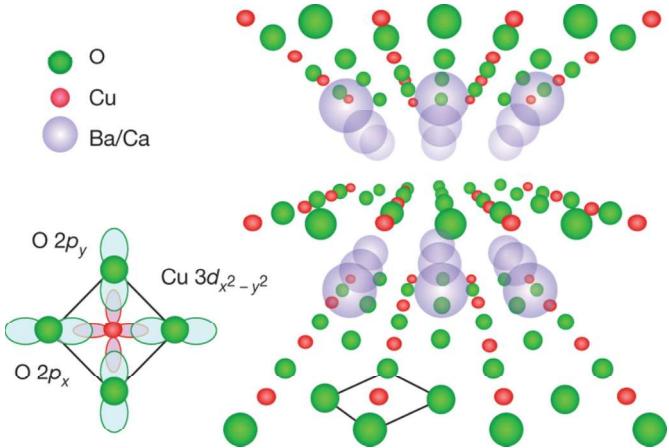
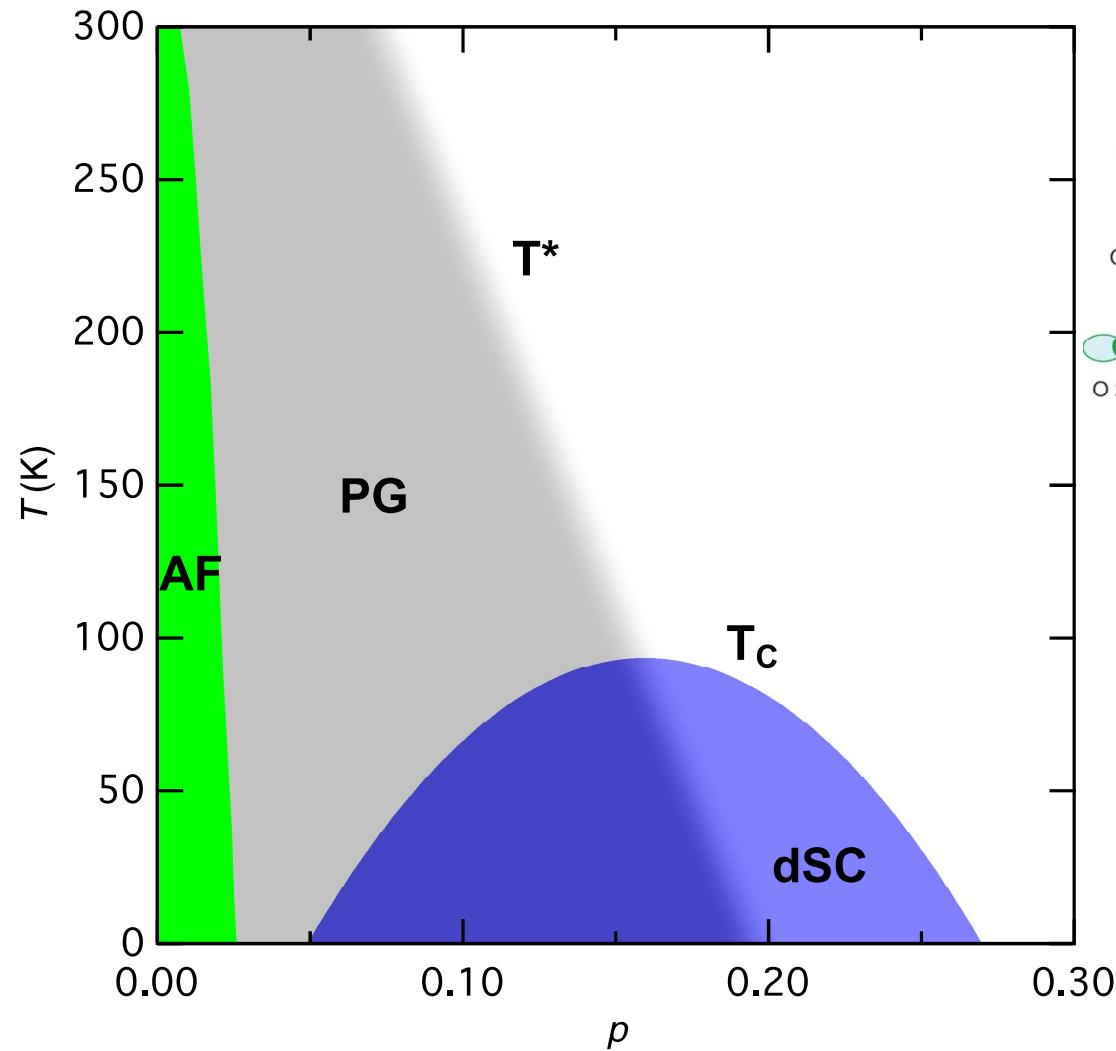


$$U \approx 3\text{eV}, \quad t \approx 400\text{meV}, \quad J \approx 150\text{meV}$$

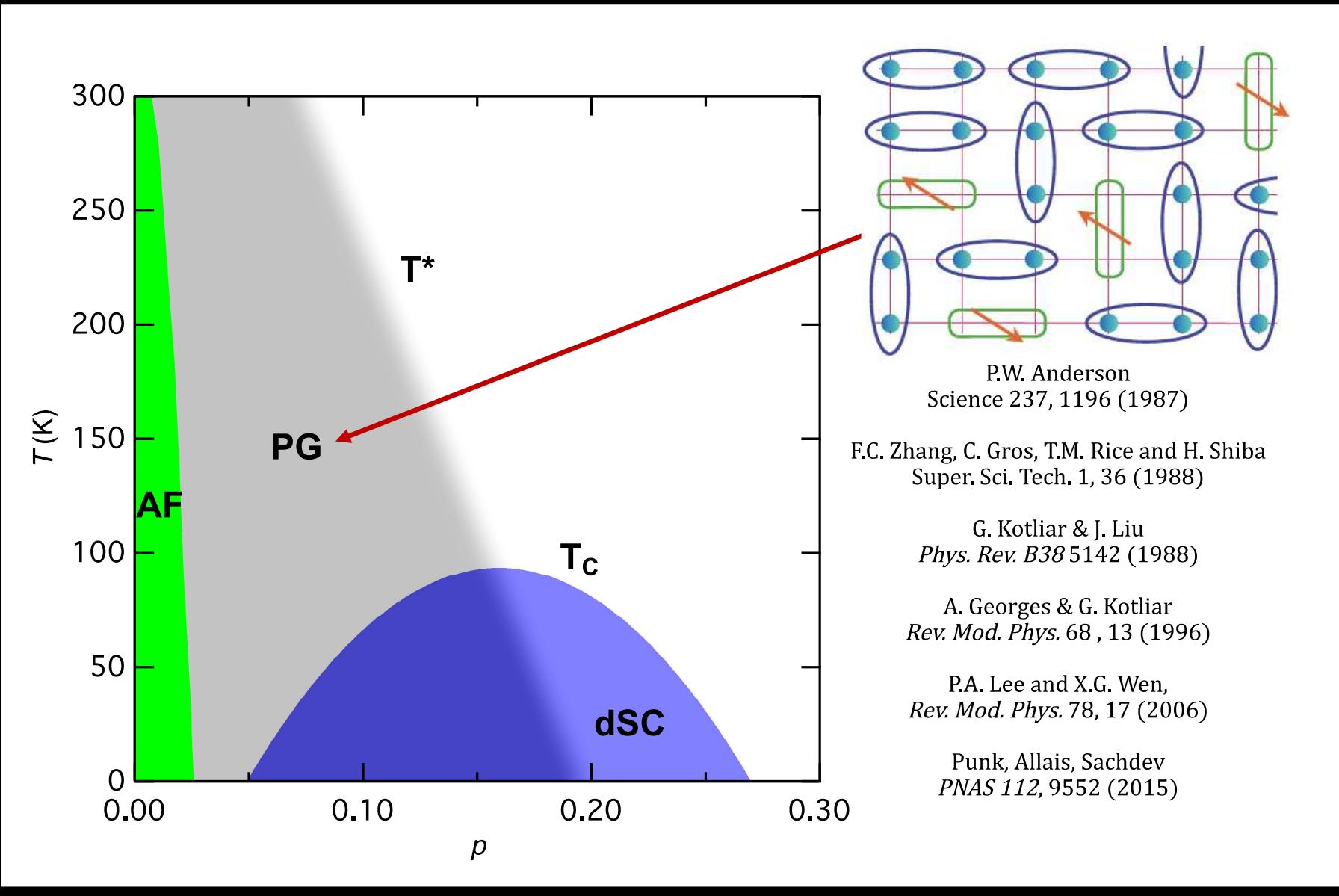
$\Rightarrow$  half-filling = correlated AF insulator



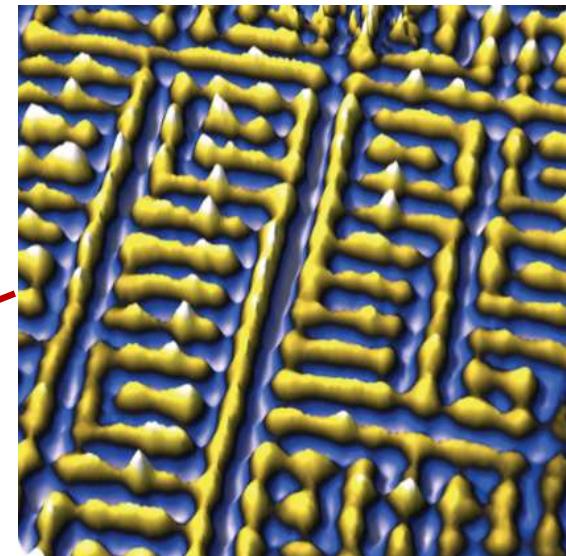
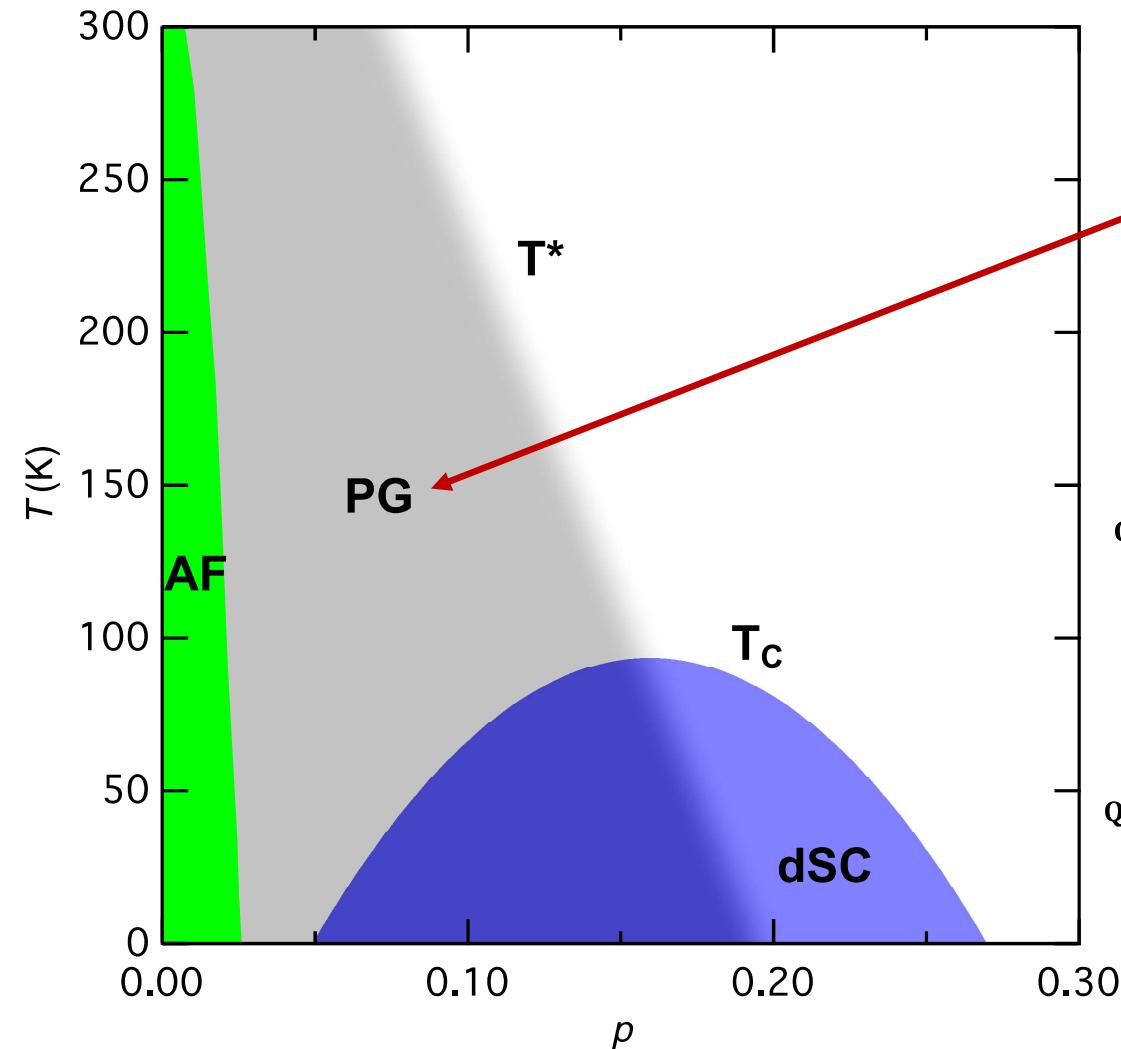
# HOLE-DOPED CuO<sub>2</sub> MOTT INSULATOR



# PSEUDOOGAP = HOLE-DOPED SPIN LIQUID ?

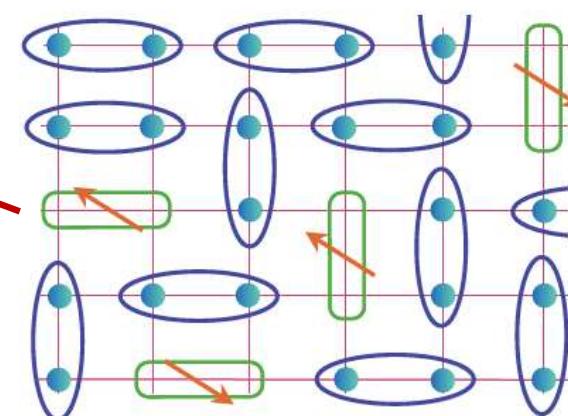
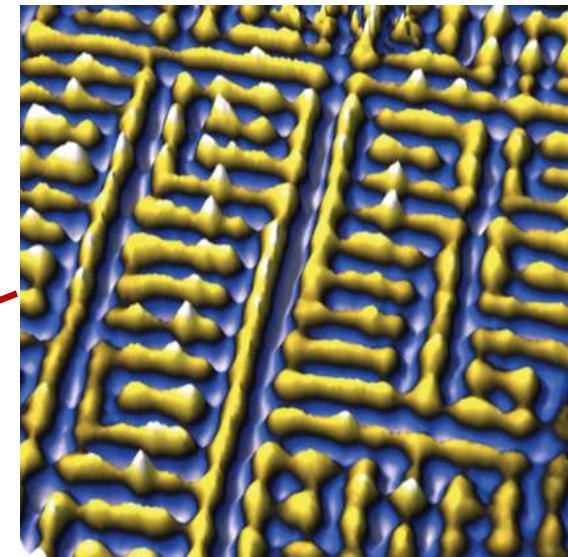
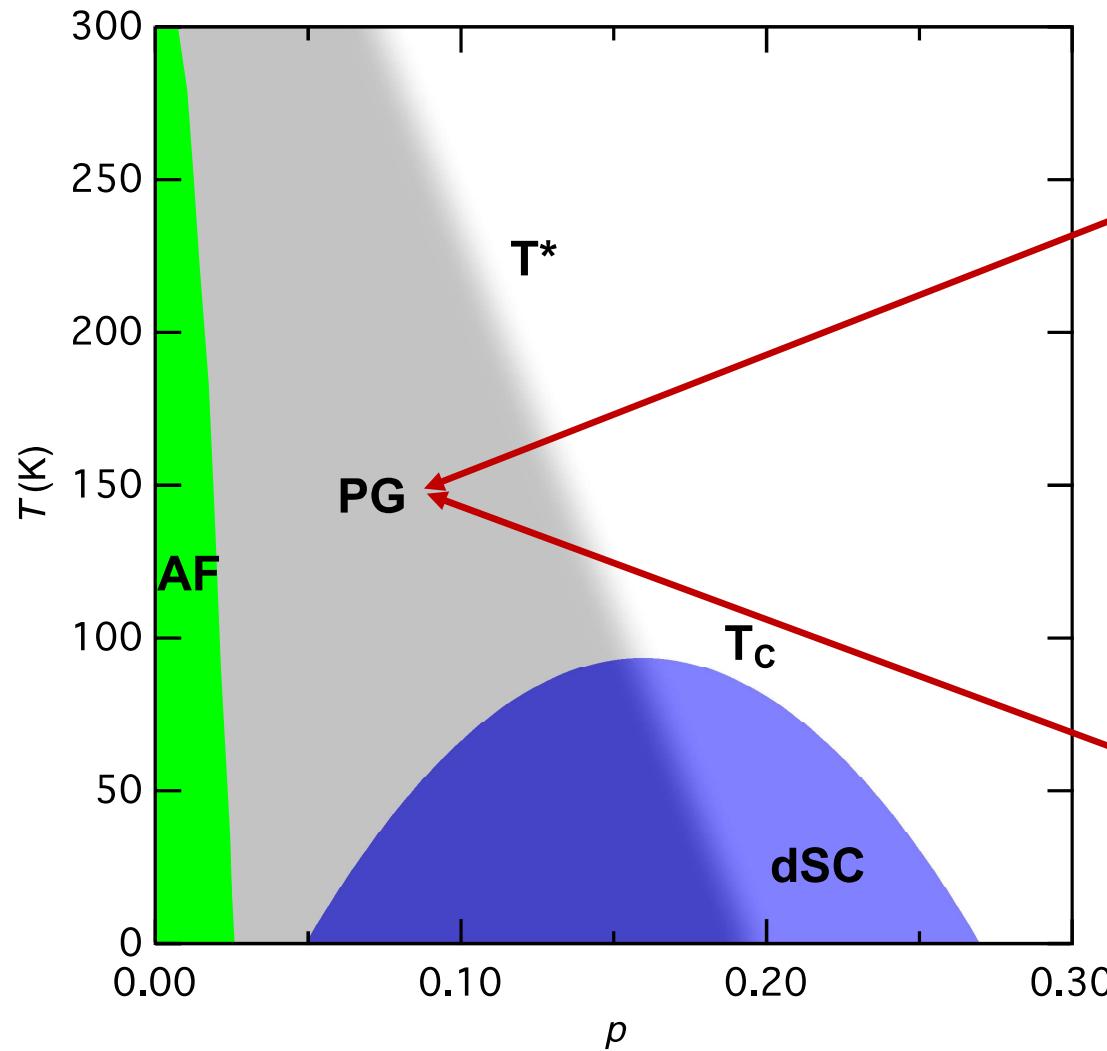


# PSEUDOOGAP = BROKEN-SYMMETRY STATE ?

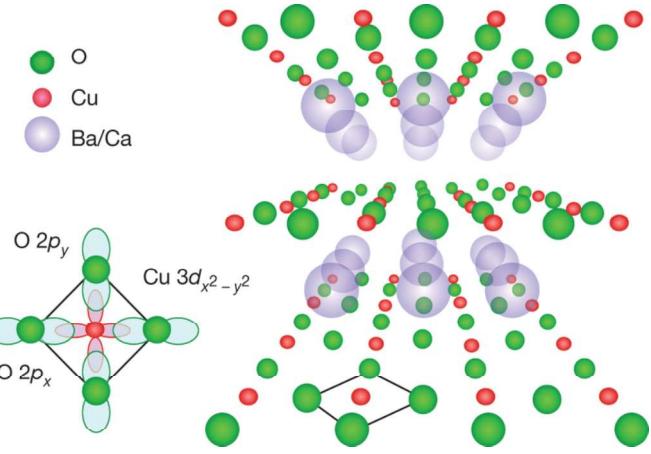
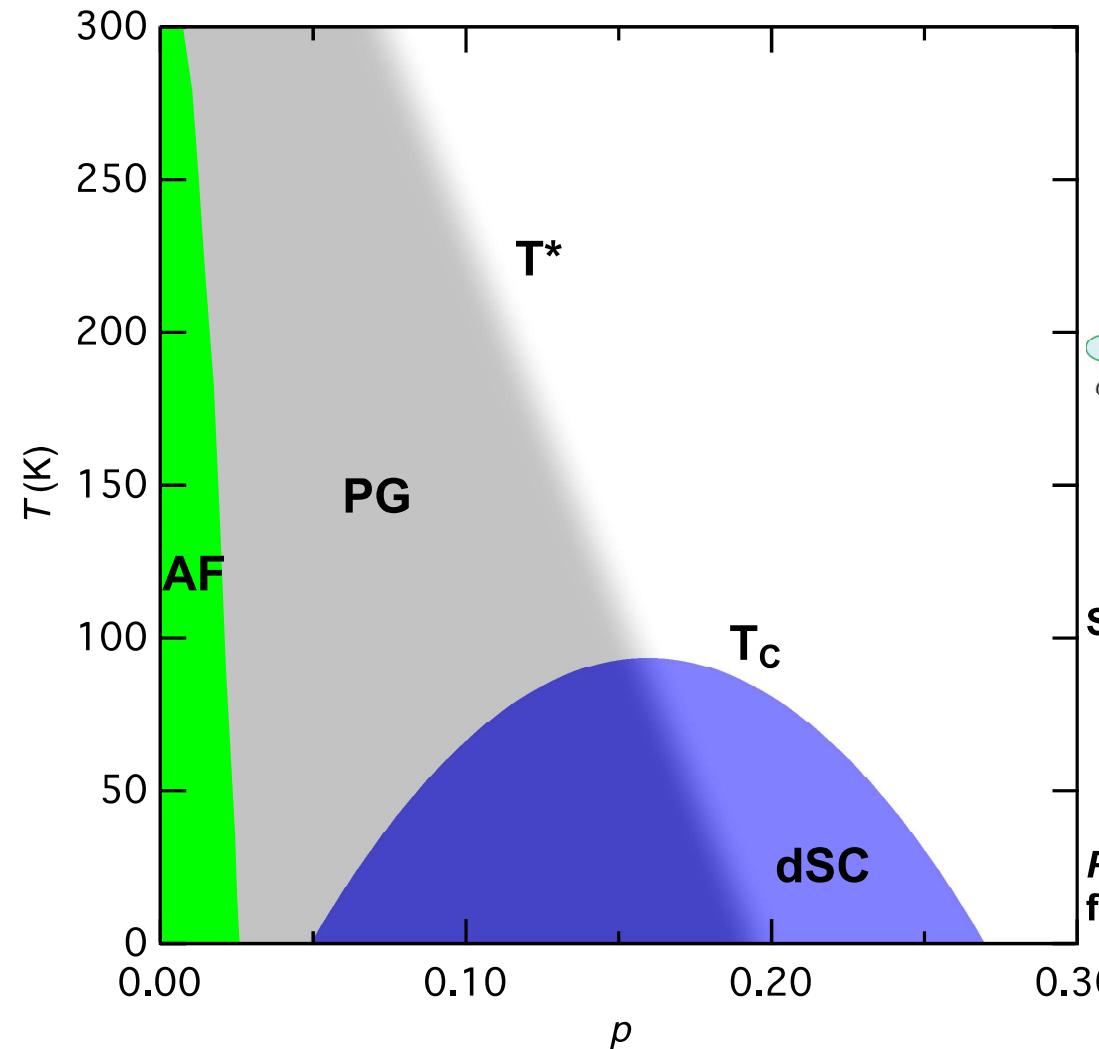


- Q=0** (2002) *Nature* 416 (6881):610-613.  
(2006) *Phys Rev Lett* 96 (19):197001  
(2008) *Nature* 455 (7211):372–375.  
(2010) *Nature* 463 (7280):519-522.  
(2013) *Nature* 498 (7452):75-77.  
(2017) *Nat Phys* 13 (3):250–254.  
(2017) *Nat Phys* 13 (11):1074–1078.
- Q=Q<sub>DW</sub>** (1995) *Nature* 375 (6532): 561–563.  
(2012) *Nat Phys* 8 (12):871–876.  
(2012) *Science* 337 (6096):821–825.  
(2014) *Science* 343 (6169):390–392.  
(2015) *Science* 350 (6263):949–952.  
(2015) *Nat. Mat.* 14 (8):796–800.

# HOLE-DOPED SPIN LIQUID + BROKEN SYMMETRY STATE ?



# CuO<sub>2</sub> STRONG-COUPLING THEORY



Strong-Coupling CuO<sub>2</sub> plane *t-J* Model

$$H = - \sum_{(i,j),\sigma} P_G t_{ij} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) P_G + J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j,$$

$P_G$  projects out doubly occupied sites from the Hilbert space.

# $\text{CuO}_2$ STRONG-COUPLING THEORY $\Rightarrow$ PAIR DENSITY WAVE

Himeda, A., Kato, T. & Ogata, M. Stripe states with oscillating  $d$ -wave superconductivity in the two-dimensional  $t$ - $t'$ - $J$  model.  
*Phys. Rev. Lett.* **88**, 117001 (2002).

Raczkowski, M. et al, Unidirectional  $d$ -wave superconducting domains in the two-dimensional  $t$ - $J$  model.  
*Phys. Rev. B* **76**, 140505 (2007).

Yang, K.-Y., Chen, W. Q., Rice, T. M., Sigrist, M. & Zhang F.-C. Nature of stripes in the generalized  $t$ - $J$  model applied to cuprate superconductors.  
*New J. Phys.* **11**, 055053 (2009).

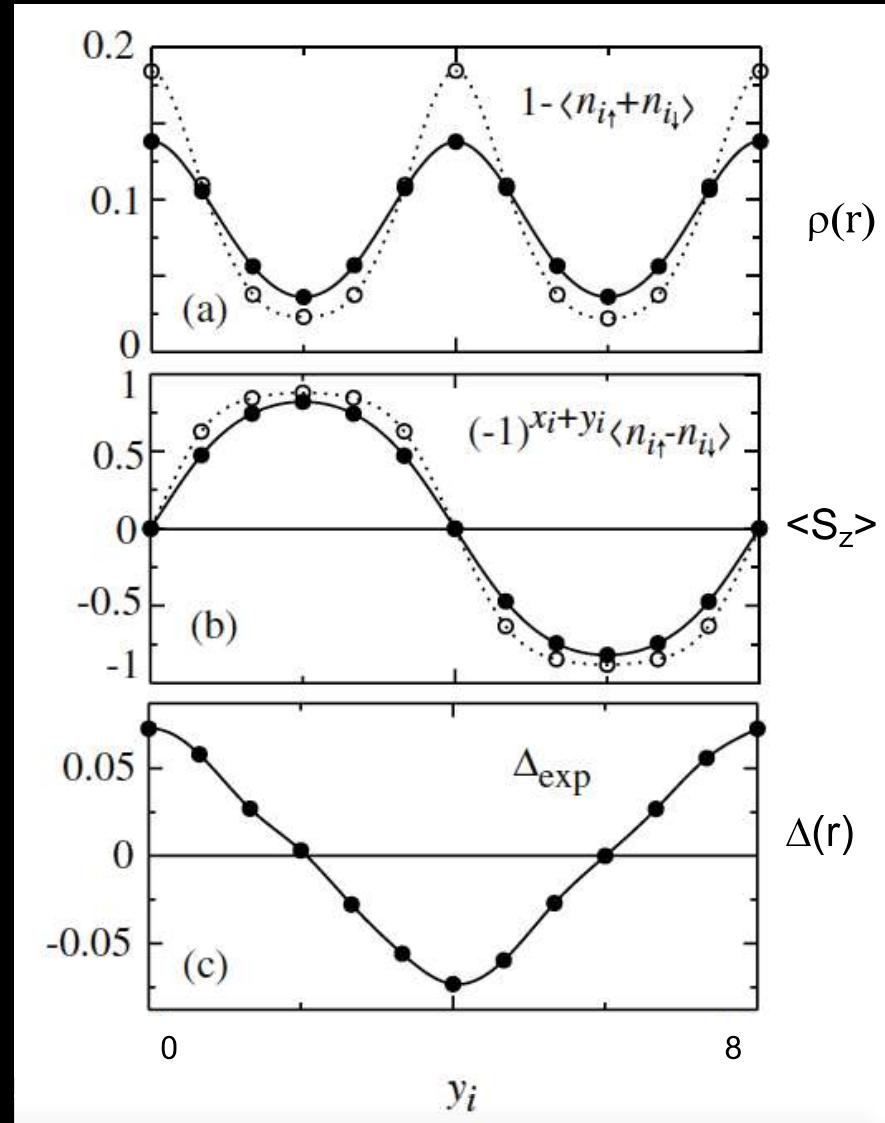
Loder, F., Graser, S., Kampf, A. P. & Kopp, T. Mean-field pairing theory for the charge-stripe phase of cuprate superconductors  
*Phys. Rev. Lett.* **107**, 187001 (2011).

Corboz, P., Rice, T. M. & Troyer, M. Competing states in the  $t$ - $J$  model: uniform  $d$ -wave state versus stripe state.  
*Phys. Rev. Lett.* **113**, 046402 (2014) .

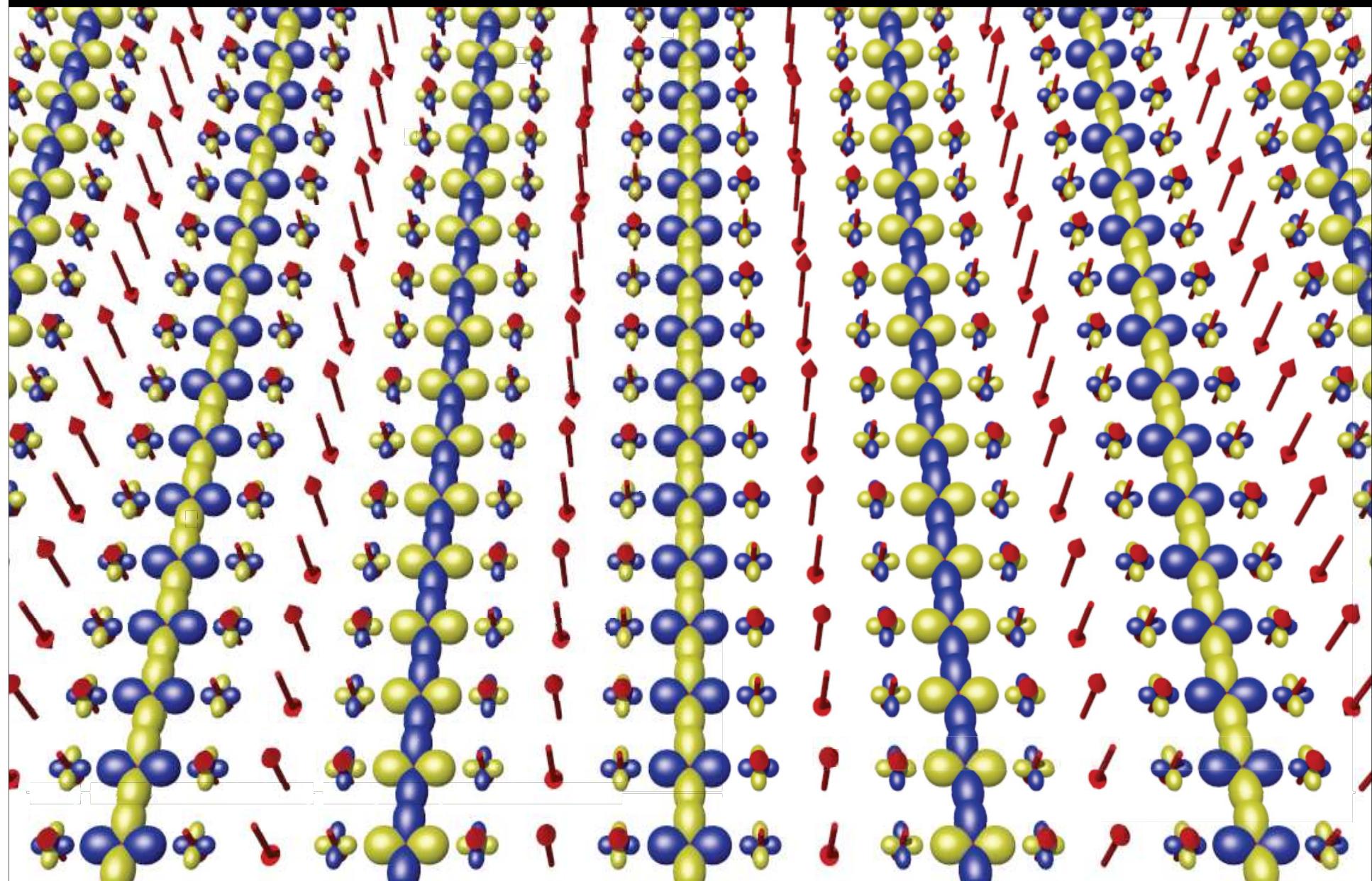
Choubey P., Wei-Lin Tu, Ting-Kuo Lee and P. J. Hirschfeld  
Incommensurate charge ordered states in the  $t$ - $t'$ - $J$  model  
*New J. Phys.* **19**, 013028 (2017)

S. Verret et al Subgap structure and pseudogap in cuprates superconductors *Phys. Rev. B* **95**, 544518 (2017)

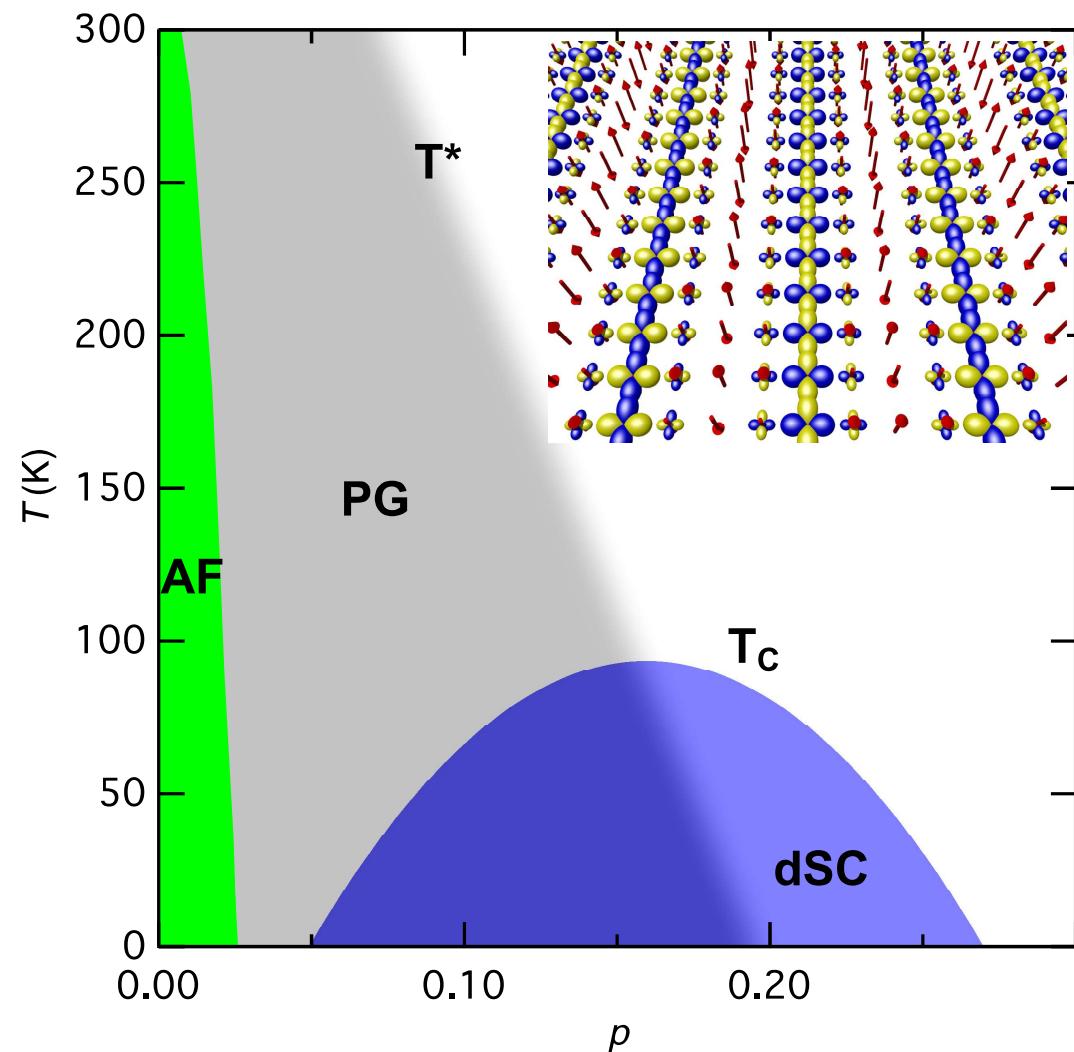
Rong-Gen Caia, Li Lia, Yong-Qiang Wang, and Jan Zaanen  
*Phys. Rev. Lett.* **119**, 181601 (2017)



# $\text{CuO}_2$ STRONG-COUPLING THEORY $\Rightarrow$ PAIR DENSITY WAVE

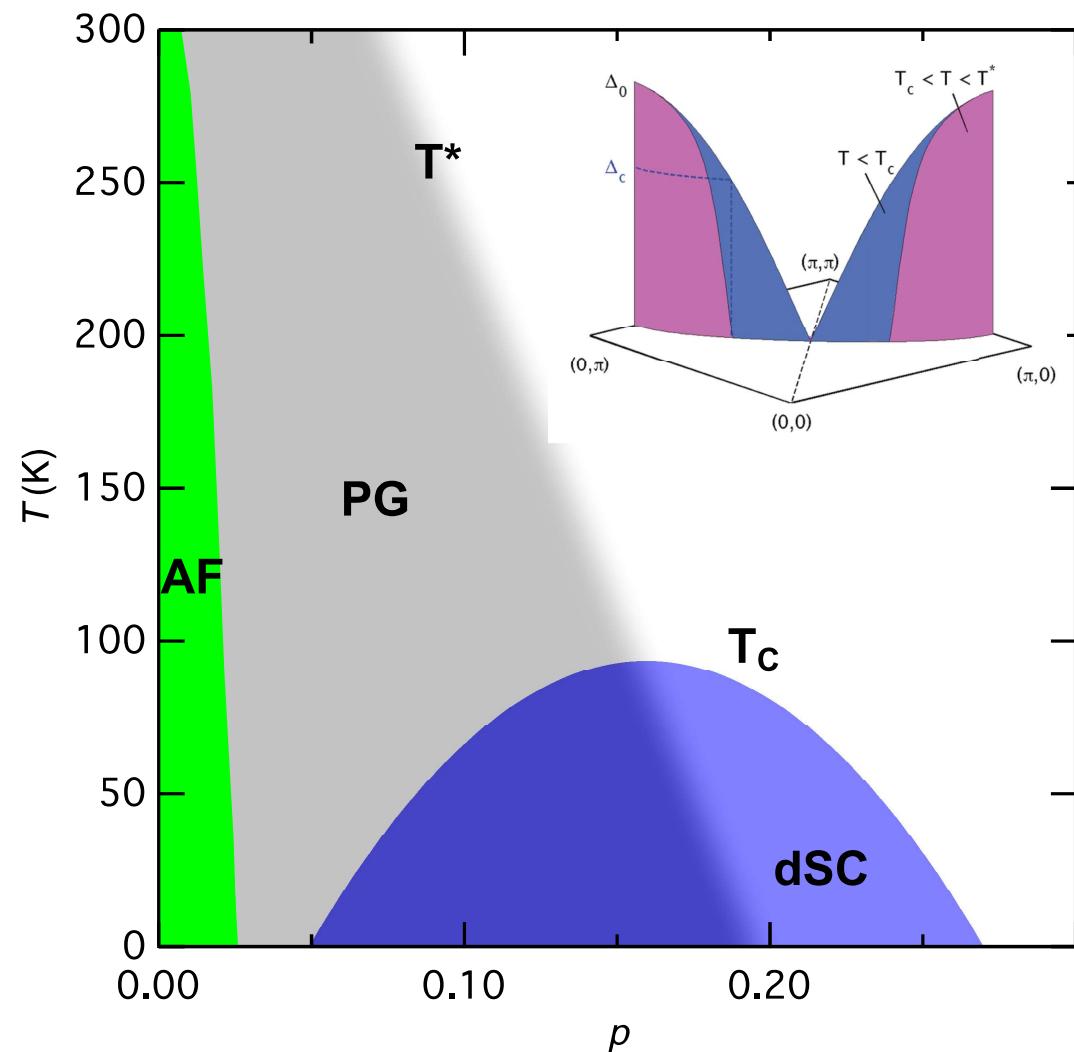


# PSEUDOOGAP $\Leftrightarrow$ PAIR DENSITY WAVE



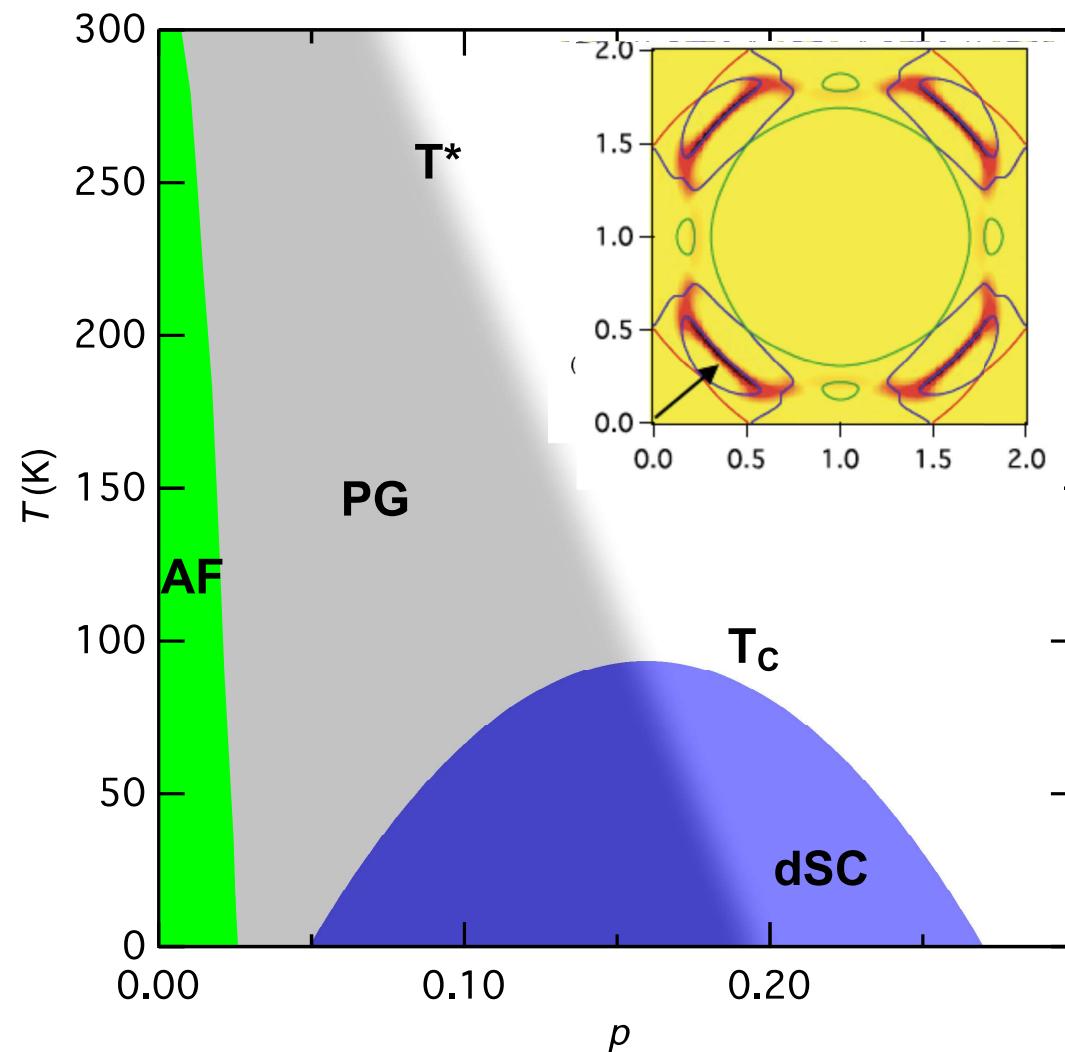
- Natural in strong coupling theory of hole-doped CuO<sub>2</sub>
- PDW free energy density very close to that of DSC

# PSEUDOOGAP $\Leftrightarrow$ PAIR DENSITY WAVE



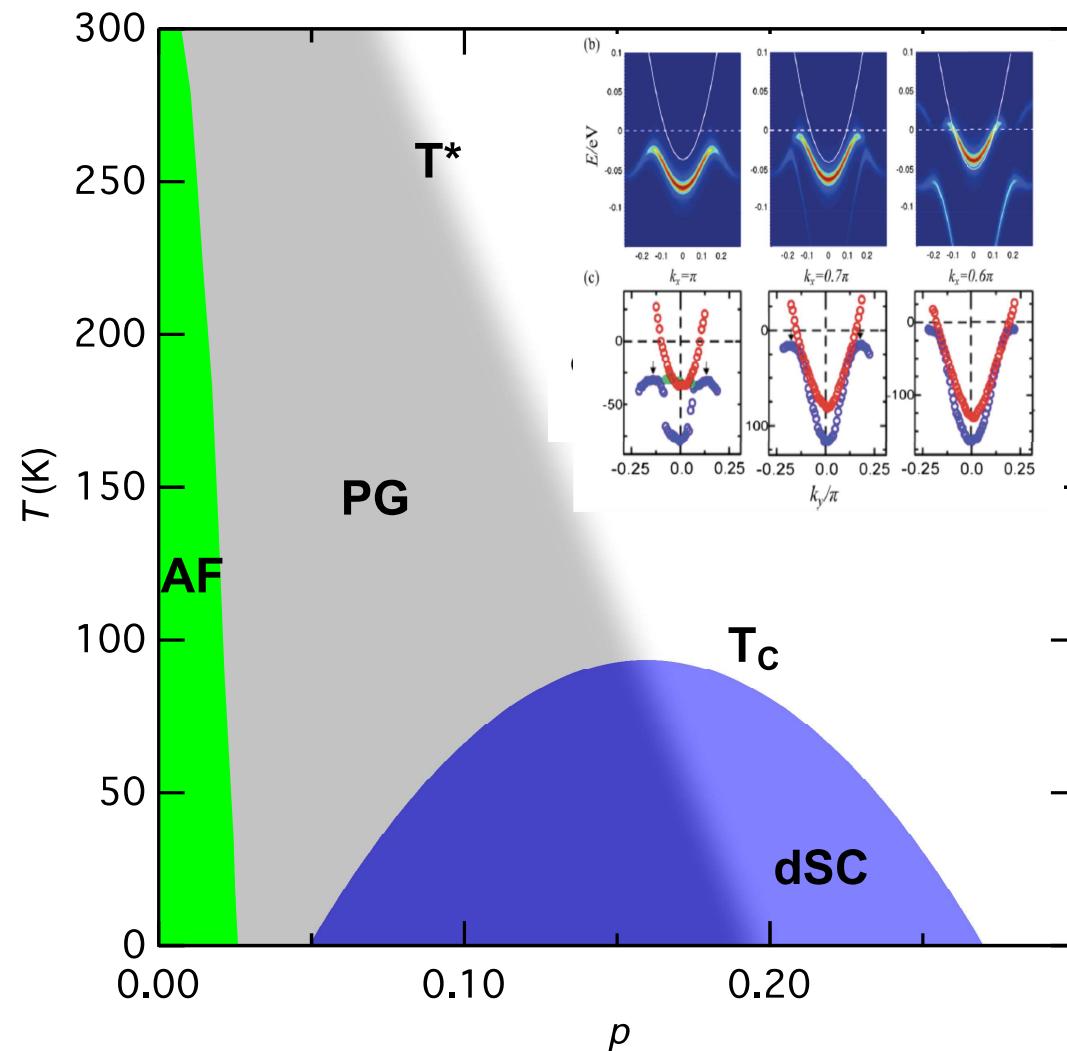
- Natural in strong coupling theory of hole-doped CuO<sub>2</sub>
- PDW free energy density very close to that of DSC
- PDW exhibits a particle-hole symmetric antinodal gap
- PDW exhibits k-space ‘Fermi Arc’ of unbound electrons

# PSEUDOOGAP $\Leftrightarrow$ PAIR DENSITY WAVE



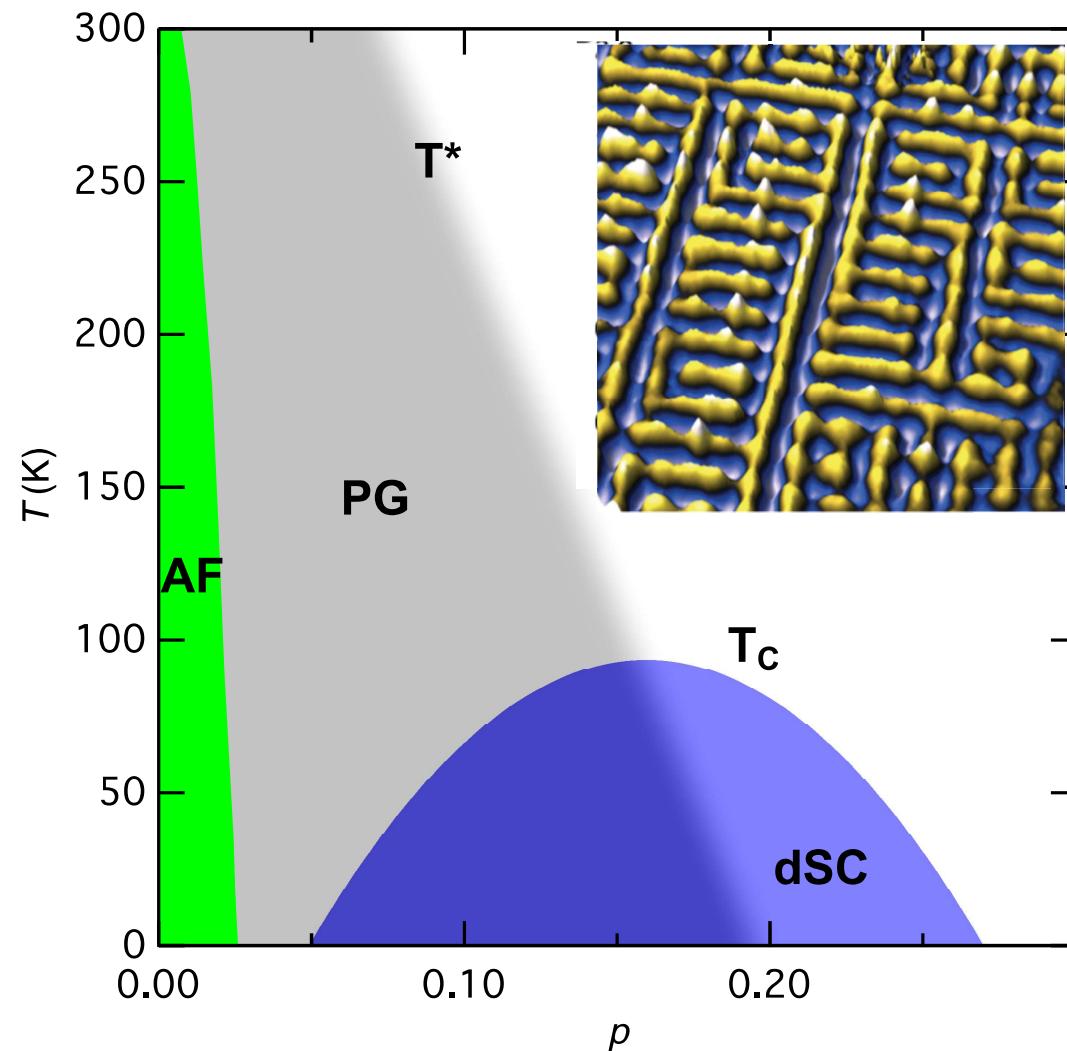
- Natural in strong coupling theory of hole-doped CuO<sub>2</sub>
- PDW free energy density very close to that of DSC
- PDW exhibits a particle-hole symmetric antinodal gap
- PDW exhibits  $k$ -space ‘Fermi Arc’ of unbound electrons
- PDW yields small electron-like pocket with correct frequency of quantum oscillations

# PSEUDOGAP $\Leftrightarrow$ PAIR DENSITY WAVE



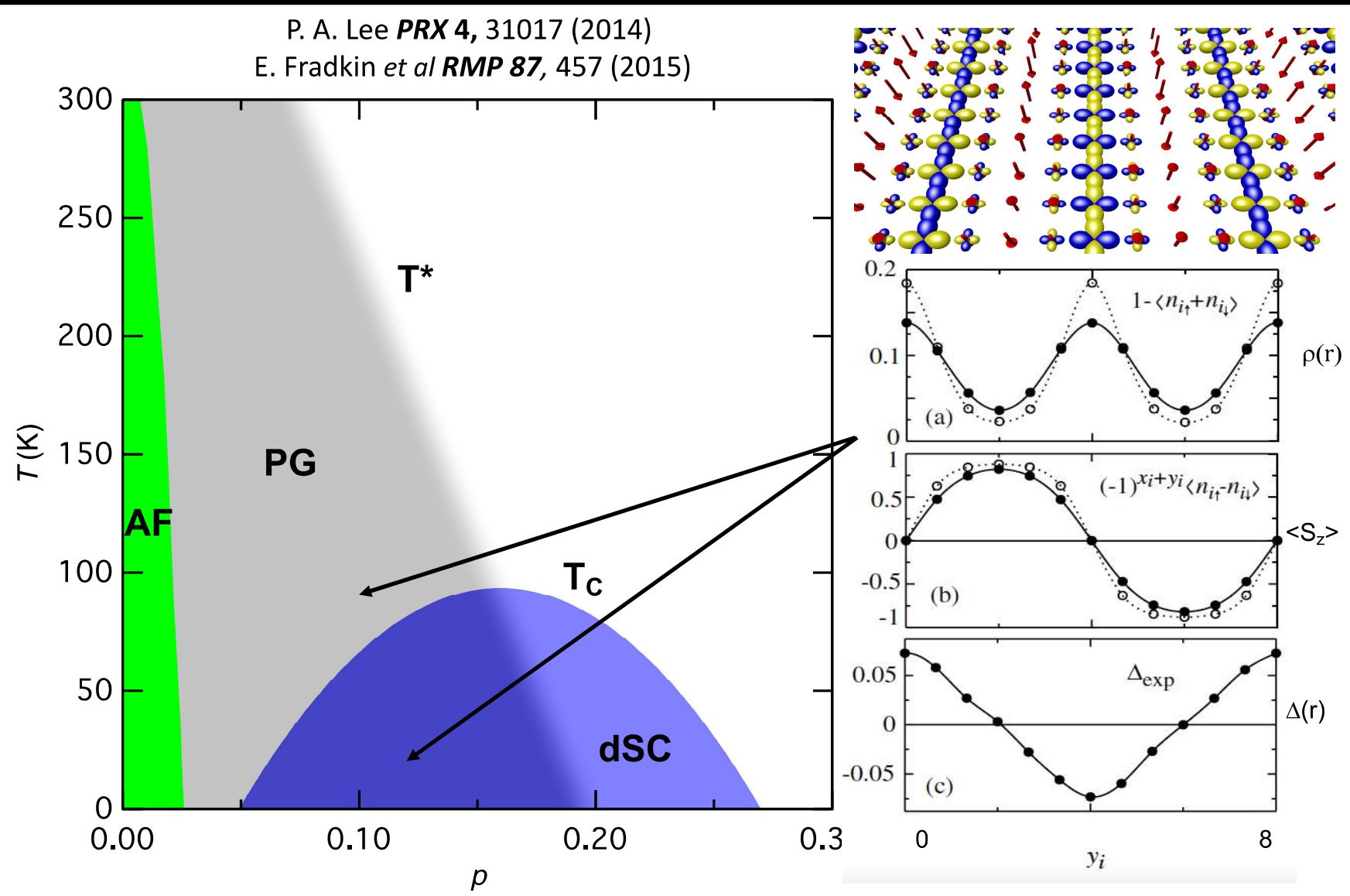
- Natural in strong coupling theory of hole-doped CuO<sub>2</sub>
- PDW free energy density very close to that of DSC
- PDW exhibits a particle-hole symmetric antinodal gap
- PDW exhibits k-space ‘Fermi Arc’ of unbound electrons
- PDW yields small electron-like pocket with correct frequency of quantum oscillations
- PDW gives the correct spectral functions for underdoped CuO<sub>2</sub>

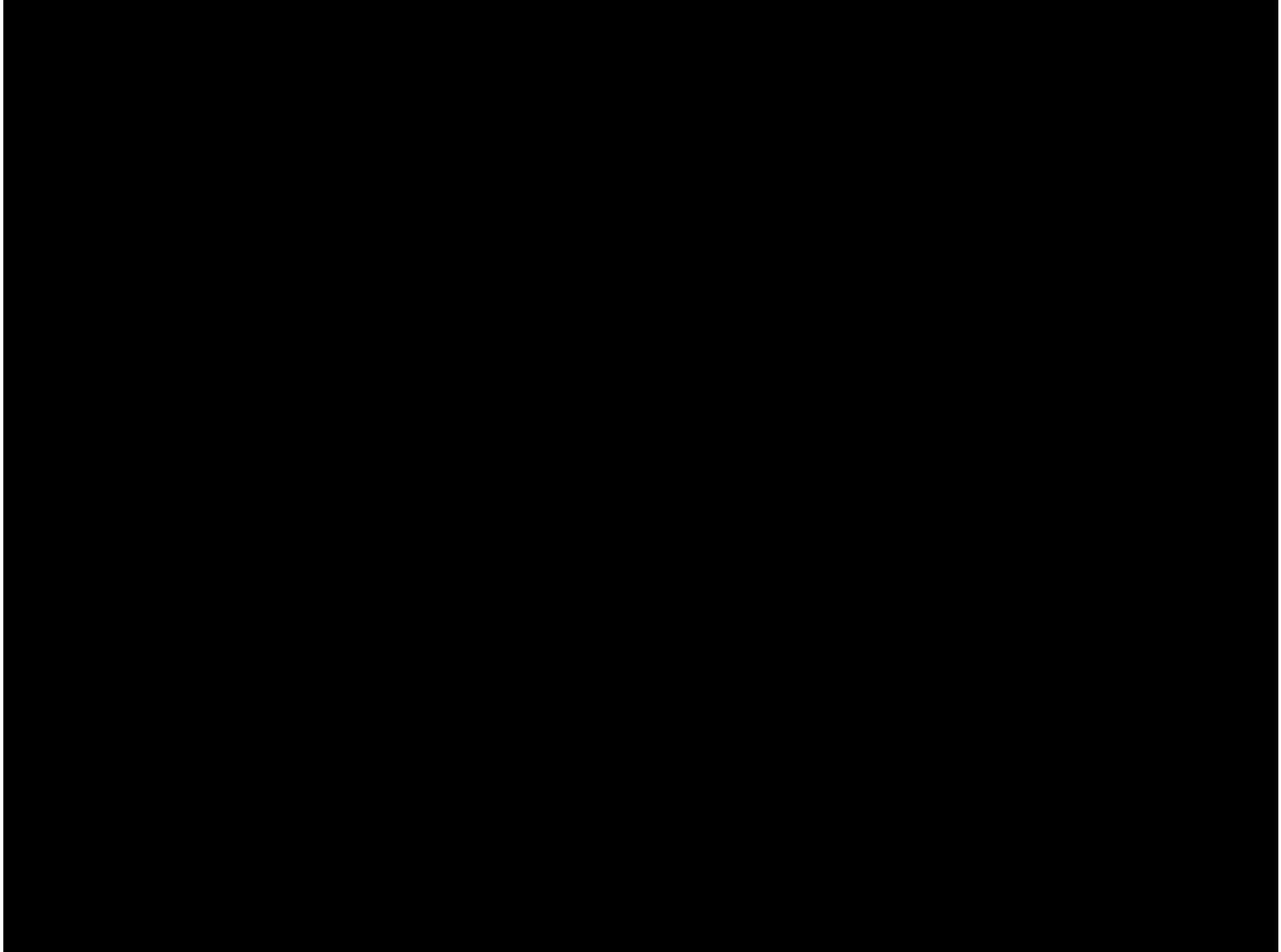
# PSEUDOOGAP $\Leftrightarrow$ PAIR DENSITY WAVE



- Natural in strong coupling theory of hole-doped CuO<sub>2</sub>
- PDW free energy density very close to that of DSC
- PDW exhibits a particle-hole symmetric antinodal gap
- PDW exhibits k-space ‘Fermi Arc’ of unbound electrons
- PDW yields small electron-like pocket with correct frequency of quantum oscillations
- PDW gives the correct spectral functions for underdoped CuO<sub>2</sub>
- PDW generates charge density modulations primarily  $2Q_P$ .

# DOES STRONG-COUPLING PDW STATE EXIST IN CUPRATES?





VISUALIZE COOPER-PAIR CONDENSATE:  $\langle c_k^\dagger c_{-k+Q_P}^\dagger \rangle$

# VISUALIZE COOPER-PAIRS ?

Pan S., Hudson E., & Davis J.C.  
*Appl. Phys. Lett.*, 73, 2992 (1998).

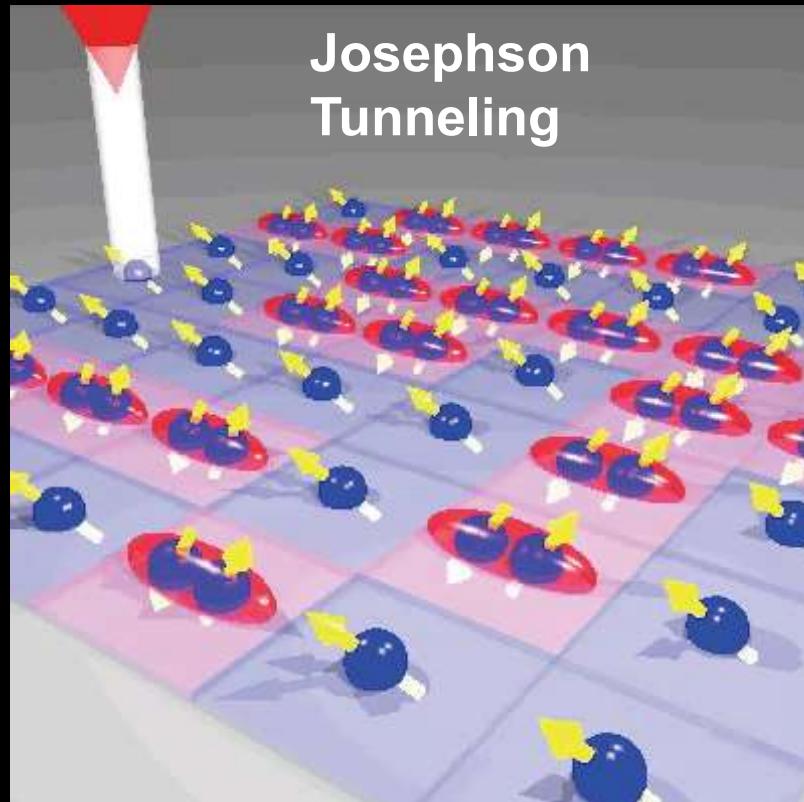
Naaman, O., Teizer, W. & Dynes, R. C.  
Fluctuation dominated Josephson tunneling with  
STM. *Phys. Rev. Lett.* 87, 097004 (2001).

Rodrigo J. G., Suderow H. & Vieira, S.  
On the use of STM superconducting tips at very low  
temperatures. *Eur. Phys. J. B* 40, 483-488 (2004).

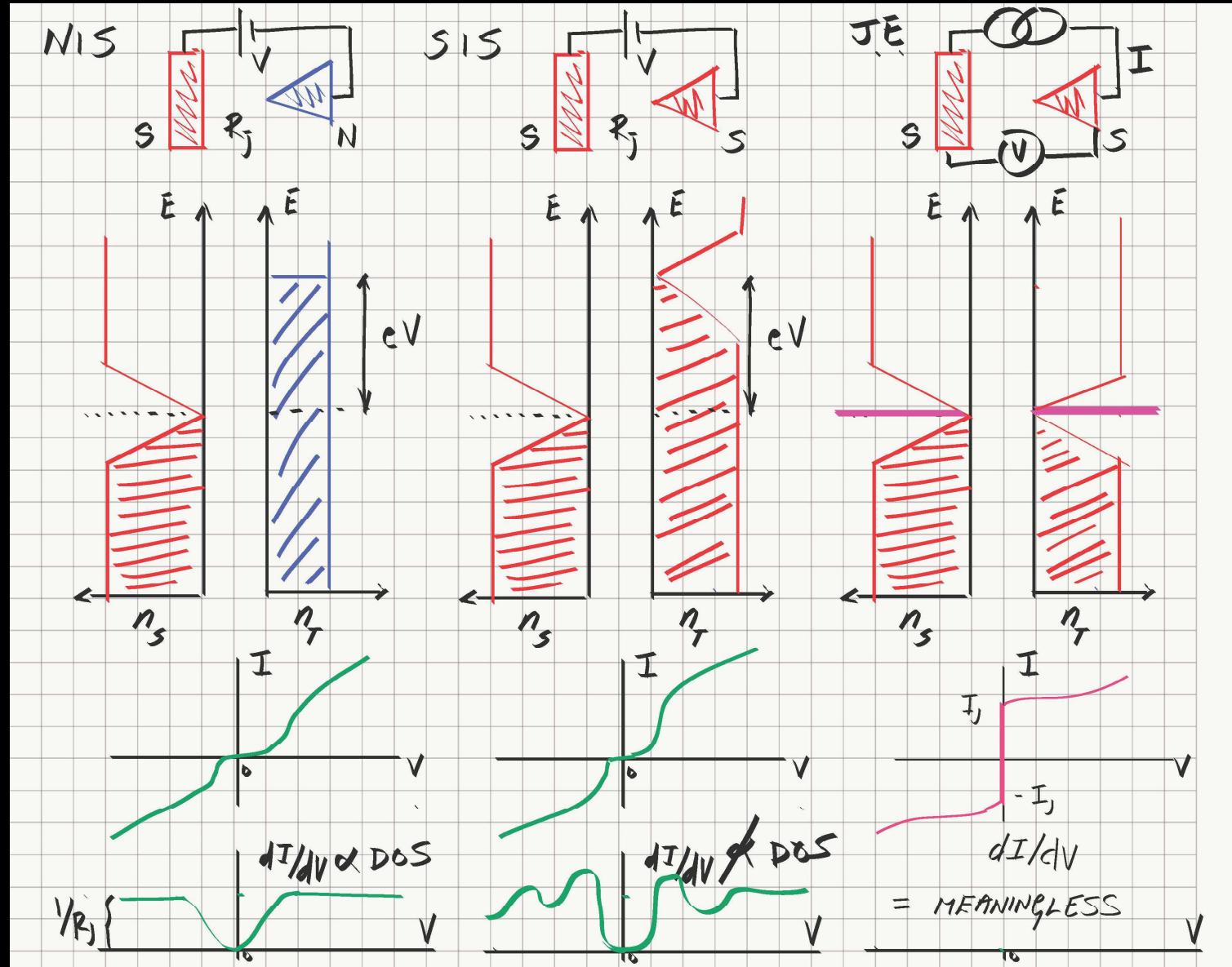
Proslier, Th. *et al.* Probing the superconducting  
condensate on nanometer scale. *Europhys. Lett.* 73,  
962-968 (2006).

Randeria M.T. *et al* Scanning Josephson Spectroscopy  
on Atomic Scale. *Phys. Rev. B* 93, 161115 (2016)

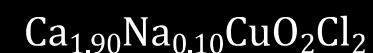
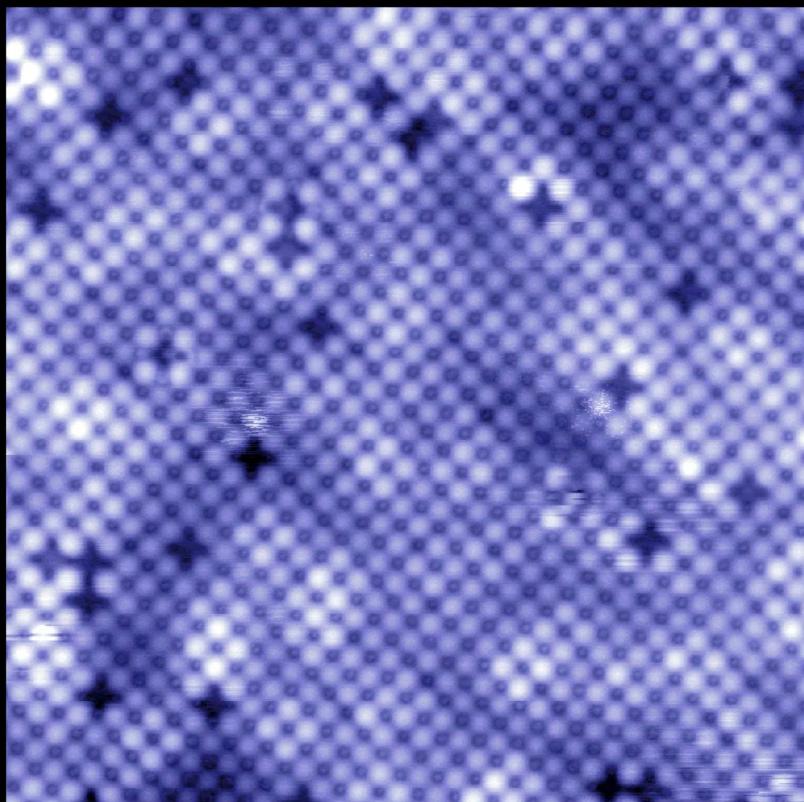
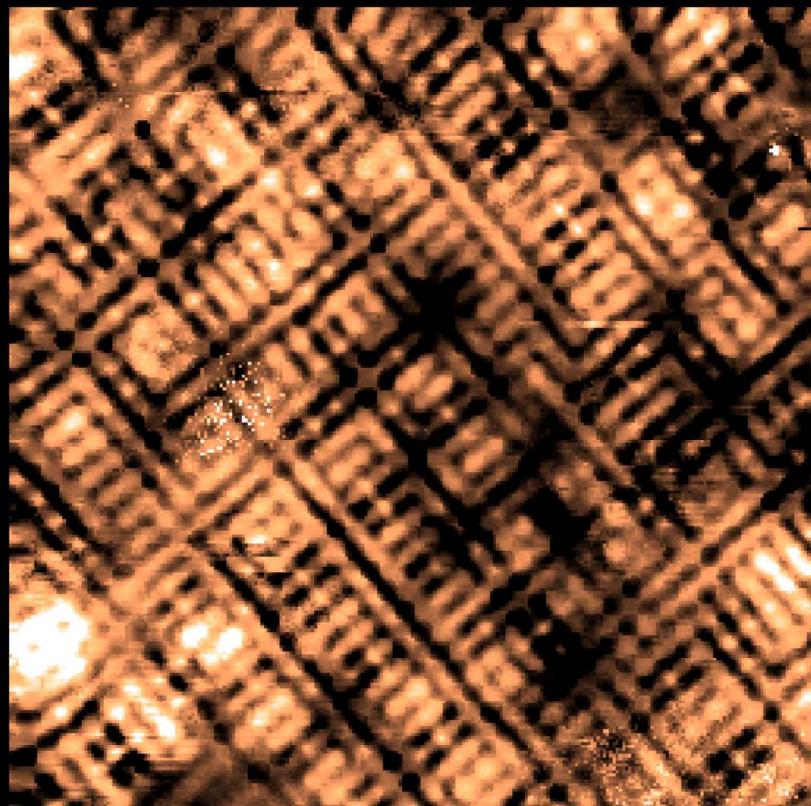
Superconducting Tip



# NIS / SIS/ JE STM



# VISUALIZE CUPRATE COOPER-PAIRS



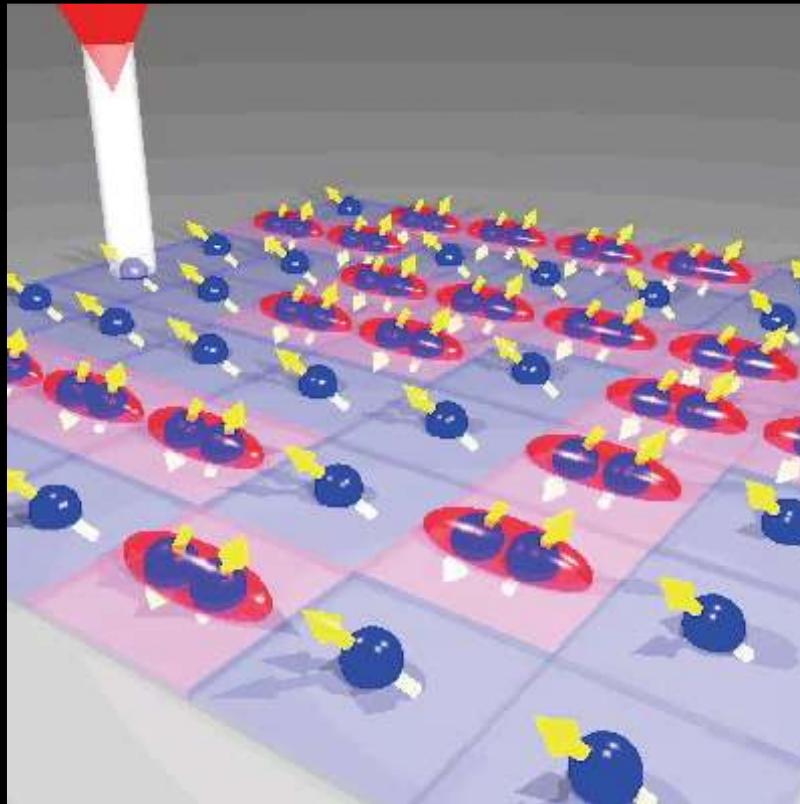
Requires SJTM with Spatial Resolution  $\sim$ 1nm

# VISUALIZE CUPRATE COOPER-PAIRS

$$I_J R = \Delta \quad \begin{matrix} \textit{Josephson} \\ \textit{(Cooper Pair)} \\ \textit{Current} \end{matrix}$$

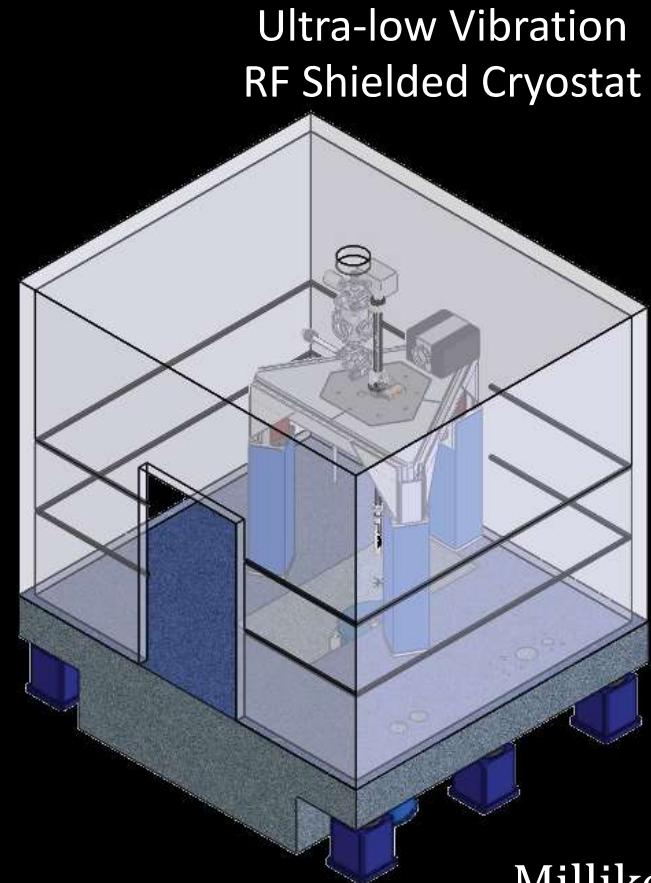
$$E_J = \frac{\hbar}{2e} I_J \quad \begin{matrix} \textit{Josephson} \\ \textit{Energy} \end{matrix}$$

$$E_J > kT \quad \begin{matrix} \textit{Phase} \\ \textit{Stabilization} \\ \textit{Temperature} \end{matrix}$$



Requires high gap  $\Delta > 10\text{meV}$  & Millikelvin Temperatures  $T \ll 1\text{K}$

# VISUALIZE CUPRATE COOPER-PAIRS

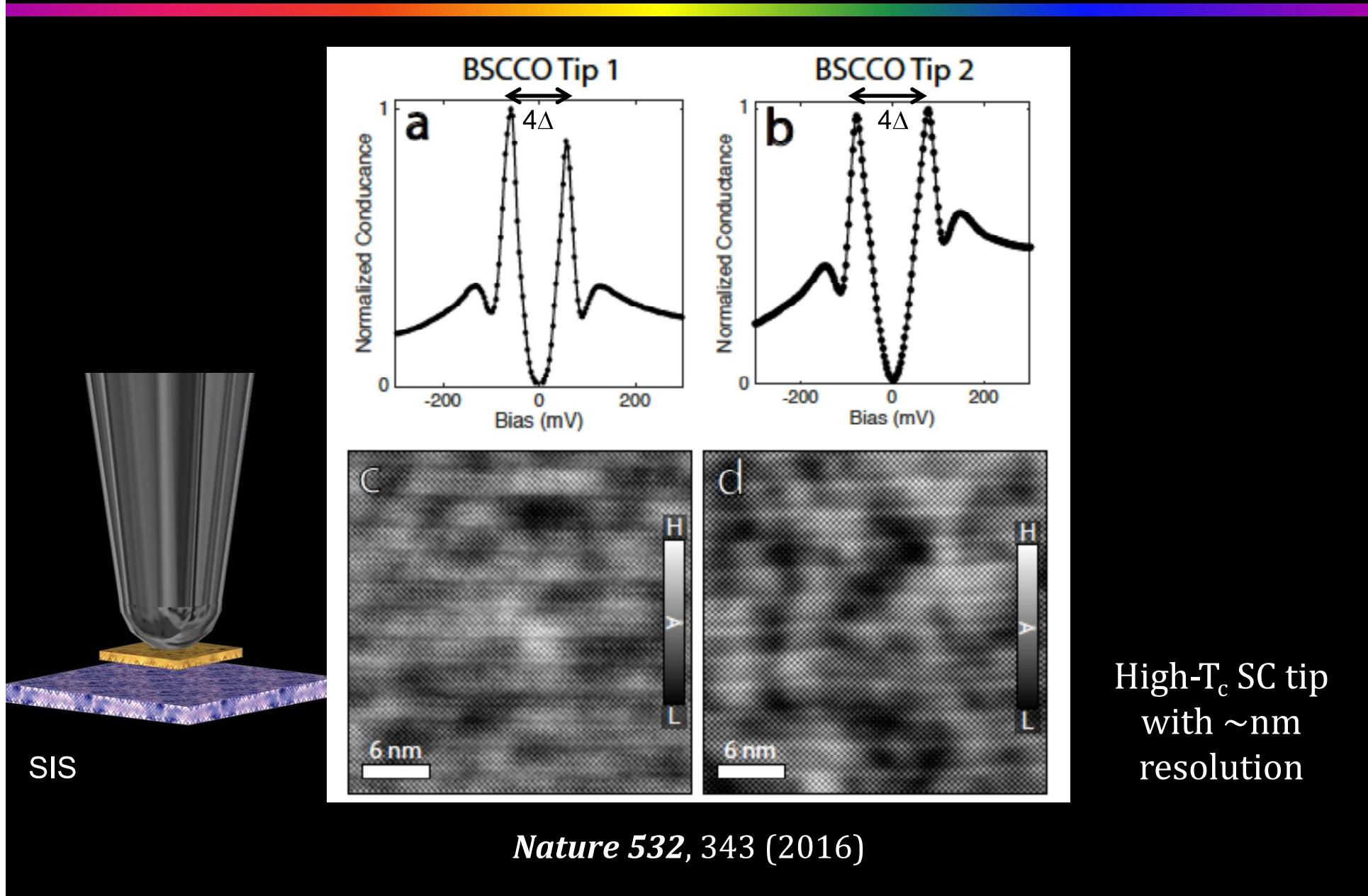


Custom Designed and Built  
mK-STM head

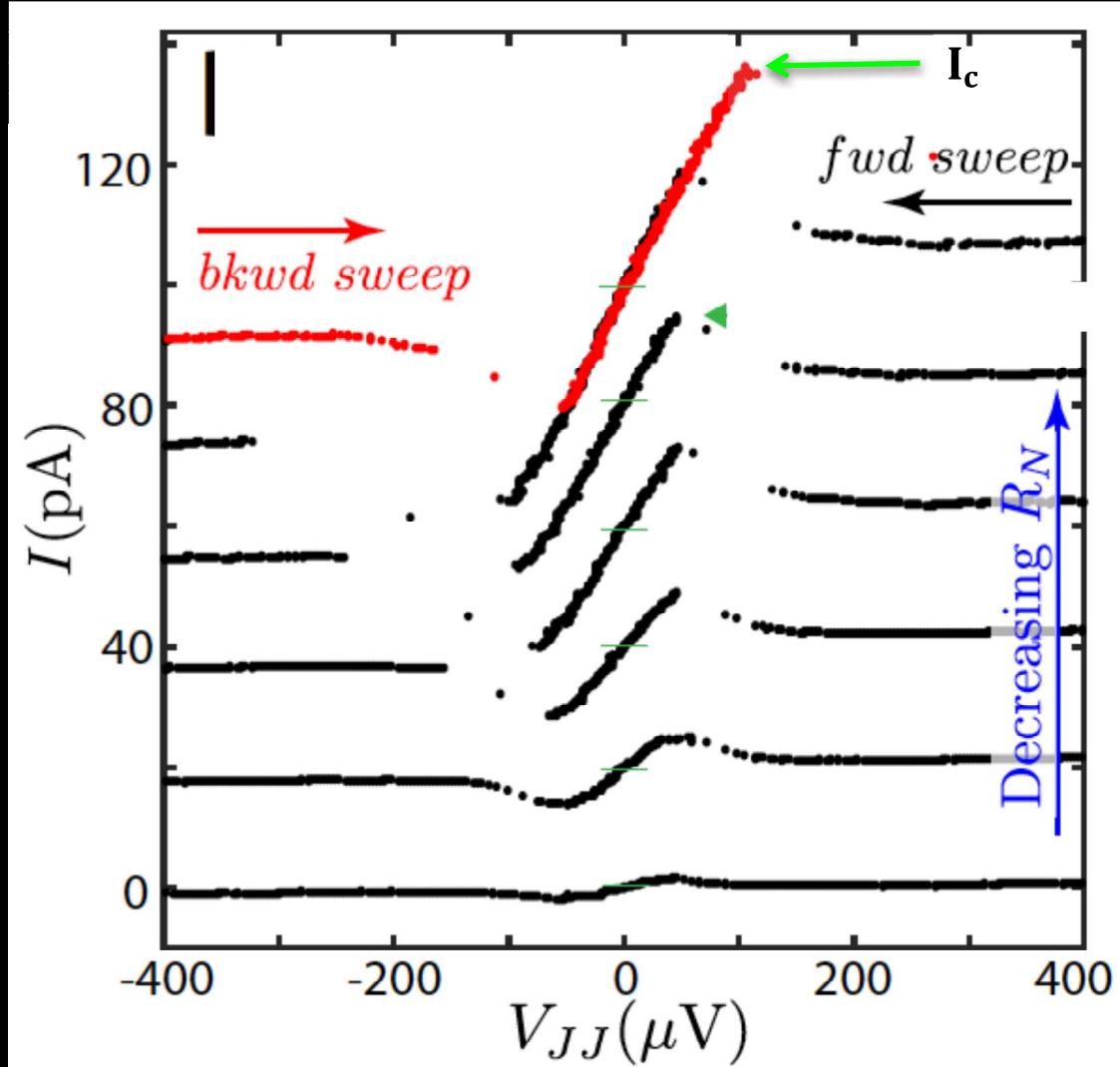


Millikelvin Operating Temperature STM

# HTS STM TIP: $\Delta \sim 25$ meV



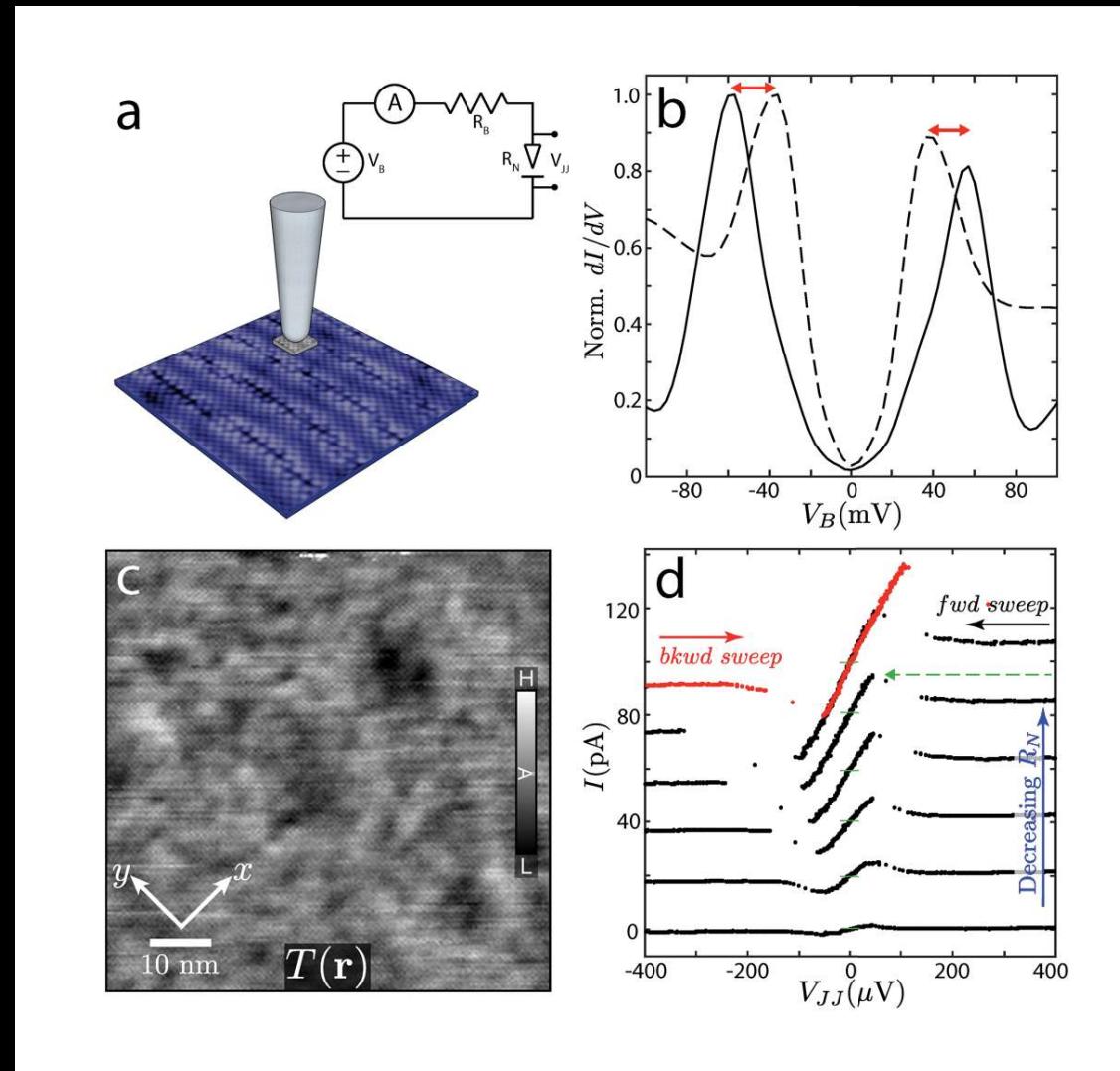
# JOSEPHSON CURRENT / HTS STM TIP



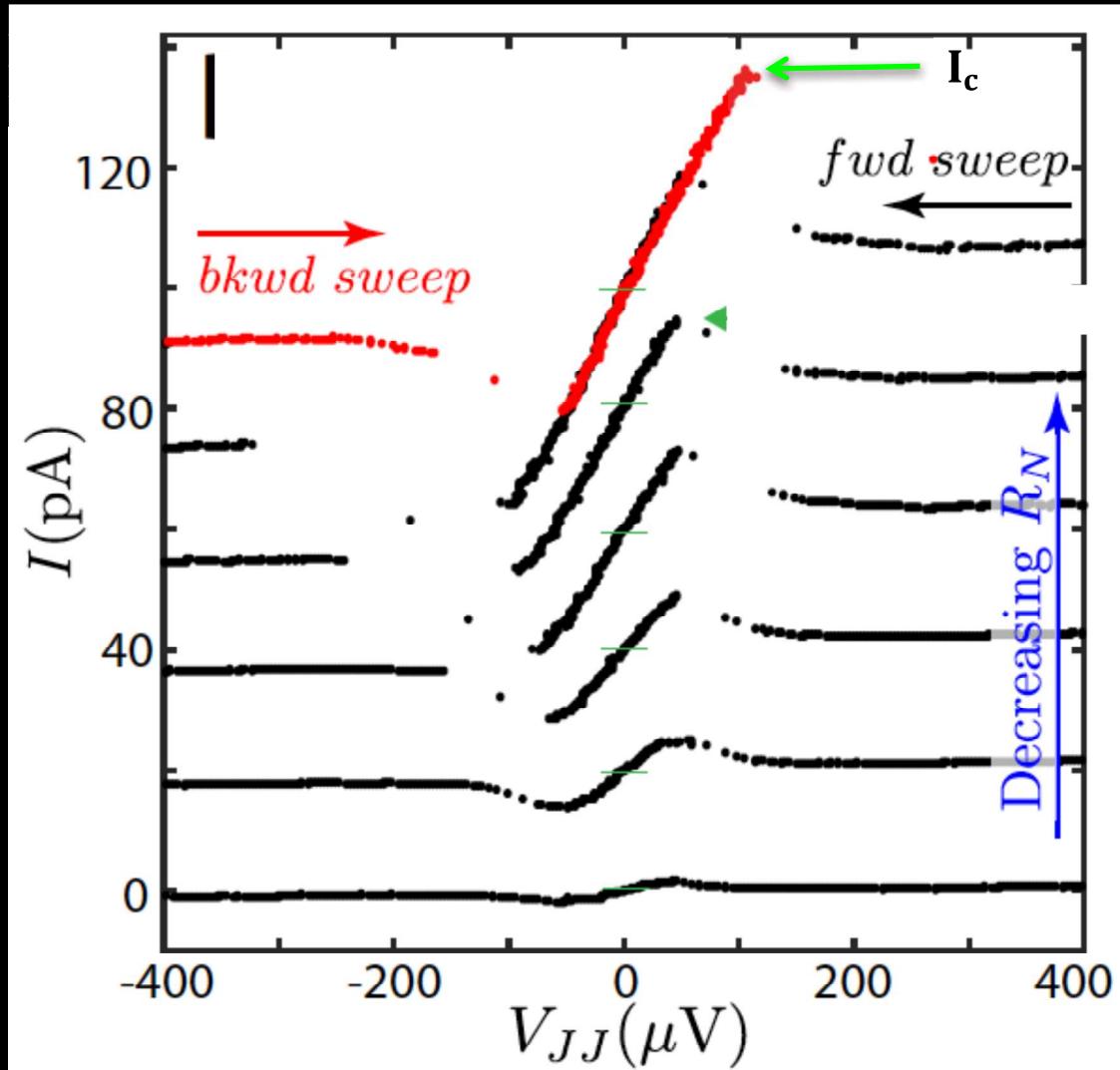
*d*-wave BSCCO tip  
 $T = 50\text{mK}$

*Nature* 532, 343 (2016)

50mK /  $\Delta_{\text{TIP}} \sim 25$  meV / nm RESOLUTION /  $R_N = 10 \text{ M}\Omega$  / 256X256



# SJTM IMAGING of $I_c(r)$ on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{x+8}$

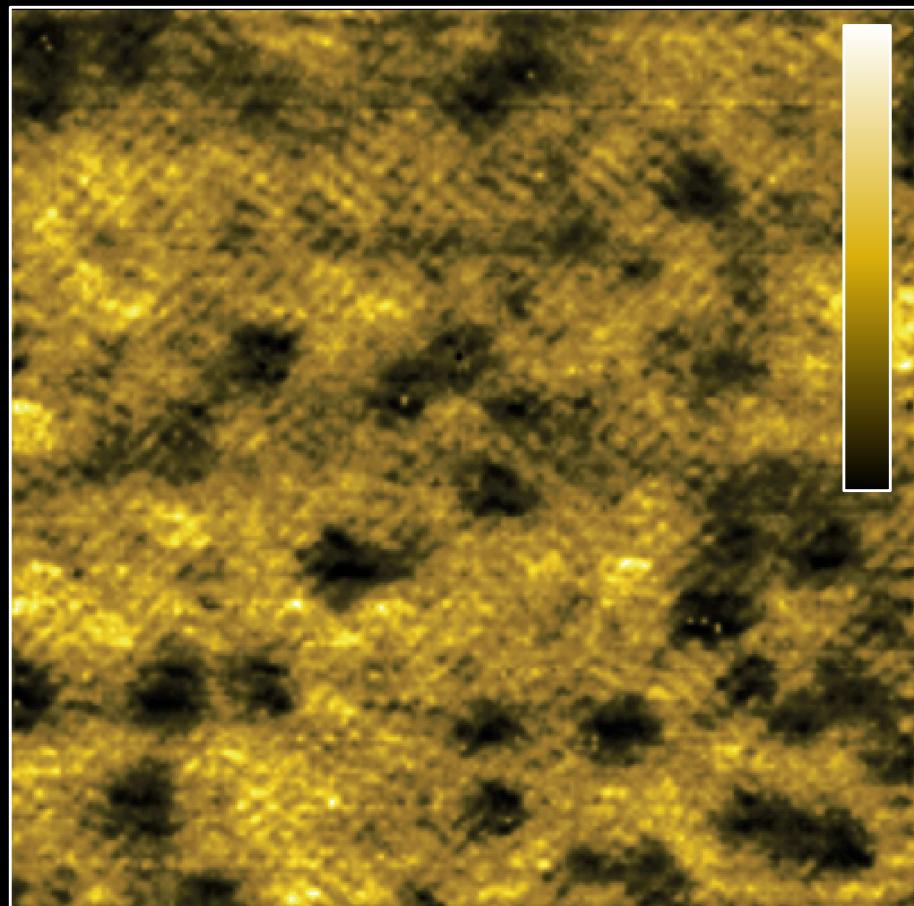


$$I_c(r) \propto I_J^2(r)$$

# SJTM IMAGING of $I_c(\mathbf{r})$ on $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{x+8}$

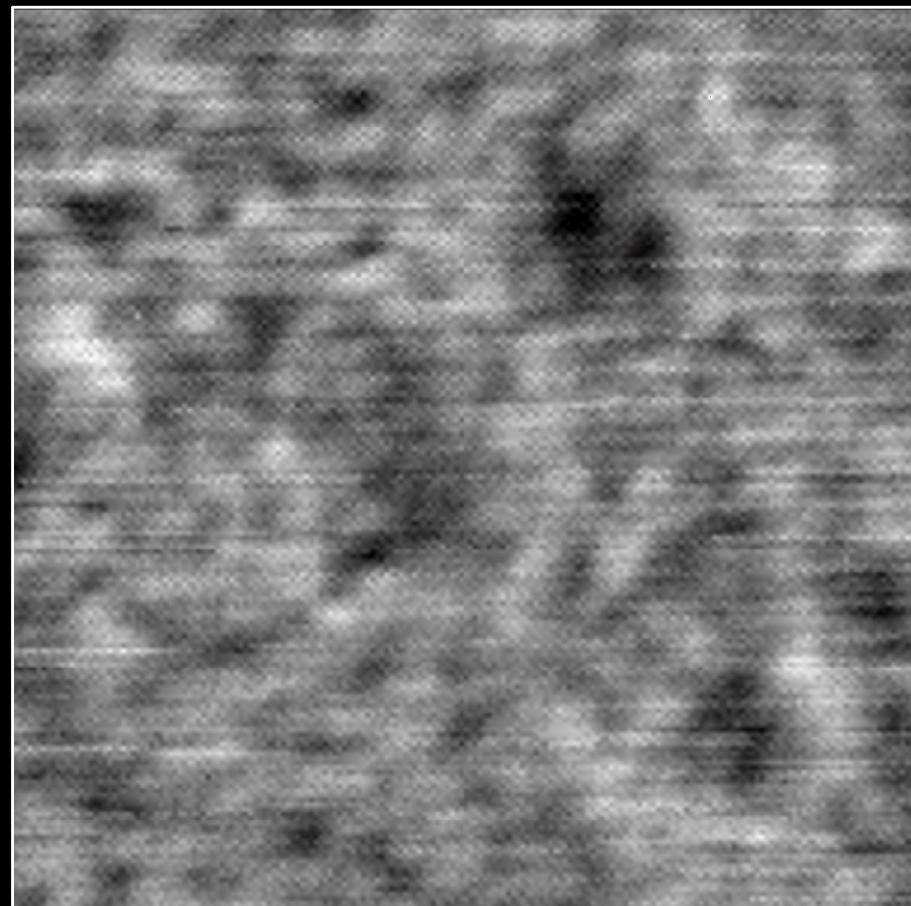
50mK

$I_c(\mathbf{r})$



Topography

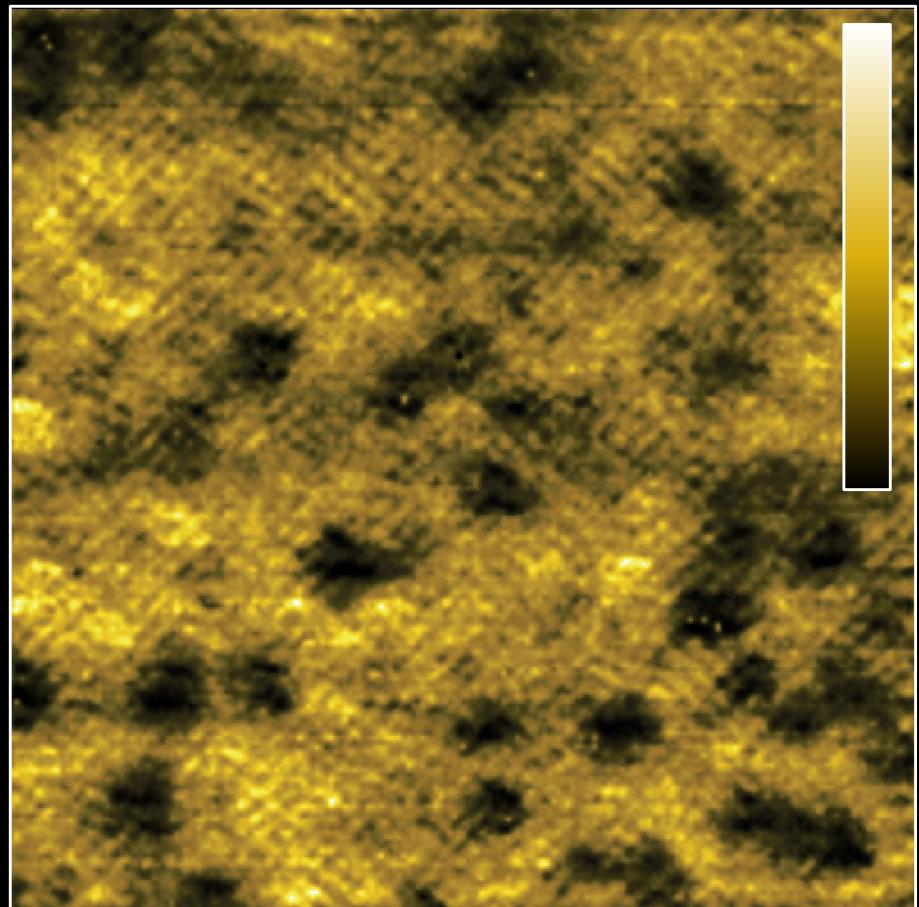
76x76nm



# VALIDATE COOPER-PAIR CONDENSATE IMAGING ?

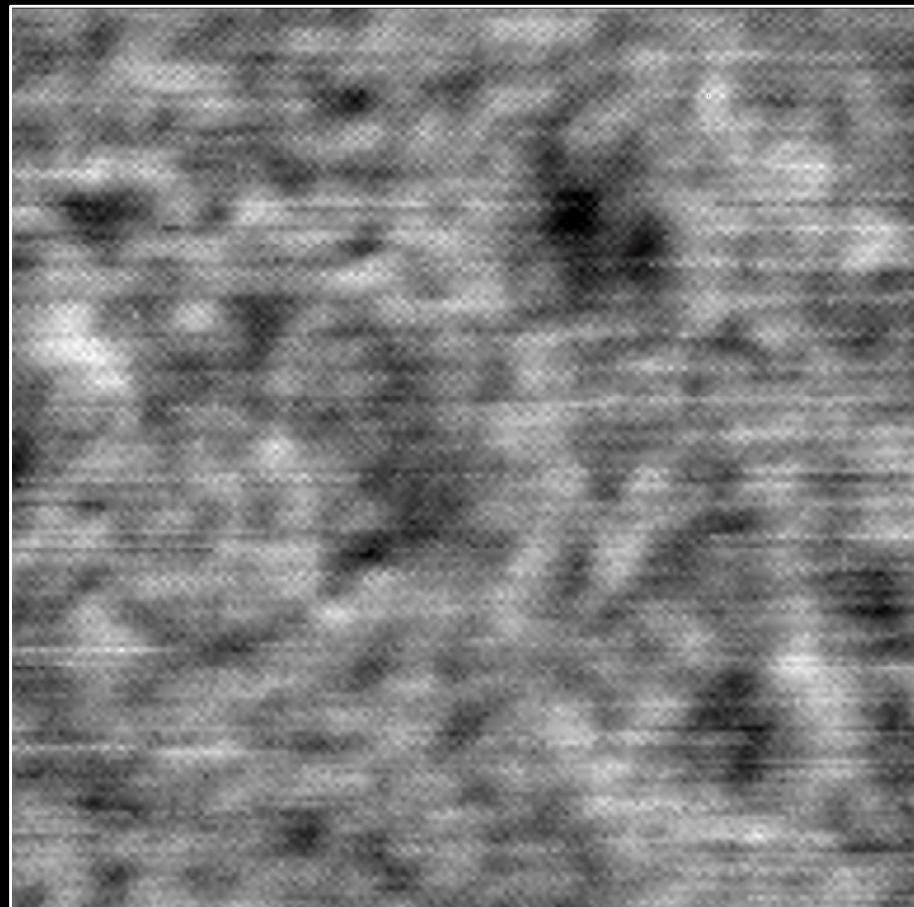
50mK

$I_c(\mathbf{r})$



Topography

76x76nm



*Nature* 532, 343 (2016)

# VALIDATE COOPER-PAIR CONDENSATE IMAGING ?

VOLUME 77, NUMBER 27

PHYSICAL REVIEW LETTERS

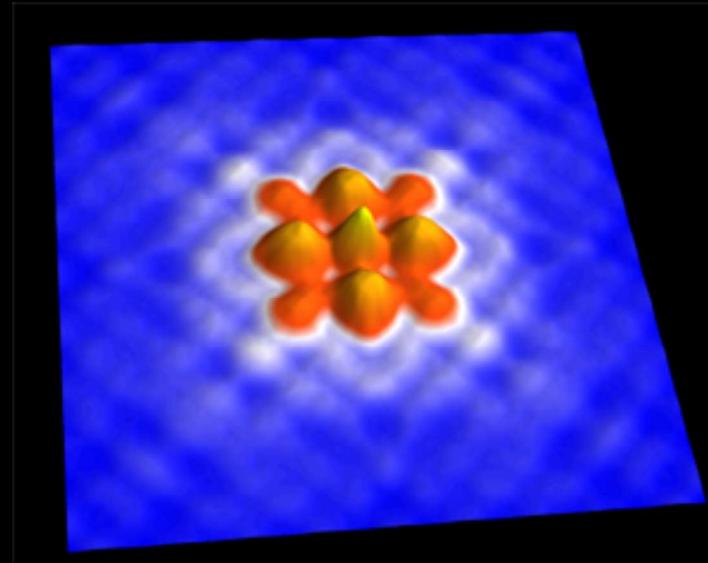
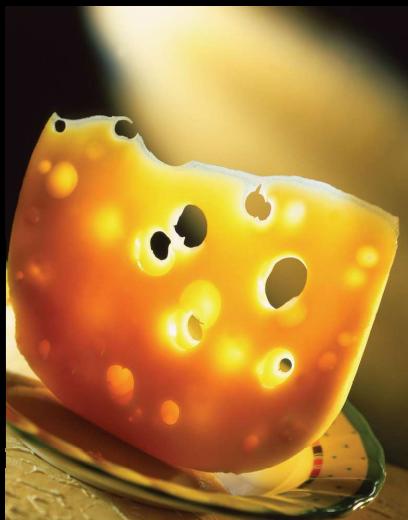
30 DECEMBER 1996

## Muon Spin Relaxation Studies of Zn-Substitution Effects in High- $T_c$ Cuprate Superconductors

B. Nachumi, A. Keren, K. Kojima, M. Larkin, G. M. Luke, J. Merrin, O. Tchernyshöv, and Y.J. Uemura  
*Physics Department, Columbia University, New York, New York 10027*

N. Ichikawa, M. Goto, and S. Uchida  
*Department of Superconductivity, University of Tokyo, Bunkyo-ku, Tokyo 113, Japan*  
(Received 12 September 1996)

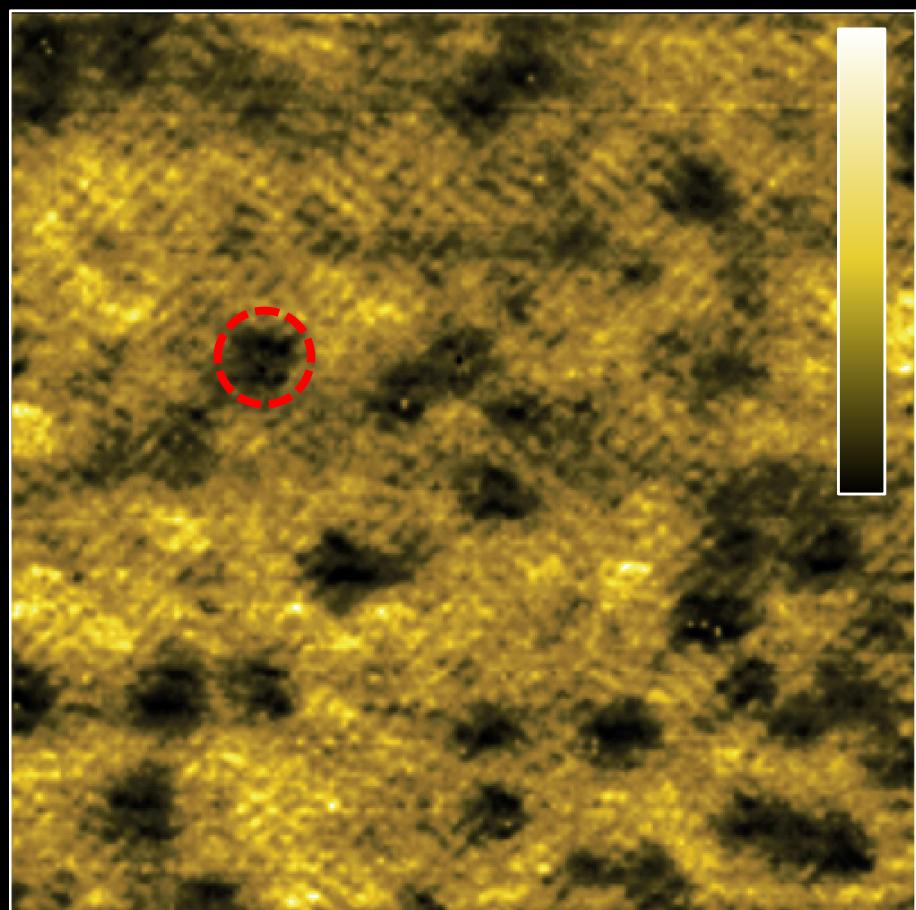
We have performed transverse-field muon spin relaxation measurements of the Zn-substituted cuprate high- $T_c$  superconductors:  $\text{La}_{2-x}\text{Sr}_x(\text{Cu}_{1-y}\text{Zn}_y)\text{O}_4$  and  $\text{YBa}_2(\text{Cu}_{1-y}\text{Zn}_y)_3\text{O}_{6.63}$ . The superconducting carrier density/effective mass  $n_s/m^*$  ratio at  $T \rightarrow 0$  decreases with increasing Zn concentration, in a manner consistent with our “swiss cheese” model in which charge carriers within an area  $\pi\xi_{ab}^2$  around each Zn are excluded from the superfluid. We discuss this result in the context of Bose condensation, pair localization, and pair breaking. [S0031-9007(96)02011-X]



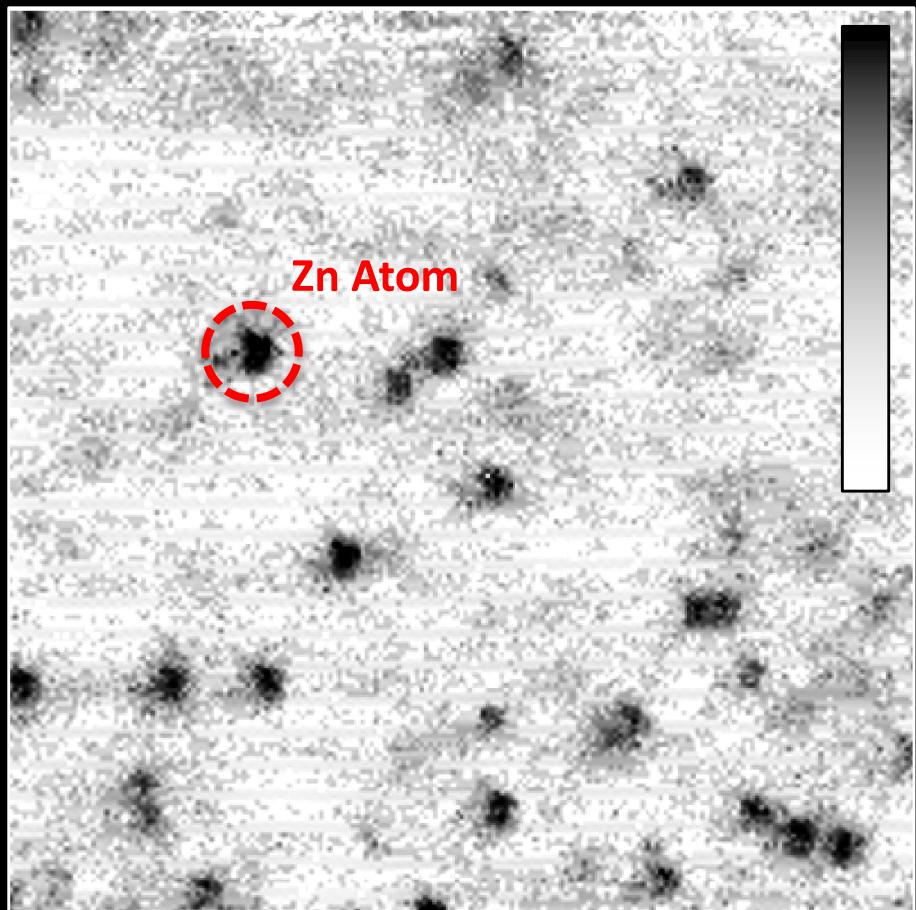
# VALIDATE COOPER-PAIR CONDENSATE IMAGING

50mK

$I_c(\mathbf{r})$



$dI/dV(\mathbf{r}, 20\text{mV})$

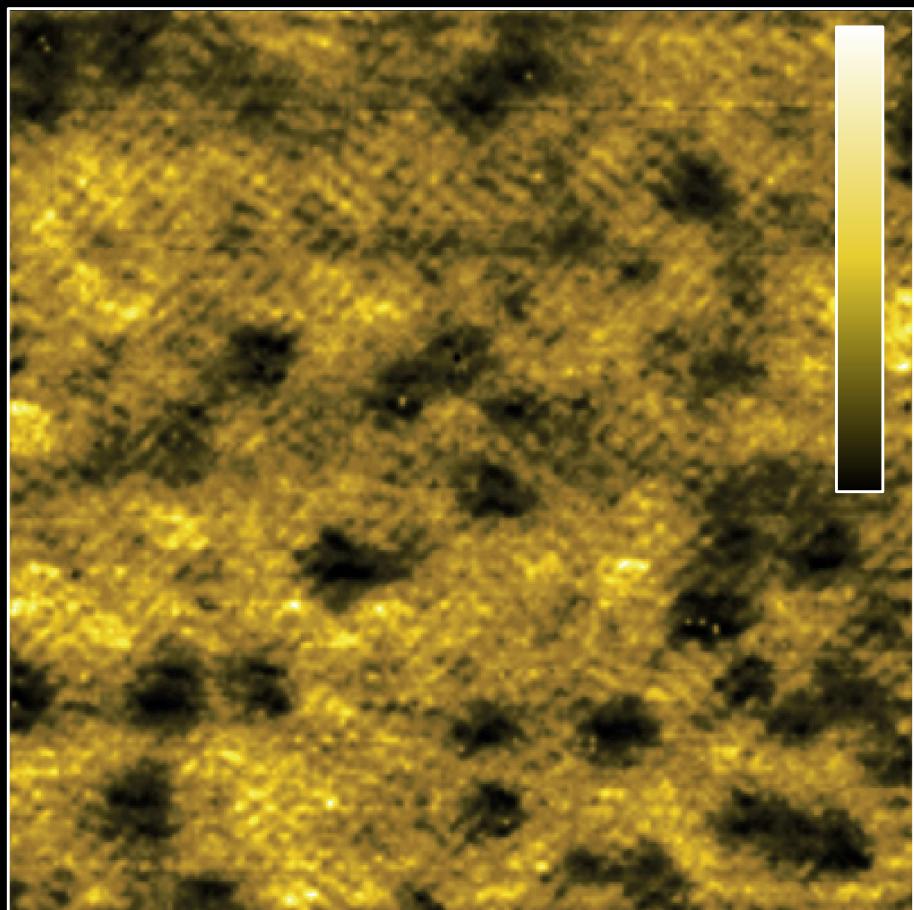


*Nature* 532, 343 (2016)

# SJTM IMAGING COOPER-PAIR CONDENSATE

50mK

$I_c(\mathbf{r})$

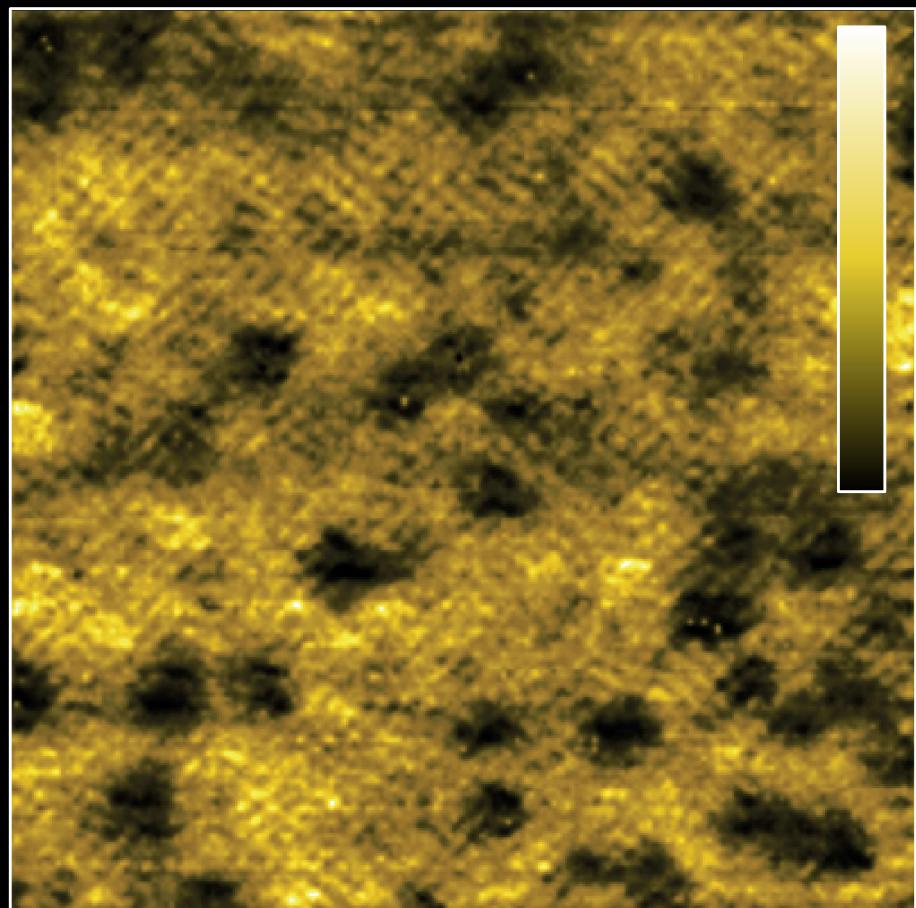


*Nature* 532, 343 (2016)

# CUPRATE COOPER-PAIR DENSITY MODULATIONS

50mK

$I_c(\mathbf{r})$



$I_c(\mathbf{q})$

(0,0.5)

(0.5,0)

+

+

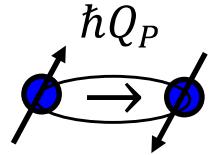
+

+

$$Q = (0,0.25)2\pi/a_0 ; (0.25,0)2\pi/a_0$$

*Nature* 532, 343 (2016)

# CUPRATE COOPER-PAIR DENSITY MODULATIONS



$$\left\langle c_{k\uparrow}^{\dagger}, c_{-k+Q_P\downarrow}^{\dagger} \right\rangle$$

$$\Delta(\mathbf{r}) = \Delta_P \left[ e^{i\mathbf{Q}_P \cdot \mathbf{r}} + e^{-i\mathbf{Q}_P \cdot \mathbf{r}} \right]$$

