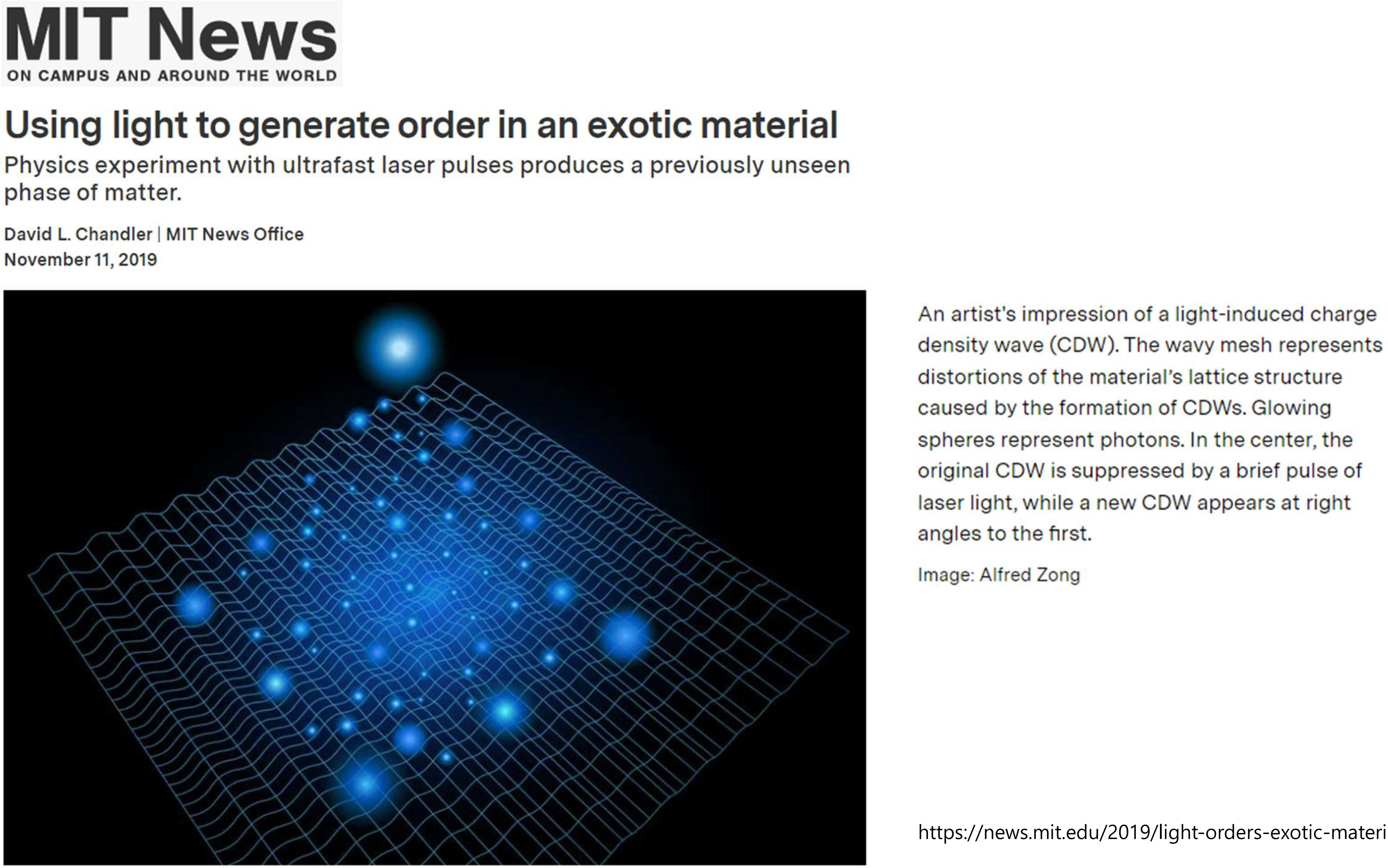


MIT News
ON CAMPUS AND AROUND THE WORLD

Using light to generate order in an exotic material

Physics experiment with ultrafast laser pulses produces a previously unseen phase of matter.

David L. Chandler | MIT News Office

November 11, 2019

An artist's impression of a light-induced charge density wave (CDW). The wavy mesh represents distortions of the material's lattice structure caused by the formation of CDWs. Glowing spheres represent photons. In the center, the original CDW is suppressed by a brief pulse of laser light, while a new CDW appears at right angles to the first.

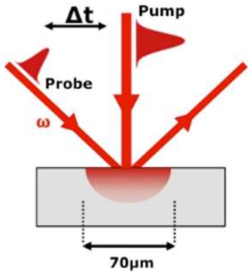
Image: Alfred Zong

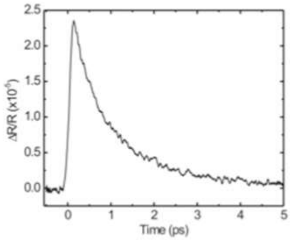
https://news.mit.edu/2019/light-orders-exotic-material-1111

Nuh Gedik group @ MIT

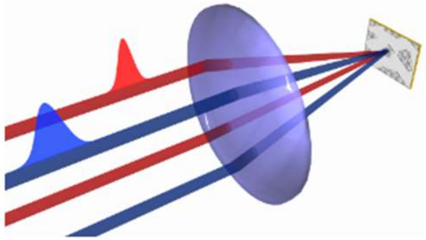
http://web.mit.edu/gediklab/research.html

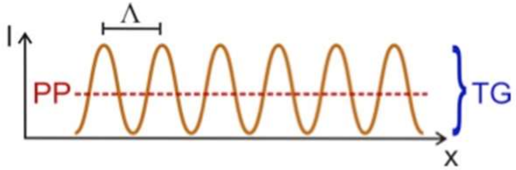
Pump Probe Spectroscopy



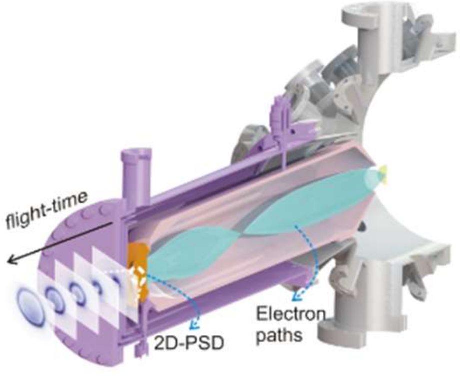


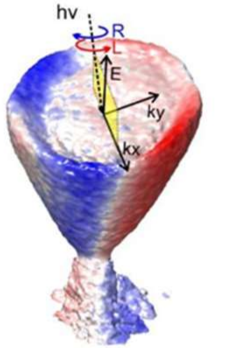
Transient Grating Spectroscopy





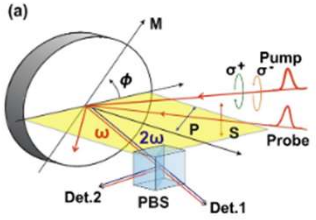
Circular Dichroism and Time-Resolved ARPES

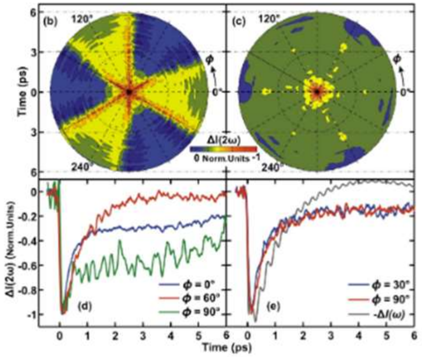




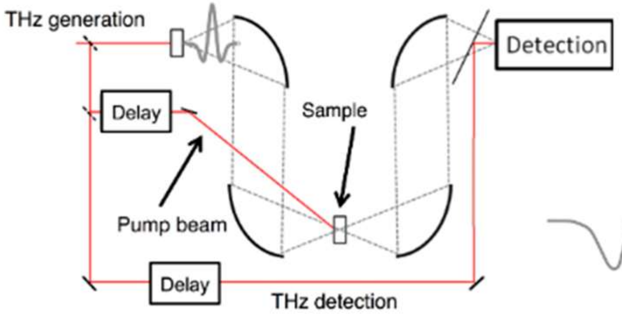
$\Delta I\,(E,k_x,k_y)=I_L-I_R$

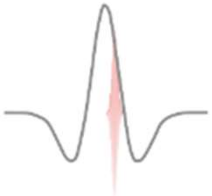
Second Harmonic Generation

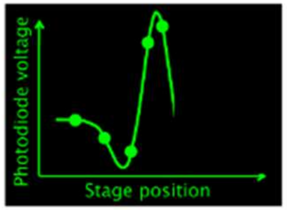




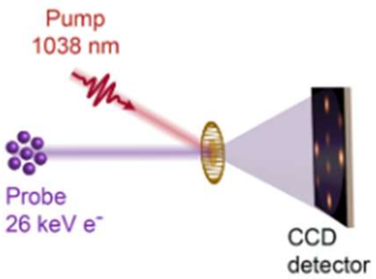
TeraHertz Time-Domain Spectroscopy





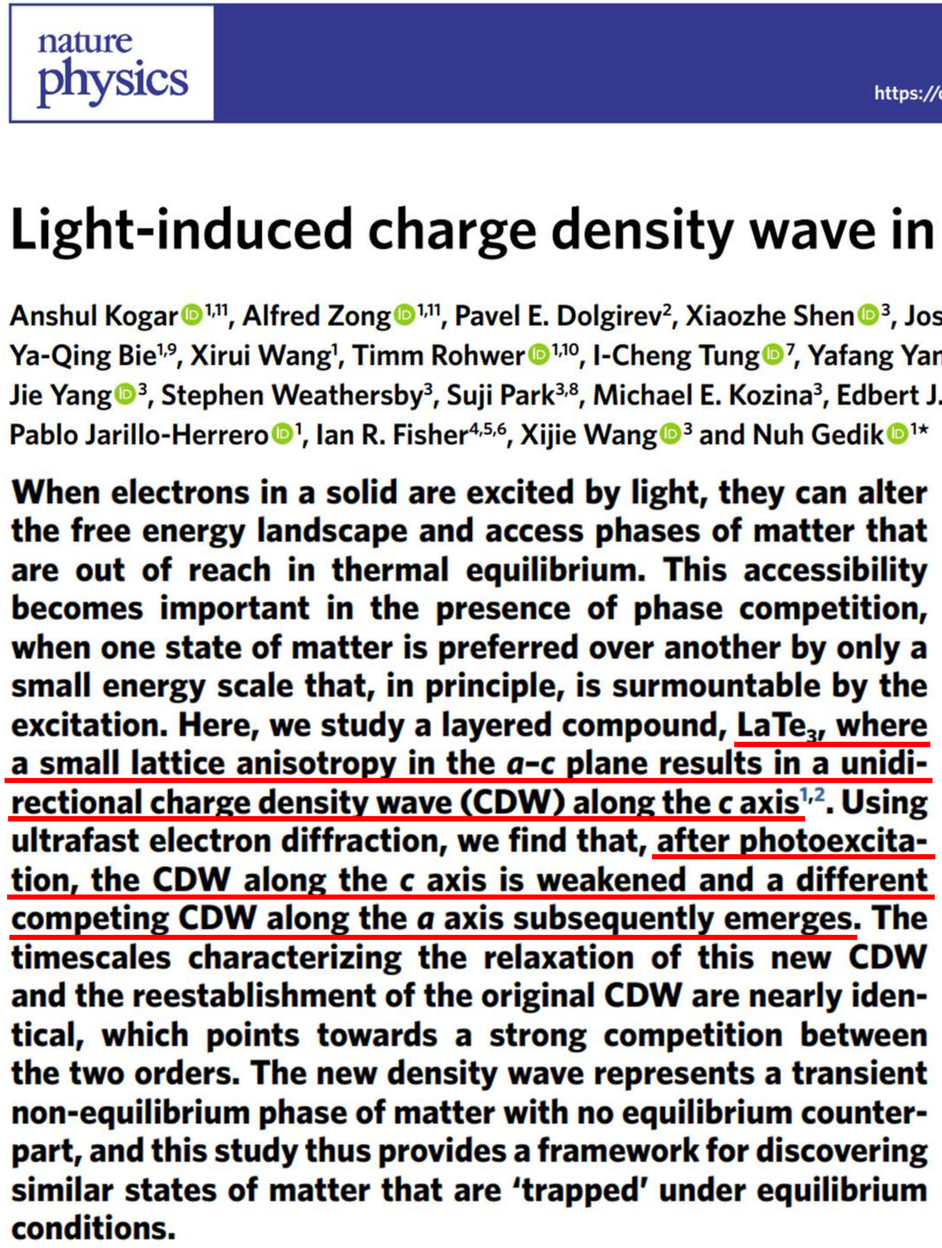


Ultrafast Electron Diffraction



Members

- 1 Professor
- 4 Postdocs
- 11 Graduate students



nature physics

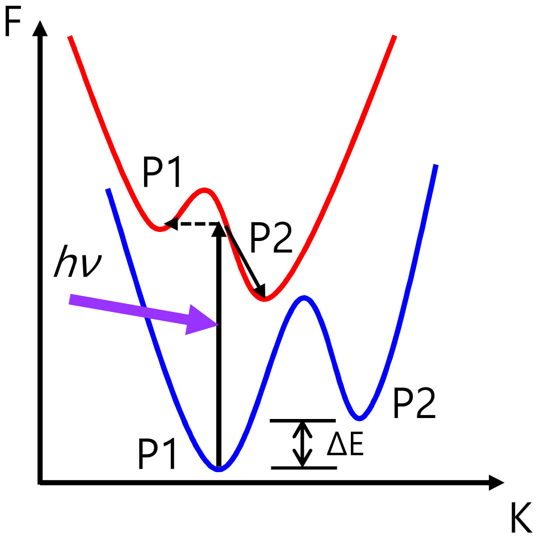
LETTERS

https://doi.org/10.1038/s41567-019-0705-3

Light-induced charge density wave in $LaTe_3$

Anshul Kogar[1,11], Alfred Zong[1,11], Pavel E. Dolgirev[2], Xiaozhe Shen[3], Joshua Straquadine[4,5,6], Ya-Qing Bie[1,9], Xirui Wang[1], Timm Rohwer[1,10], I-Cheng Tung[7], Yafang Yang[1], Renkai Li[3], Jie Yang[3], Stephen Weathersby[3], Suji Park[3,8], Michael E. Kozina[3], Edbert J. Sie[4,6], Haidan Wen[7], Pablo Jarillo-Herrero[1], Ian R. Fisher[4,5,6], Xijie Wang[3] and Nuh Gedik[1*]

When electrons in a solid are excited by light, they can alter the free energy landscape and access phases of matter that are out of reach in thermal equilibrium. This accessibility becomes important in the presence of phase competition, when one state of matter is preferred over another by only a small energy scale that, in principle, is surmountable by the excitation. Here, we study a layered compound, $LaTe_3$, where a small lattice anisotropy in the *a*–*c* plane results in a unidirectional charge density wave (CDW) along the *c* axis[1,2]. Using ultrafast electron diffraction, we find that, after photoexcitation, the CDW along the *c* axis is weakened and a different competing CDW along the *a* axis subsequently emerges. The timescales characterizing the relaxation of this new CDW and the reestablishment of the original CDW are nearly identical, which points towards a strong competition between the two orders. The new density wave represents a transient non-equilibrium phase of matter with no equilibrium counterpart, and this study thus provides a framework for discovering similar states of matter that are 'trapped' under equilibrium conditions.

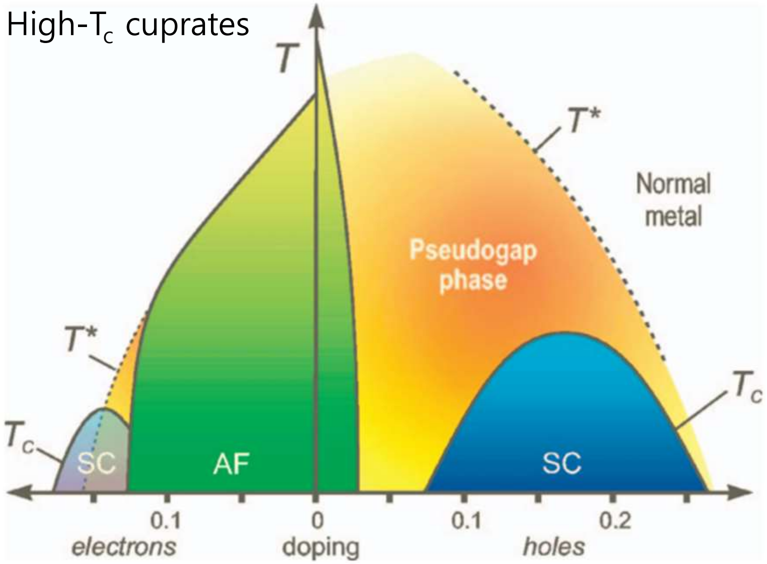


Competition or cooperation between proximal phases

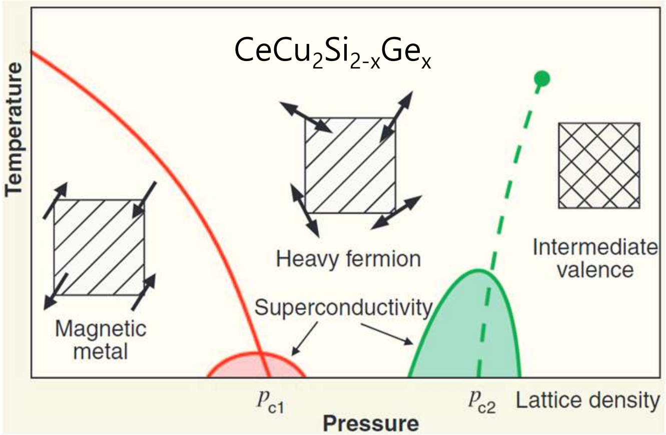
- ^{3}He: FM spin fluctuations vs p-wave superfluidity
- f-electron systems: heavy fermion SC vs AFM order
- Cu- and Fe-based superconductors: SC vs charge & spin orders
- CMR manganites: FM metal vs AF-CO insulator

→ mesoscopic phase separation when $\Delta E \lesssim T$

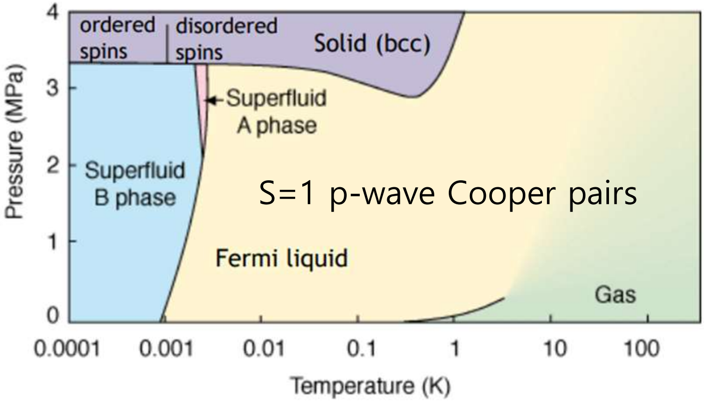
High-T_c cuprates



Fischer et al., Rev. Mod. Phys. **79**, 353 (2007)

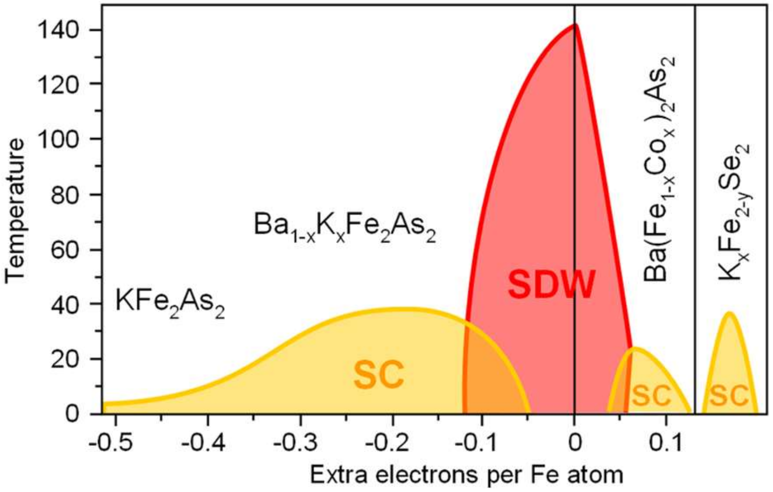


Yuan et al., Science **302**, 2104 (2003)

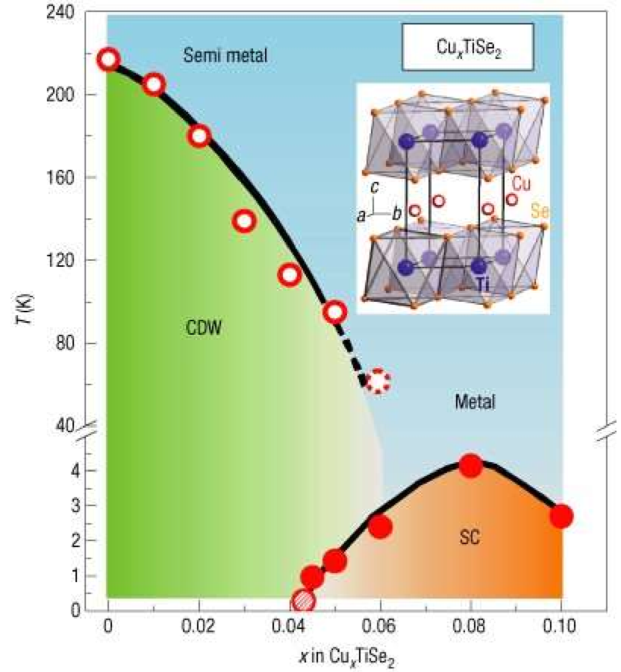


S=1 p-wave Cooper pairs

Vollhardt & Wölfle, *The Superfluid Phases of Helium 3* (1990)

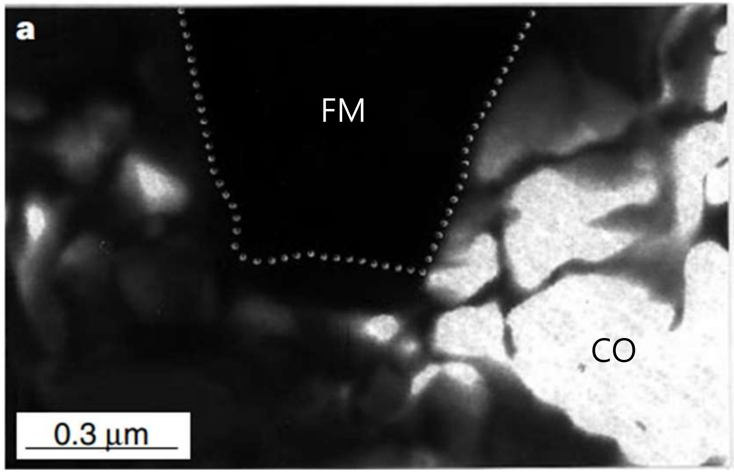


Kordyuk, Low Temp. Phys. **38**, 888 (2012)



Morosan et al., Nat. Phys. **2**, 544 (2006)

Dark-field image of $La_{5/8-y}Pr_yCa_{3/8}MnO_3$



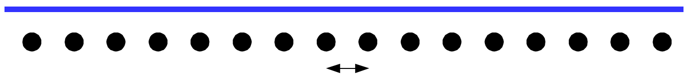
Uehara et al., Nature **399**, 560 (1999)

What is a charge density wave (CDW)?

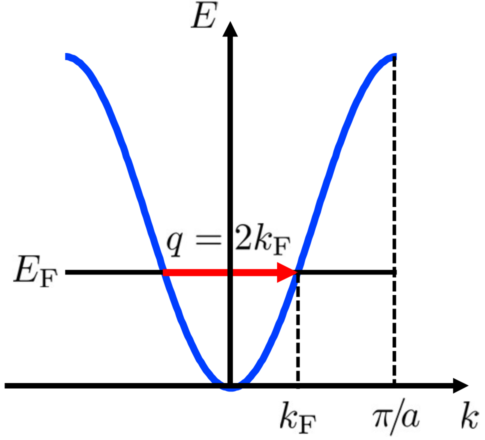
Usually, conduction electrons in a solid are in a quantum liquid state., i.e., spatially homogeneous.
But, the Peierls mechanism in a 1D system can bring some modulation in electron density.

Normal state T > T_{CDW}

electron density $\rho(x) = \rho_0$

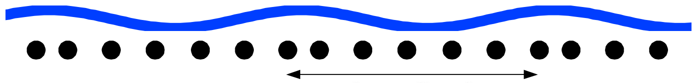


lattice parameter a



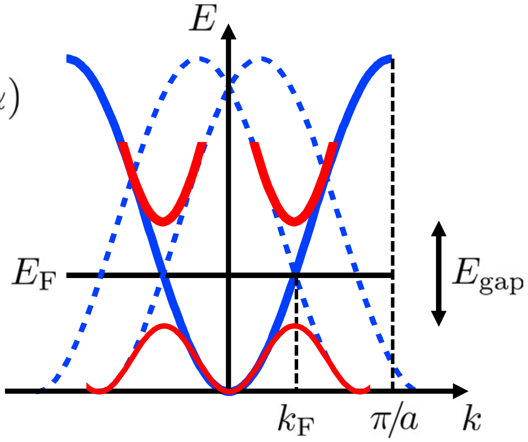
CDW state T < T_{CDW}

electron density $\rho = \rho_0 + \rho_1 \cos(qx + \alpha)$



lattice parameter $a' = 2\pi/q$

Gap opening due to BZ folding
& band mixing.

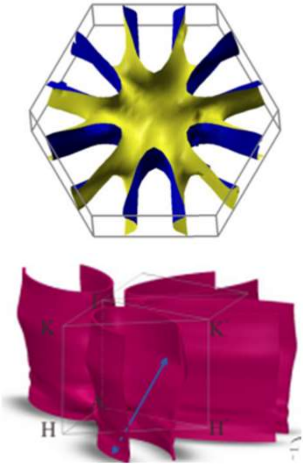


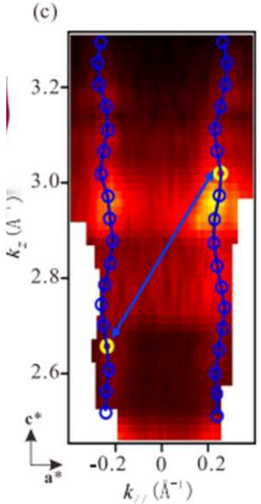
How about in real 3D materials?

Fermi-surface nesting: parallel regions with finite area on Fermi surfaces

→ Kohn anomaly: effective screening of phonons with $\mathbf{q}_{\mathrm{CDW}}$ (phonon softening)

$$\epsilon(\mathbf{q},\omega) = 1 + \chi(\mathbf{q},\omega) = 1 - V_{\mathbf{q}} \sum_{\mathbf{k}} \frac{n_{\mathbf{k+q}} - n_{\mathbf{k}}}{\hbar\omega + E_{\mathbf{k+q}} - E_{\mathbf{k}}}$$

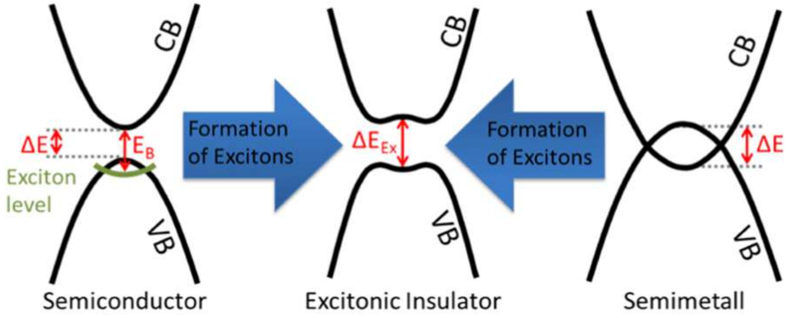


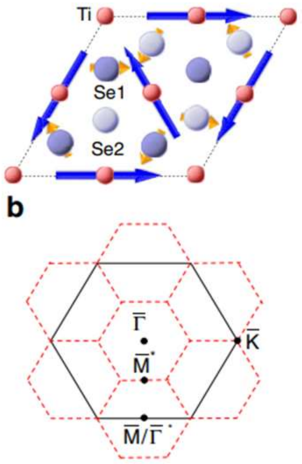


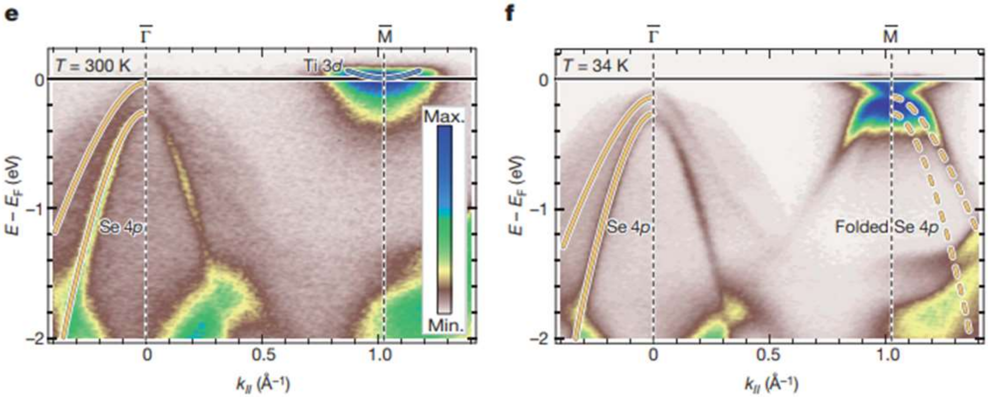
Other mechanisms for CDW

※ Excitonic insulator or band-type Jahn-Teller mechanism

T. Rohwer et al, Nature **471**, 490 (2011)







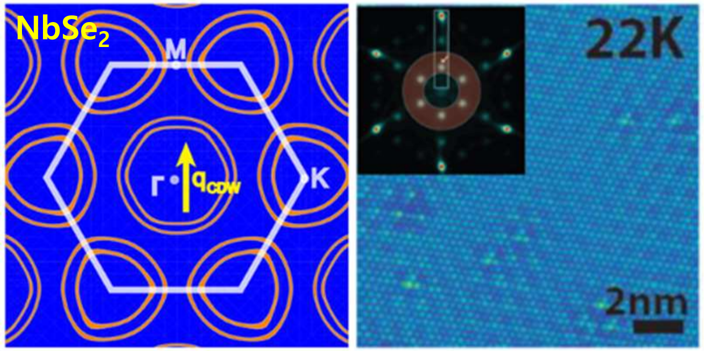
※ Saddle point @ EF in quasi-2D materials (VHS)

$$\epsilon(\mathbf{q},\omega) = 1 + \chi(\mathbf{q},\omega) = 1 - V_{\mathbf{q}} \sum_{\mathbf{k}} \frac{n_{\mathbf{k+q}} - n_{\mathbf{k}}}{\hbar\omega + E_{\mathbf{k+q}} - E_{\mathbf{k}}}$$

$$\sum_{\mathbf{k}} \longrightarrow \int dE D(E)$$

※ Strong **q**-dependent el-ph coupling

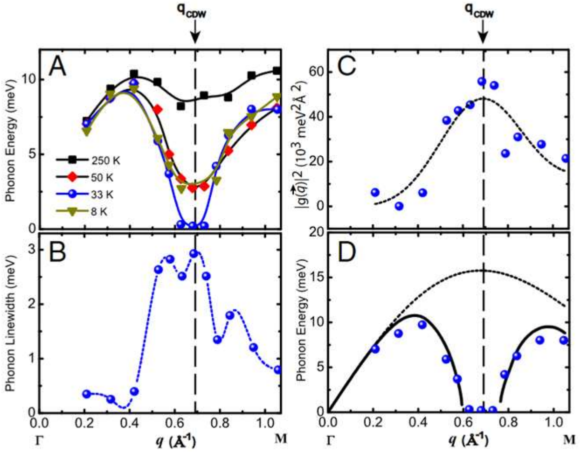
X. Zhu et al., PNAS **112**, 2367 (2015)



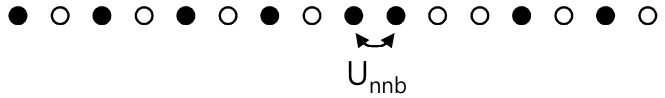
DFT

STM (FFT)

CDW without FSN



※ Different from charge order caused by nnb Coulomb repulsions not related with phonons eg. Fe_3O_4, $La_{0.5}Ca_{0.5}MnO_3$, etc.



※ None of the before, still unknown

Methods

Sample preparation:

Mechanical exfoliation down to 60 nm checked by AFM,

then transferred to 10-nm-thick Si_3N_4 window

MeV UED @ SLAC:

pump laser: 800 nm, 80 fs Ti:sapphire laser, incidence angle 5°, 500 x 500 μm^2 beam size on a sample (FWHM)

probe e^--beam: 3.1 MeV, 180 Hz, 90 x 90 μm^2 beam size on a sample (FWHM)

detector: P43 phosphor screen for electrons, EMCCD (Andor iXon Ultra 888)

Δt = 300 fs ← THz streaking

keV UED @ homelab:

pump laser: 1038 nm, 190 fs, 10 kHz Yb:KGW laser, 500 x 500 μm^2 beam size on a sample (FWHM)

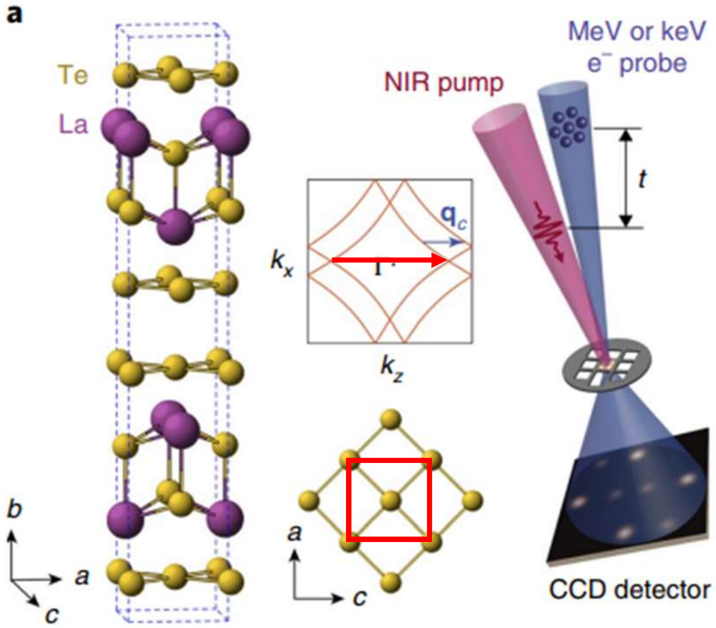
probe e^--beam: photoelectron by 4th harmonic (260 nm) laser, 26 kV DC acceleration, 270 x 270 μm^2 beam size on a sample (FWHM)

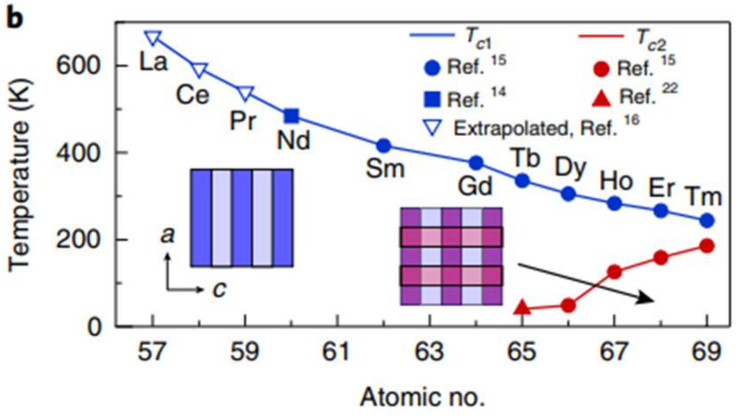
detector: Al-coated P46 phosphor screen, CCD (PI-MAX II)

Δt = 1 ps ← resolution-limited CDW peak intensity reduction

Light induced charge density wave in LaTe$_3$

Gedik group @ MIT, Nat. Phys. **16**, 159 (2020)

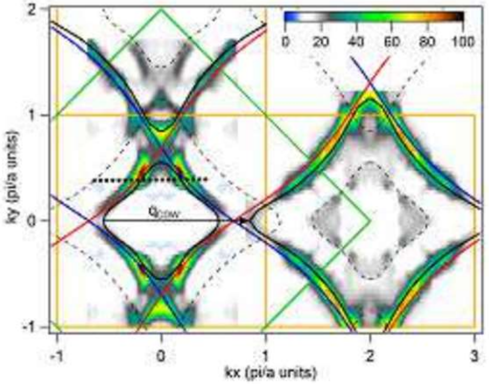


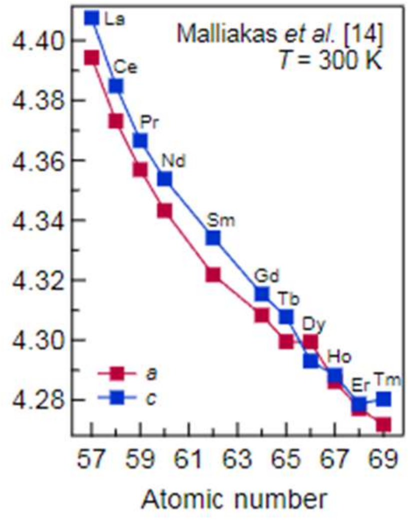


Phys. Rev. B **90**, 085105 (2014).

CeTe$_3$ FS map

Phys. Rev. Lett. **93**, 126405 (2004)

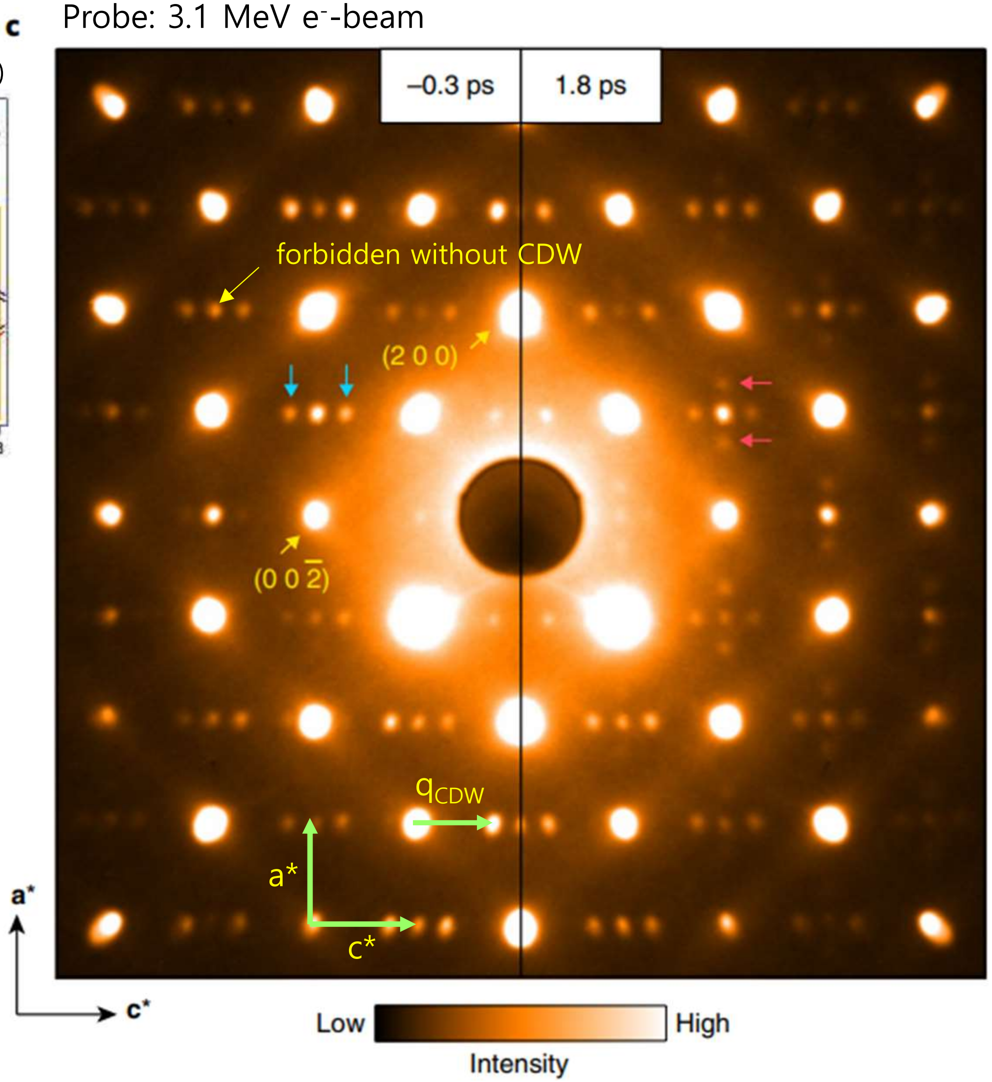




J. Am. Chem. Soc. **128**, 12612 (2006)

pump: 800 nm, 80 fs laser

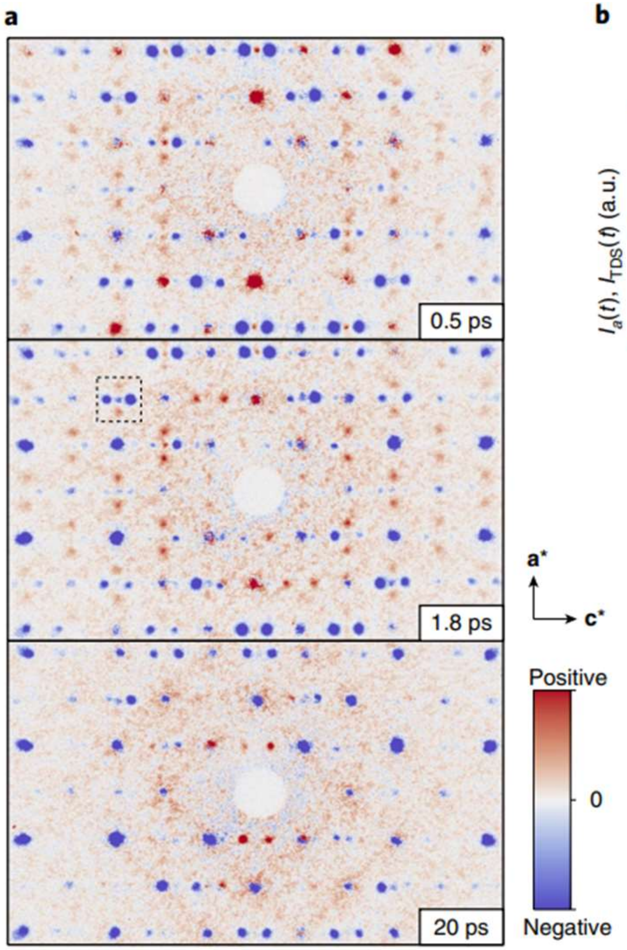
Probe: 3.1 MeV e⁻-beam



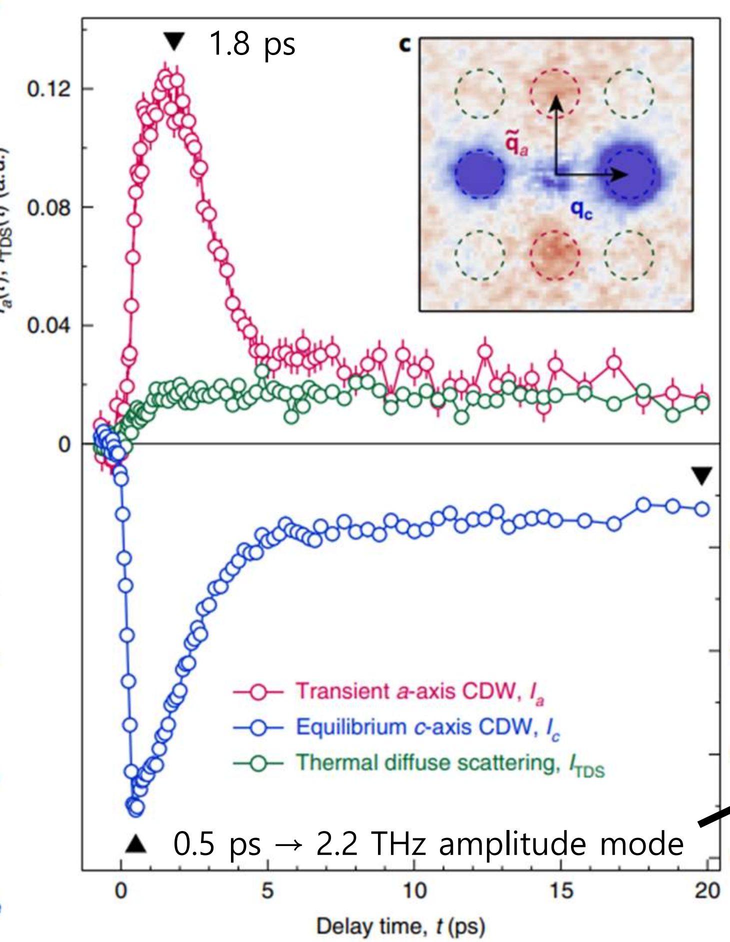
a, c value changes after pumping < 0.02%

Ultrafast dynamics of CDW peaks of $\mathrm{LaTe_3}$

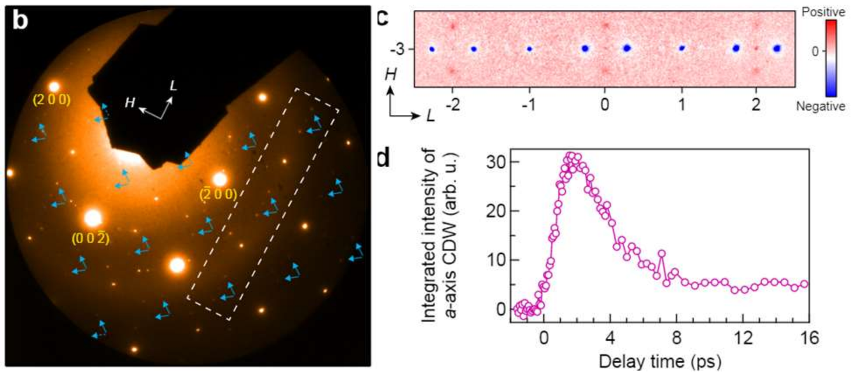
Intensity difference (off – on)

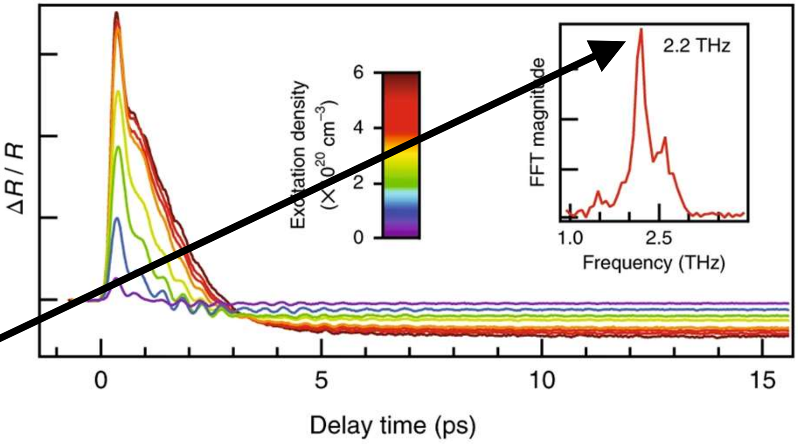


Delay-time scan of integrated intensity within r_q = 1.5 FWHM



Comparison with 26 keV UED

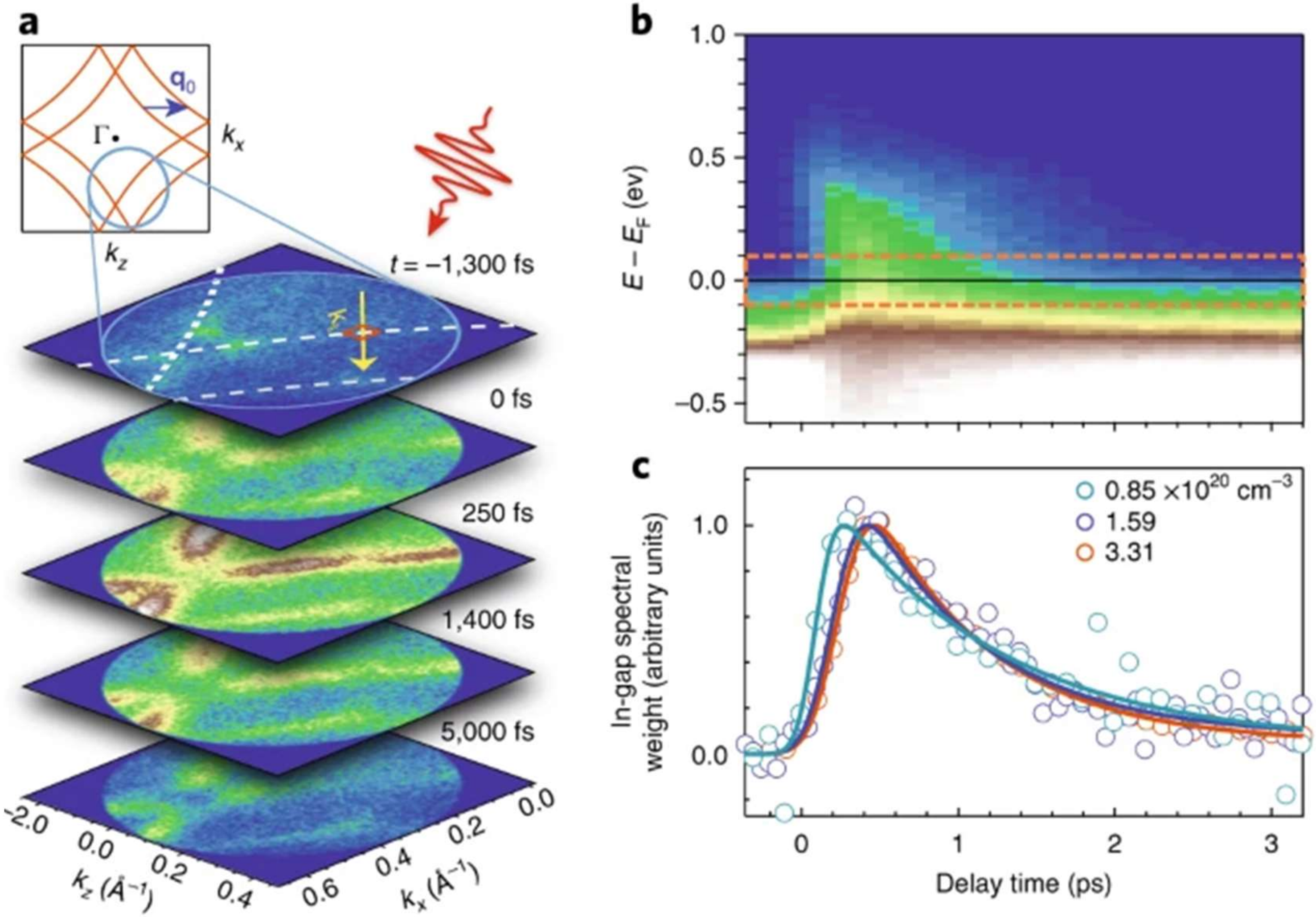




Nat. Phys. **15**, 27 (2019)

tr-ARPES FS maps showing CDW gap dynamics

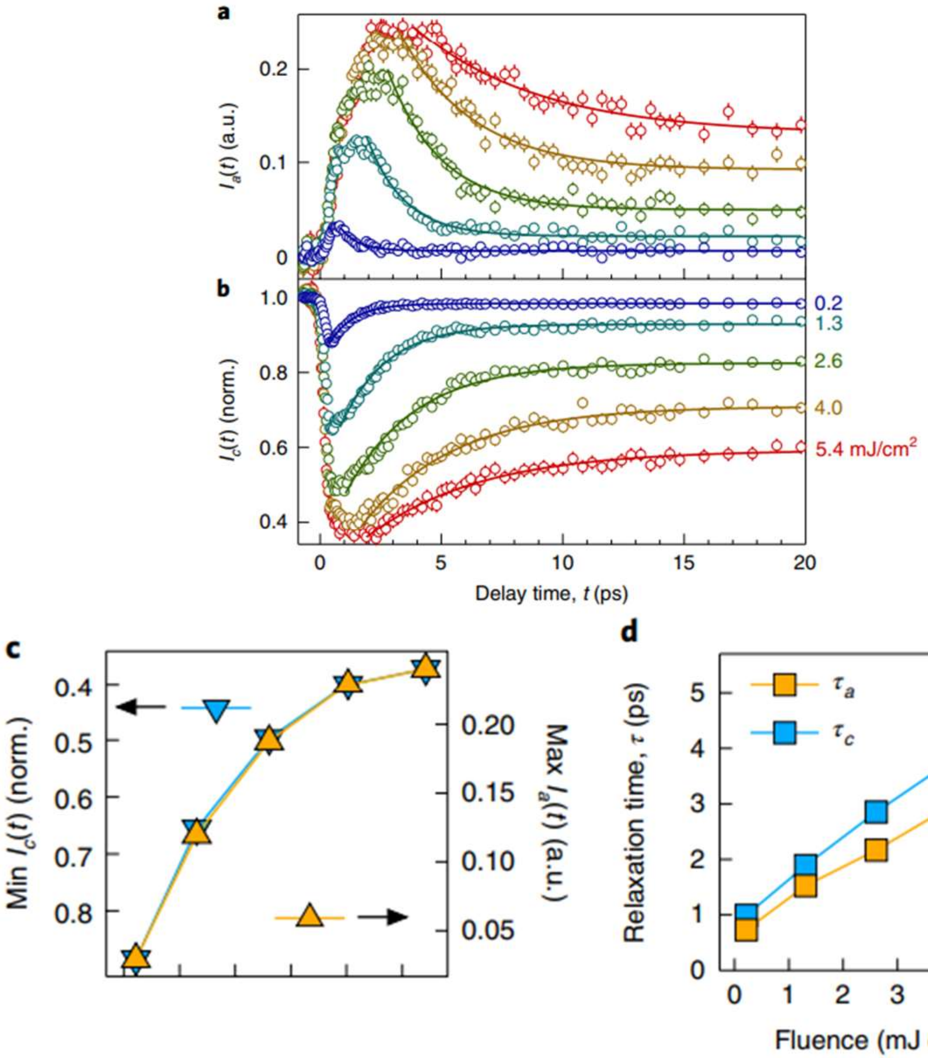
Gedik group @ MIT, Nat. Phys. **15**, 27 (2019)

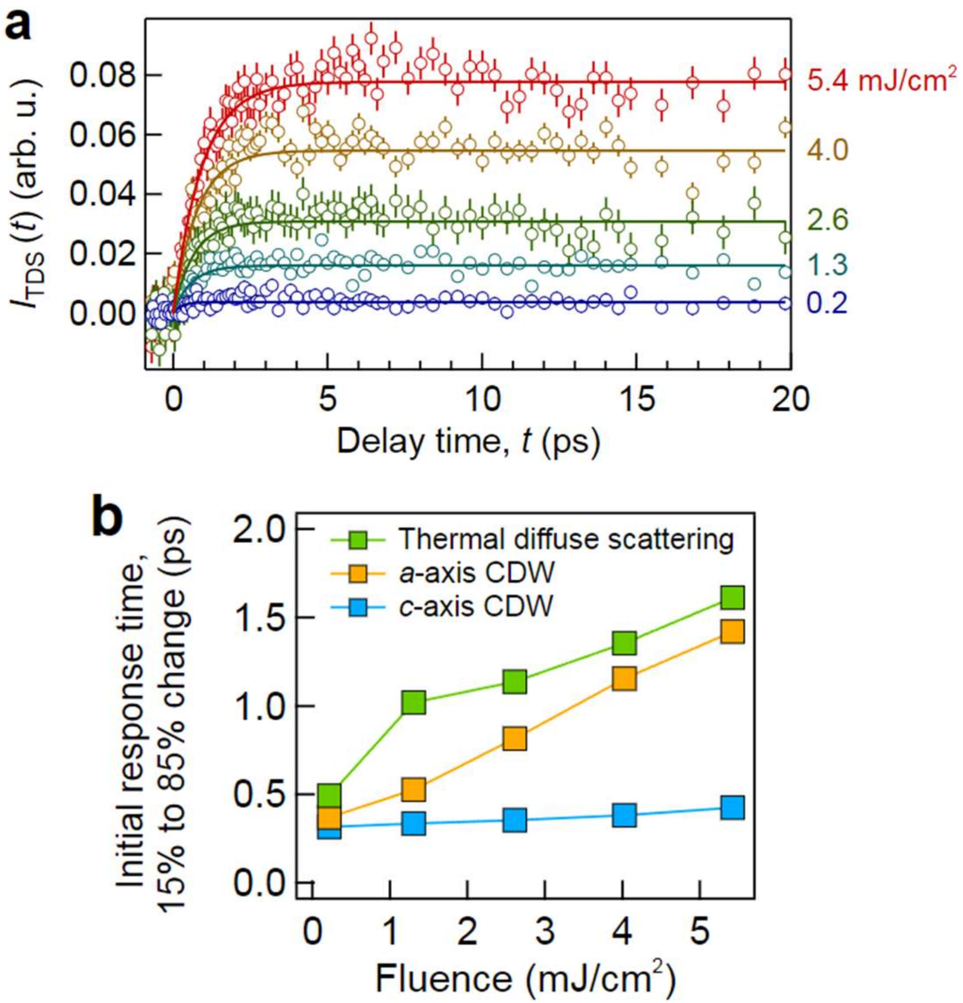


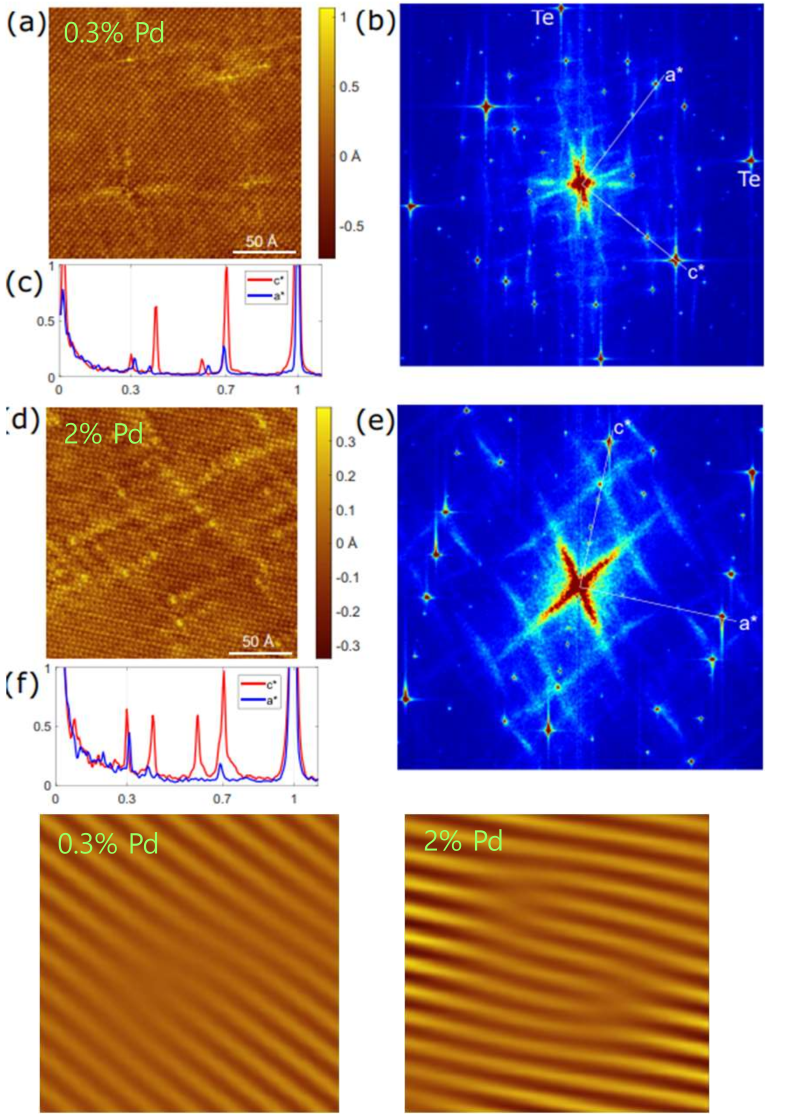
excitation density: $F = \dfrac{E}{\hbar\omega} \cdot (1 - R) \cdot \dfrac{1}{\pi(w_x/2)(w_y/2)d}$

Perfect anti-correlation between a-CDW and c-CDW intensity changes

decay time $\tau_a \approx$ recovery time τ_c → same mechanism & phase competition

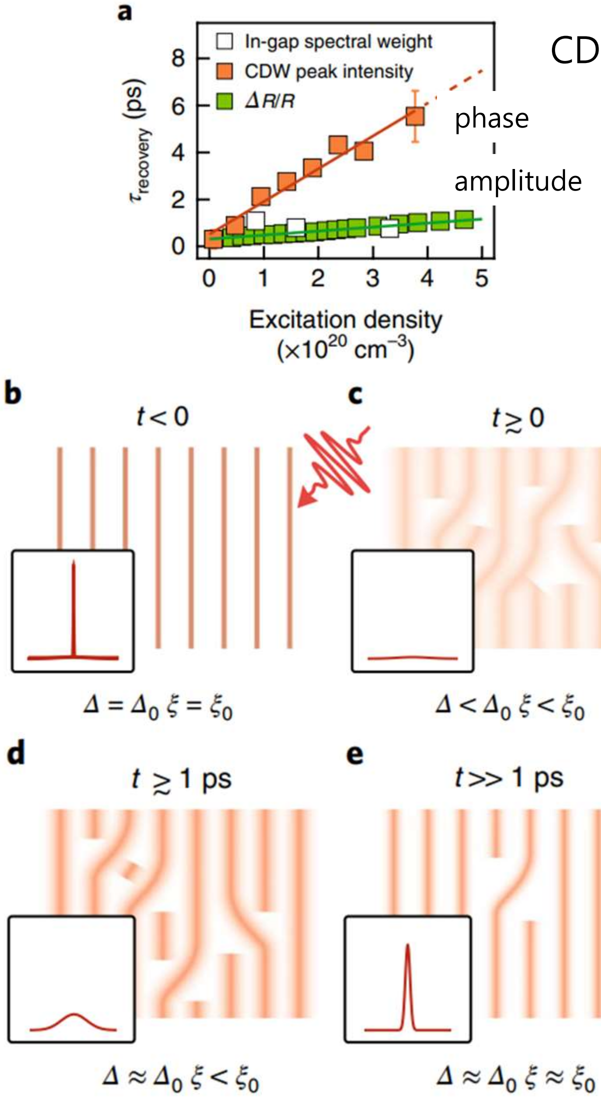






Pd defect induced CDW dislocation & a-CDW

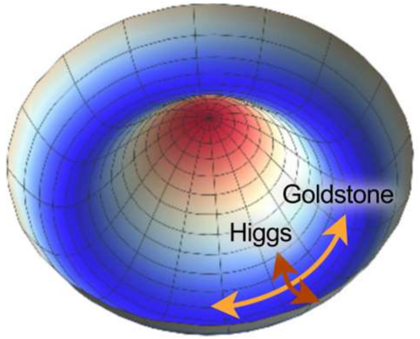
Phys. Rev. B **100**, 235446 (2019)

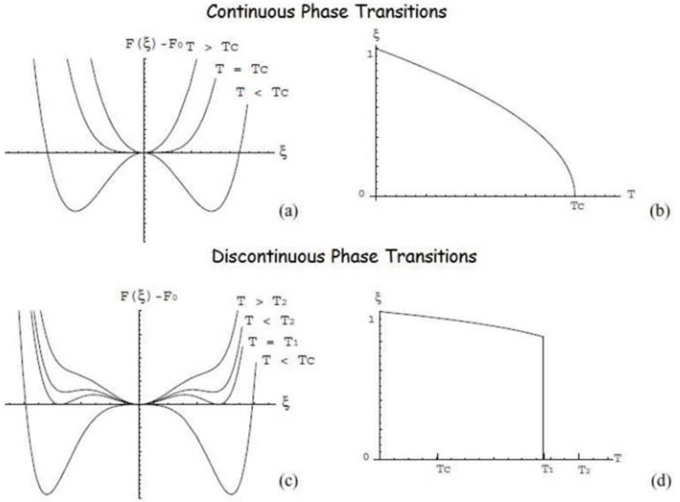


Nat. Phys. **15**, 27 (2019)

CDW PM τ_{rec} ~ 5 x CDW AM τ_{rec}

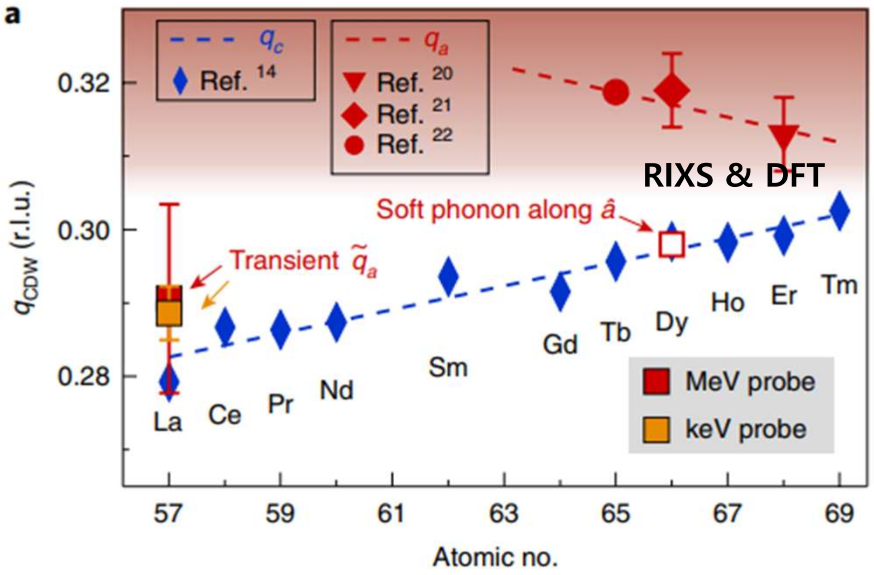
$$\mathcal{L} = \frac{1}{2}(\partial_\mu \phi)^2 - \frac{1}{2}m^2\phi^2 - \frac{\lambda}{4!}\phi^4, \quad \phi = |\phi|e^{i\theta}$$





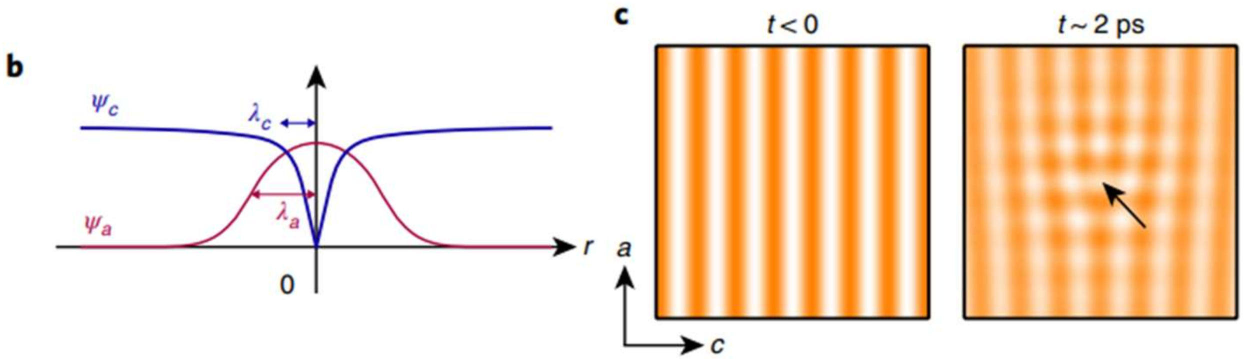
Landau theory of phase transitions

Transient nature of q_a in $LaTe_3$ different from ground state q_a in other $RETe_3$



Once c-CDW forms, gap opening changes nesting conditions for a-CDW.

If c-CDW melts, gap closing causes competition between c- & a-CDW with $q_a \approx q_c$.



Origin of transient CDW: topological defect/anti-defect pair generation
c-CDW is topologically inhibited, which allows a-CDW.

Ginzburg-Landau free energy density

$$\mathcal{F} = r_c|\psi_c|^2 + \frac{\beta_c|\psi_c|^4}{2} + \kappa_c|\nabla_{\mathbf{r}}\psi_c|^2 + r_c|\psi_c|^2 + \frac{\beta_c|\psi_c|^4}{2} + \kappa_c|\nabla_{\mathbf{r}}\psi_c|^2 + \eta|\psi_c|^2|\psi_a|^2$$

Minimize $\int d^2\mathbf{r}\mathcal{F}(\mathbf{r})$

$$\rightarrow \quad -\kappa_c\nabla_{\mathbf{r}}^2\psi_c + r_c\psi_c + \beta_c|\psi_c|^2\psi_c + \eta|\psi_a|^2\psi_c = 0$$

$$-\kappa_a\nabla_{\mathbf{r}}^2\psi_a + r_a\psi_a + \beta_a|\psi_a|^2\psi_a + \eta|\psi_c|^2\psi_a = 0$$

by putting $\psi_c(r,\phi) = \psi_c^\infty f(r)e^{im\phi}$ and $\psi_a(r,\phi) = \psi_a^\infty g(r)$