Ising domain wall networks from intertwined CDWs in single-layer $TiSe_2$

Maria N Gastiasoro

Donostia International Physics Center

MINISTERIO DE CIENCIA E INNOVACIÓN

Financiado por la Unión Europea
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Plan de Recuperación, Transformación y Resiliencia

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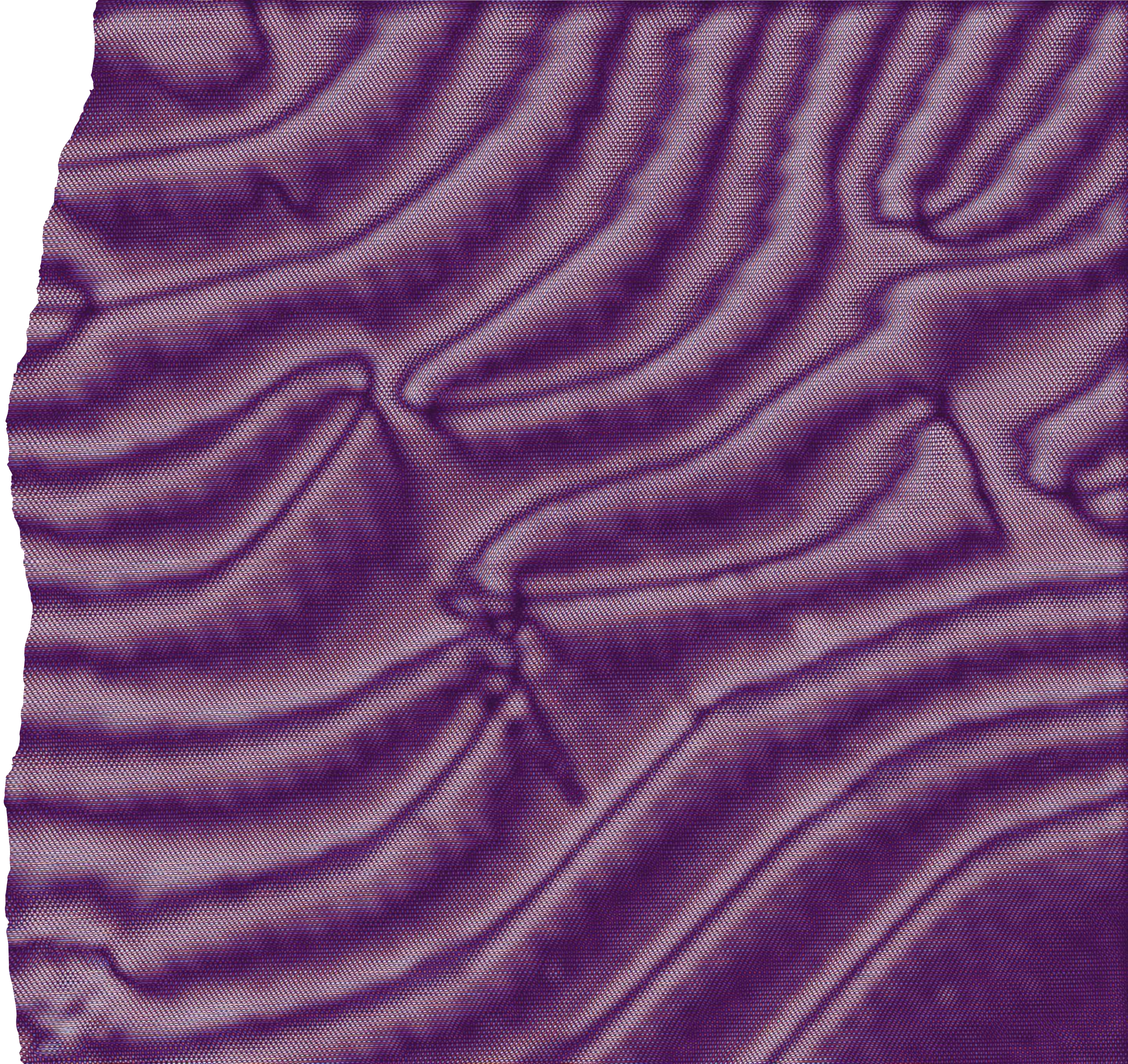
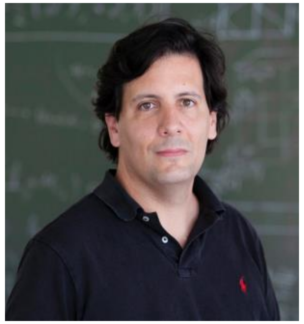


Acknowledgements

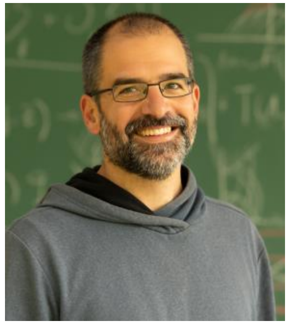
Experiments



M. M. Ugeda

W. Wan, P. Dreher

Theory



F. de Juan

D. Munoz-Segovia

W. Wan *et al*, arXiv:2411.05725 (2024)

MNG and F. de Juan, *in preparation*

Ramon y Cajal Fellowship RYC2021-031639-I

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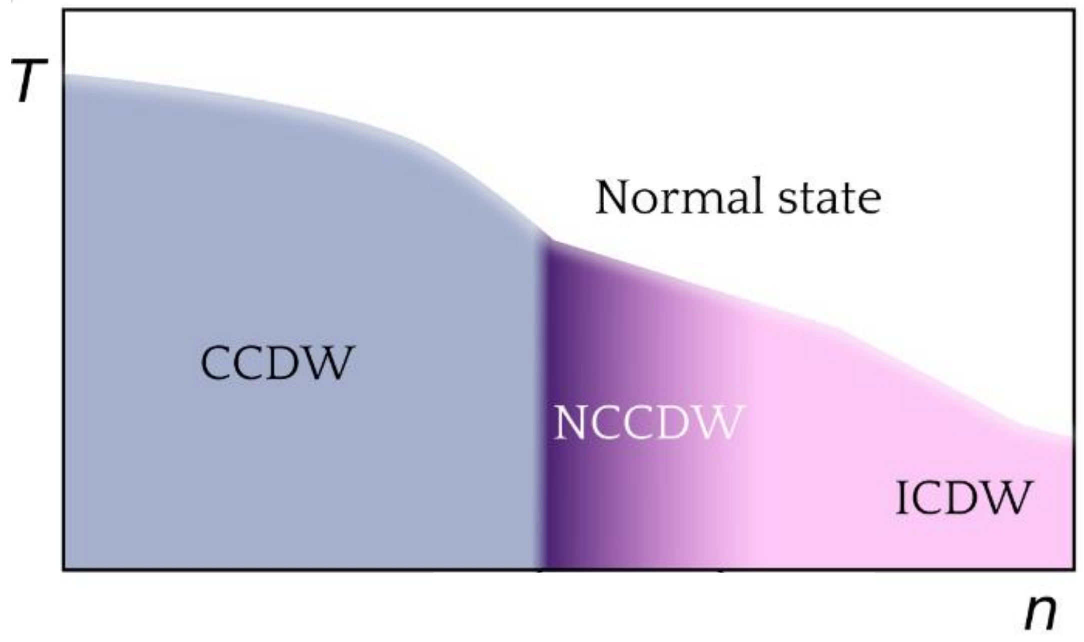
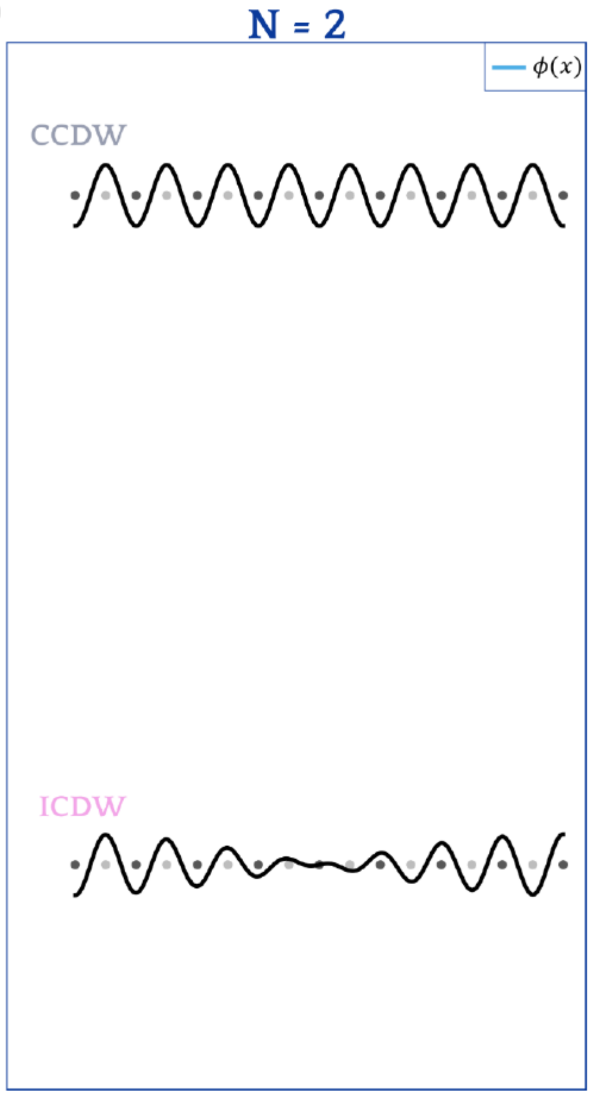
C-**NC**-I CDW transition with Q=G/N



$G/2 = \pi/a$

Q=G/2

Q_I= Q +δ



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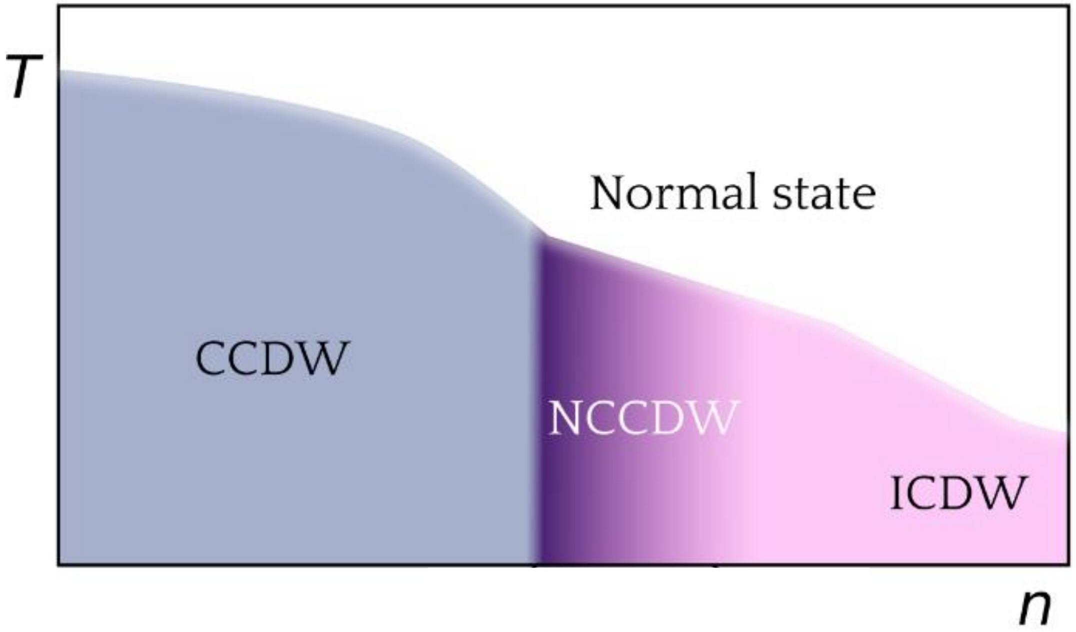

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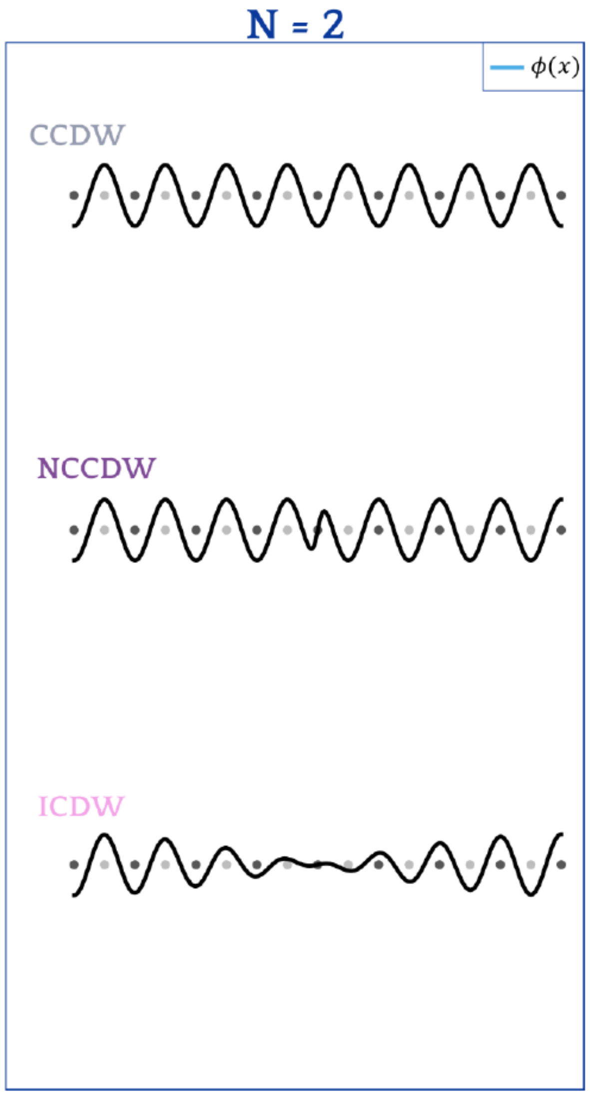

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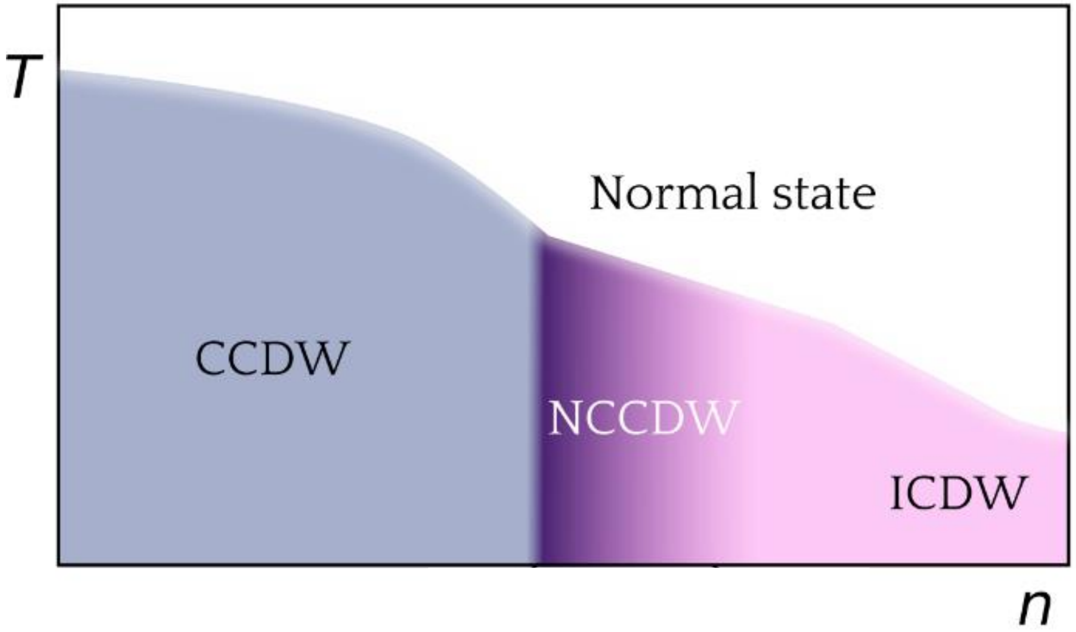


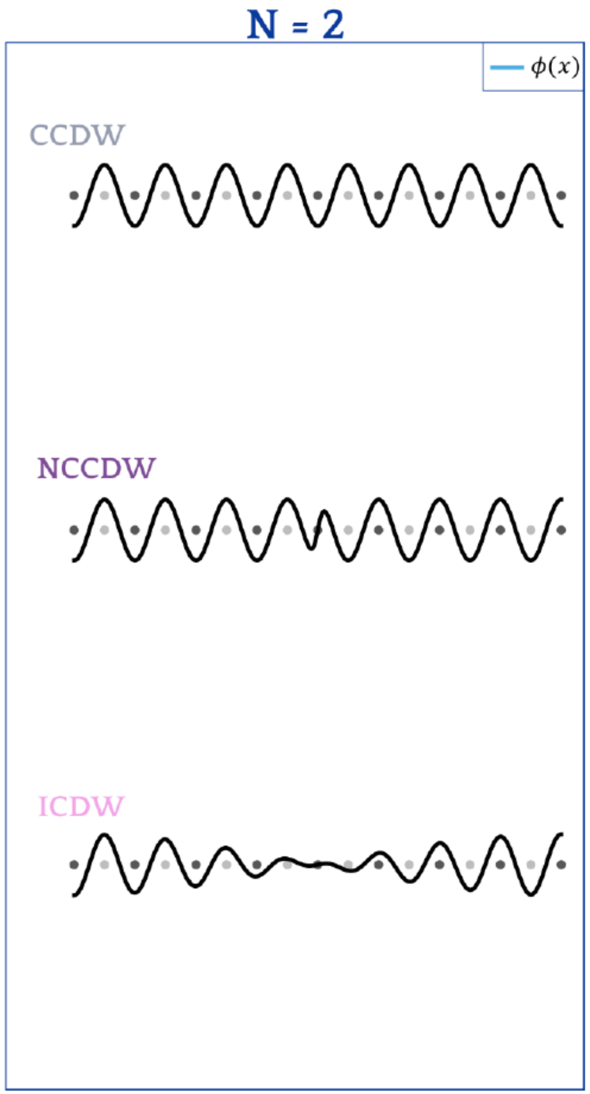
$G/2 = \pi/a$ $\rho(x) = \Re[\phi(x)e^{iQx}]$

Translations represented by sign change → *Real* order parameter $\phi(x)$!

$$\phi(x+a) = e^{iQa}\phi(x) = e^{i\pi}\phi(x) = -\phi(x)$$

Q=G/2

$Q_I = Q + \delta$



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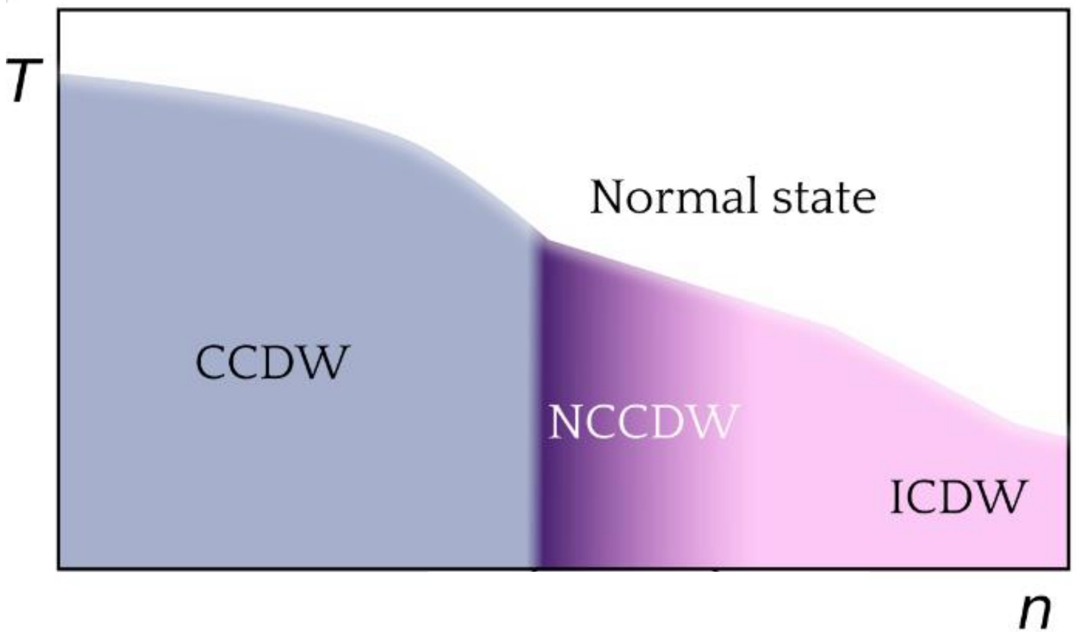


$$G/2 = \pi/a \qquad \rho(x) = \Re[\phi(x)e^{iQx}] \rightarrow \phi(x)\cos(\frac{\pi}{a}x)$$

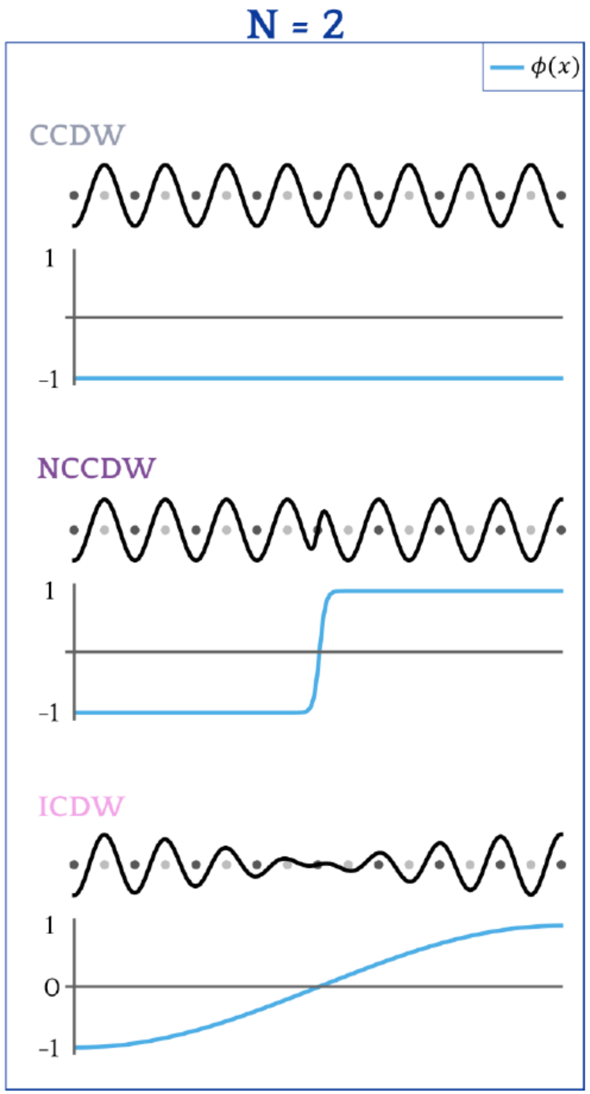
Translations represented by sign change → *Real* order parameter $\phi(x)$!

$$\phi(x+a) = e^{iQa}\phi(x) = e^{i\pi}\phi(x) = -\phi(x)$$

Q=G/2

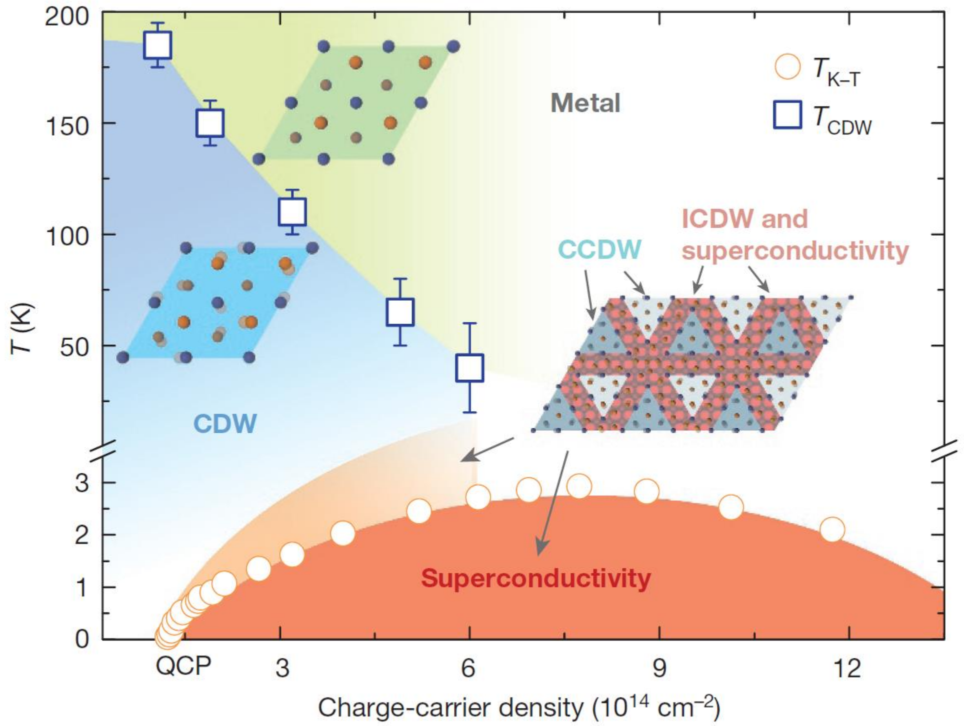
Ising domain

Q_I= Q +δ



W. L. McMillan, Phys. Rev. B (1976), K. Nakanishi & H. Shiba, J. Phys. Soc. Jap. (1977)...

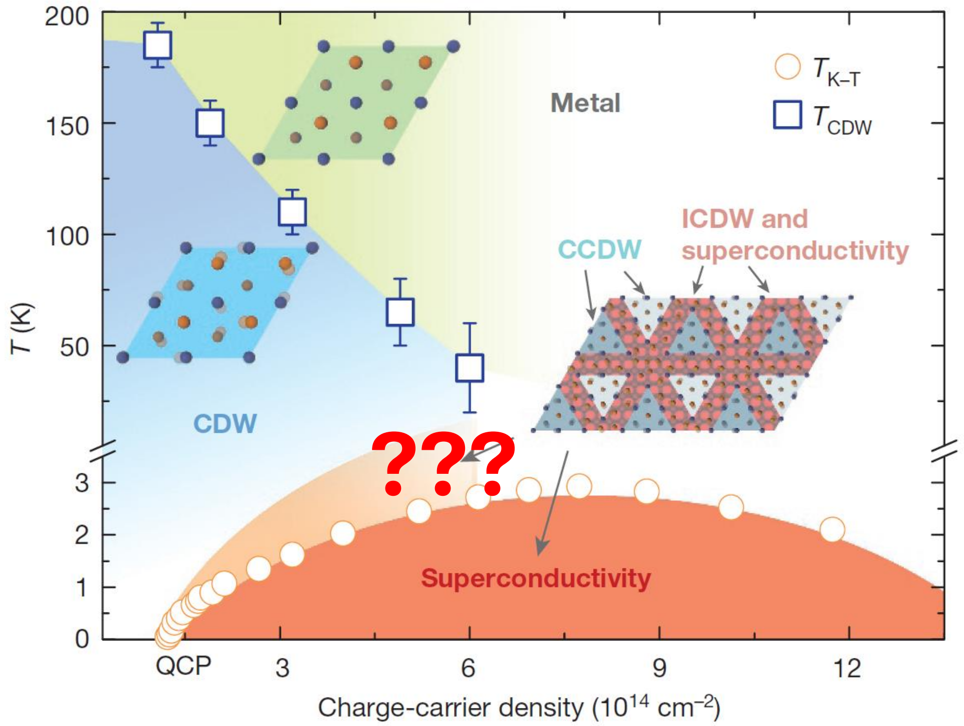
Bulk $TiSe_2$: example of CDW with N=2



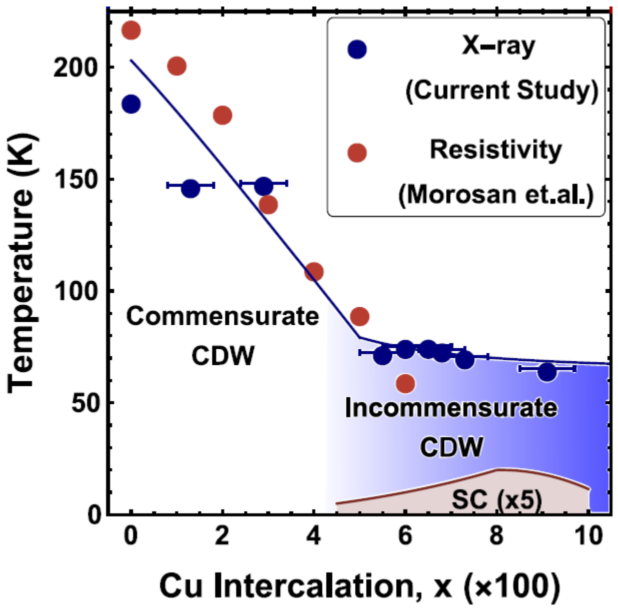
Castro-Neto, Nature (2016)

ICDW state and DWs may play an important role in the emergence of SC

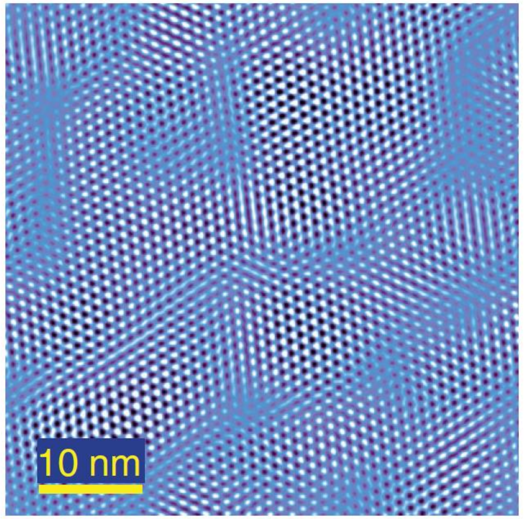
Bulk $TiSe_2$: example of CDW with N=2



Castro-Neto, Nature (2016)



Out-of-plane ICDW



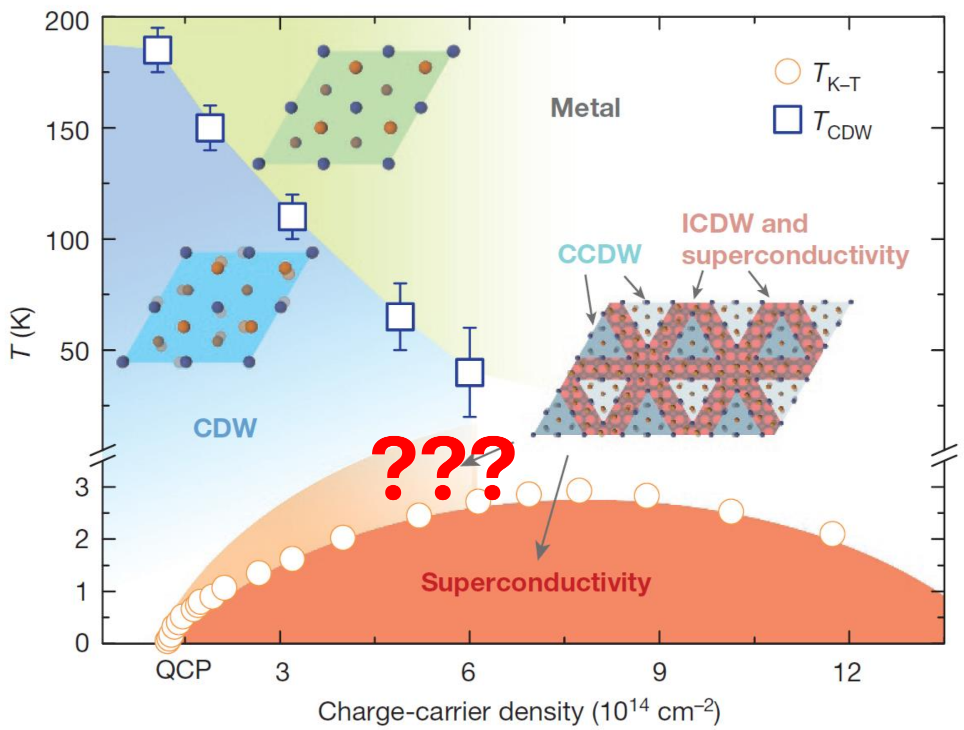
In-plane ICDW, CCDW domain

Domains in Cu doped bulk samples. CDW OPs?

ICDW state and DWs may play an important role in the emergence of SC

Kogar, PRL (2017), Novello PRL (2017), Yan PRL (2017), Spera PRB (2019), Pasztor PRR (2019)...

Bulk $TiSe_2$: example of CDW with N=2



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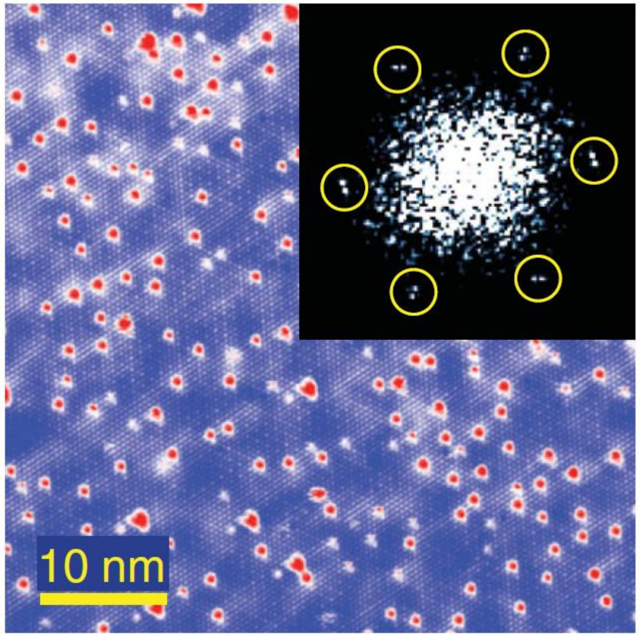
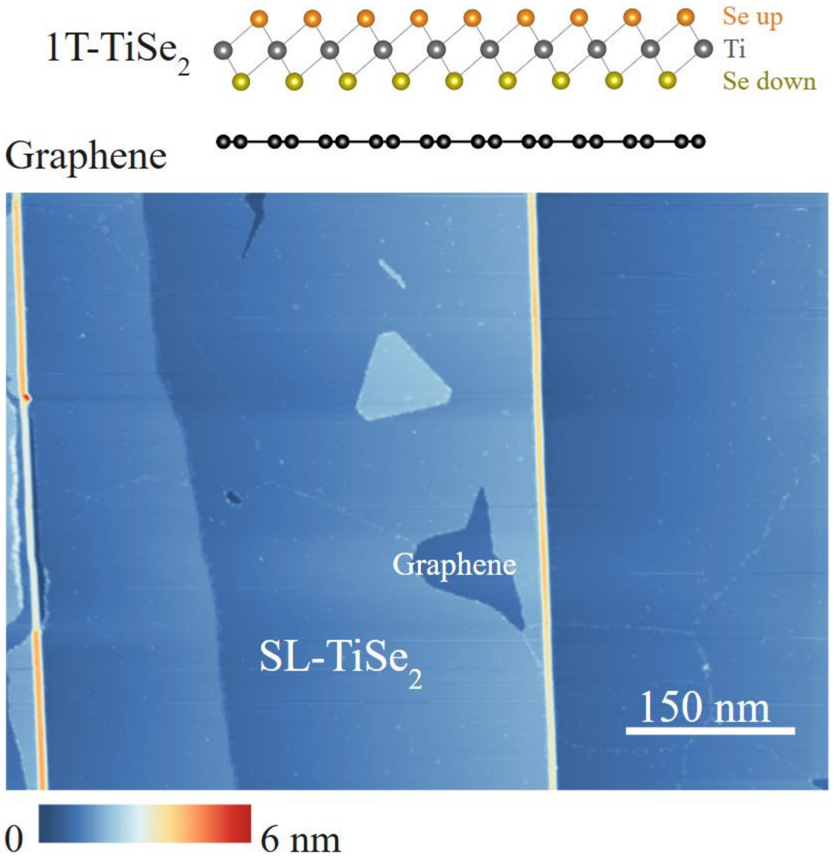


Domains in Cu doped bulk samples. CDW OPs? Role of disorder in domain formation?

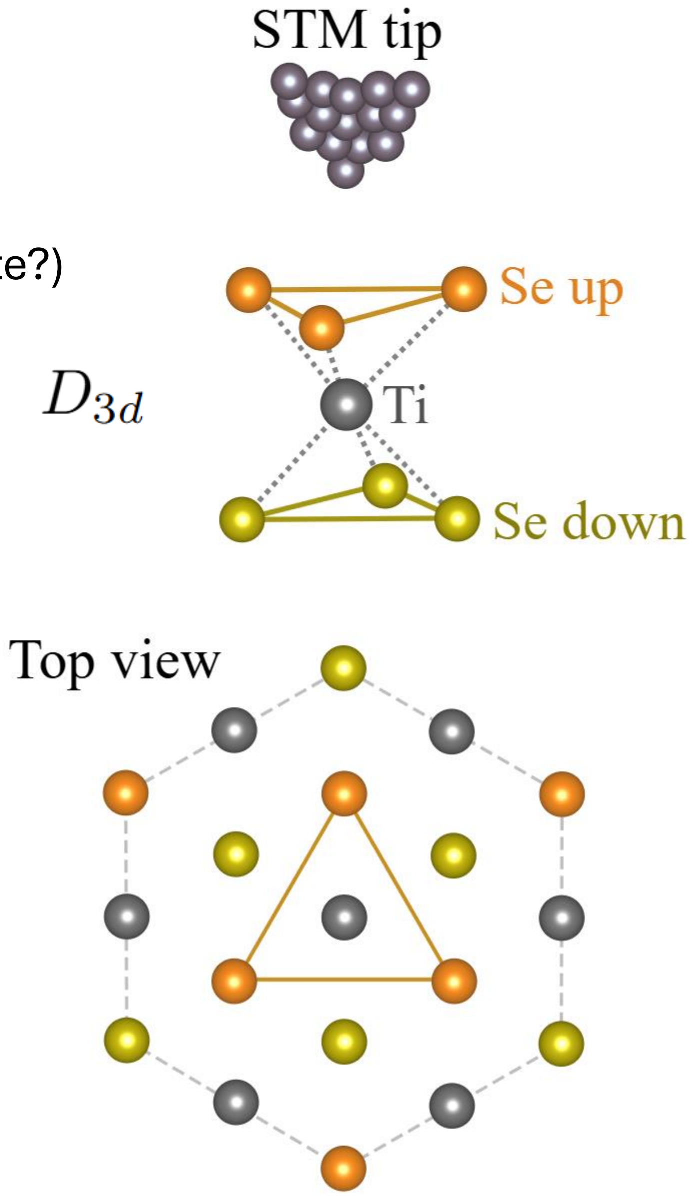
Kogar, PRL (2017), Novello PRL (2017), Yan PRL (2017), Spera PRB (2019), Pasztor PRR (2019)...

Monolayer $TiSe_2$ (Ugeda's lab)



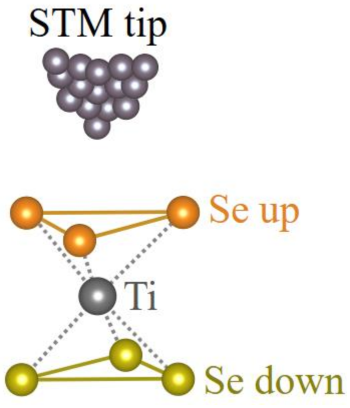
Small electron doping
(charge transfer from substrate?)

Single-crystal domains of hundreds of nm
Density of defects below 10^{12} cm^{-2}

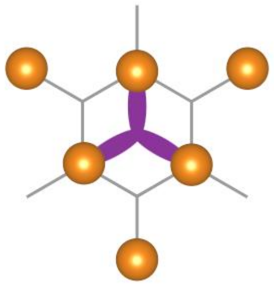


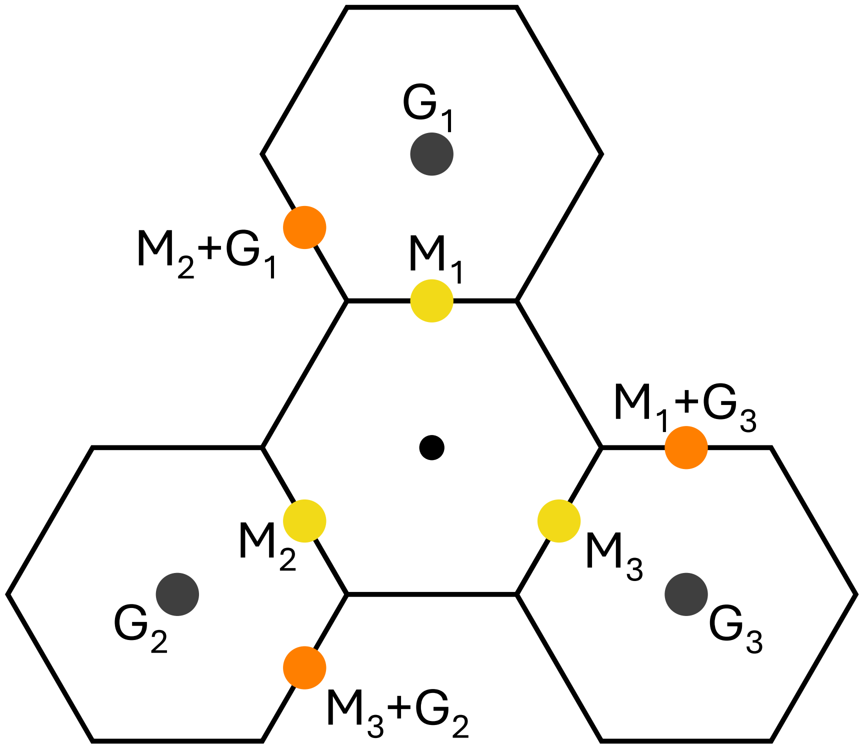
CDW in small scales: 2x2 triple-Q

$$\vec{Q}_n = \vec{G}_n/2 = \vec{M}_n$$



$T > T_c$





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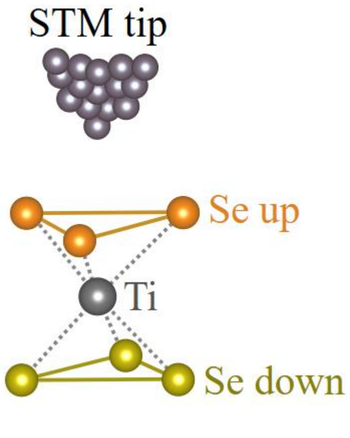

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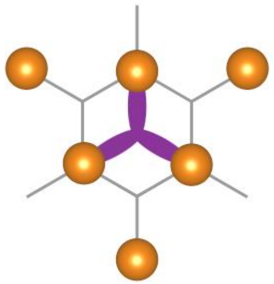

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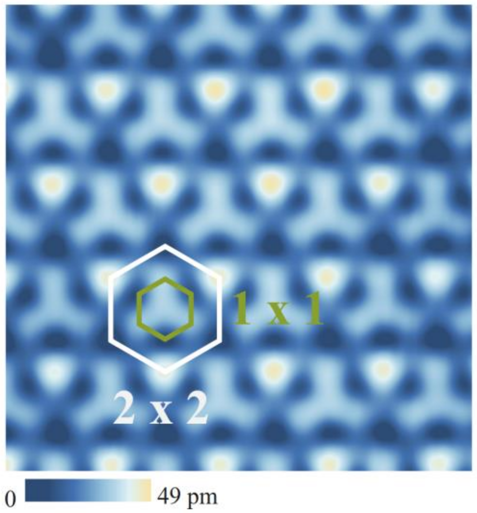

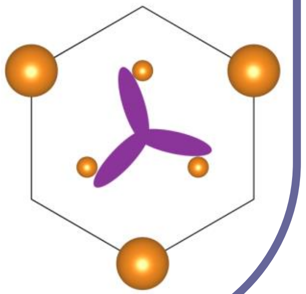

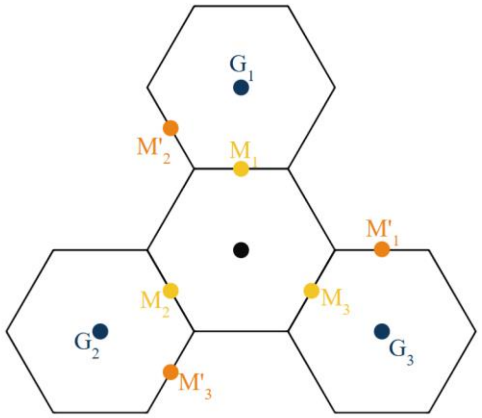

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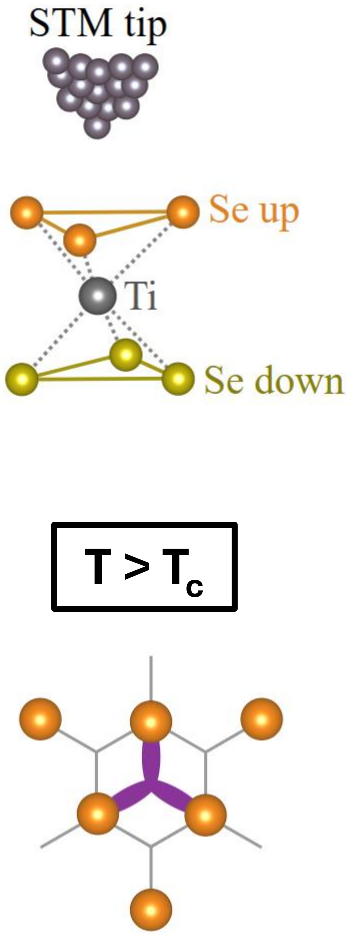

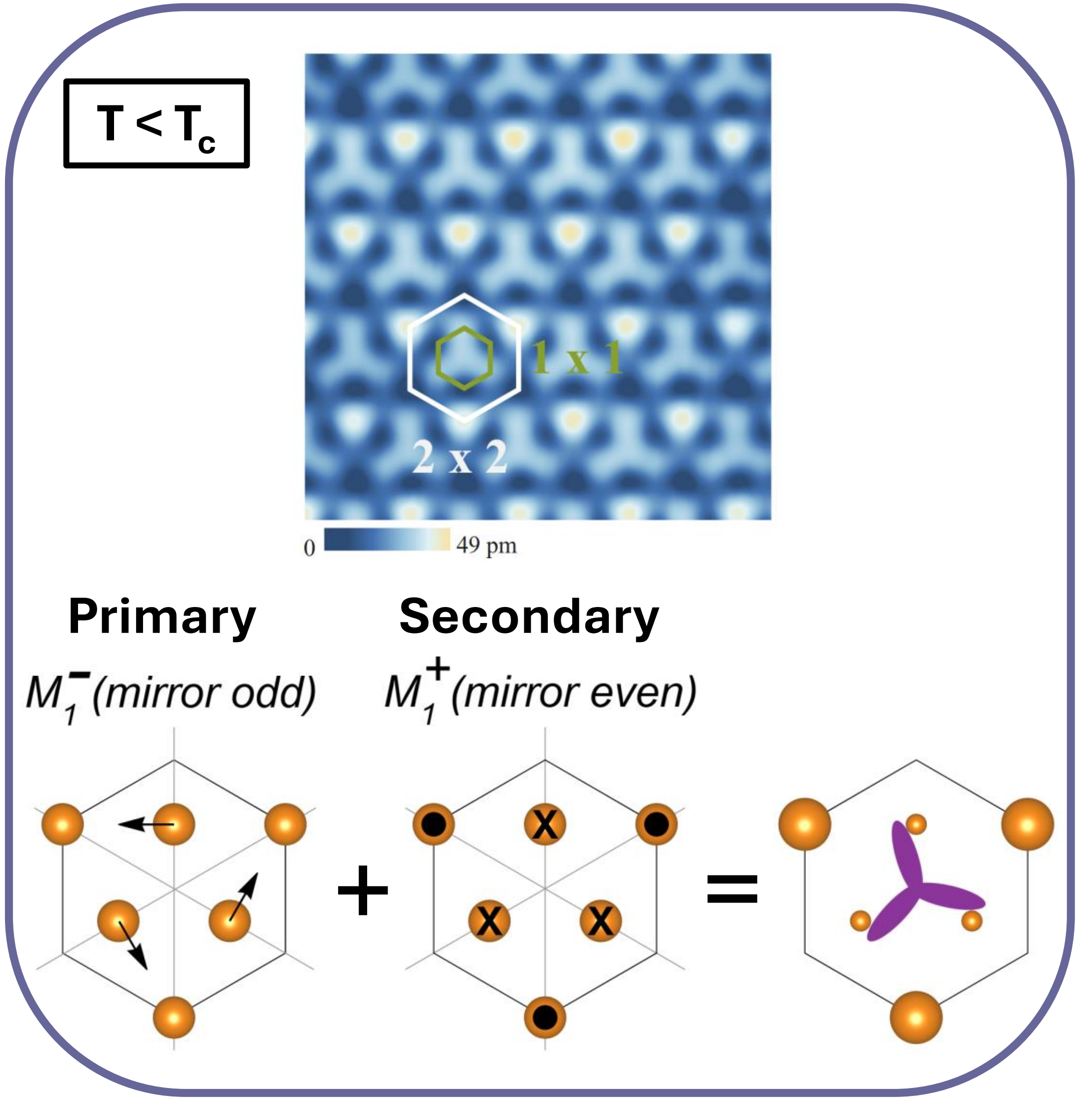

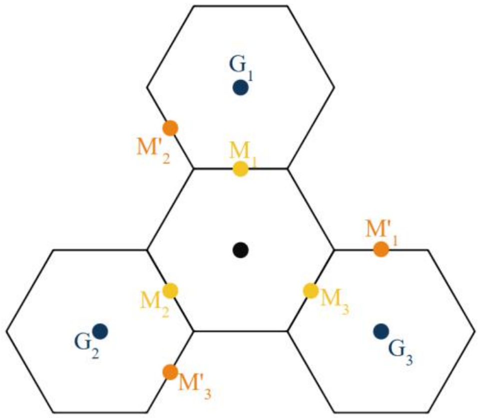

$$\rho_{2\times2}(\vec{x}) = 2\Re\sum_{n}\left[A_n^{M}(\vec{x})e^{-i\vec{M}_n\vec{x}} + A_n^{M'}(\vec{x})e^{-i\vec{M}'_n\vec{x}}\right]$$

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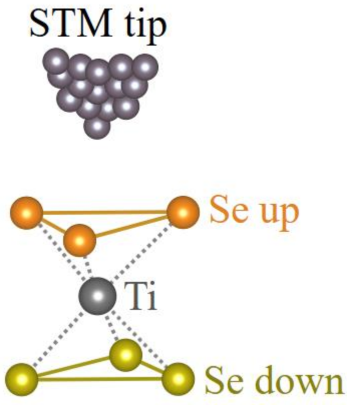
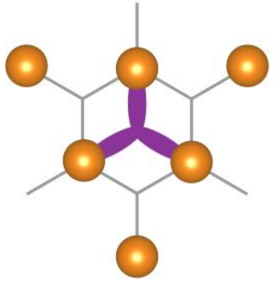


T > T$_c$



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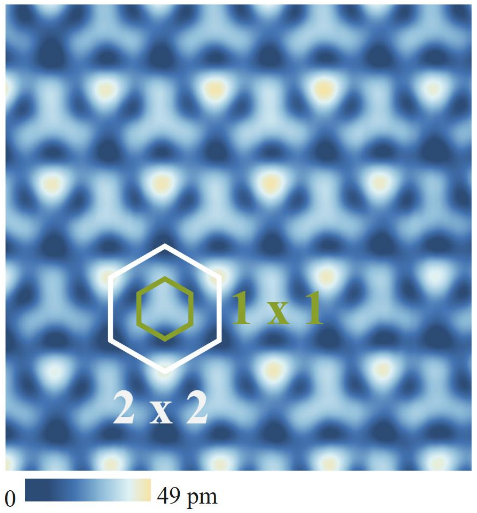
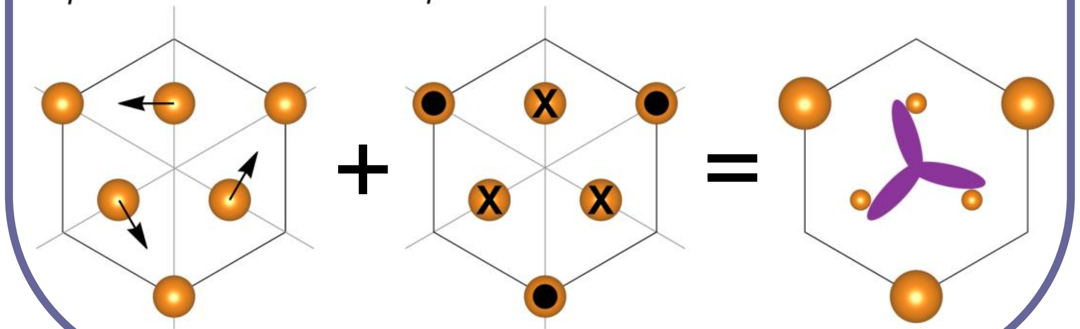


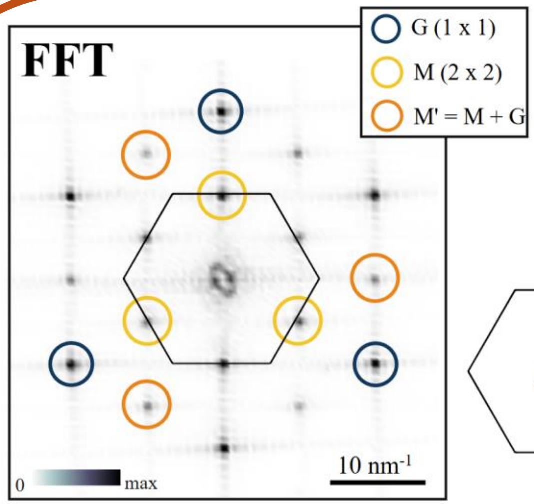
Primary

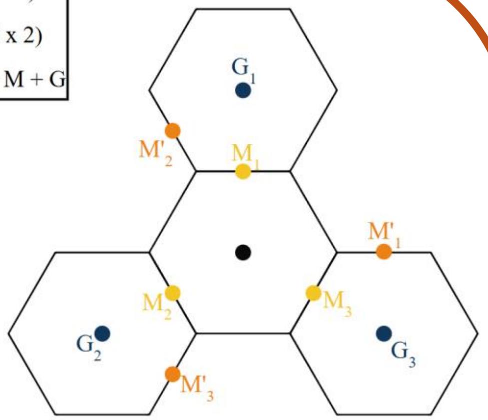
M_1^- *(mirror odd)*

Secondary

M_1^+ *(mirror even)*







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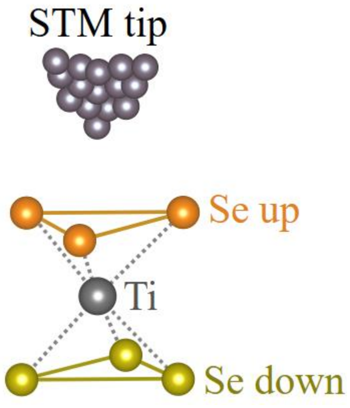

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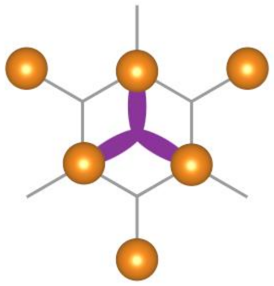

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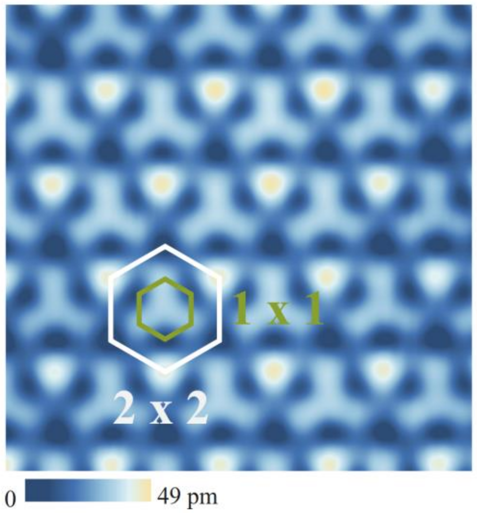
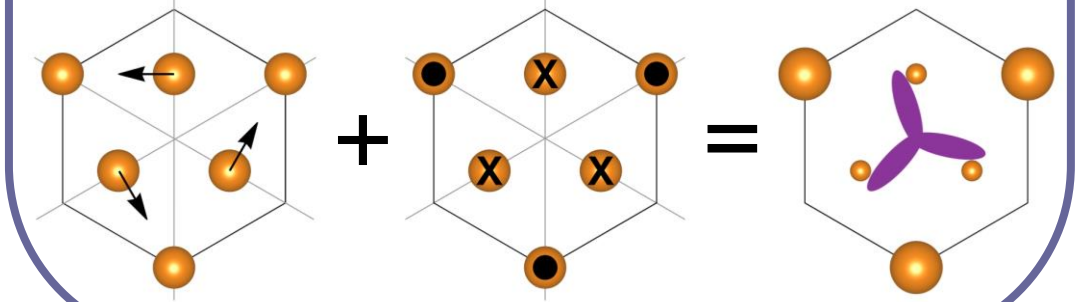
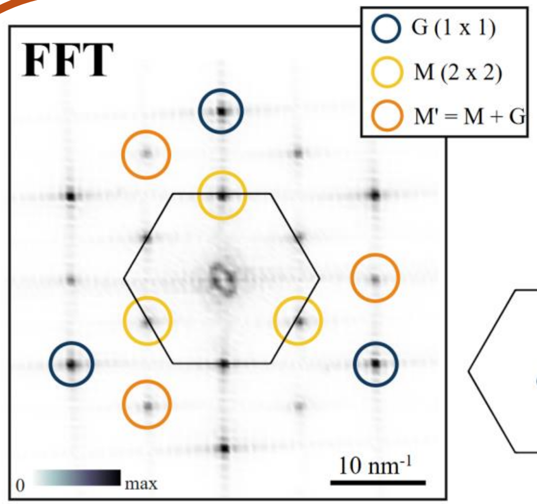
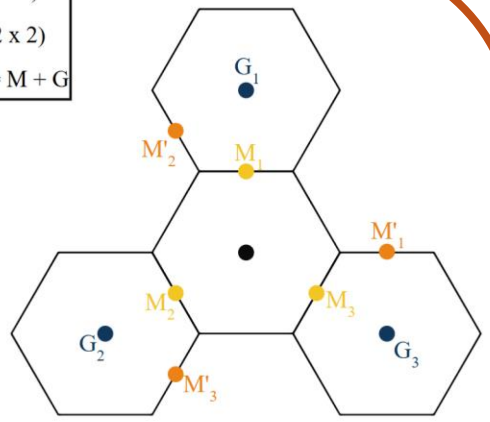


Primary M_1^- *(mirror odd)*

Secondary M_1^+ *(mirror even)*







Under σ$_v$: $e^{i\vec{M}_n\vec{x}} \rightarrow e^{i\vec{M}_n\vec{x}}$

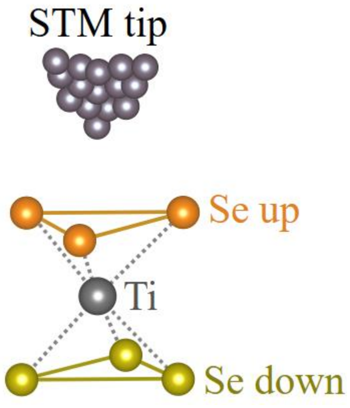
$$\Re A_n^M, \Im A_n^M \rightarrow M_1^+$$

Secondary !!!

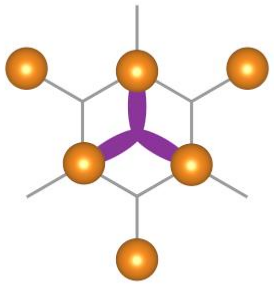
$$\rho_{2\times2}(\vec{x}) = 2\Re\sum_n \left[A_n^M(\vec{x})e^{-i\vec{M}_n\vec{x}} + A_n^{M'}(\vec{x})e^{-i\vec{M}'_n\vec{x}}\right]$$

CDW in small scales: 2x2 triple-Q

$$\vec{Q}_n = \vec{G}_n/2 = \vec{M}_n$$



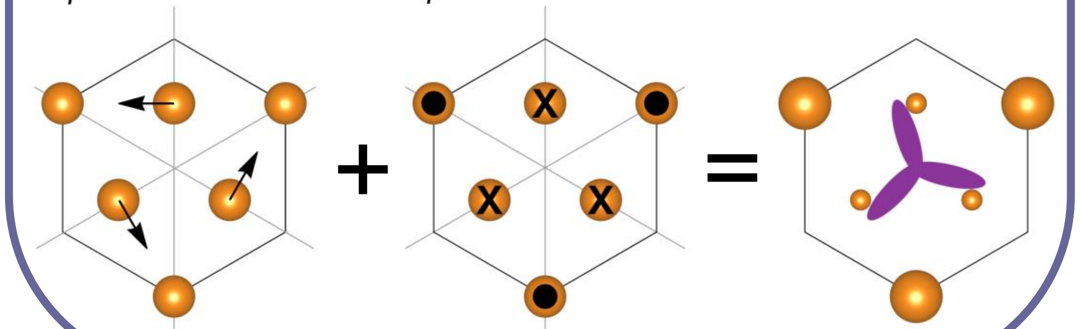
$T > T_c$

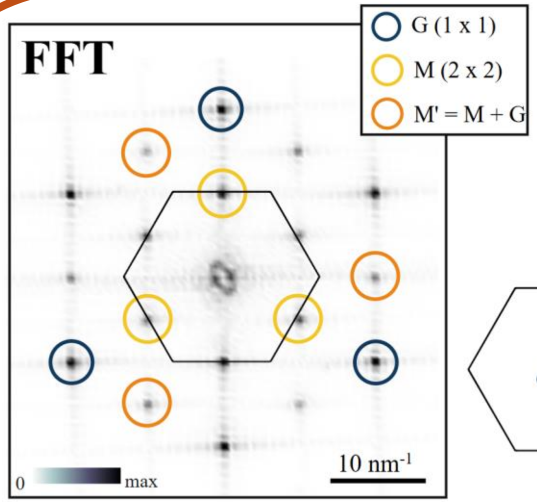


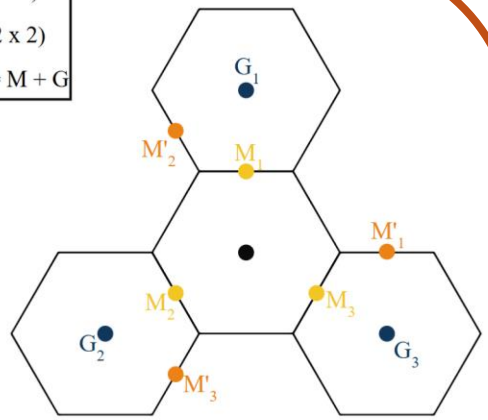
$T < T_c$

Primary M_1^- *(mirror odd)*

Secondary M_1^+ *(mirror even)*



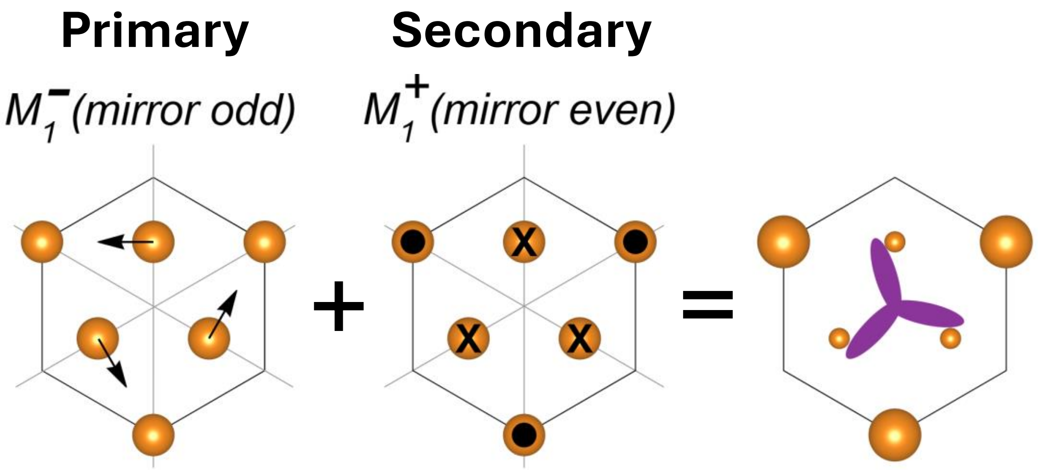


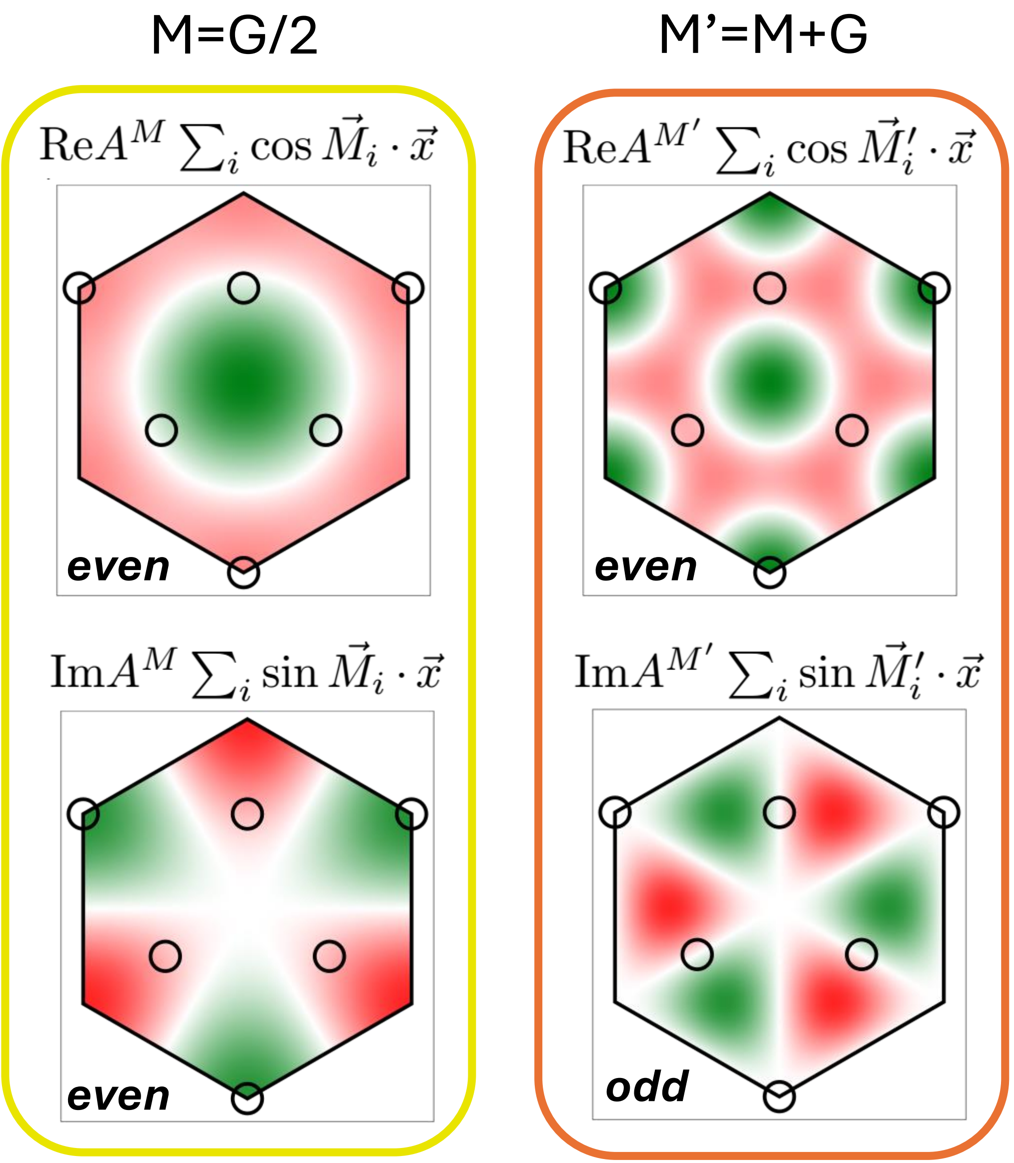


Under σ_v:

$$e^{i\vec{M}_n\vec{x}} \to e^{i\vec{M}_n\vec{x}}$$
$$e^{i\vec{M}'_n\vec{x}} \to e^{-i\vec{M}'_n\vec{x}}$$

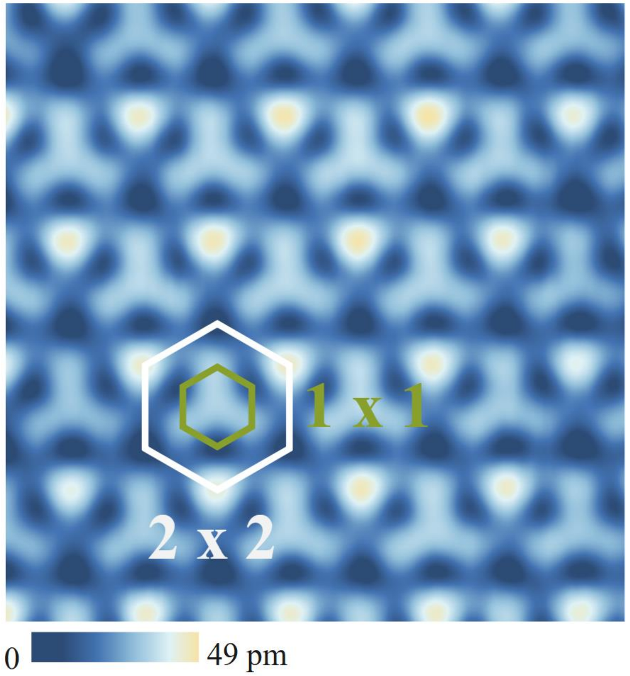
$$\Re A_n^M,\ \Im A_n^M,\ \Re A_n^{M'} \to M_1^+$$
$$\Im A_n^{M'} \to M_1^- \ !!!$$


Primary
Secondary
M_1^- (mirror odd)
M_1^+ (mirror even)
X
X
X
X
+
=


M=G/2
M'=M+G
$\mathrm{Re}A^M \sum_i \cos \vec{M}_i \cdot \vec{x}$
even
$\mathrm{Im}A^M \sum_i \sin \vec{M}_i \cdot \vec{x}$
even
$\mathrm{Re}A^{M'} \sum_i \cos \vec{M}'_i \cdot \vec{x}$
even
$\mathrm{Im}A^{M'} \sum_i \sin \vec{M}'_i \cdot \vec{x}$
odd

Extract CDW order parameters: $\Delta_n(x)$ and $\phi_n(x)$

Measured by STM



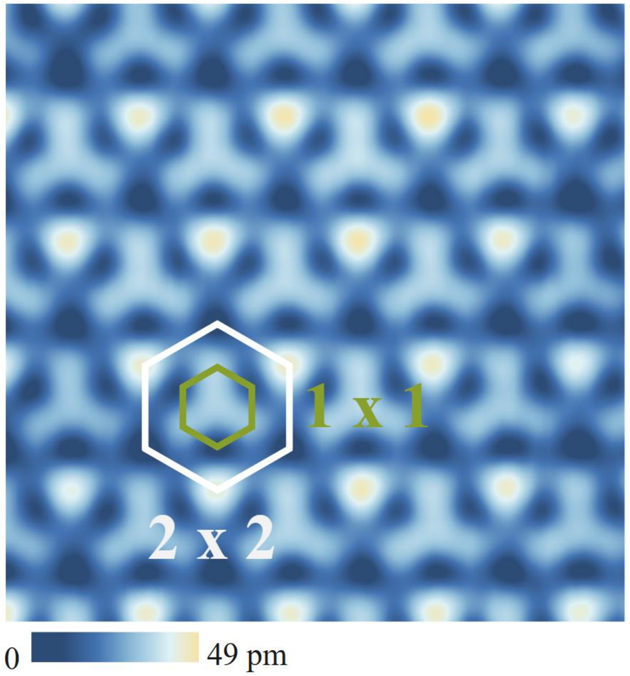
Implement Lawler-Fujita to extract with well defined phases A_n^q

$$M_1^- \text{ (primary)} \rightarrow \Delta_n = \Im A_n^{M'}$$

$$M_1^+ \text{ (secondary)} \rightarrow \phi_n = f(\Re A_n^M, \Im A_n^M, \Re A_n^{M'})$$

Extract CDW order parameters: $\Delta_n(x)$ and $\phi_n(x)$

Measured by STM



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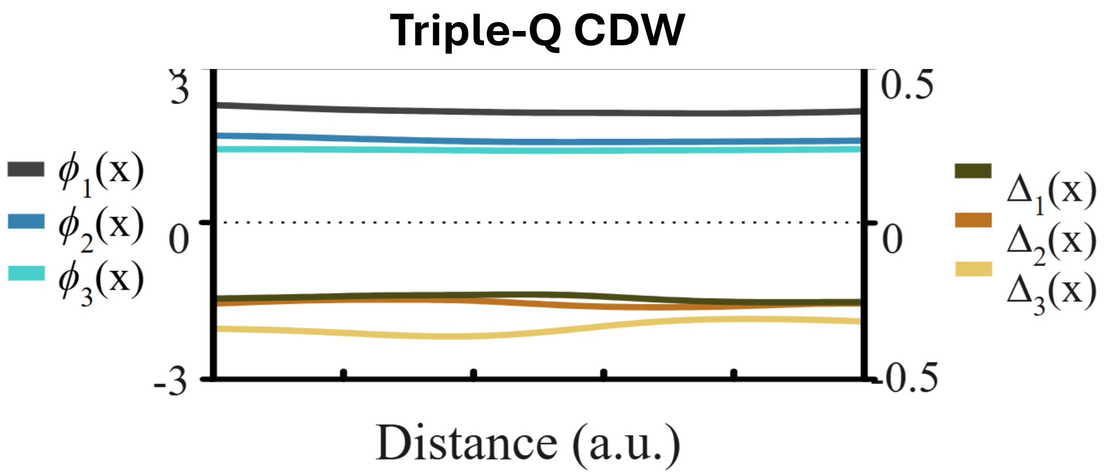

Extract CDW order parameters: $\Delta_n(x)$ and $\phi_n(x)$

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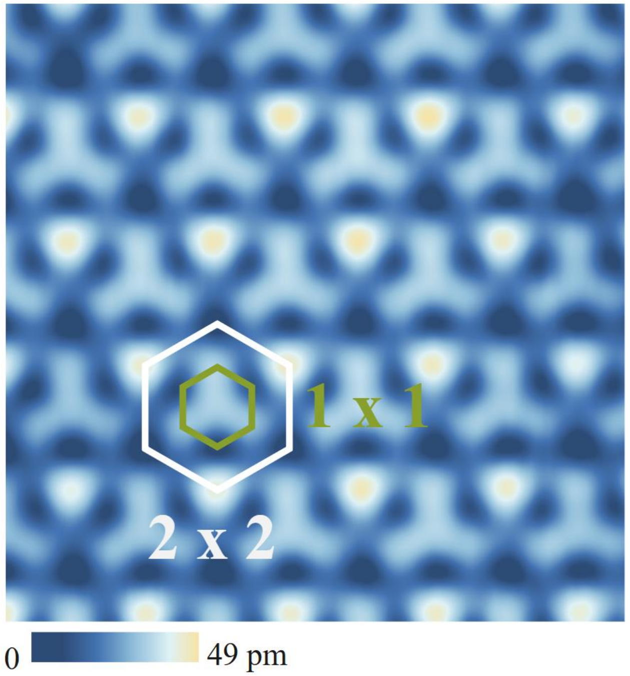

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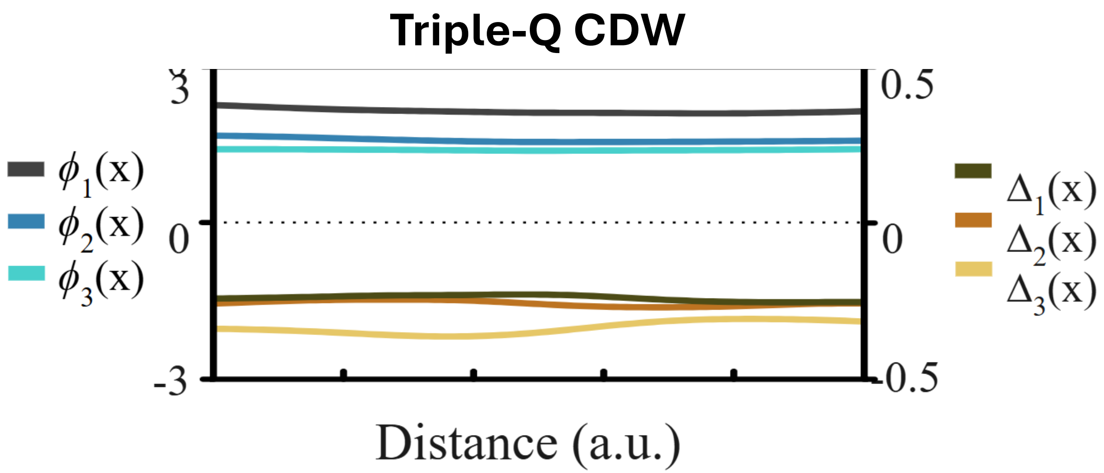
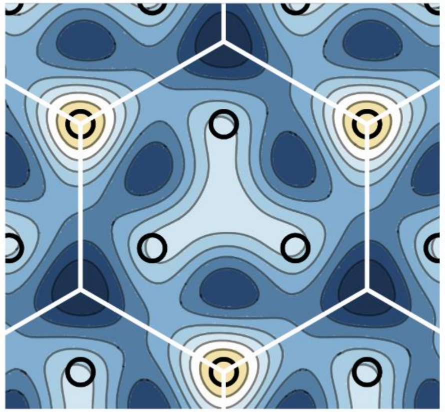
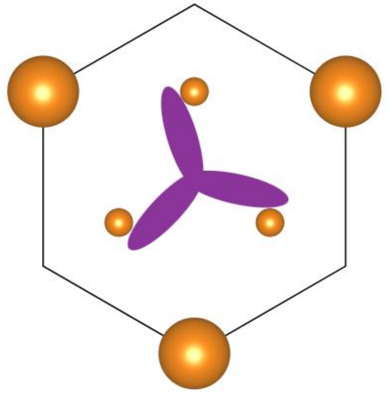


Reconstructed with extracted Δ_n and ϕ_n





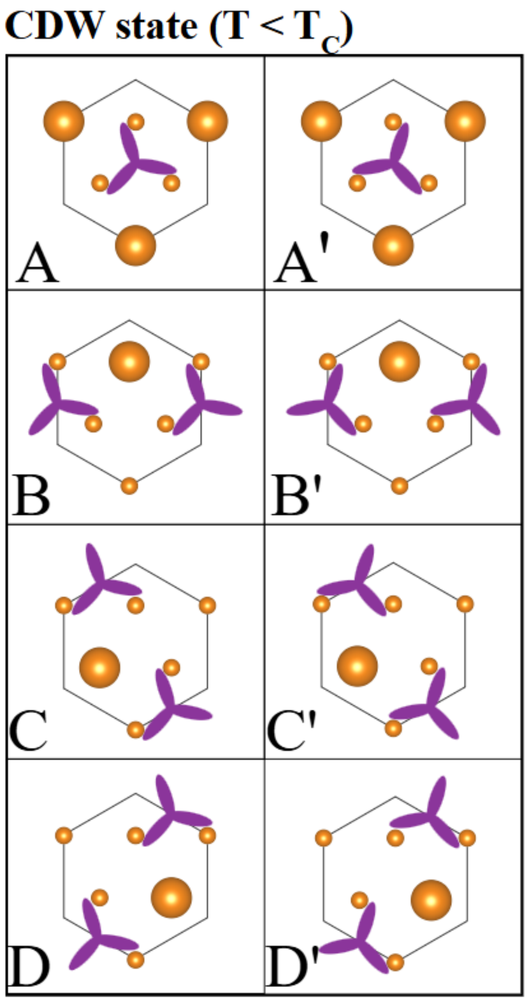
CCDW state

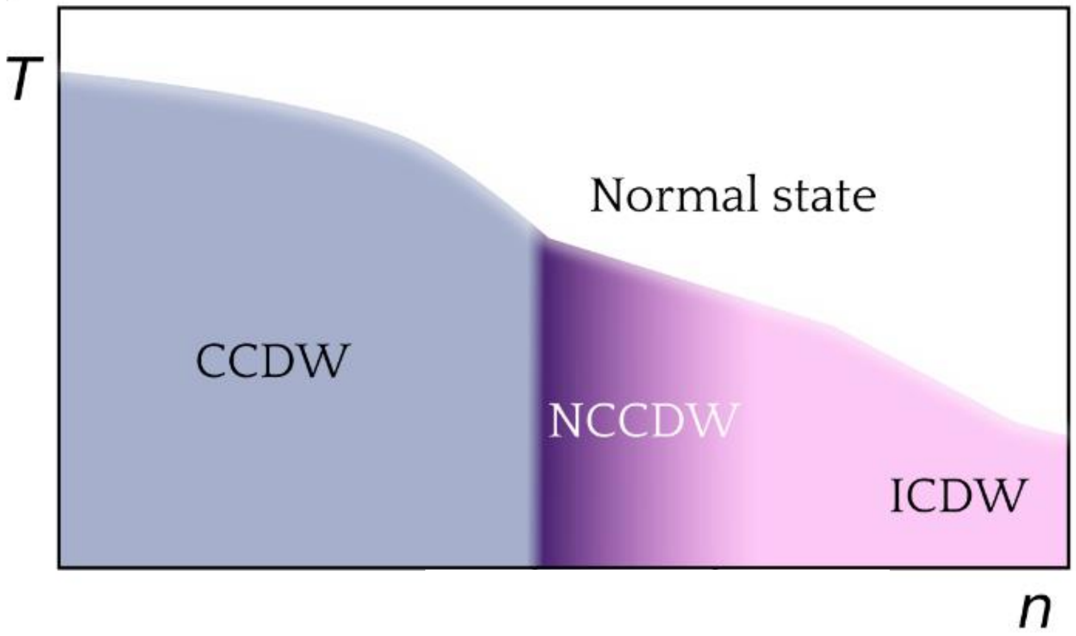
4 choices for the propeller center in the CDW unit cell & chirality:

8 degenerate CCDW ground states

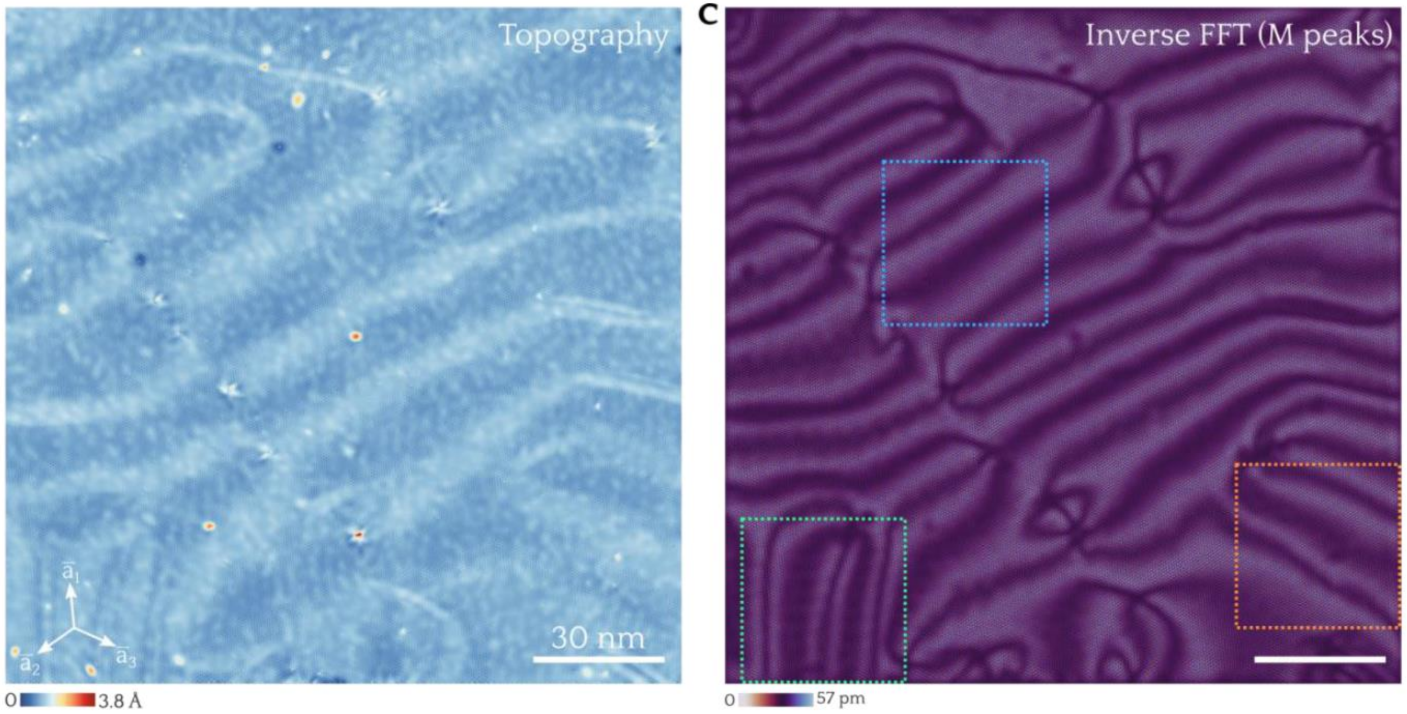
$$\vec{\Delta}(\vec{x}) = (\Delta_1(\vec{x}), \Delta_2(\vec{x}), \Delta_3(\vec{x}))$$

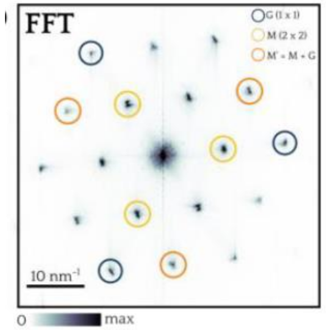
A (- - -)	A' (+ + +)
B (- + +)	B' (+ - -)
C (+ - +)	C' (- + -)
D (+ + -)	D' (- - +)
$\Delta_1\Delta_2\Delta_3 < 0$	$\Delta_1\Delta_2\Delta_3 > 0$





CDW over large scales: NC-CDW phase

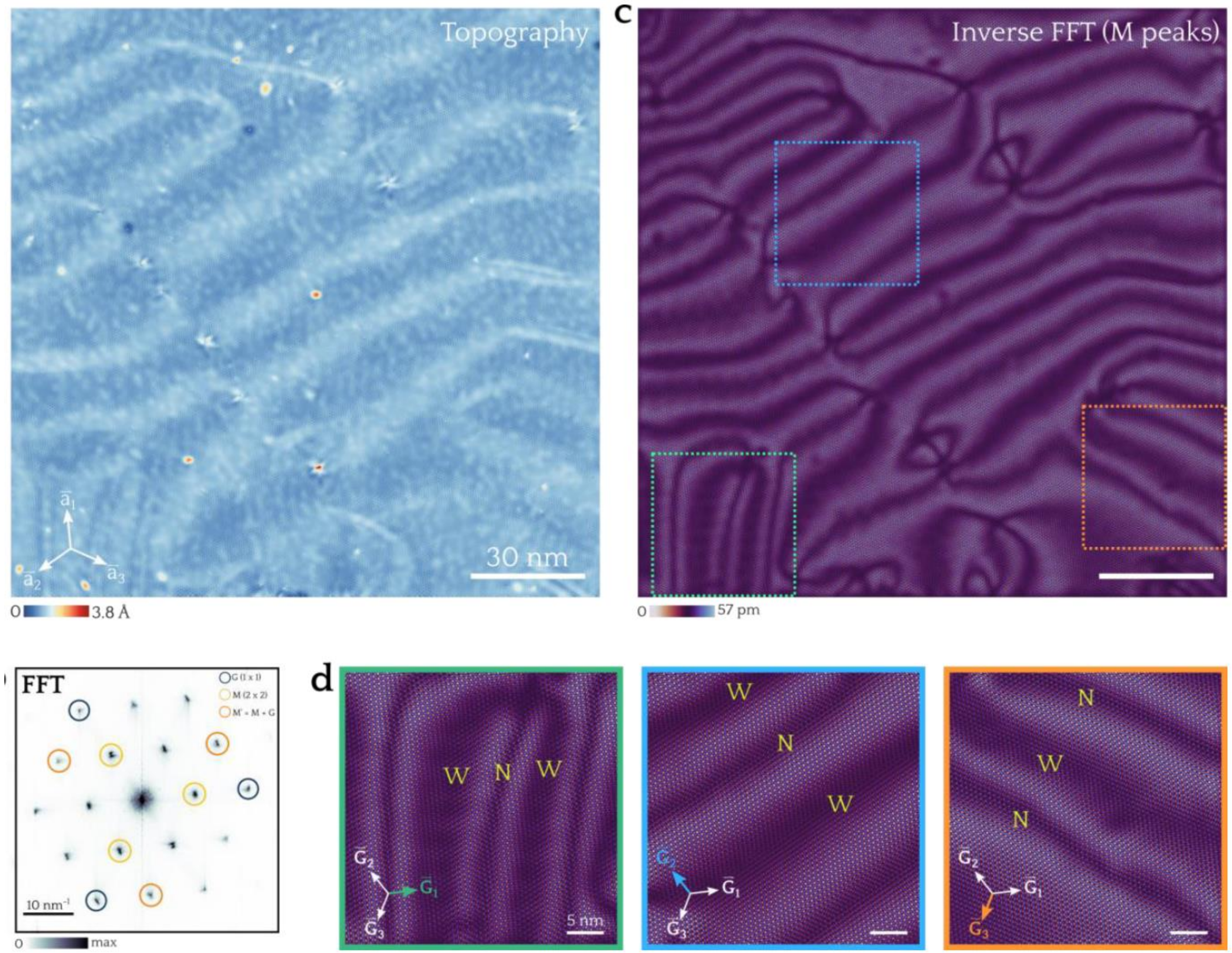




Highly inhomogeneous 2x2 CDW with LR ordering of domains & FT same as for single domain → NC phase

Modulation of λ~20 nm, 1D stripes (not previously believed 2D network)

CDW over large scales: NC-CDW phase

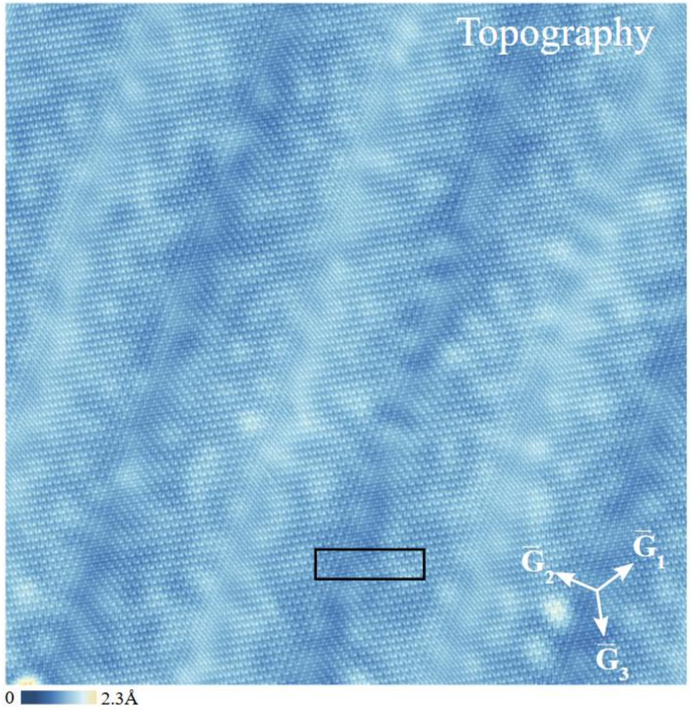


Highly inhomogeneous 2x2 CDW with LR ordering of domains & FT same as for single domain → NC phase

Modulation of λ~20 nm, 1D stripes (not previously believed 2D network)

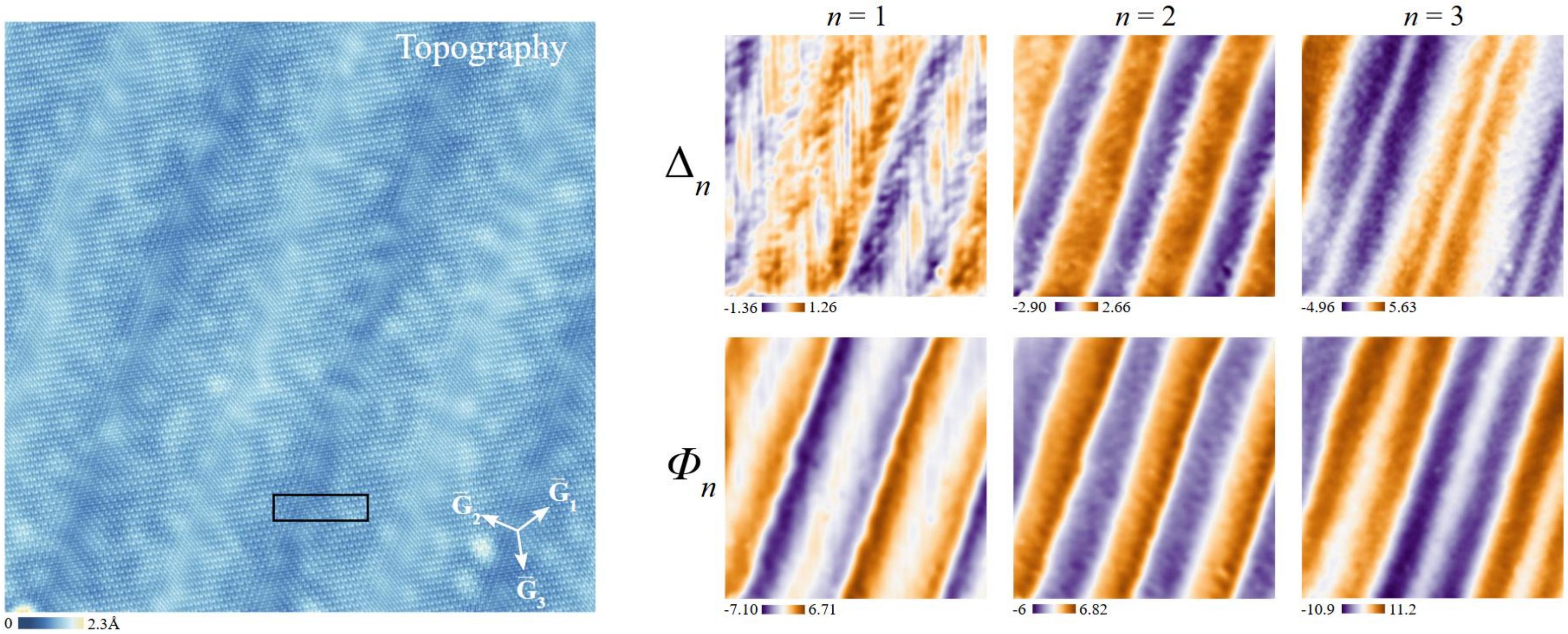
Three types of 'super-domains' with $\vec{q}_{\mathrm{NC}}$ roughly parallel to $\vec{Q}_n$

Intertwinned CDW order parameters



$\vec{q}_{\mathrm{NC}} \sim \vec{Q}_2$ super-domain

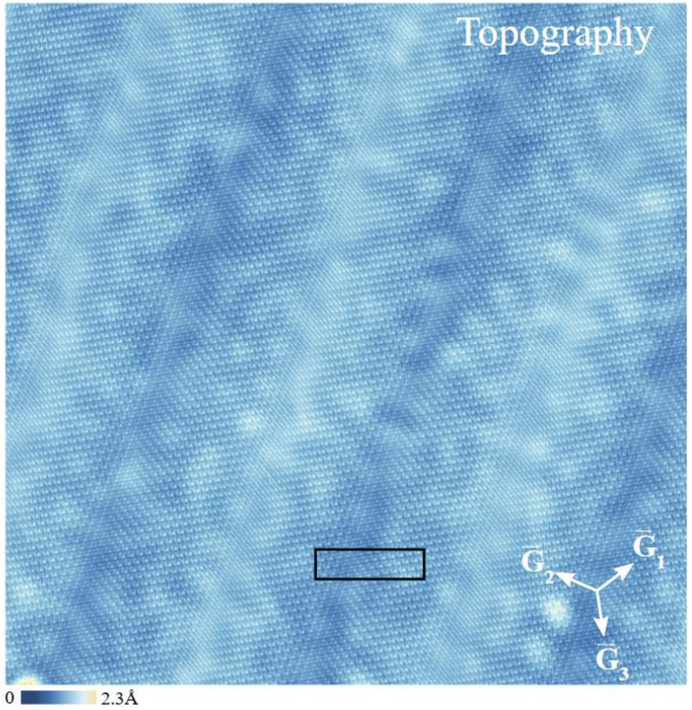
Intertwinned CDW order parameters



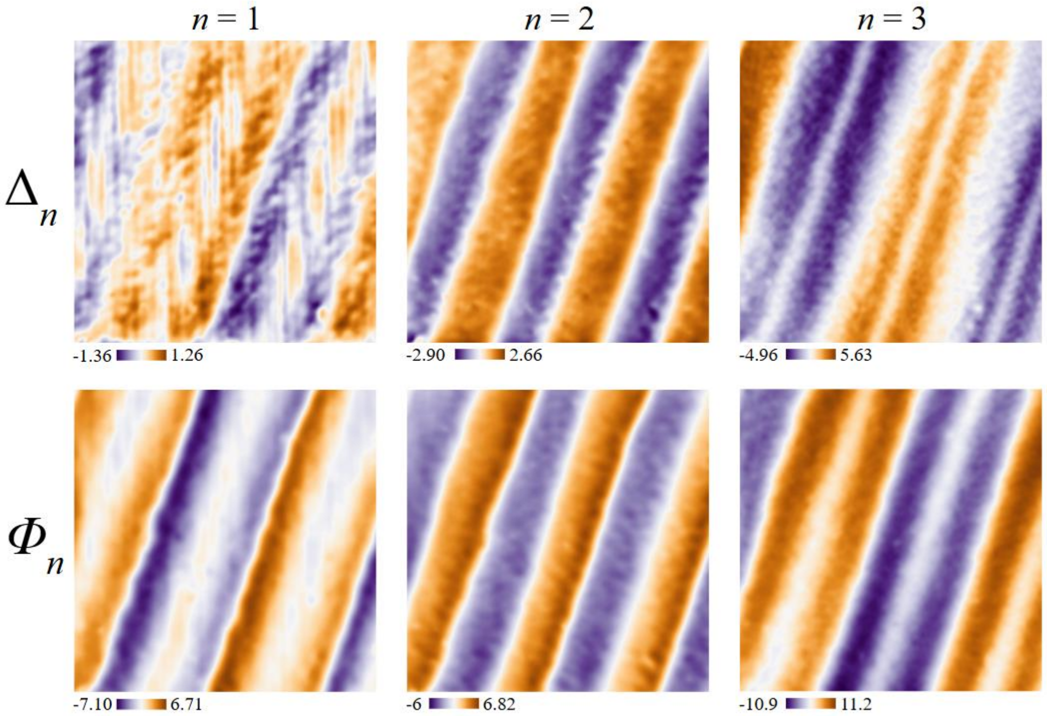
$\vec{q}_{\rm NC} \sim \vec{Q}_2$ super-domain

$$\Delta_1 \sim \sin(\vec{q}_{\rm NC}\vec{x}) + c\sin(3\vec{q}_{\rm NC}\vec{x})$$
$$\Delta_2 \sim \sin(2\vec{q}_{\rm NC}\vec{x})$$
$$\Delta_3 \sim \cos(\vec{q}_{\rm NC}\vec{x}) - c\cos(3\vec{q}_{\rm NC}\vec{x})$$
$$\phi_n \propto -\Delta_n$$

Intertwinned CDW order parameters



$\vec{q}_{\mathrm{NC}} \sim \vec{Q}_2$ super-domain



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$$\Delta_3 \sim \cos(\vec{q}_{\mathrm{NC}}\vec{x}) - c\cos(3\vec{q}_{\mathrm{NC}}\vec{x})$$
$$\phi_n \propto -\Delta_n$$

'ABCD' pattern:
four CCDW states with

$$\Delta_1\Delta_2\Delta_3 < 0$$

NC-CDW state

8 degenerate CCDW ground states

$$\vec{\Delta}(\vec{x}) = (\Delta_1(\vec{x}), \Delta_2(\vec{x}), \Delta_3(\vec{x}))$$

A (- - -)	A' (+ + +)
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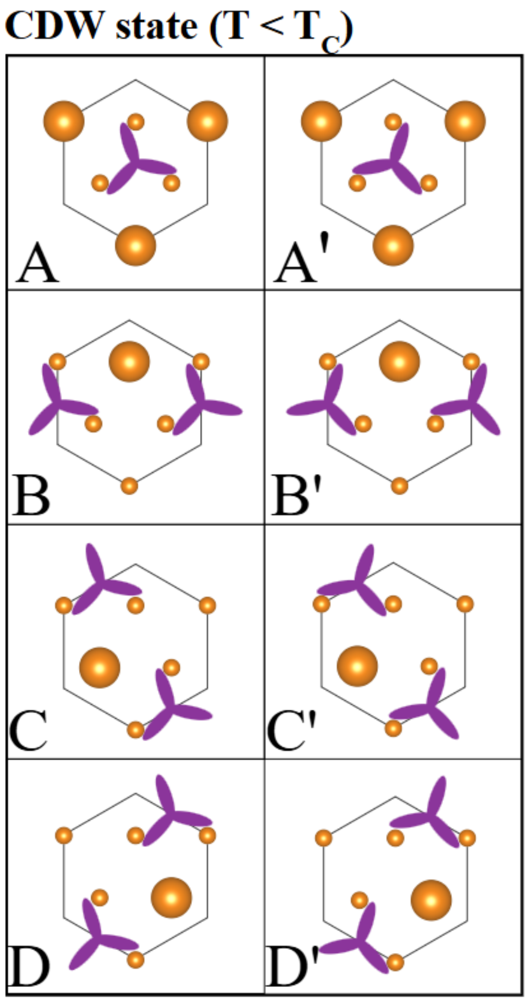
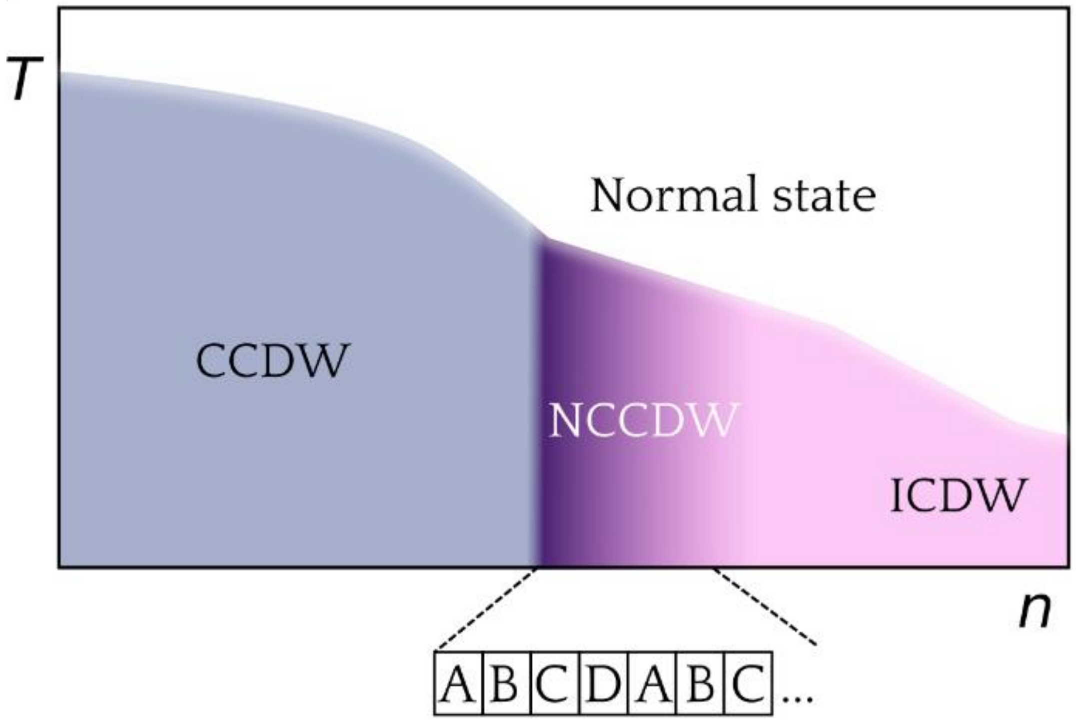




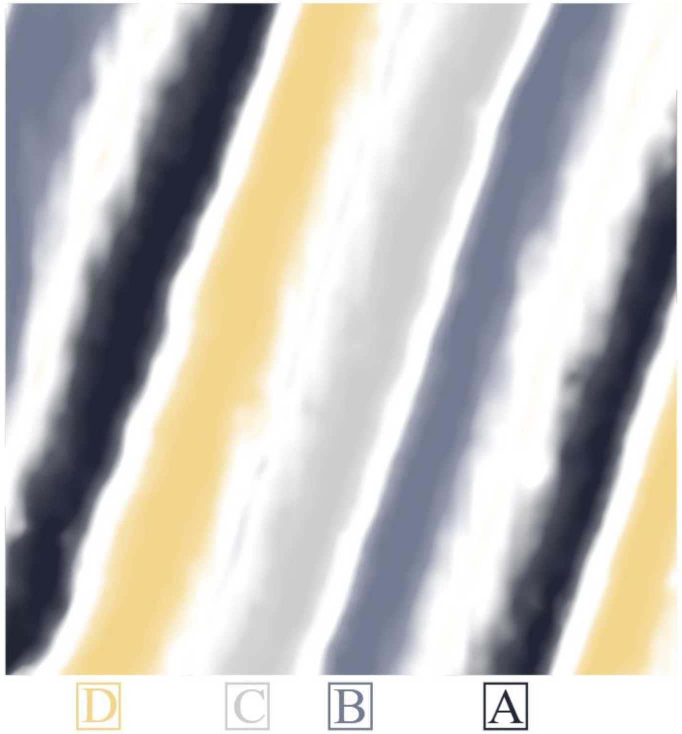
Sign changes of OPs shift center of the CDW by one atomic unit cell: e.g. B to C translation by a_3

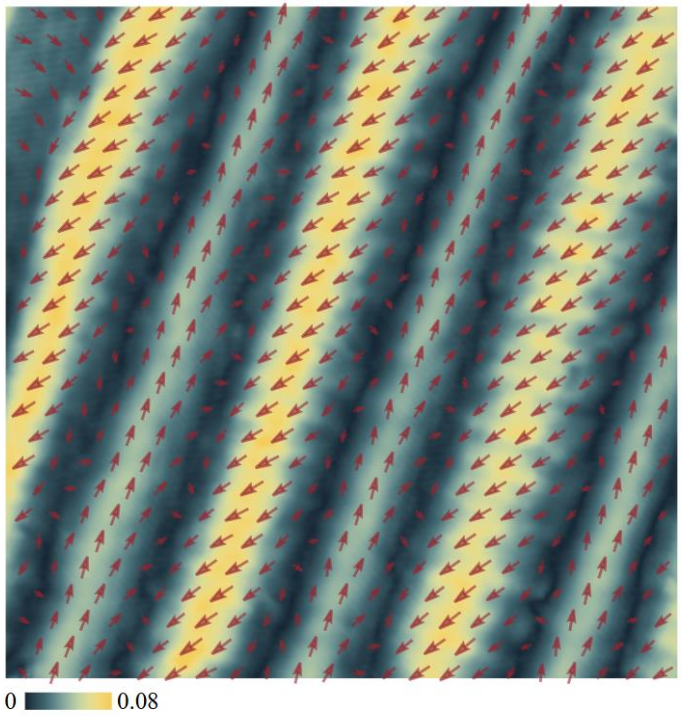
Nematic 1x1 component

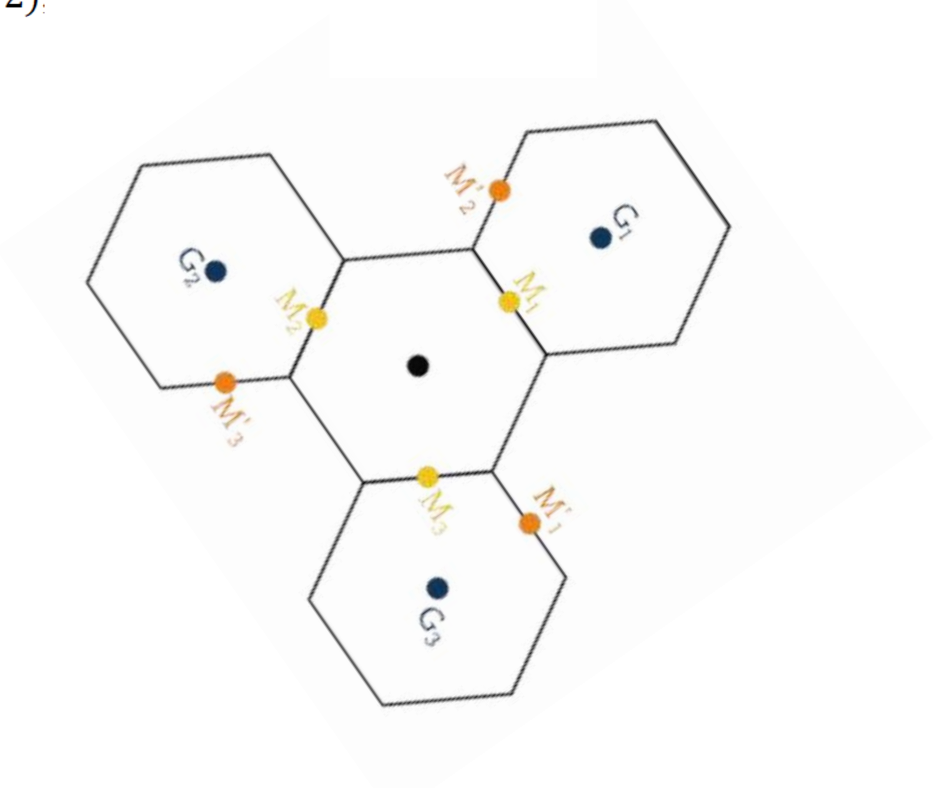
$$\rho_{2\times 2}(\vec{x}) = 2\Re \sum_n \left[A_n^M(\vec{x}) e^{-i\vec{M}_n \vec{x}} + A_n^{M'}(\vec{x}) e^{-i\vec{M}'_n \vec{x}} \right]$$

$$\boxed{\rho_{1\times 1}(\vec{x}) = 2\Re \sum_n A_n^G(\vec{x}) e^{-i\vec{G}_n \vec{x}}}$$

$$(N_x, N_y) = \Im\left(\frac{-\sqrt{3}}{2}(A_2^G - A_3^G), A_1^G - A_2^G/2 - A_3^G/2\right)$$







Vanishes T>Tc & selects axis T<Tc
Strongest in the domain walls
where CDW is weakest

$$\vec{N}_{\text{wide}} \sim \vec{G}_1$$

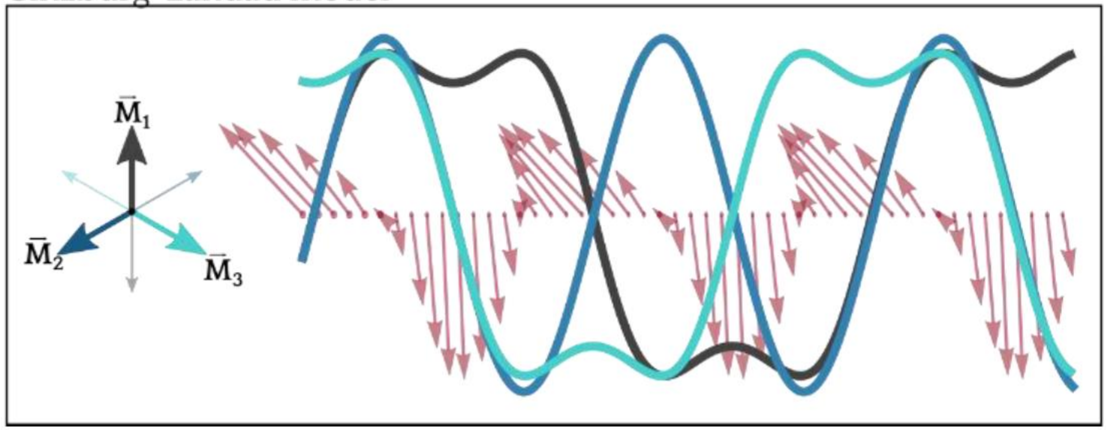
$$\vec{N}_{\text{narrow}} \sim \vec{G}_3$$

Origin of the Ising domain wall phenomenology

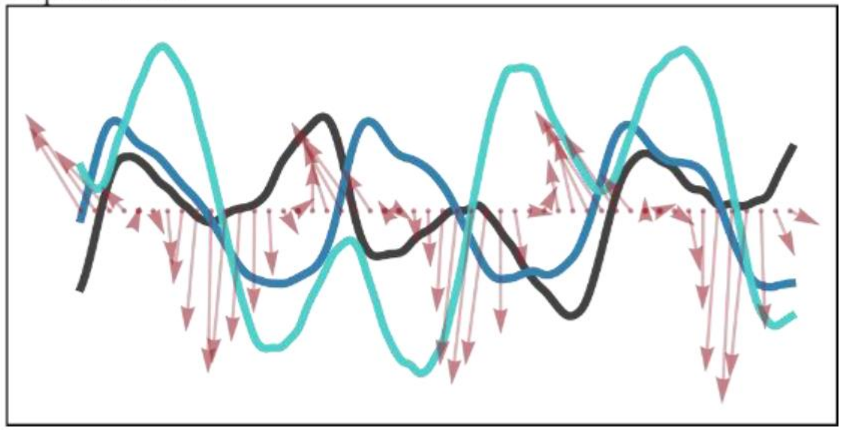
Ginzburg-Landau theory of coupled Δ_n (primary) and ϕ_n (secondary)

$$\mathcal{F} = \mathcal{F}_{\mathrm{CCDW}} + \mathcal{F}_{\partial}^{\Delta} + \mathcal{F}_{\partial}^{\Delta\phi} + \mathcal{F}_{N}$$

Ginzburg-Landau model



Experiment



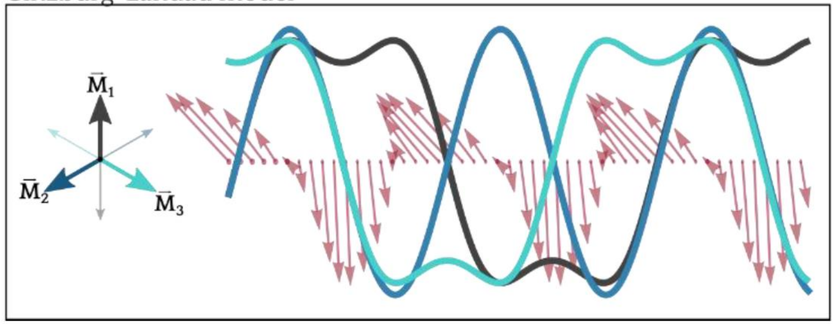
Origin of the Ising domain wall phenomenology

$$\mathcal{F} = \mathcal{F}_{\text{CCDW}} + \mathcal{F}_{\partial}^{\Delta} + \mathcal{F}_{\partial}^{\Delta\phi} + \mathcal{F}_N$$

- ϕ_n crucial (harmonic content generated by cubic terms):

$$\mathcal{F}_{\text{CCDW}} = \sum_n \left[a_\Delta \Delta_n^2 + c_\Delta \Delta_n^4 + a_\phi \phi_n^2 + \underline{b_\phi \phi_1 \phi_2 \phi_3} + c_\phi \phi_n^4 + \underline{b_{\Delta\phi} \phi_n \Delta_{n+1} \Delta_{n+2}}\right]$$

Ginzburg-Landau model



Origin of the Ising domain wall phenomenology

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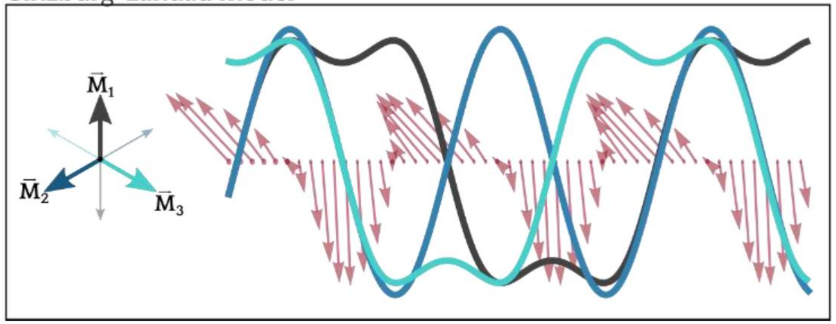
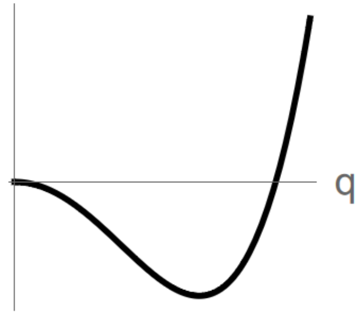
- Derivative terms driving I-CDW (no dephasing so not McMillan term):

$$\mathcal{F}_{\partial}^{\Delta(I)} = \sum_n \Delta_n \left(d_1^\Delta \partial_x^2 + d_3^\Delta \partial_x^4\right)\Delta_n \text{ with } d_1^\Delta, d_3^\Delta > 0$$

Ginzburg-Landau model

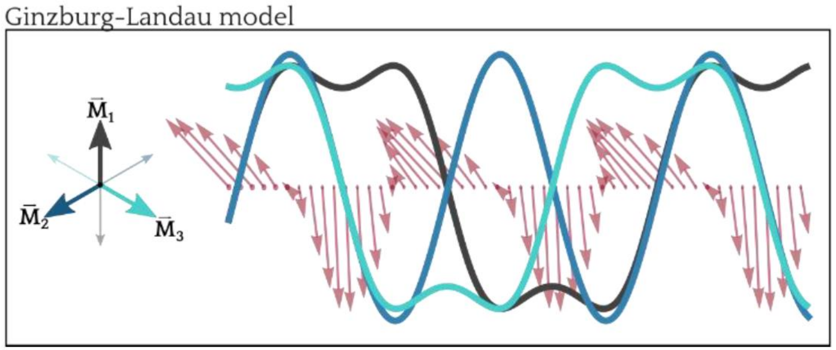


$-d_1^\Delta q^2 + d_3^\Delta q^4$



Origin of the Ising domain wall phenomenology

$$\mathcal{F} = \mathcal{F}_{\mathrm{CCDW}} + \mathcal{F}_{\partial}^{\Delta} + \cancel{\mathcal{F}_{\partial}^{\Delta\phi}} + \mathcal{F}_N$$



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- Derivative terms driving I-CDW (no dephasing so not McMillan term):

$$\mathcal{F}_{\partial}^{\Delta(I)} = \sum_n \Delta_n \left(d_1^\Delta \partial_x^2 + d_3^\Delta \partial_x^4\right) \Delta_n \text{ with } d_1^\Delta, d_3^\Delta > 0$$

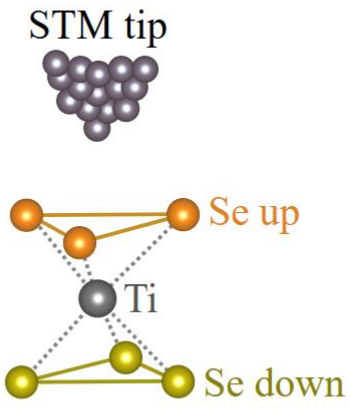
- Alternation of nematic direction component in consecutive domain wall

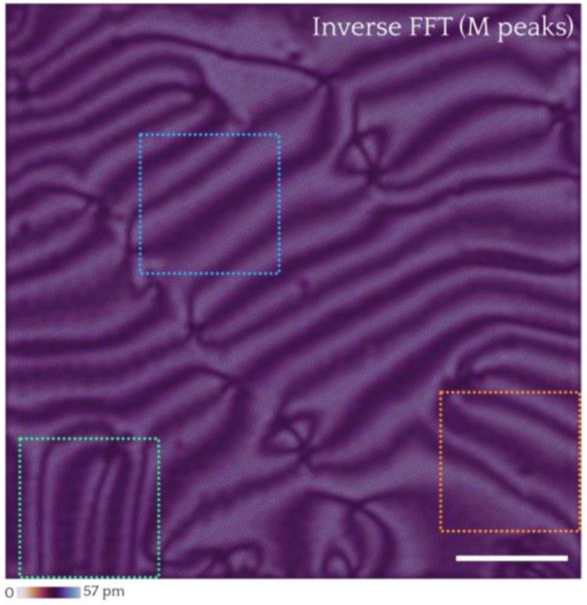
$$\mathcal{F}_N = a_N \left(-\tfrac{\sqrt{3}}{2}\left(\Delta_2^2 - \Delta_3^2\right), \Delta_1^2 - \tfrac{1}{2}\left(\Delta_2^2 + \Delta_3^2\right)\right) \cdot (N_x, N_y)$$

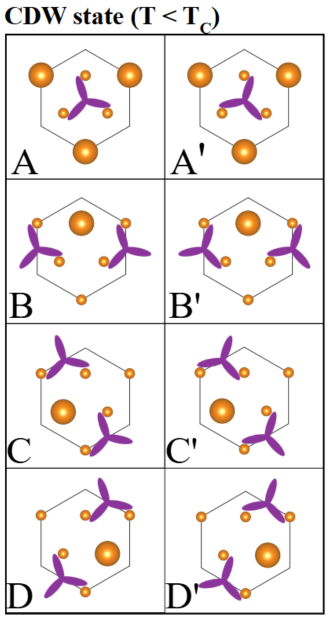
Conclusions

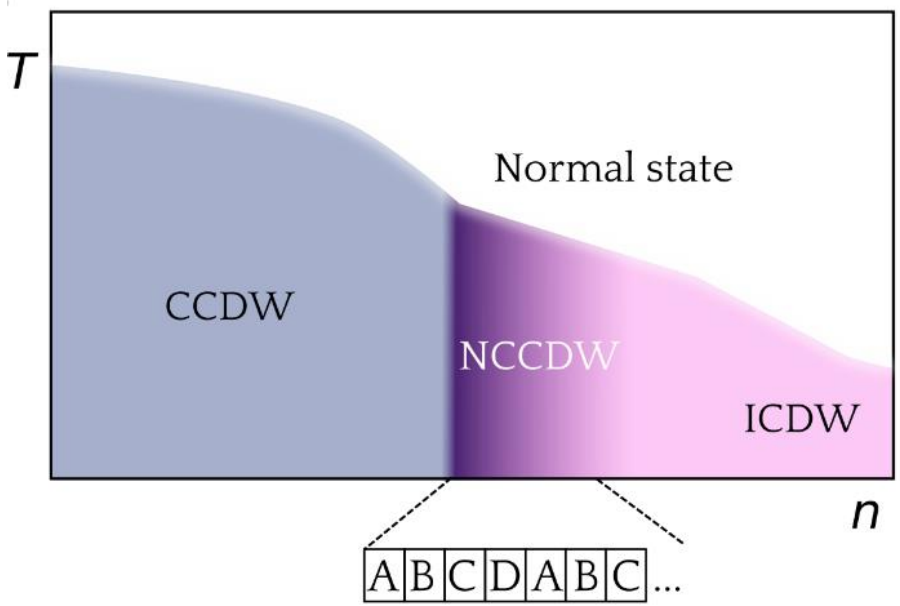
W. Wan *et al*, arXiv:2411.05725 (2024)
MNG and F. de Juan, *in preparation*

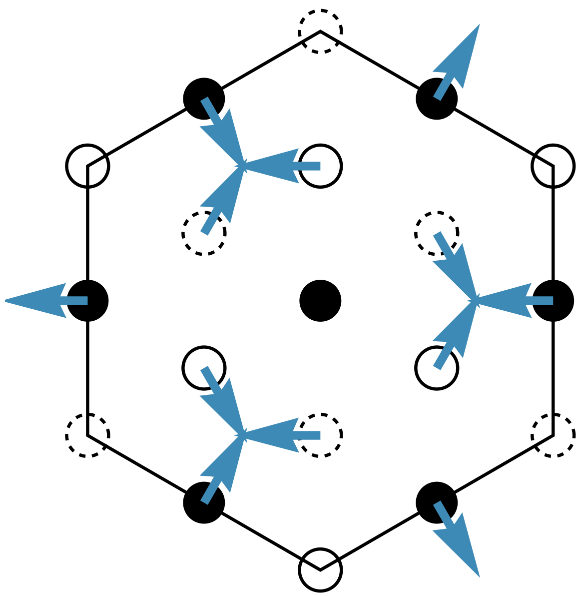
- NC-CDW in monolayer $TiSe_2$: coupled Δ_n (primary) and ϕ_n (secondary) with Ising DWs
- Identified minimal GL terms: (i) secondary OP crucial (ii) nematic component at DWs
- Outlook: Revisit bulk! Chirality? Implications for SC ?





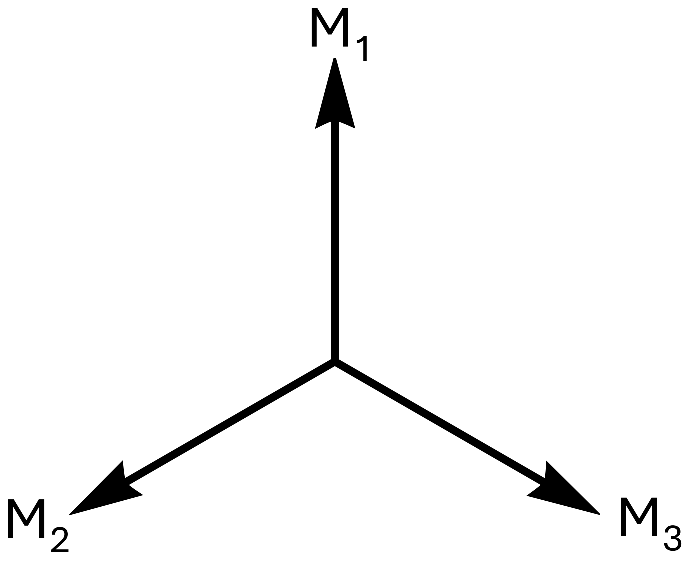




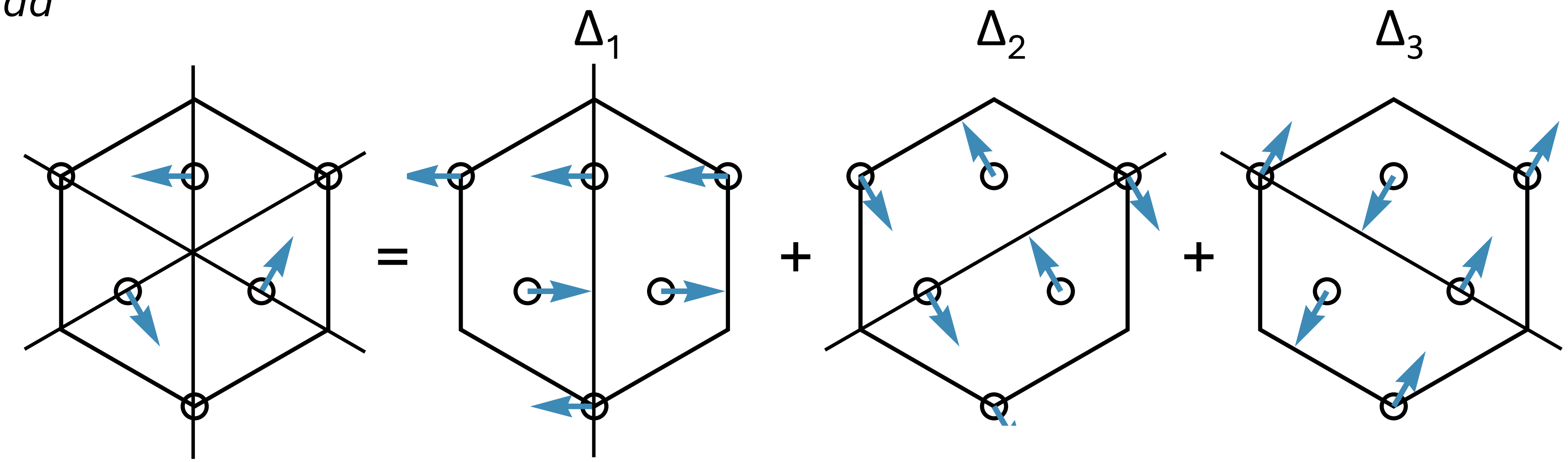


Triple-Q CDW ($M_i = G_i/2$)

$$\vec{\Delta}(\vec{x}) = (\Delta_1(\vec{x}), \Delta_2(\vec{x}), \Delta_3(\vec{x}))$$



mirror *odd*



Minimize F by expanding the OPs in harmonics

$$\phi_i(x) = \sum_n \phi_{i,n} e^{in\eta x}$$

$$\Delta_i(x) = \sum_n \Delta_{i,n} e^{in\eta x}$$

$$N_j^G(x) = \sum_n N_{j,n}^G e^{in\eta x}$$

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Domain-like Incommensurate Charge-Density-Wave States and the First-Order Incommensurate-Commensurate Transitions in Layered Tantalum Dichalcogenides. I. 1T-Polytype

Kazuo NAKANISHI and Hiroyuki SHIBA

Institute for Solid State Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo 106

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