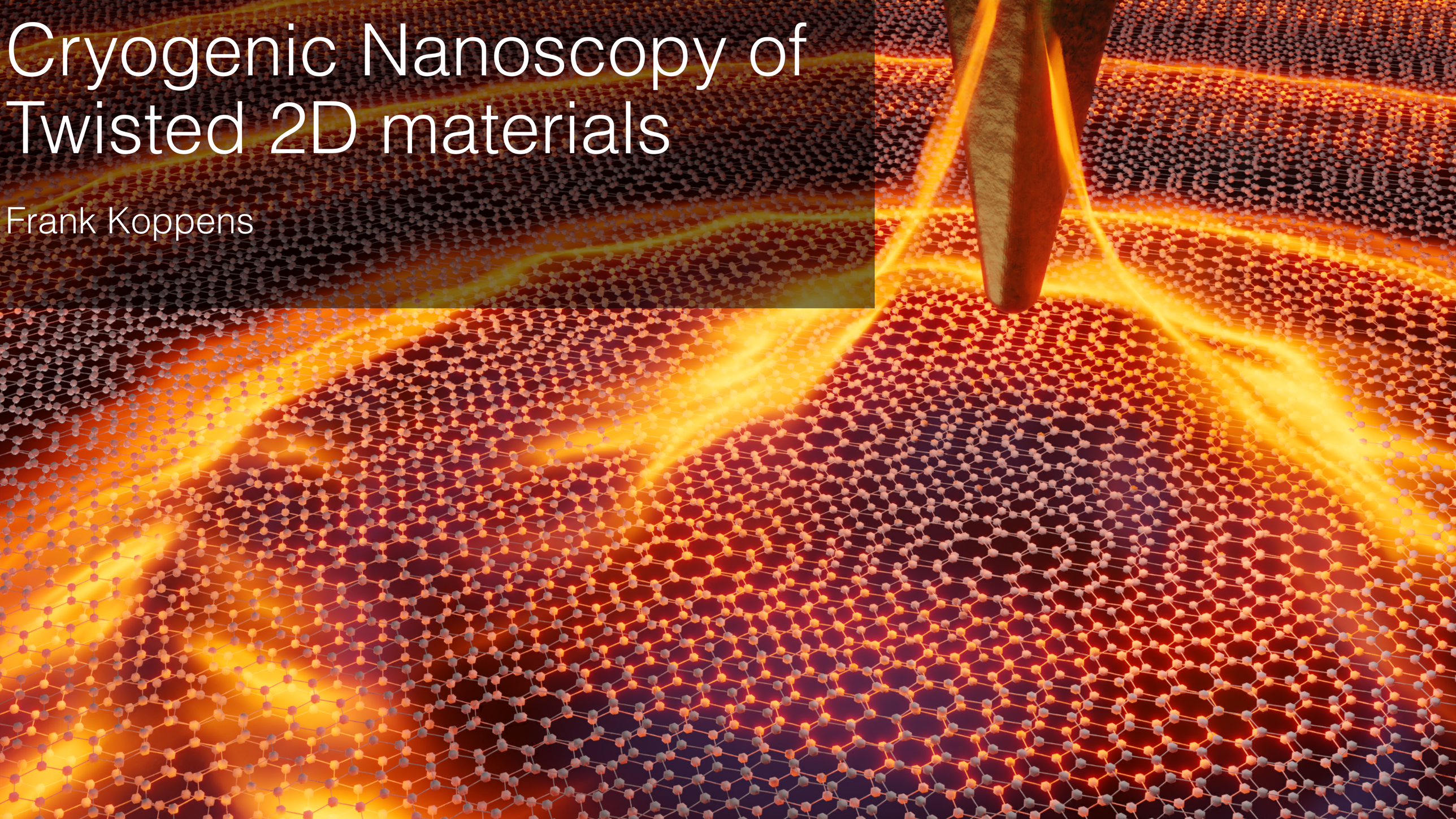
Cryogenic Nanoscopy of Twisted 2D materials

Frank Koppens



ICFO

- 10km south of Barcelona
- 20 years old
- 400 Researchers
- 23 research groups

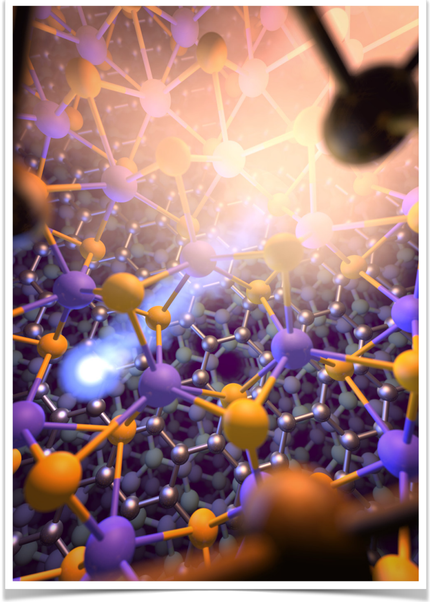
- Opto-electronics
- Nano-photonics
- Non-linear optics
- Quantum optics
- Medical optics
- Bio-physics

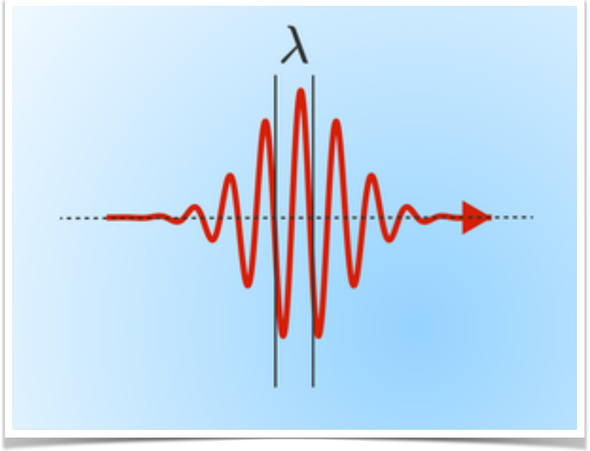
How to survive pandemic

Nanoscale → Micronscale

Electron: nanometer

Photon:
500 nanometer - 500 micrometer
(VIS - THz)



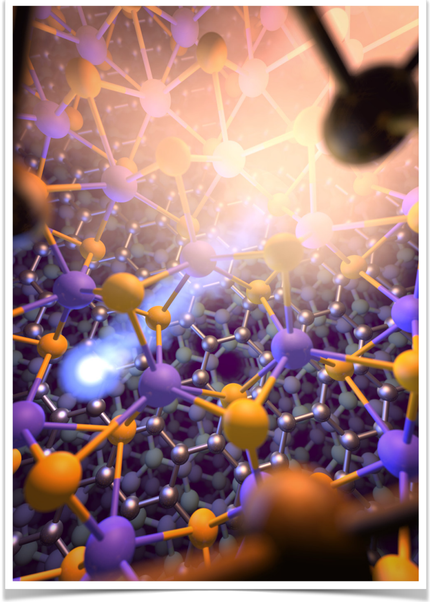


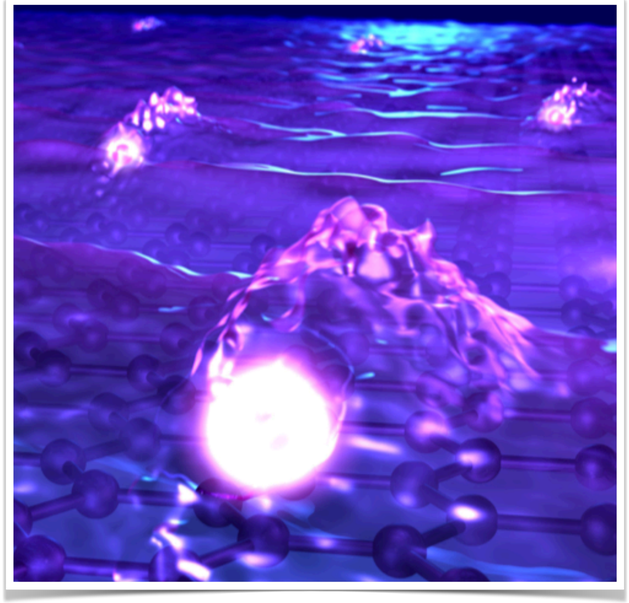
Nanoscale → Micronscale

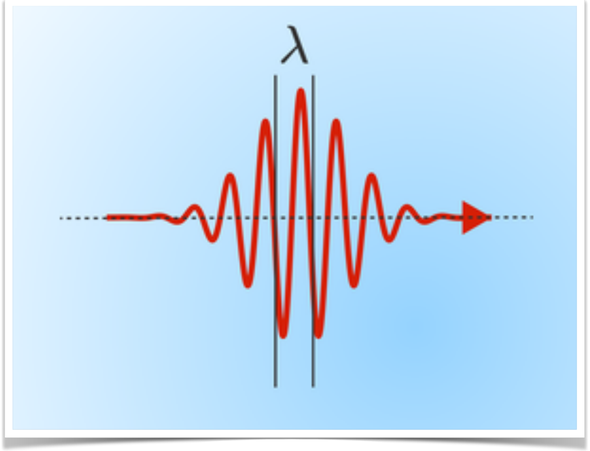
Electron: nanometer

Polaritons: 20 nanometer

Photon:
500 nanometer - 500 micrometer
(VIS - THz)







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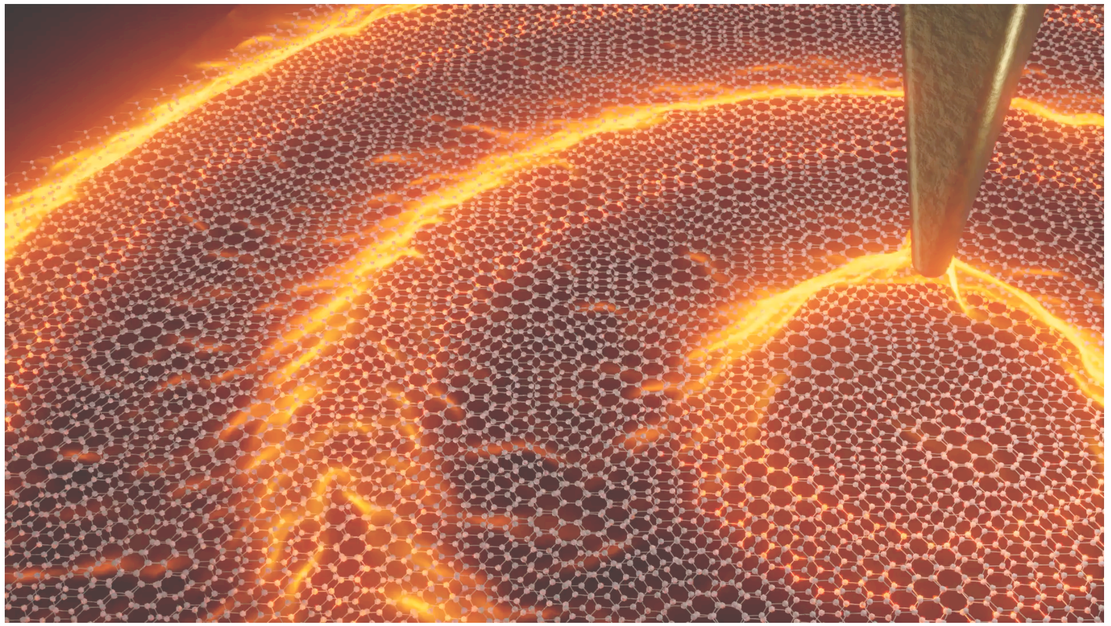
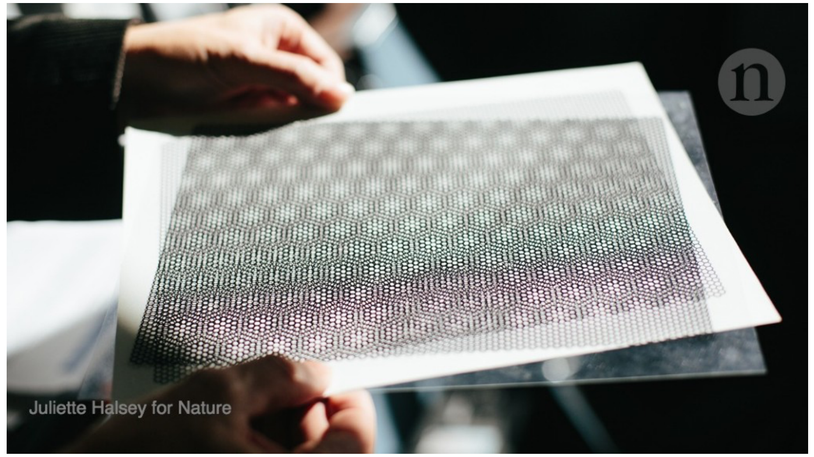
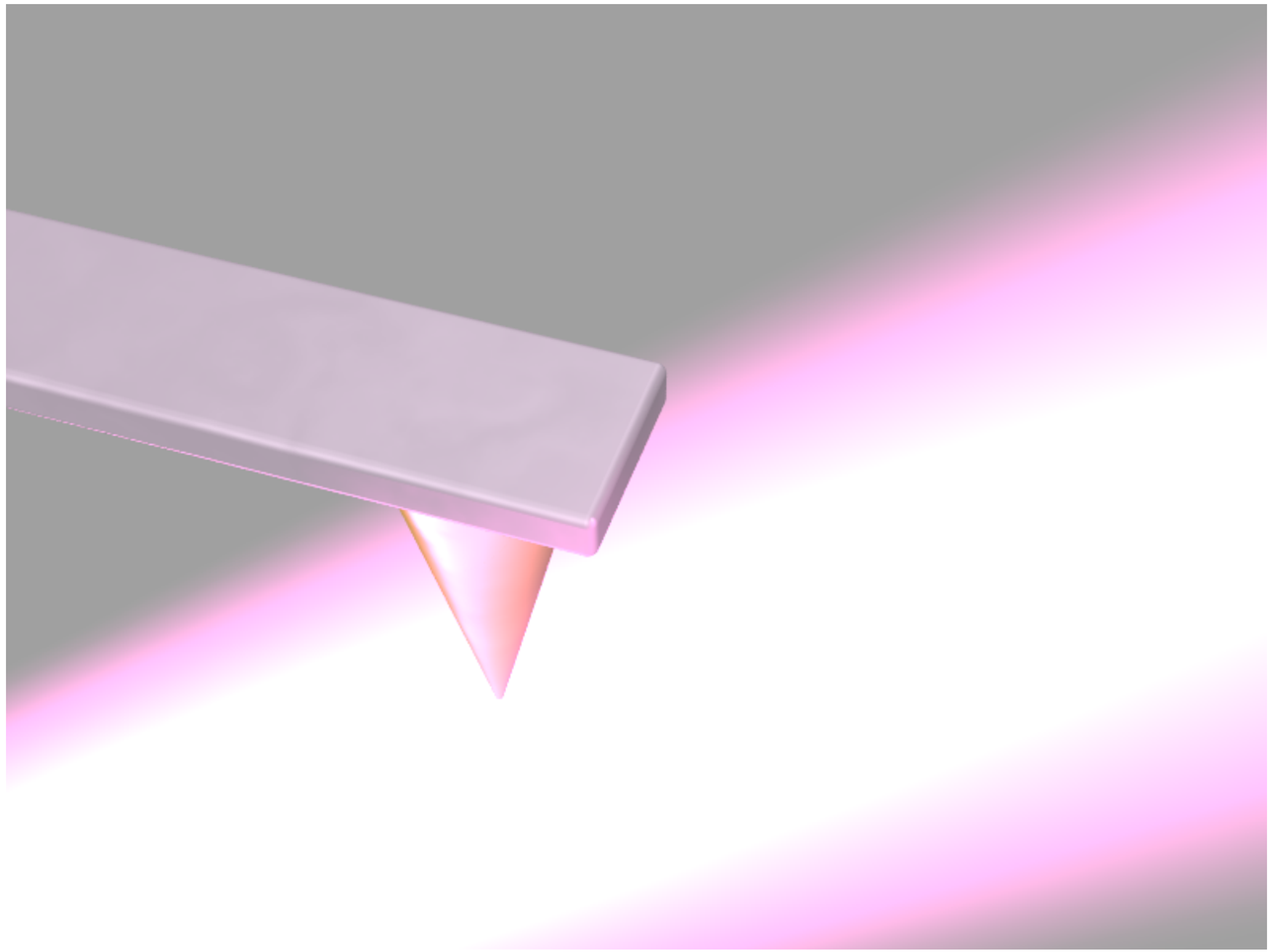




Near-field microscopy

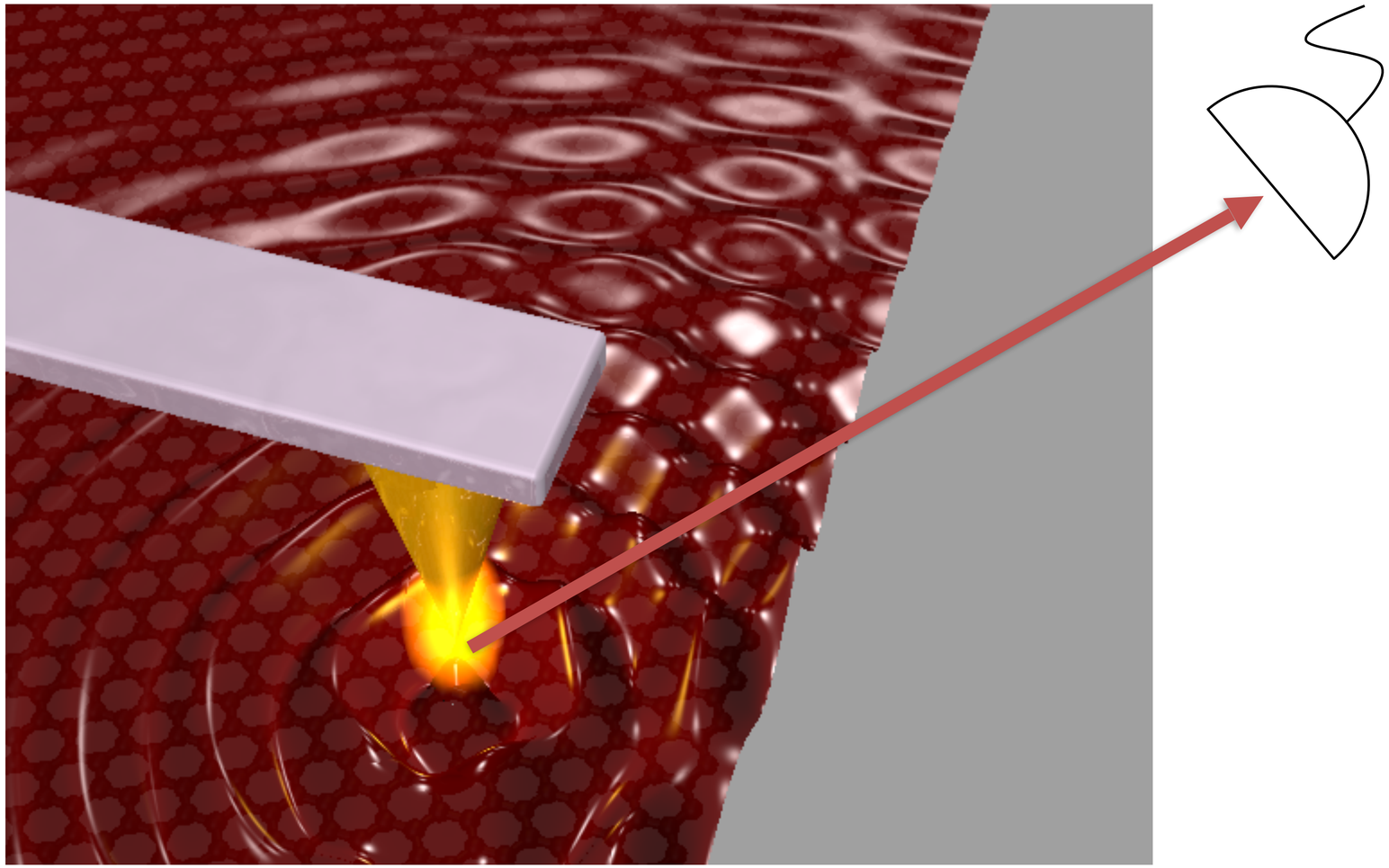
Mid-infrared or terahertz light (wavelength 2-120μm, E=0.01-0.5eV) couples with an atomic force microscope tip



Pioneered by groups of Hillenbrand, Koppens, Basov

See e.g. Chen et al, Nature 2012. Fei et al, Nature 2012

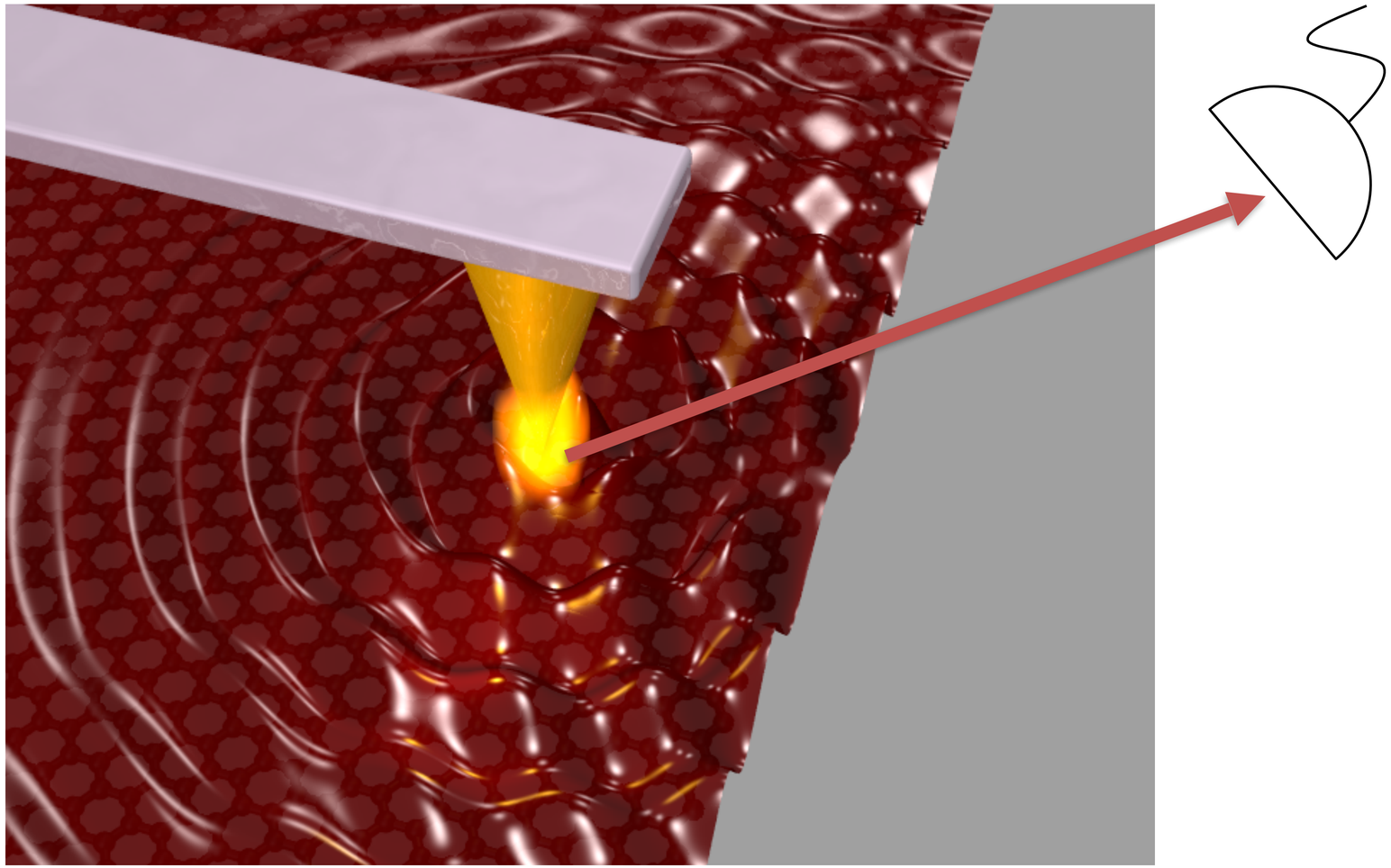
Scattered light from the tip apex is collected in the far-field



Pioneered by groups of Hillenbrand, Koppens, Basov

See e.g. Chen et al, Nature 2012. Fei et al, Nature 2012

The device is moved and the plasmon is excited at a different position



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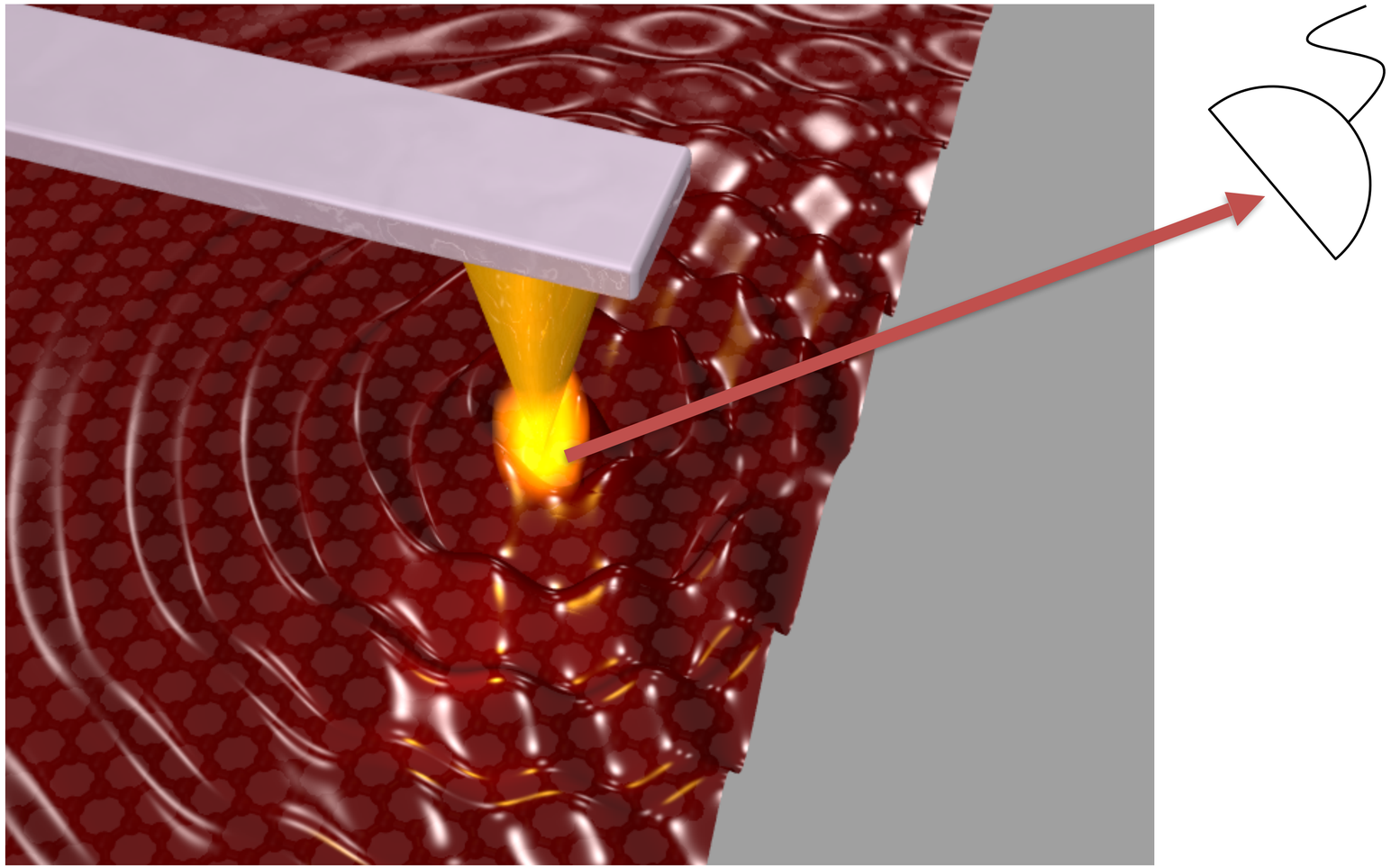
See e.g. Chen et al, Nature 2012. Fei et al, Nature 2012

Scattering near-field microscopy:
probe light of VIS-IR-Terahertz with 20nm resolution

Probe optical conductivity with 20nm resolution
$\sigma(\omega,q)$

Probe propagating collective excitations

e.g. plasmons
acoustic plasmons
phonon polaritons



See also groups of Hillenbrand, Basov, …

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Scattering near-field microscopy:
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with 20nm resolution
$\sigma(\omega,q)$

Probe propagating collective
excitations

e.g. plasmons
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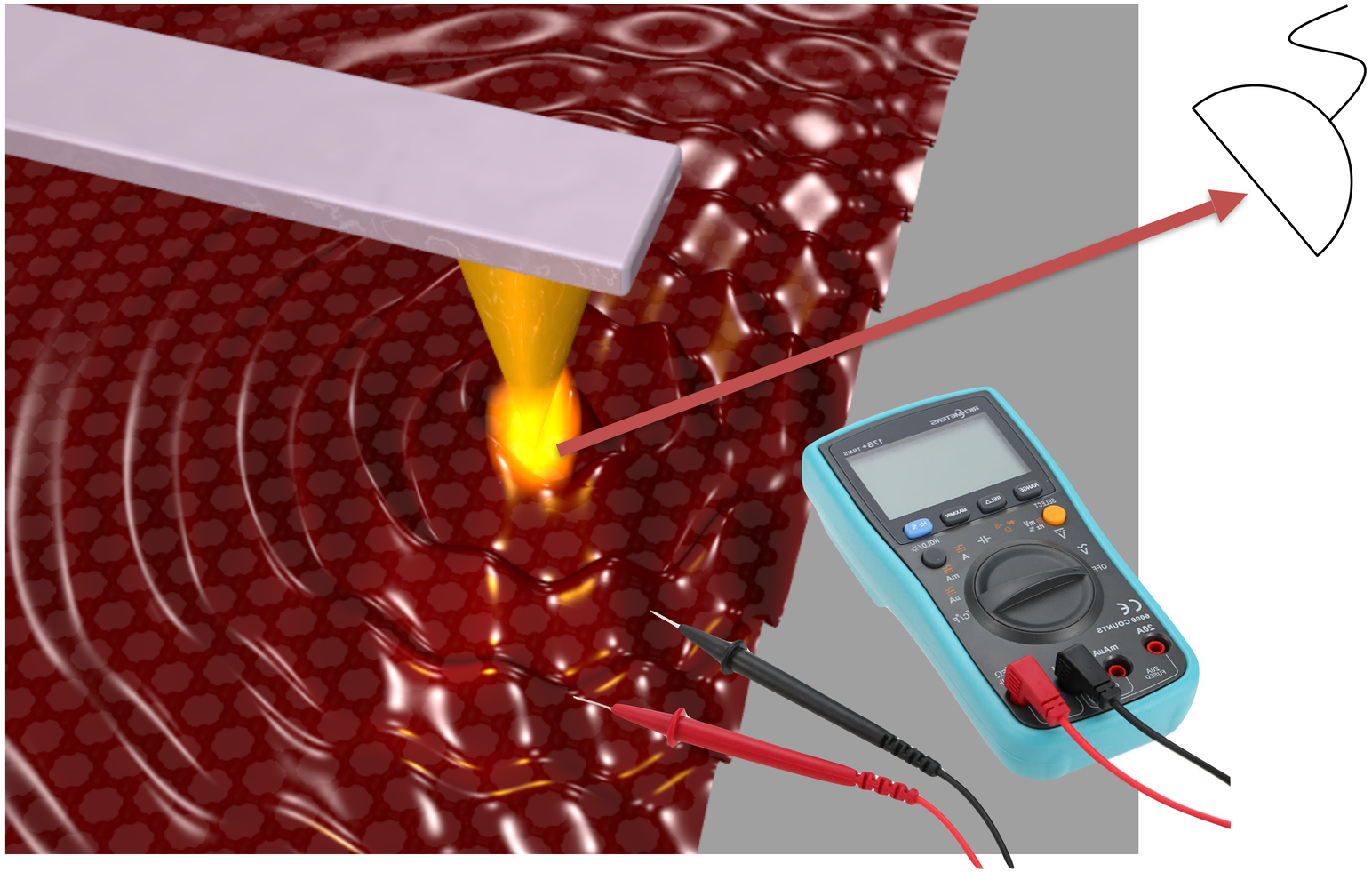
Photocurrent nanoscopy:
Seebeck
Domains
Density profiles

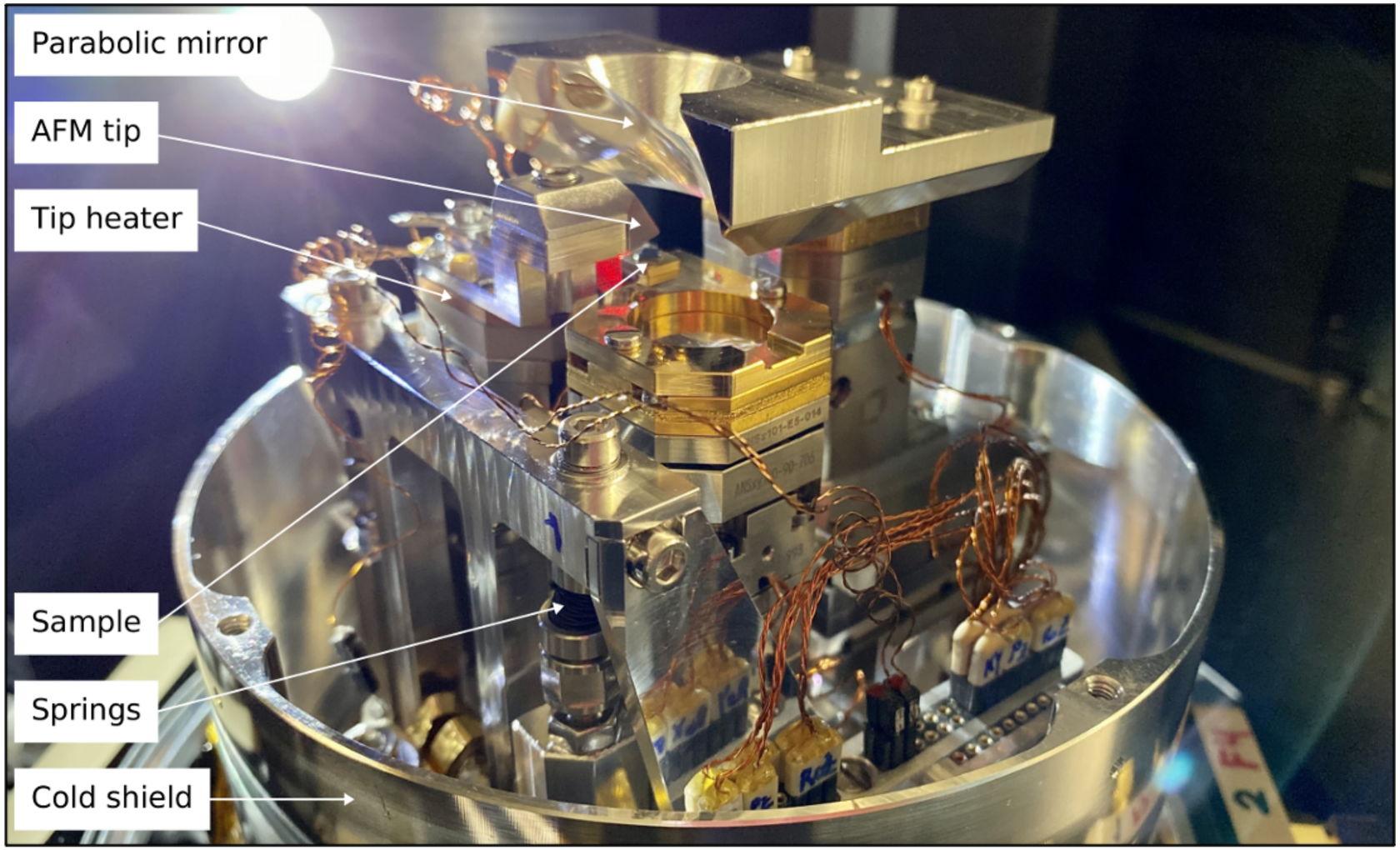


See also groups of Hillenbrand, Basov, …

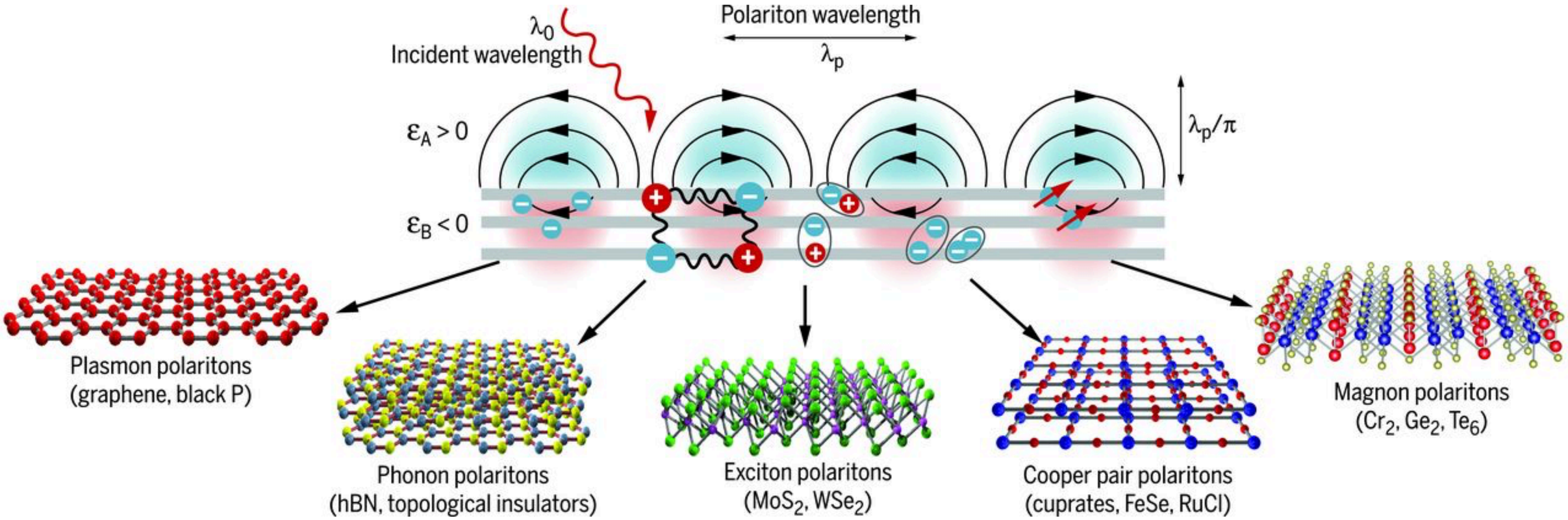
See e.g. Chen et al, Nature 2012. Fei et al, Nature 2012

Cryogenic near-field microscopy

Temperature 6K



Polaritons in 2D materials

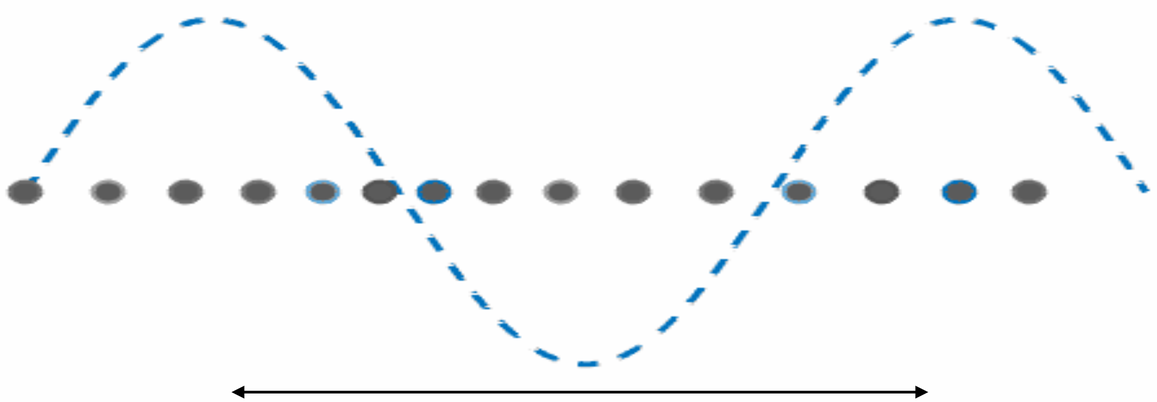


See reviews:

Low et al., Polaritons in layered two-dimensional materials, Nature Materials (2016).

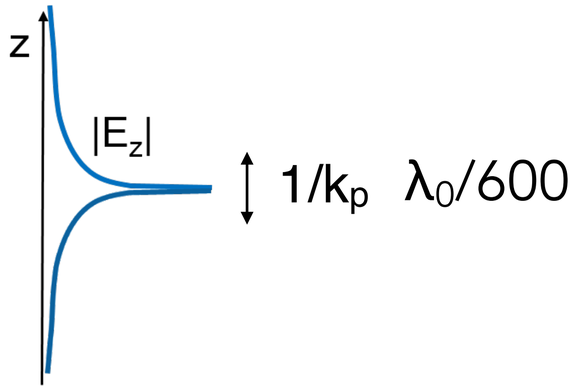
Basov et al., Science (2016)

Graphene "Dirac" plasmons



$\lambda_{plasmon}=1/k_p$

$\lambda_{photon}/200$



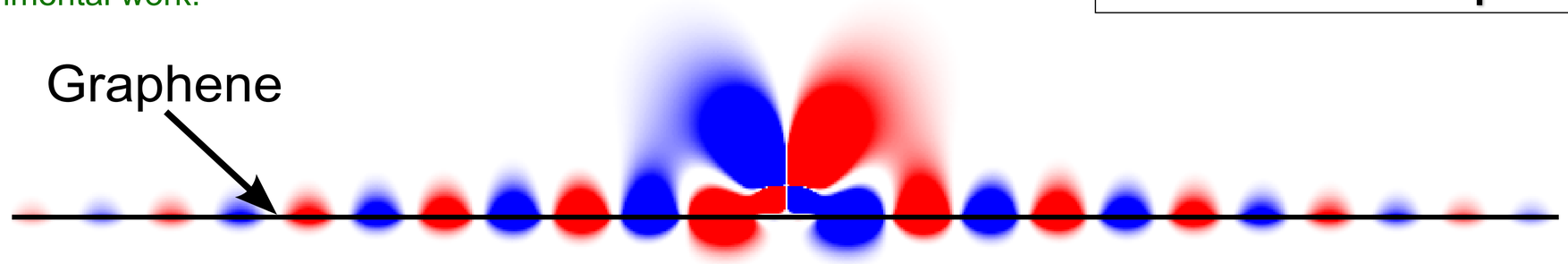
Many body excitation

Long-range Coulomb interaction

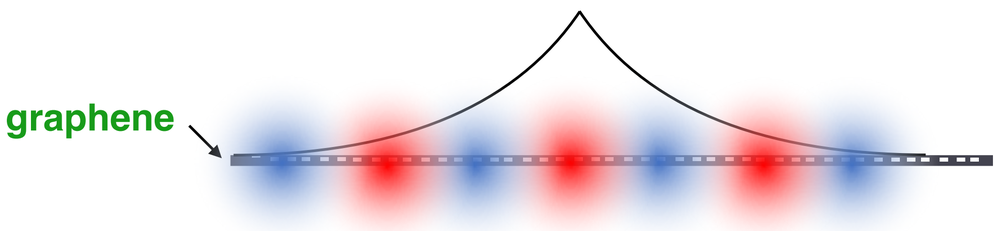
Can couple to light

Pioneering theory work:
Jablan et al, PRB (2009)
Hwang et al, PRB (2007)
Polini et al, PRB (2008)
Wunsch et al., NJP (2008)
Koppens, Abajo, Nano Letters (2012)

Pioneering experimental work:
Basov
Hillenbrand
Atwater
Halas
Mortenson
Pruneri
Altug
etc etc.

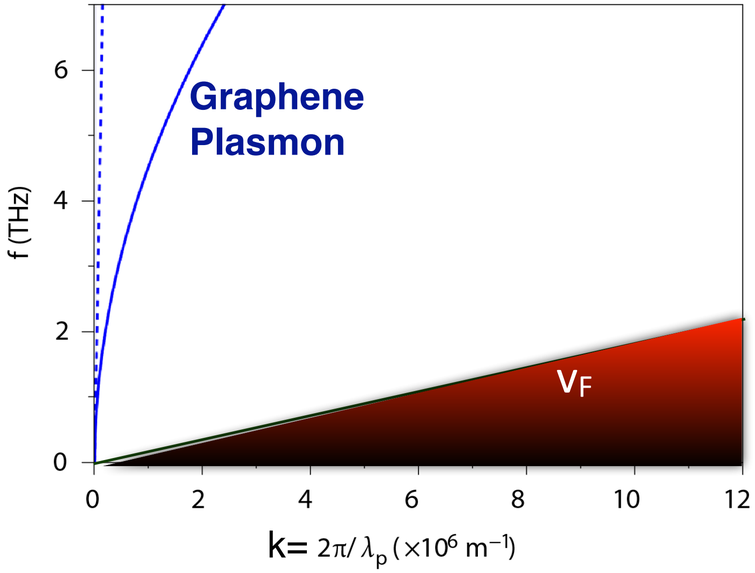


Normal plasmon

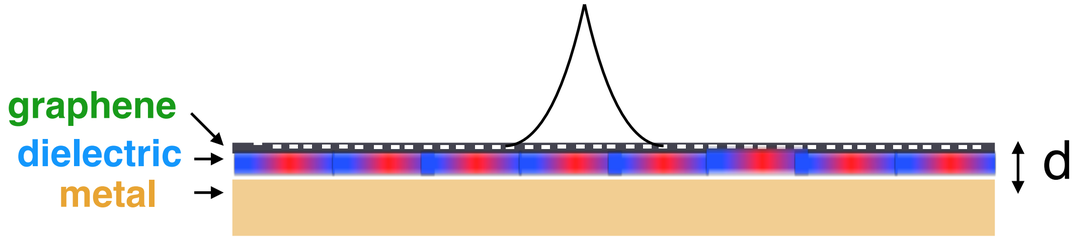


velocity ~c/100

$$\omega \propto \sqrt{q}$$



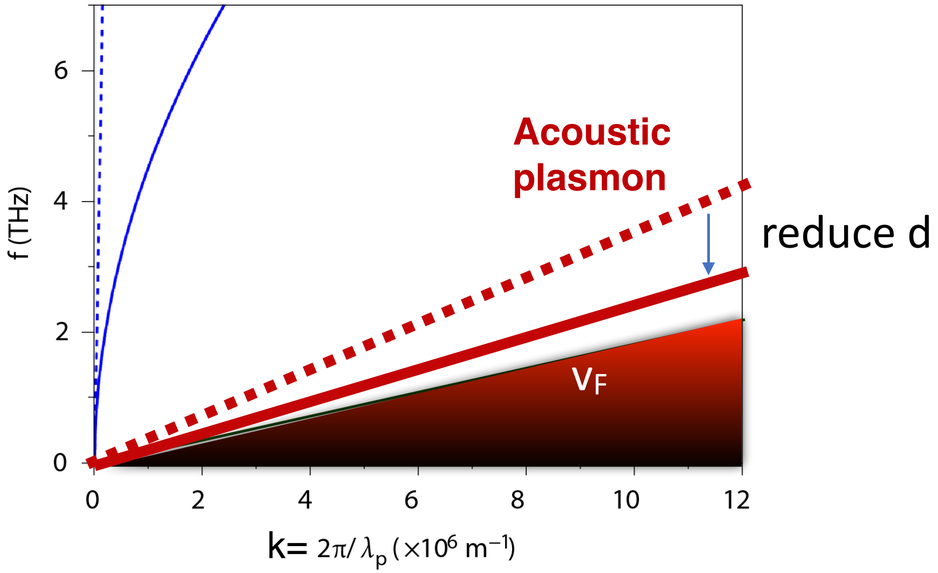
Screened plasmon: short-range interactions



velocity ~c/300

Alcaraz, Science 2018

$$\omega \approx q v_F$$



Acoustic plasmon:

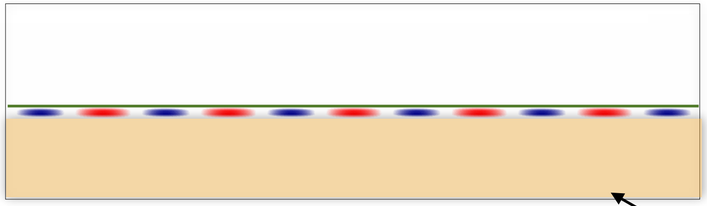
Alonso-Gonzalez et al., Nature Nanotech (2017)

Confinement down to one atom

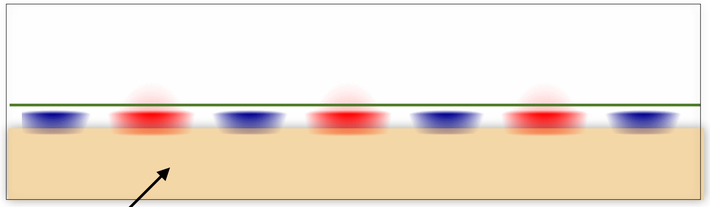
Alcaraz, Science 2018

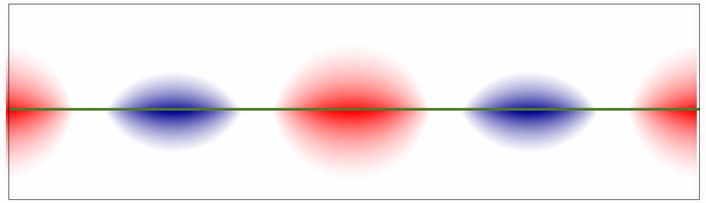
Graphene - insulator - metal

d=0.3 nm



d=5 nm

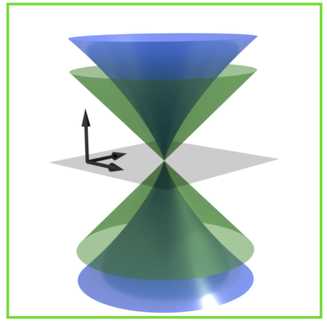




Propagation speed ~c/300

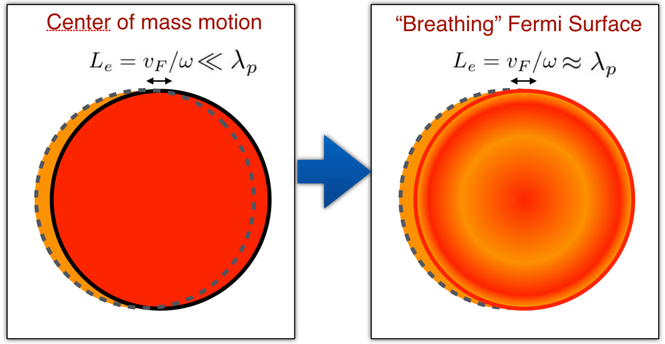
Metal

Fermi velocity renormalisation

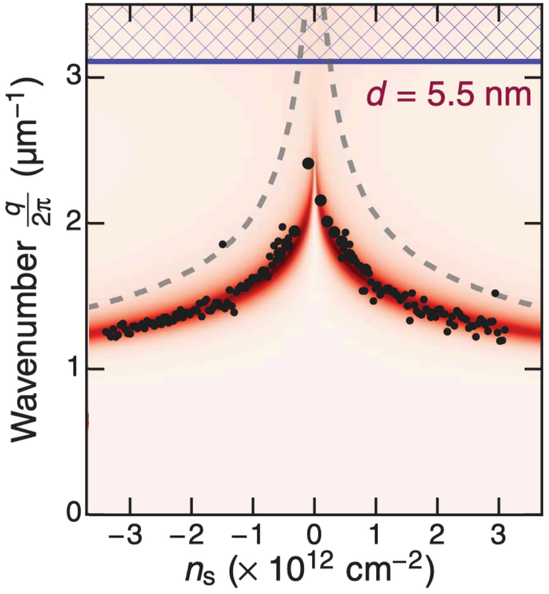


See e.g.
Elias et al., Nat.Phys. 2011

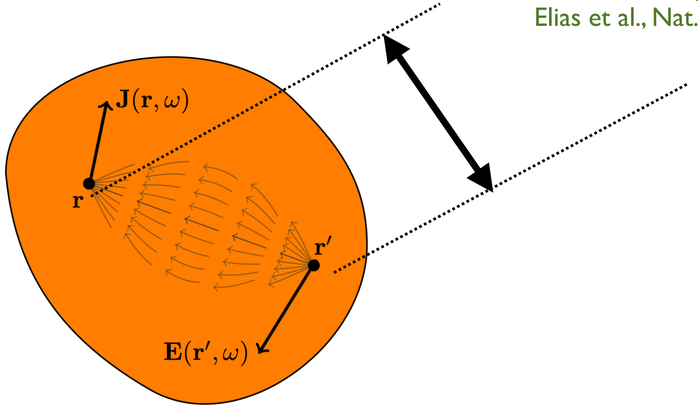
Compressibility



Graphene non-local effects
(incl. e-e interactions, compressibility)



Lundeberg et al., Science 2017

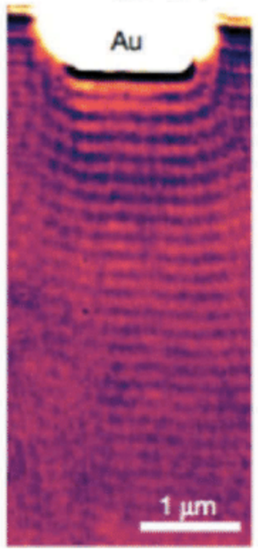


Thesis: Christensen (Mortensen group)

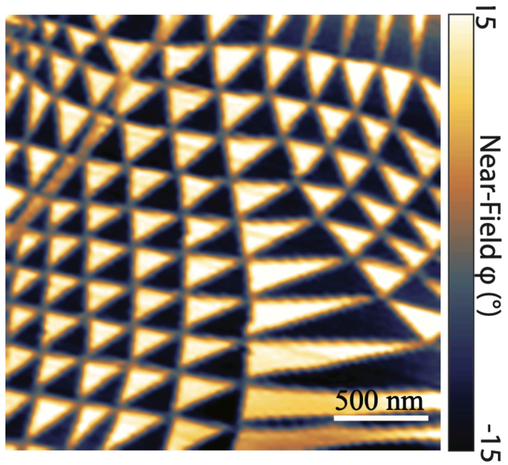
$$\sigma(\omega, q)$$

Scattering optical near-field microscopy

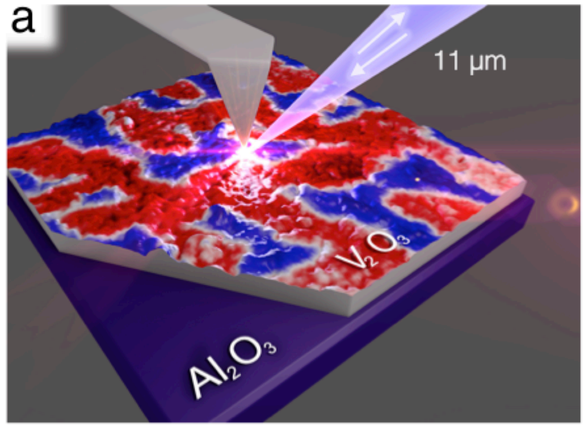
Infrared Propagating plasmons



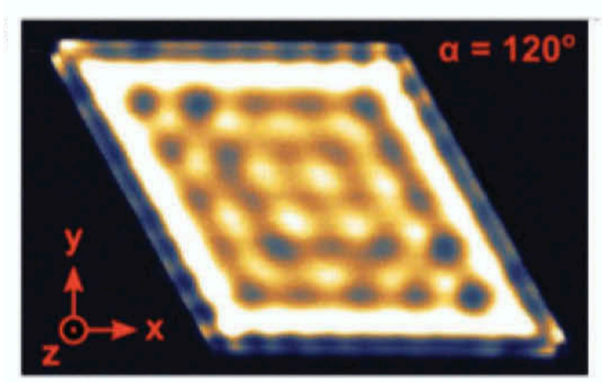
Twisted graphene



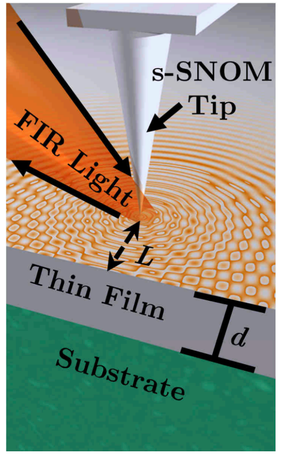
Correlated materials



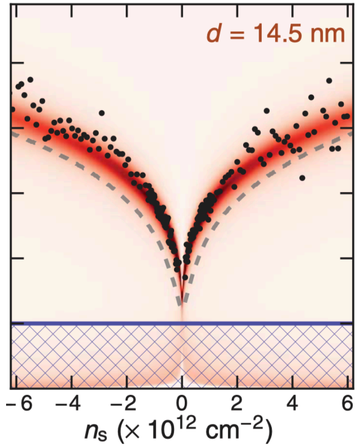
Propagating phonon polaritons



Superconductors



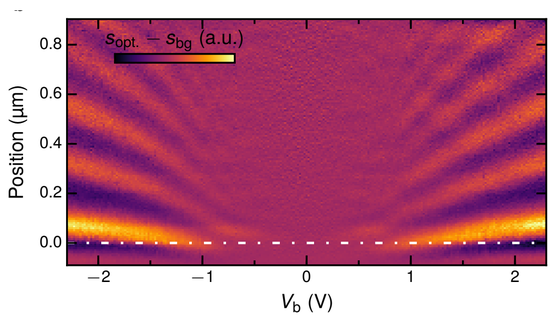
Quantum non-local
Electron interactions

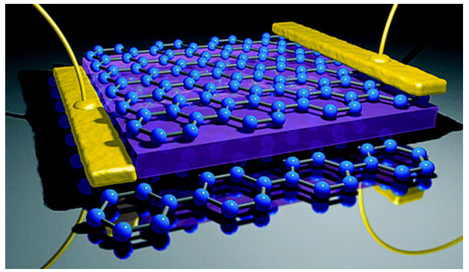


See also groups of Hillenbrand, Basov, etc. …

Plasmons: Lego toolbox for light manipulation at atomic scale

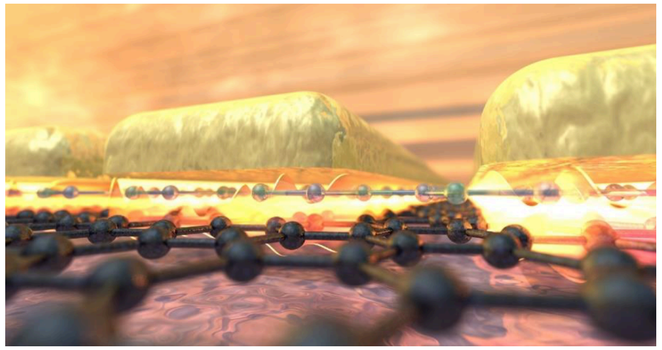
Plasmons in double layer graphene quantum tunnelling device





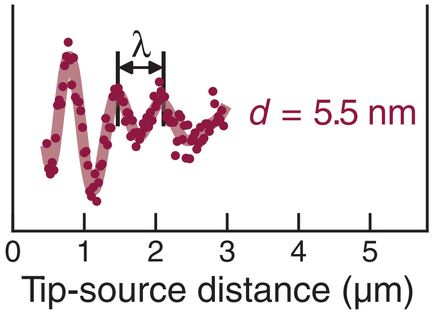
Woessner et al., ACS Photonics 2017

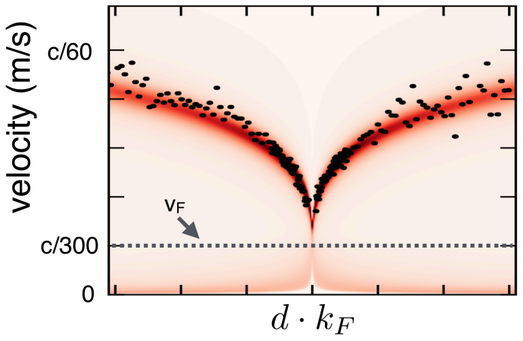
Confine plasmons to one atom



Alcarez et al. Science 2018

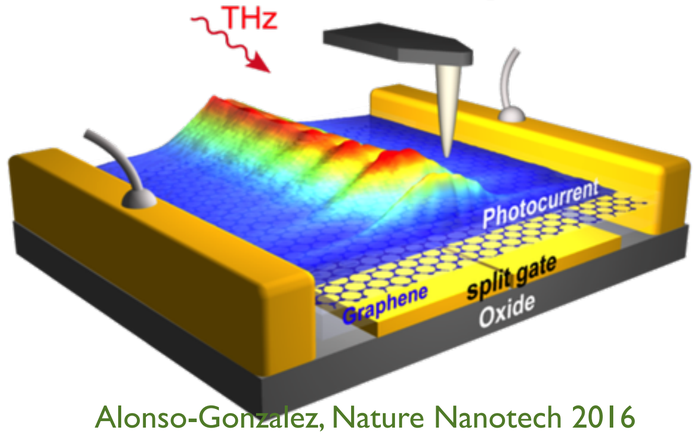
Acoustic THz plasmons in graphene:
Extreme non-local effects. Electron interaction effects.





Lundeberg et al., Science 2017

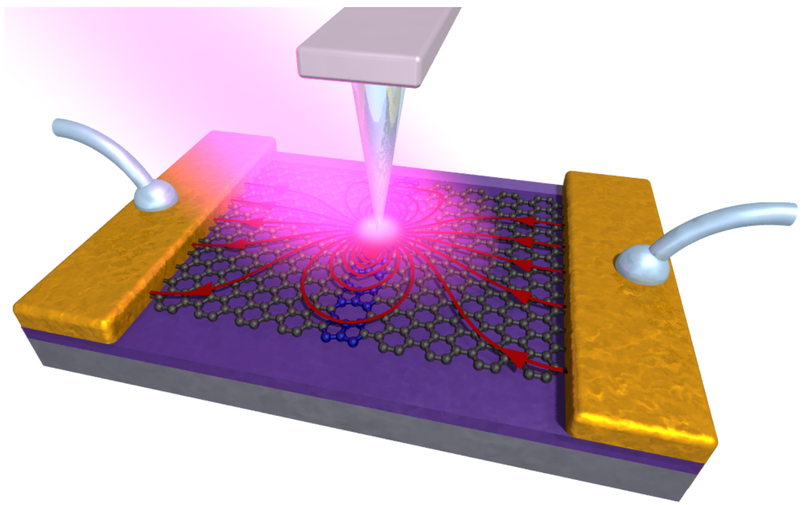
Infrared+THz detection/generation



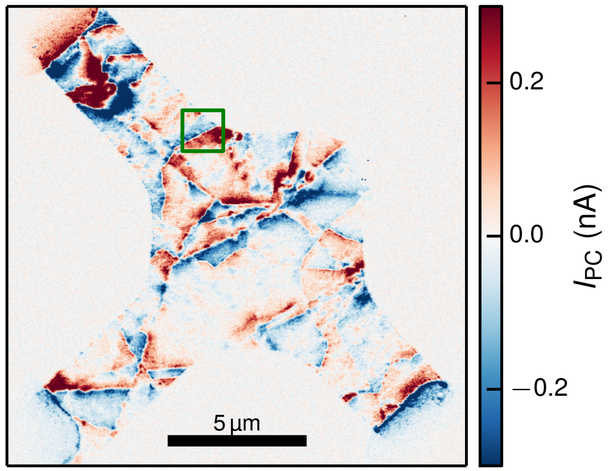
Alonso-Gonzalez, Nature Nanotech 2016
Lundeberg, Nature Materials 2016

Near-field photodetection for MIR/THz

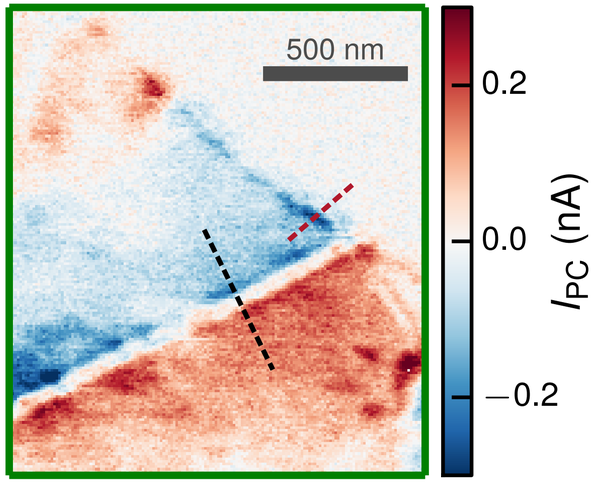
Excitation 10.000 nm



CVD Graphene (mobility ~4000cm²/Vs)

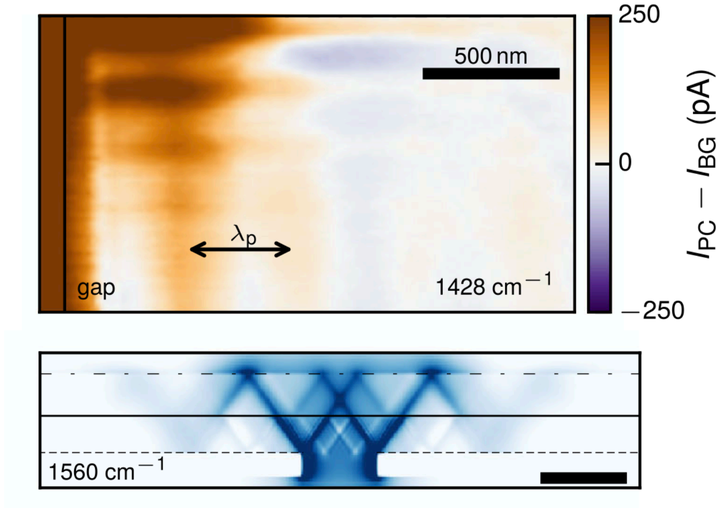


Grain boundaries



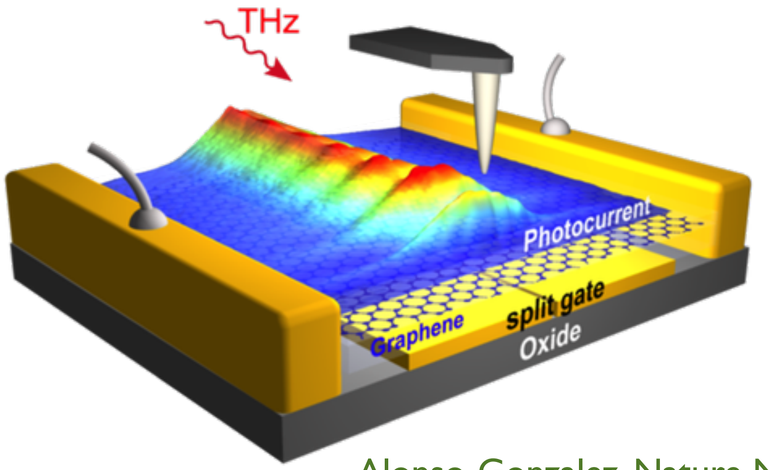
Woessner, Alonso-Gonzalez et al., Nature Com 2016

Electrical Phonon polariton detection



Woessner, 2D materials and applications 2017

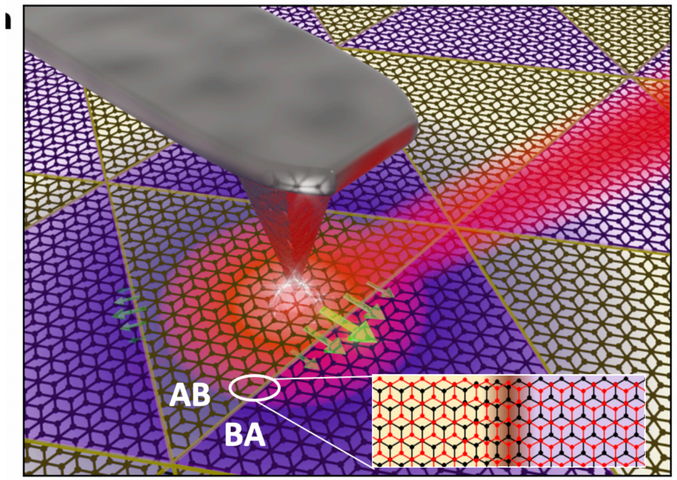
Electrical Plasmon polariton detection



Alonso-Gonzalez, Nature Nanotech 2016
Lundeberg, Nature Materials 2016

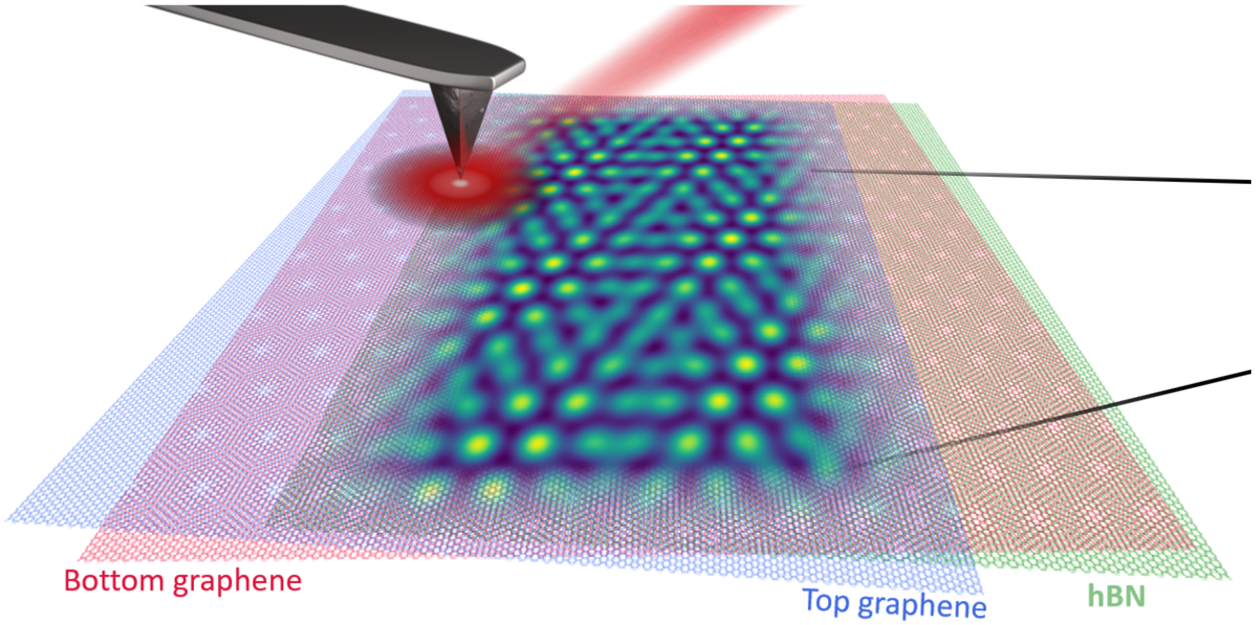
Light-matter interactions in twisted graphene

Nano-imaging of photoresponse inside a moiré unit cell

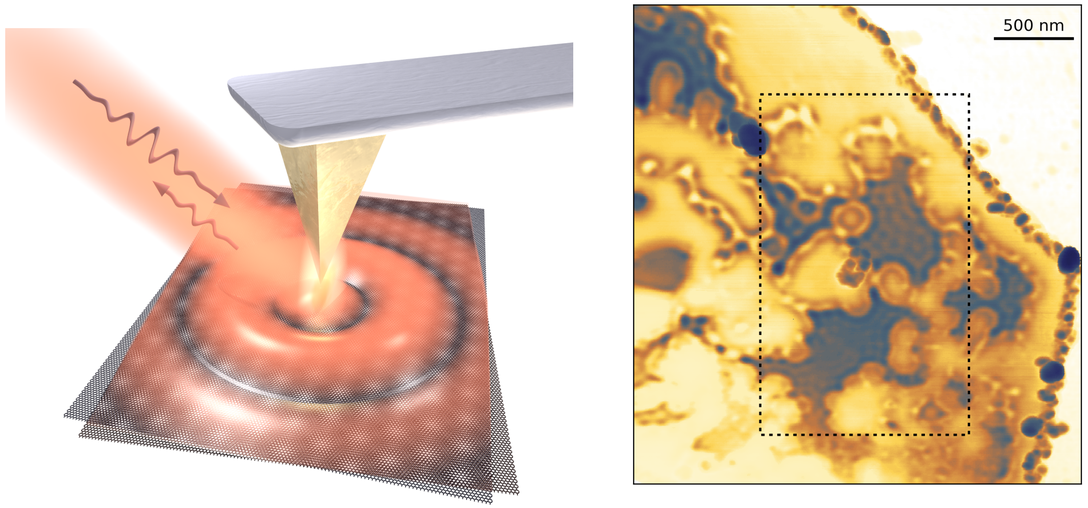


Hesp et al., Nature Com 2020

Supermoire of twisted graphene aligned on hBN

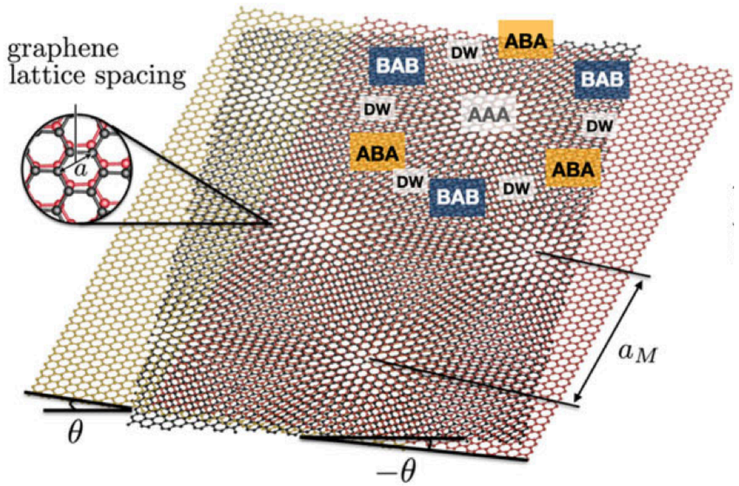


Interband plasmons in twisted graphene close to the magic angle

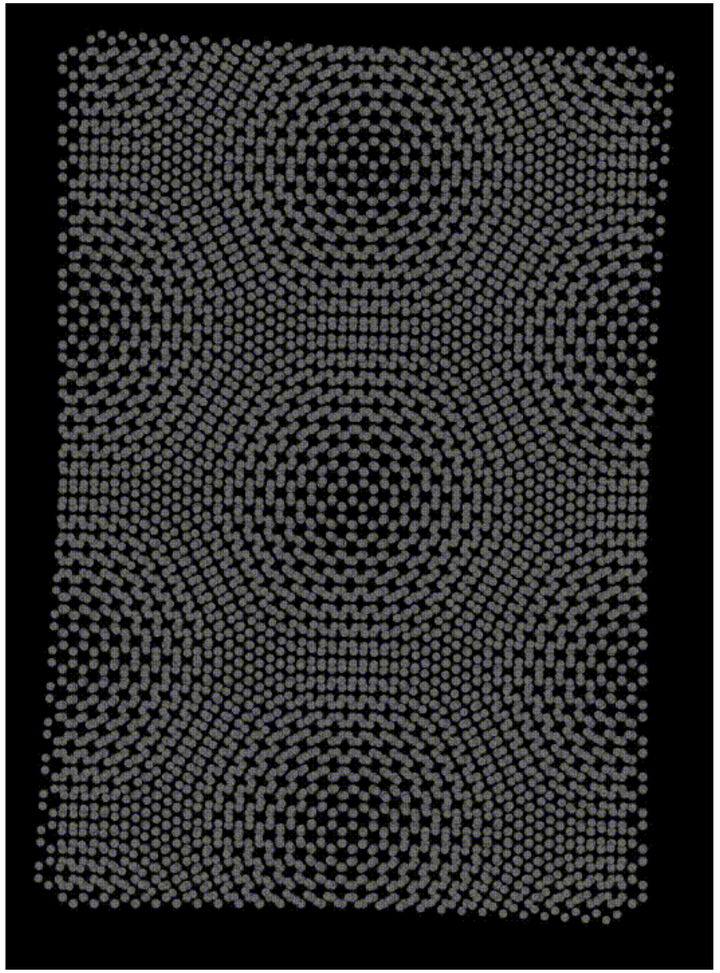


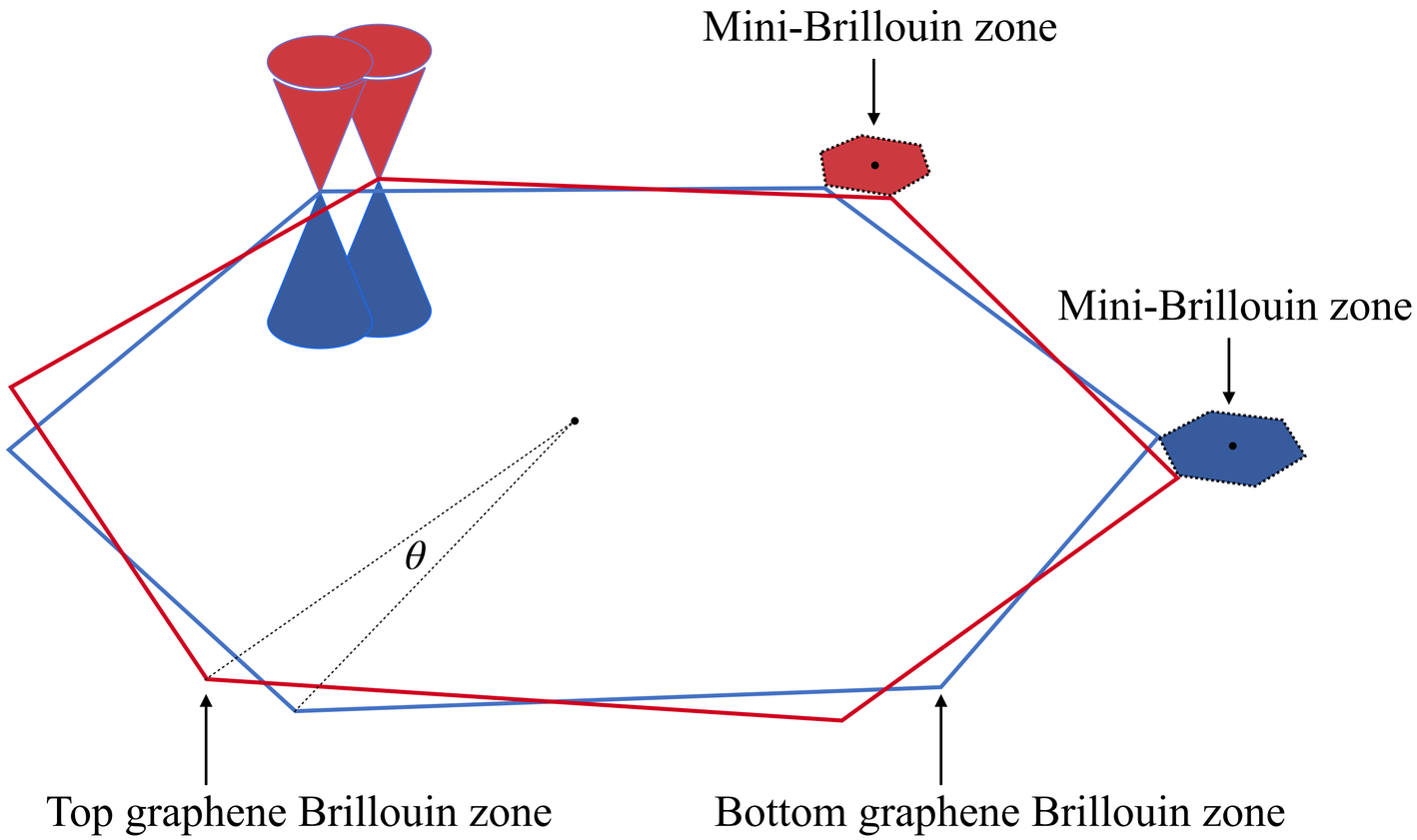
Hesp et al., Nature Physics 2021

Photocurrent nanoscopy of twisted trilayer graphene

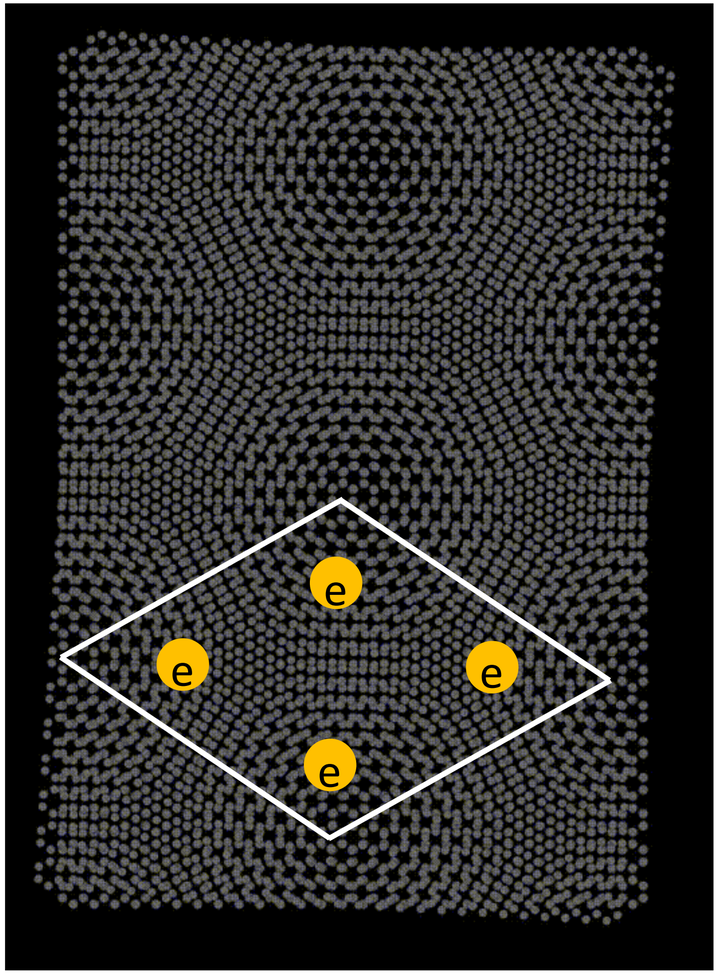


Slowing down electrons in graphene using moiré engineering

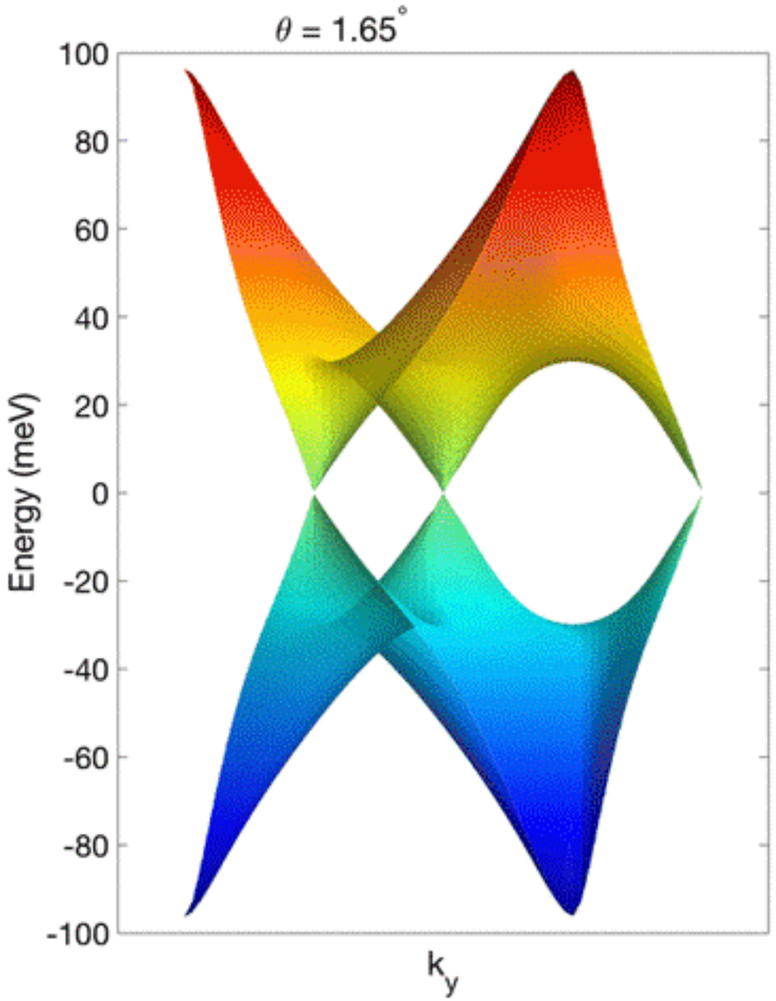




Slowing down electrons in graphene using moiré engineering

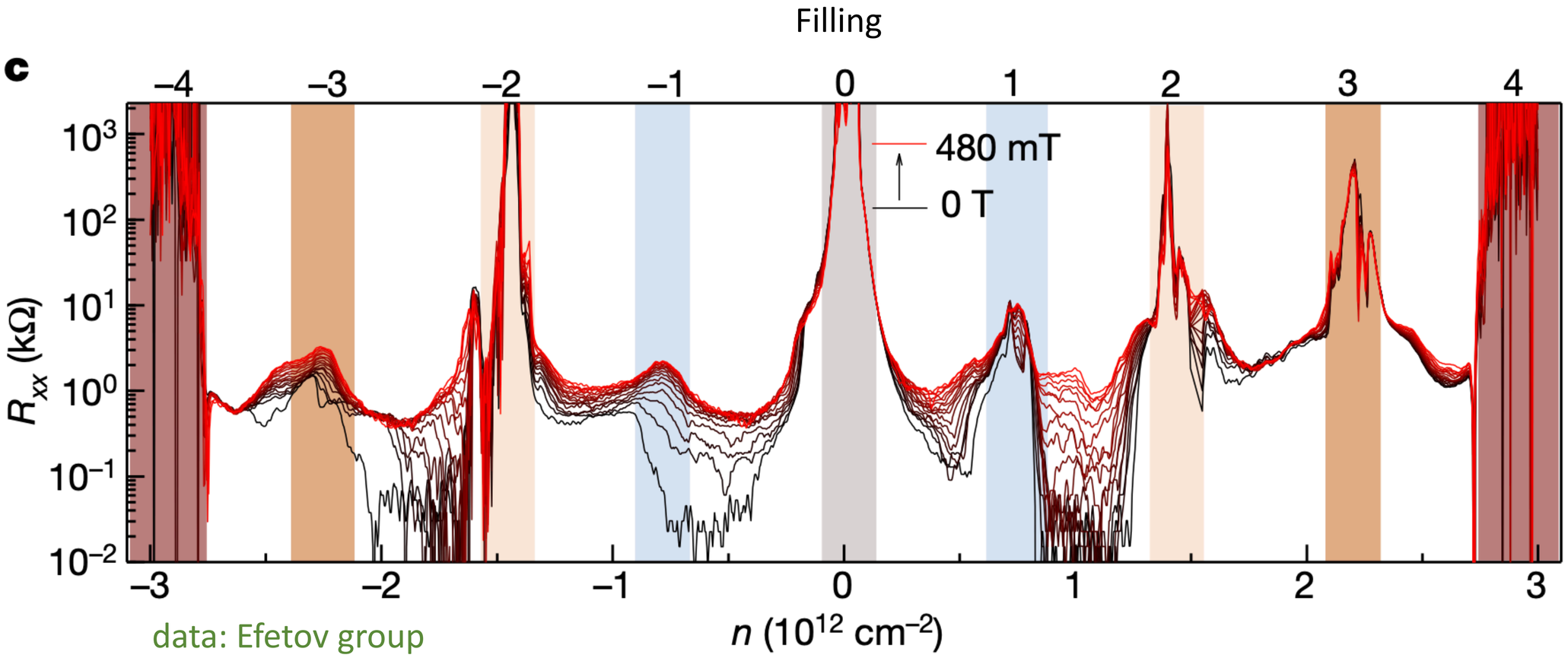


Four-fold degenerate electrons (spin/valley)



Bistrizer & MacDonald, *PNAS* **108**, 30 (2011)
Cao et. al., *Nature* **556**, 43-50 (2018)
Cao et. al., *Nature* **556**, 80-84 (2018)

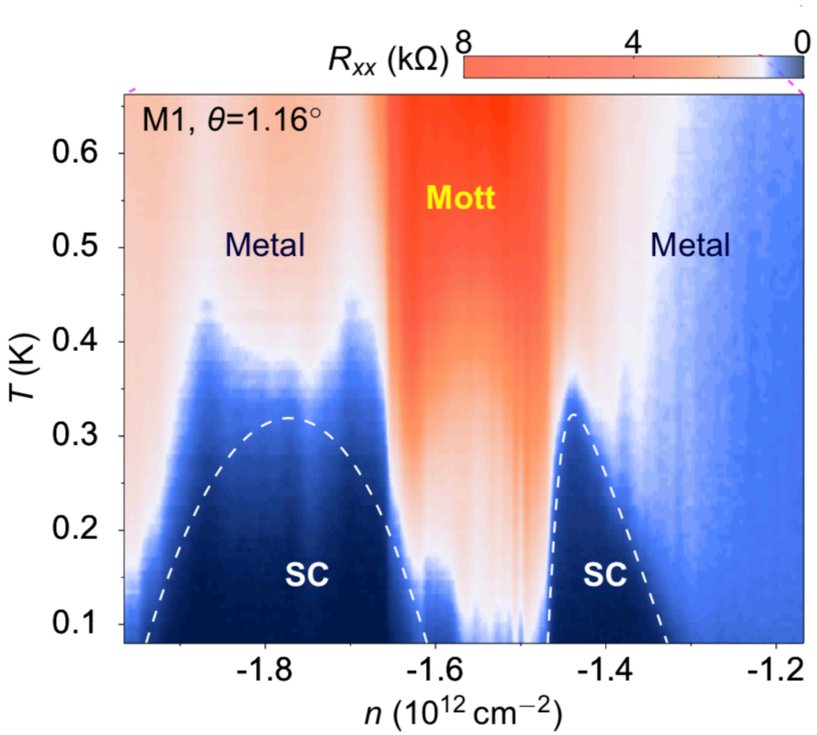
Example: transport in magic-angle graphene



data: Efetov group

Slowing down electrons in graphene using moiré engineering

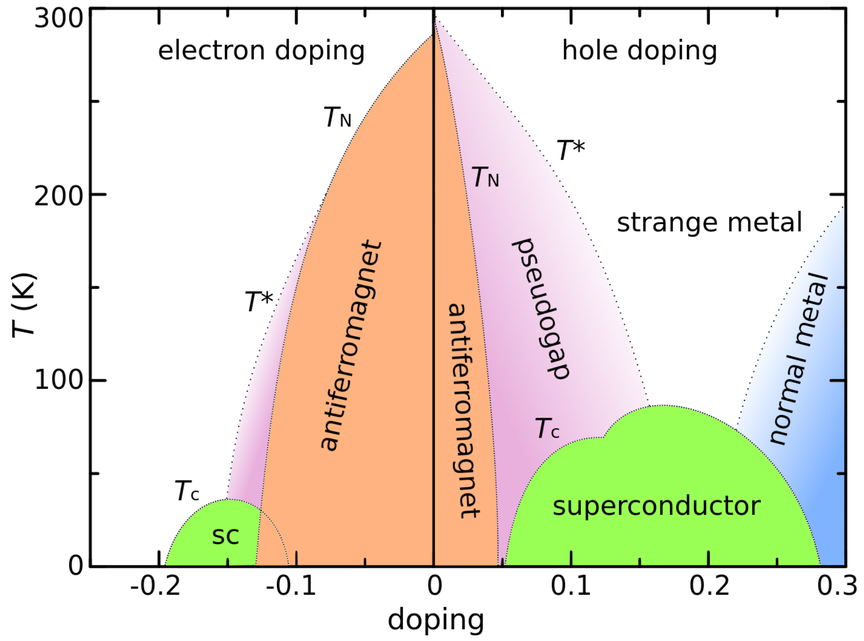
Magic angle twisted bilayer graphene



Back-gate tuneable!

Cao et. al., *Nature* **556**, 43-50 (2018)
Cao et. al., *Nature* **556**, 80-84 (2018)

Copper-oxide high-T_c superconductors



C-L Song et. al., *Science* **332**, 6036 (2011)

Emergent quantum phases in moiré materials

Graphene-based

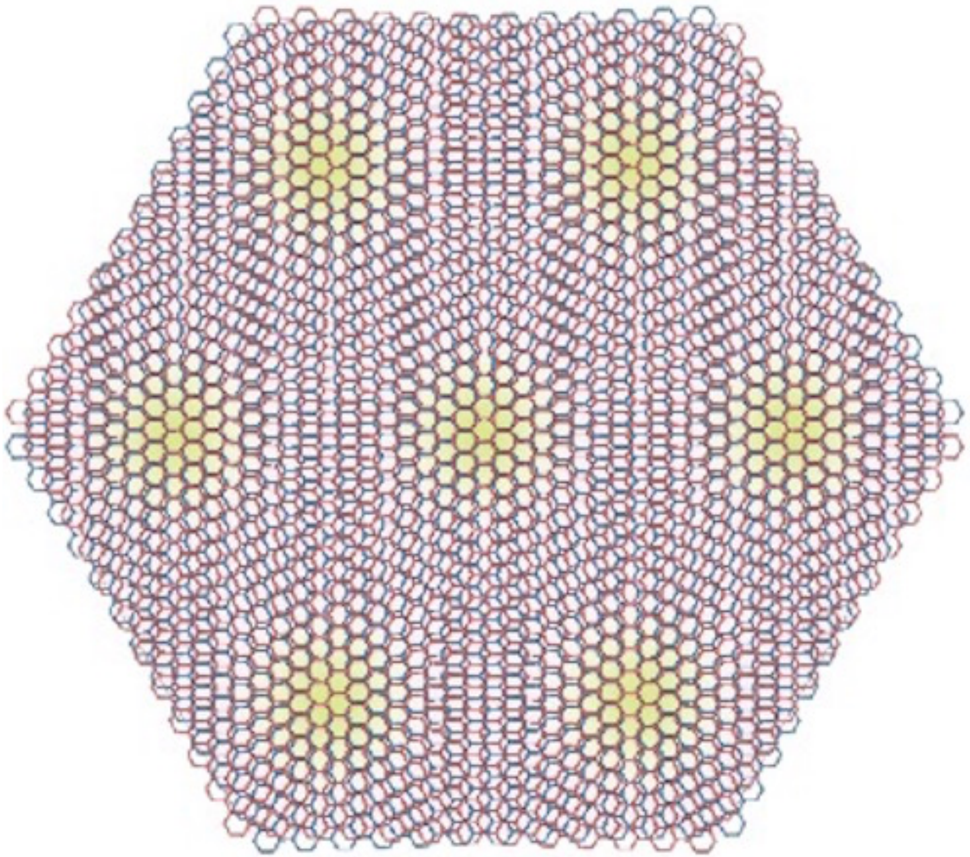
- **Magic angle twisted bilayer graphene**: (Unconventional) superconductivity, orbital magnetism, Chern insulators, correlated insulators, nematic orders, strange metal phase.
- **Magic angle twisted trilayer graphene**: Spin-triplet superconductivity.
- **Twisted double-bilayer graphene**: Tunable spin-polarizes correlated insulators, ferromagnetic metal.
- **Twisted mono-bilayer graphene**: Zero-field orbital Chern insulators.
- **ABC Trilayer aligned with hBN**: Correlated insulators, superconductivity.

Semiconductor-based

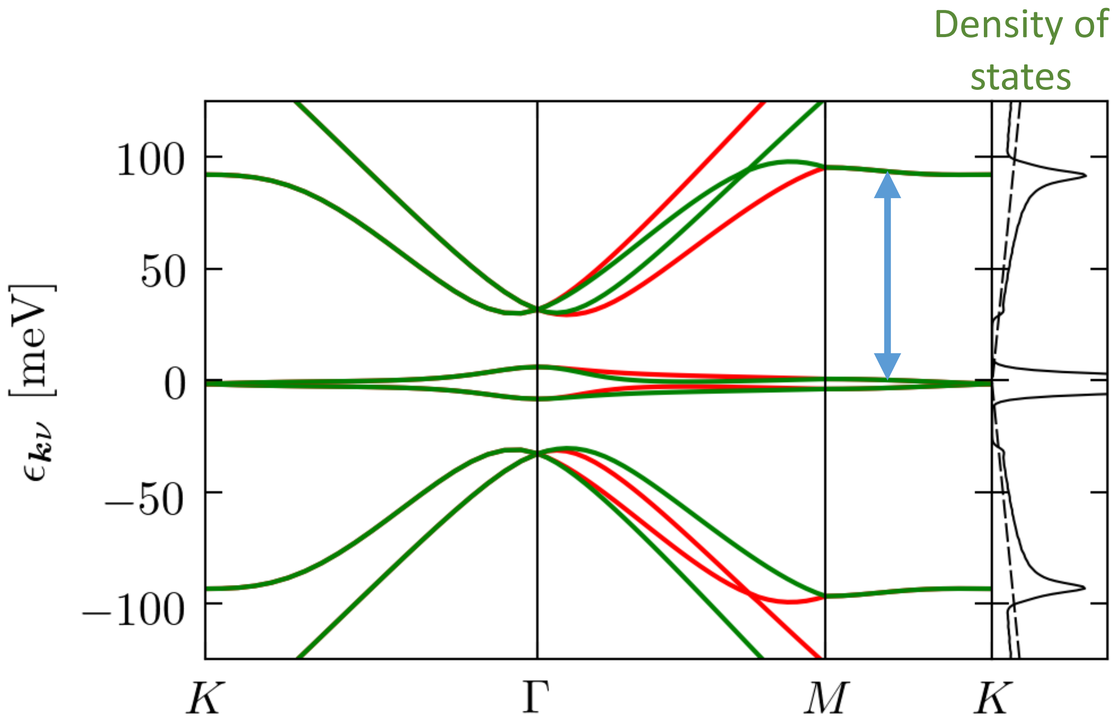
- **Twisted 2D semiconductors**: Wigner crystal, Mott insulator.
- **Twisted WTe2**: Luttinger liquid.
- …

Optical transitions of magic angle graphene

Twist angle ~1.3°
Moire period ~15nm



Accessible with infrared light



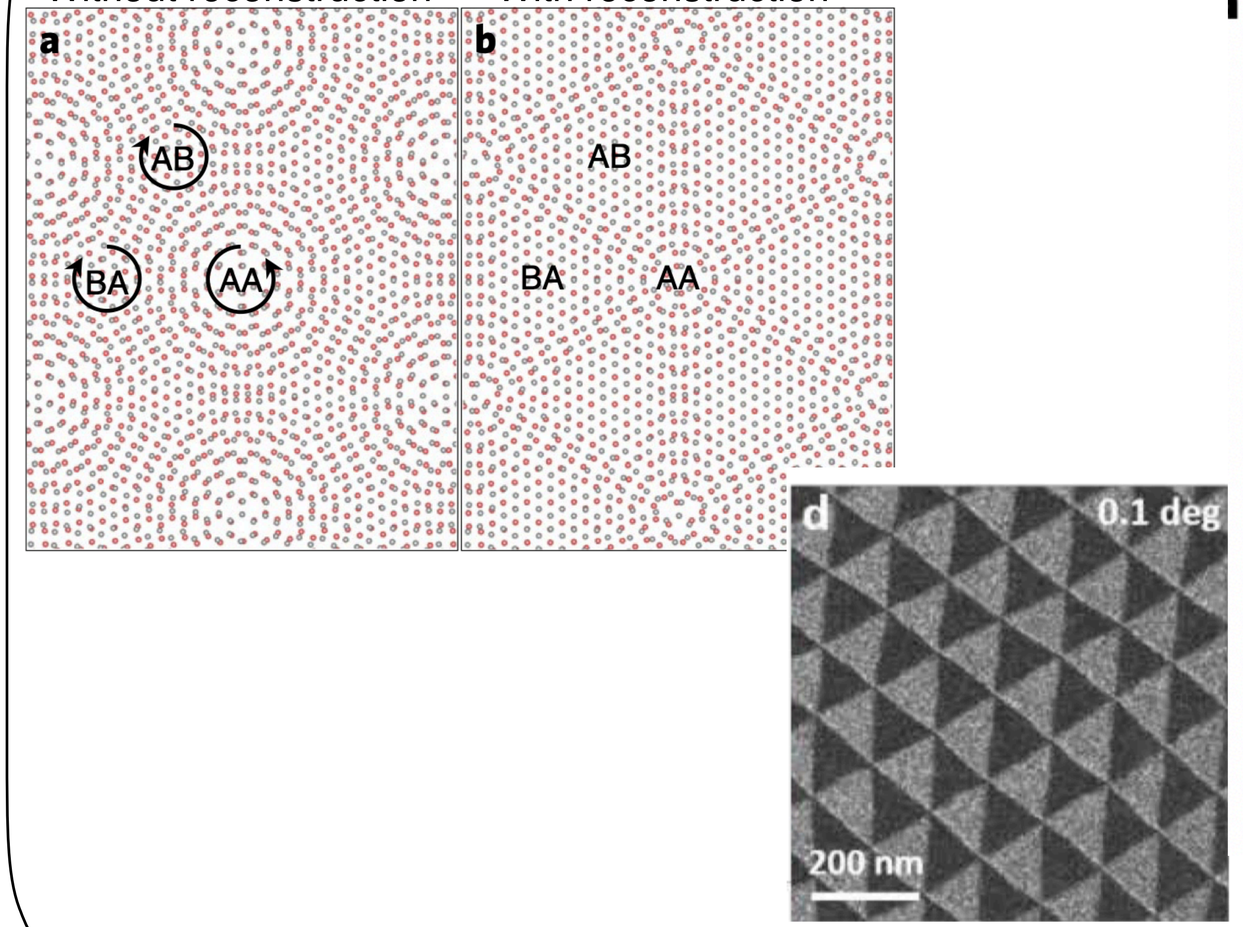
~100meV

P. Novelli, I. Torre, F.H.L. Koppens, F. Taddei, and M. Polini, Phys. Rev.)

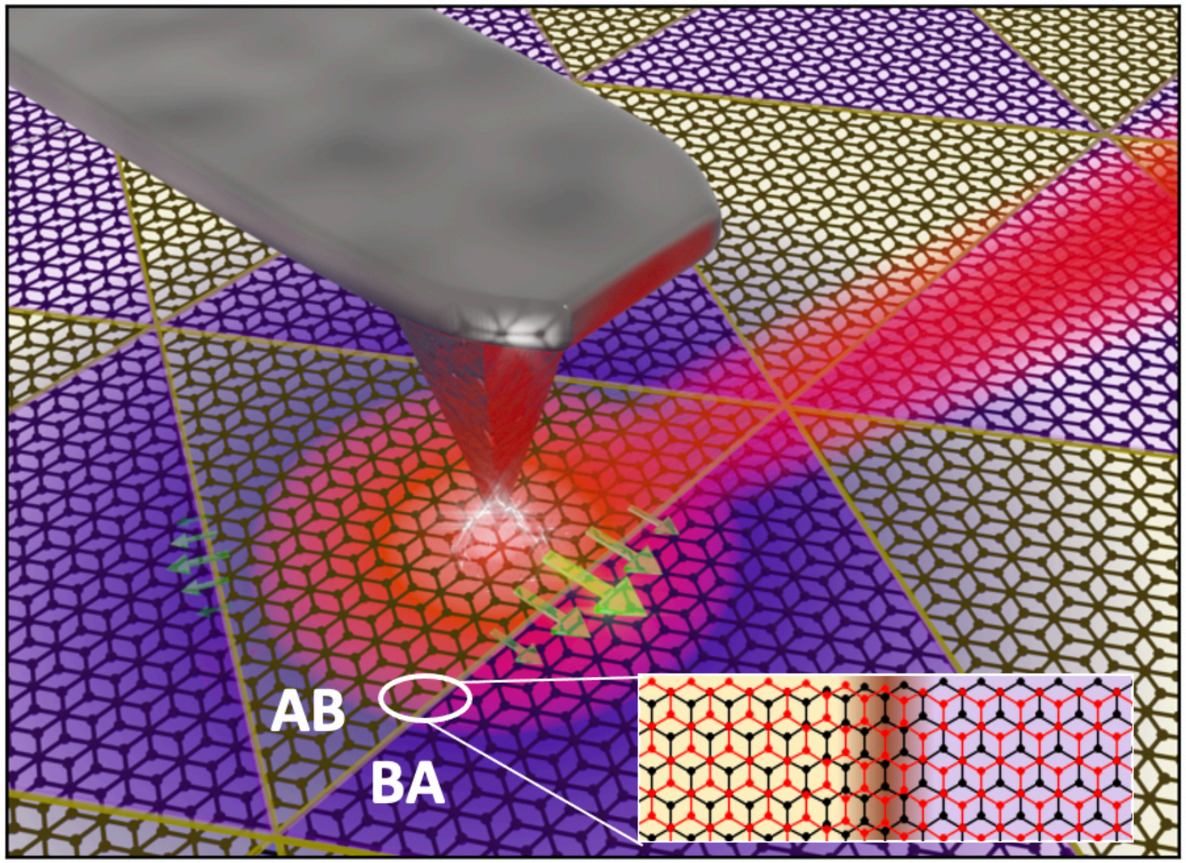
Probing photo-thermoelectric response inside moiré unit cell

Angle < 0.2°
Moire > 70 nm

Without reconstruction With reconstruction

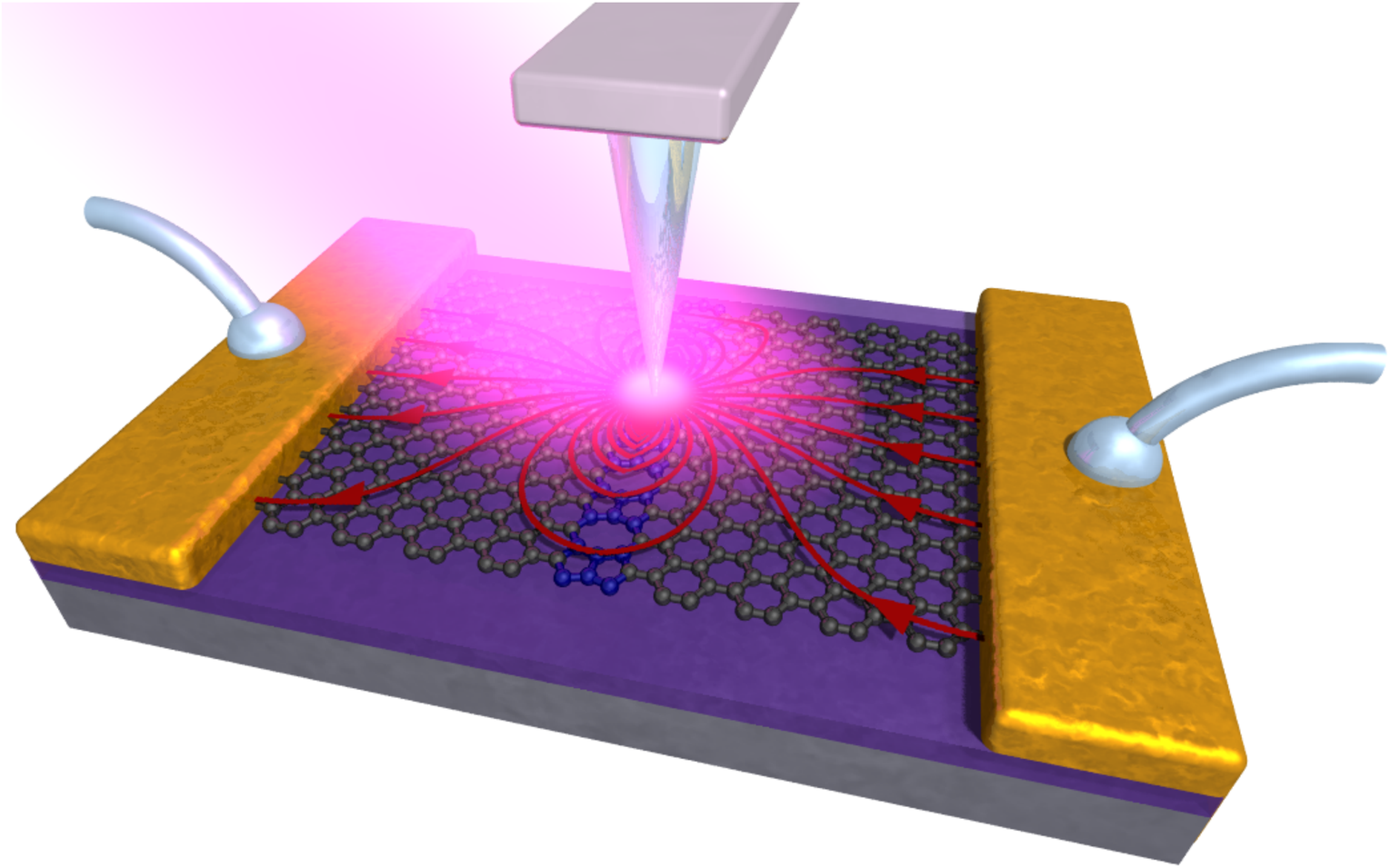


Yoo et al., Nat. Materials 2019



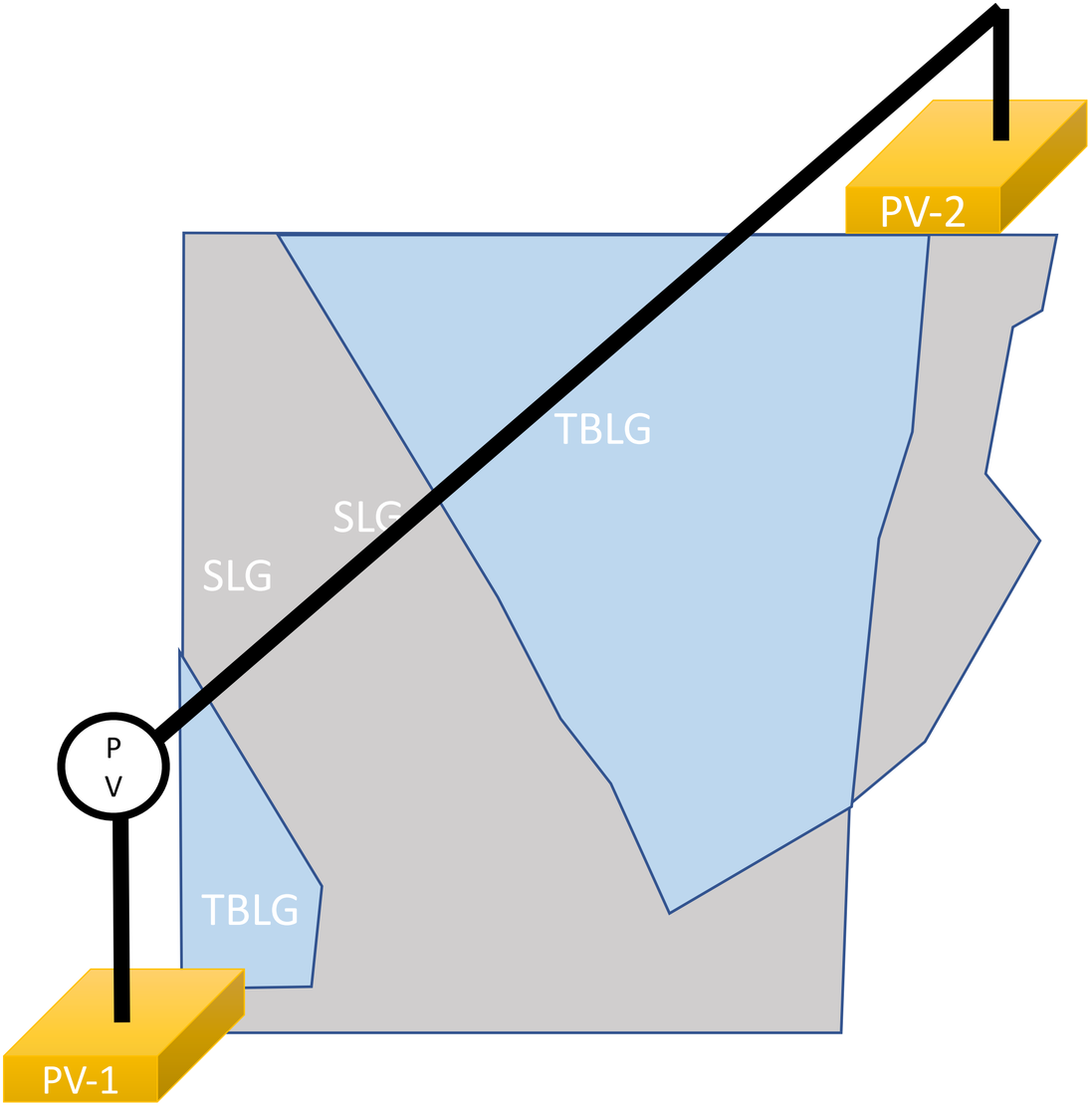
Hesp et al., Nature Communications 2021
Sunku et al. (Basov group)., Nature Communications 2021

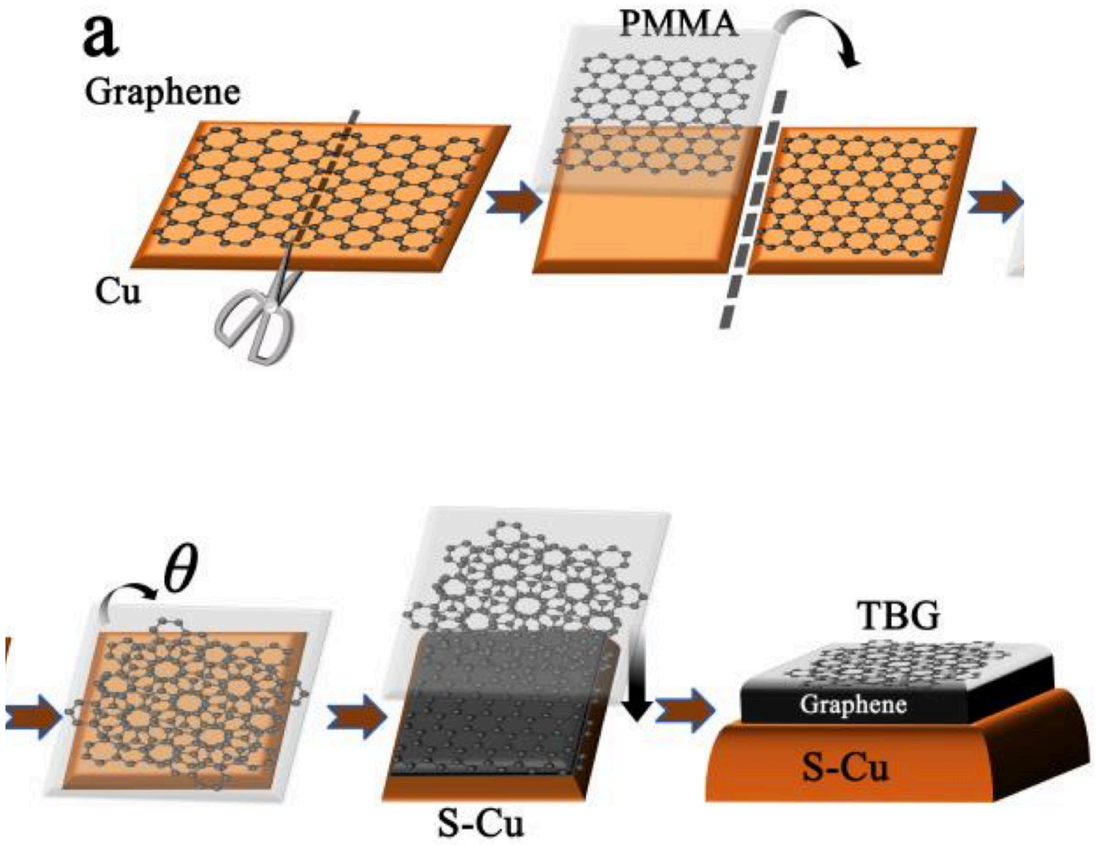
Small-angle twisted graphene (<0.1°)



Illumination: infrared light 10.6 um (116 meV)

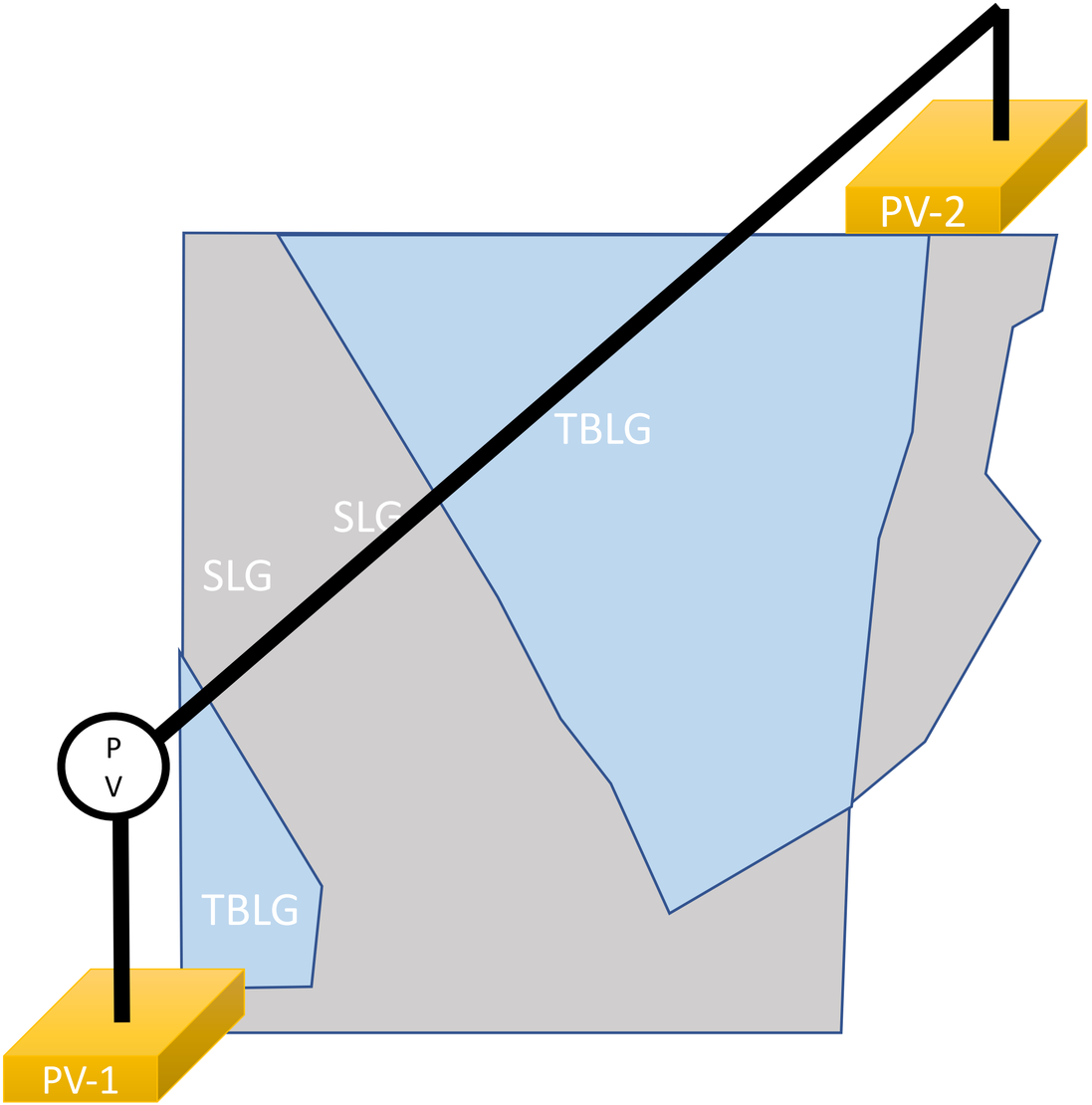
Near-field photocurrent: Small-angle twisted graphene (<0.1°)

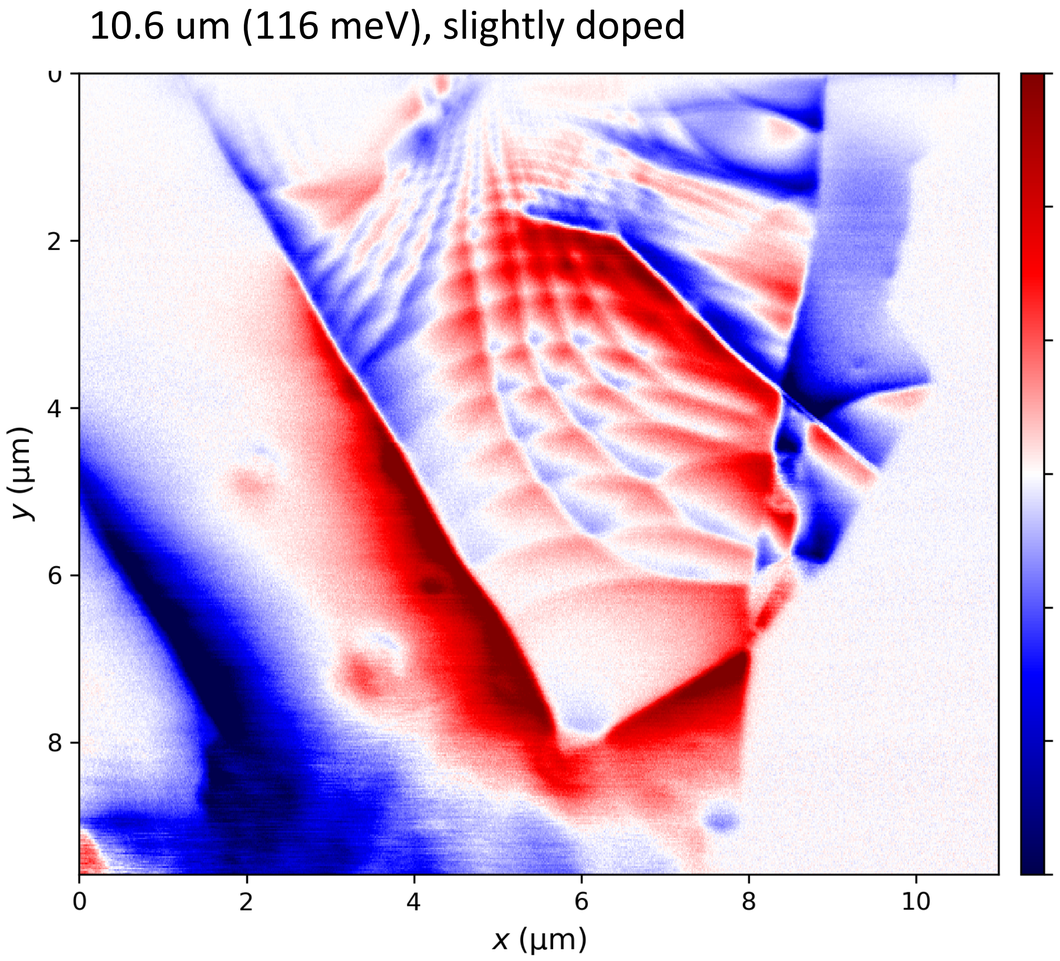




Hesp et al., Arxiv 2011.05060. Nature Communications 2021

Near-field photocurrent: Small-angle twisted graphene (<0.1°)





Hesp et al., Arxiv 2011.05060. Nature Communications 2021

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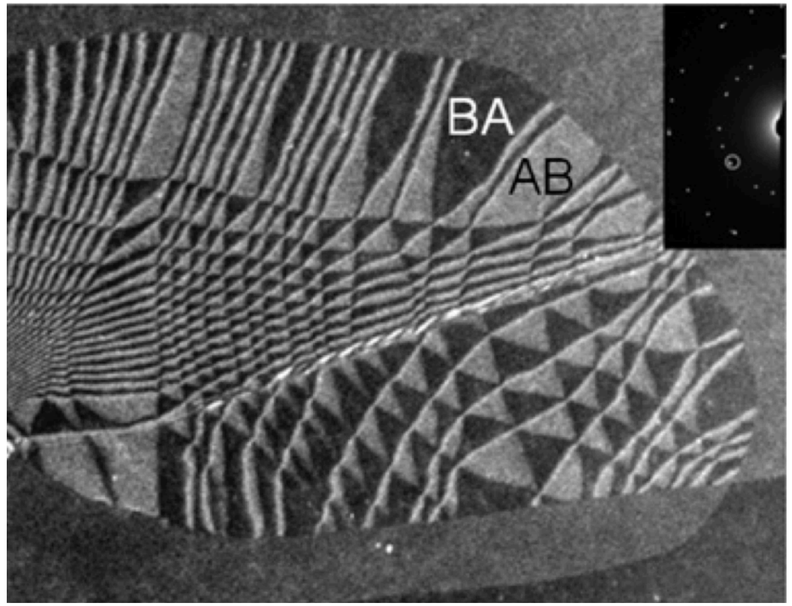

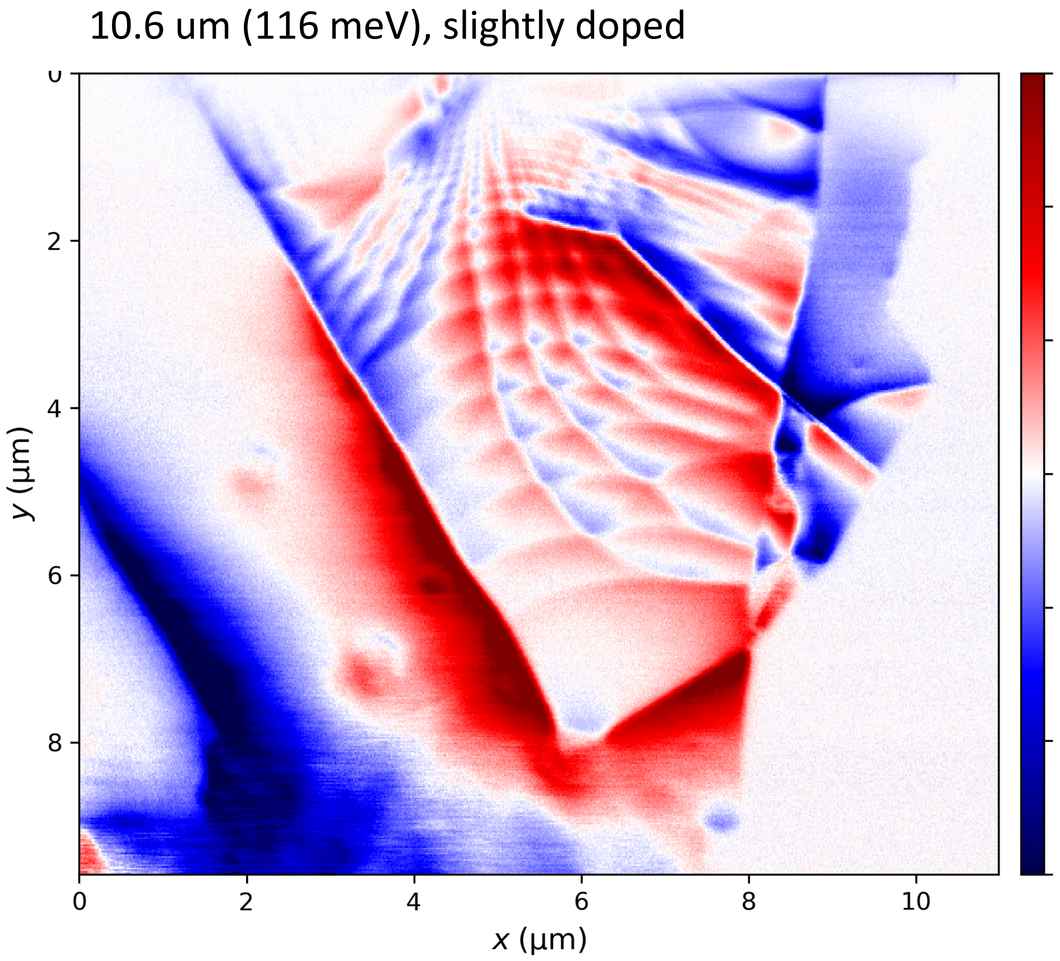

Hesp et al., Arxiv 2011.05060. Nature Communications 2021

Photo-thermoelectric model

Thermo-electric field E_T:

$$E_T = S\nabla T$$

Current generated:

$$I = \frac{1}{R}\int_0^L E_T dx$$

$$\mathcal{R}(x) = -\frac{1}{\kappa W}\int dx' \frac{L_{\text{cool}}}{2} e^{-\frac{|x'-x|}{L_{\text{cool}}}} \partial_x S(x')$$

Photo-thermoelectric model

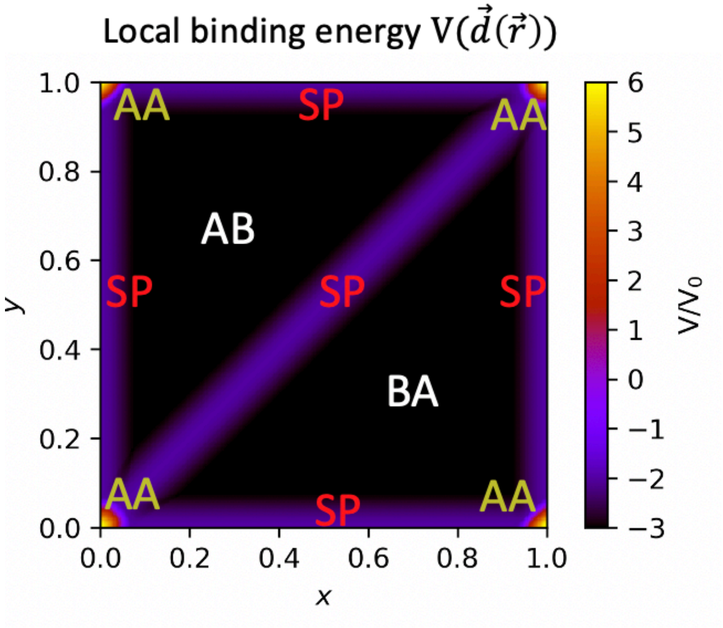

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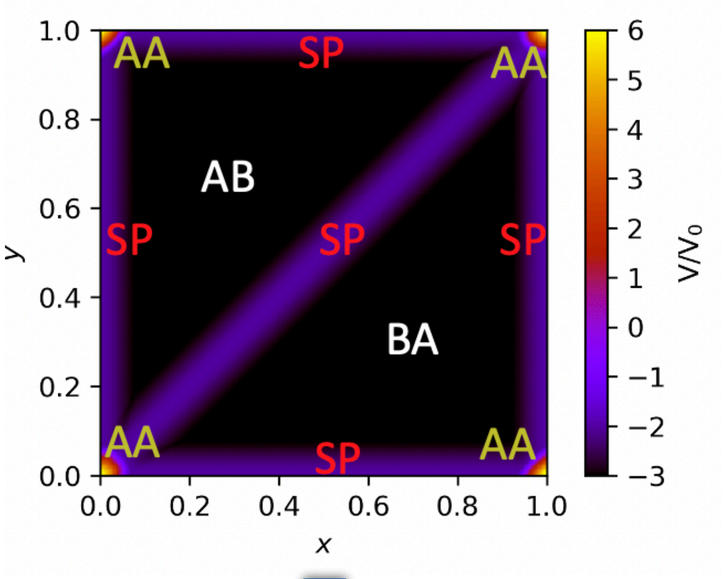
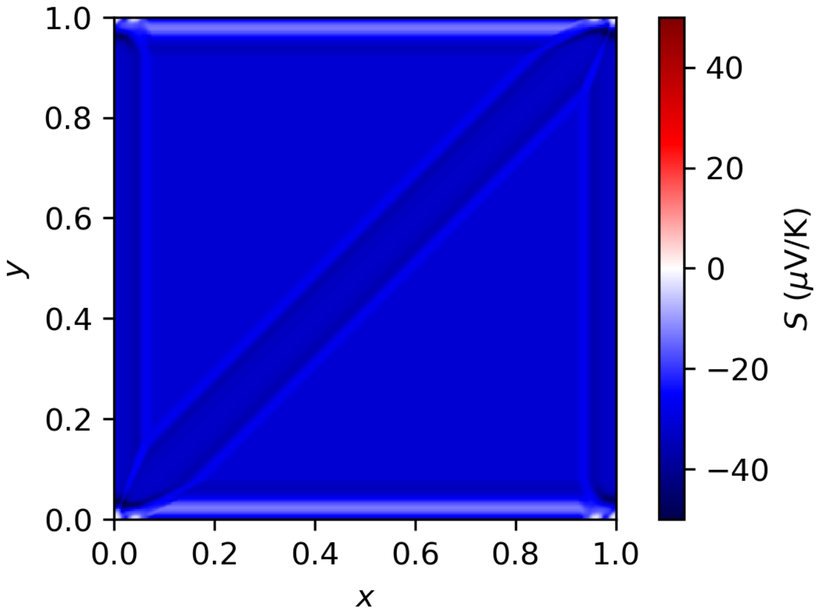
Photo-thermoelectric model

Local binding energy $V(\vec{d}(\vec{r}))$



Seebeck $S(\vec{d}(\vec{r}))$



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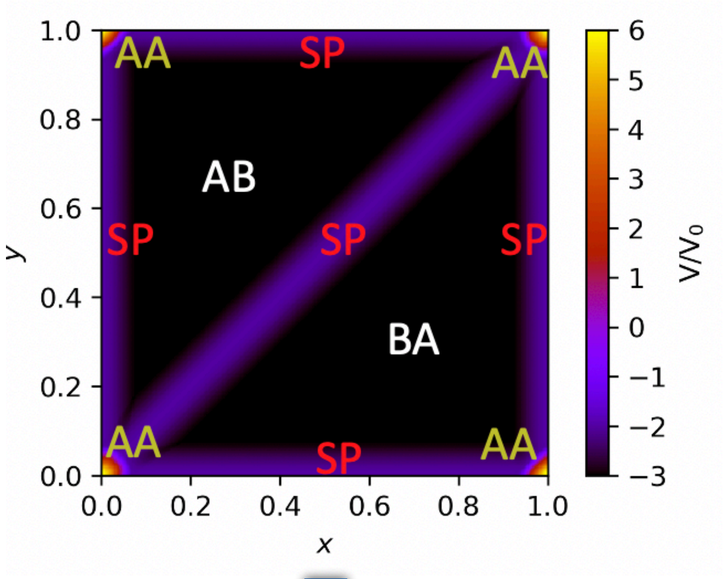

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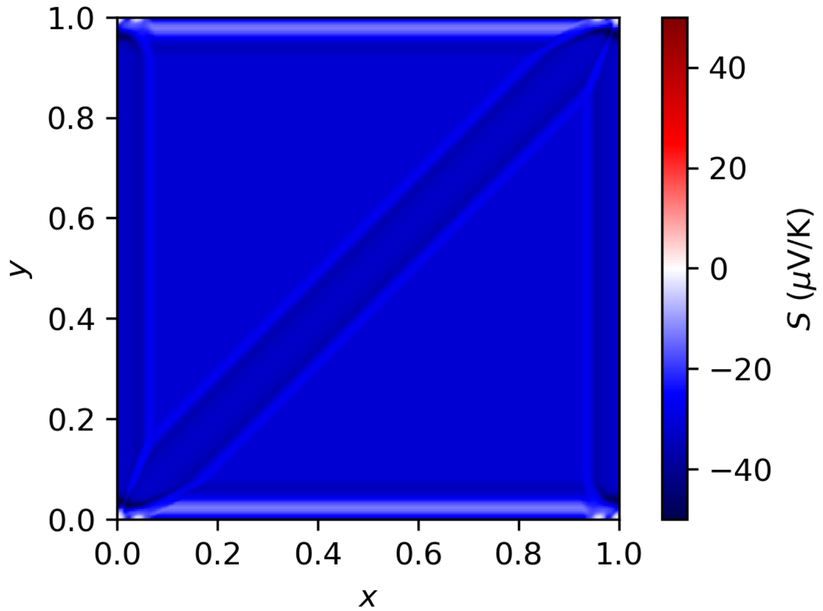

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Seebeck coefficient

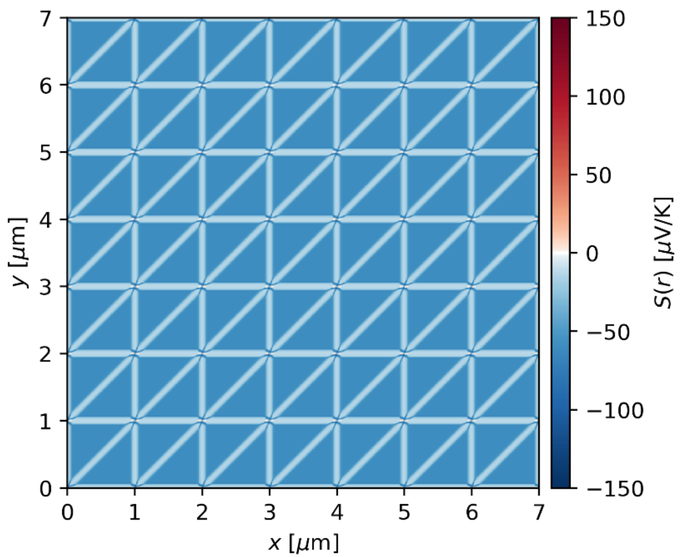
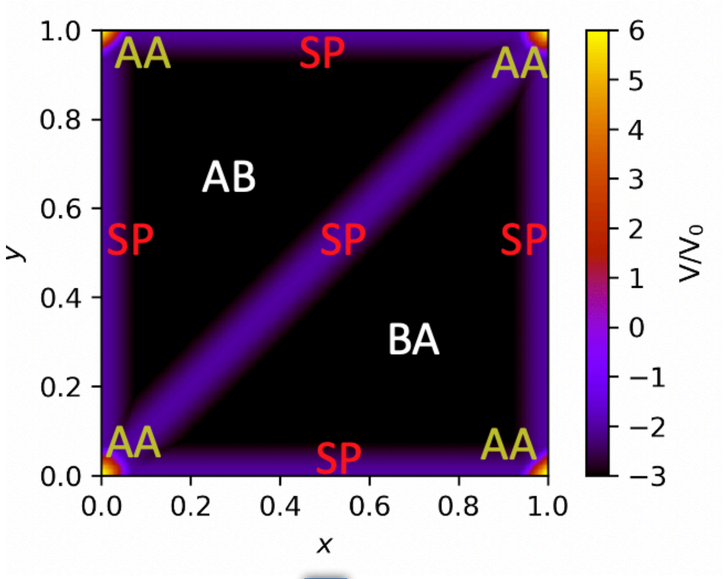


Photo-thermoelectric model

Local binding energy $V(\vec{d}(\vec{r}))$



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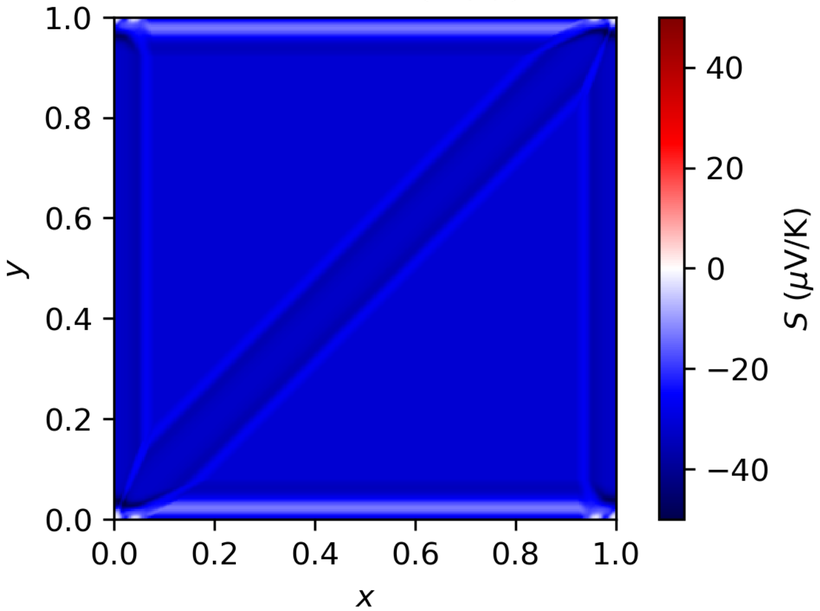
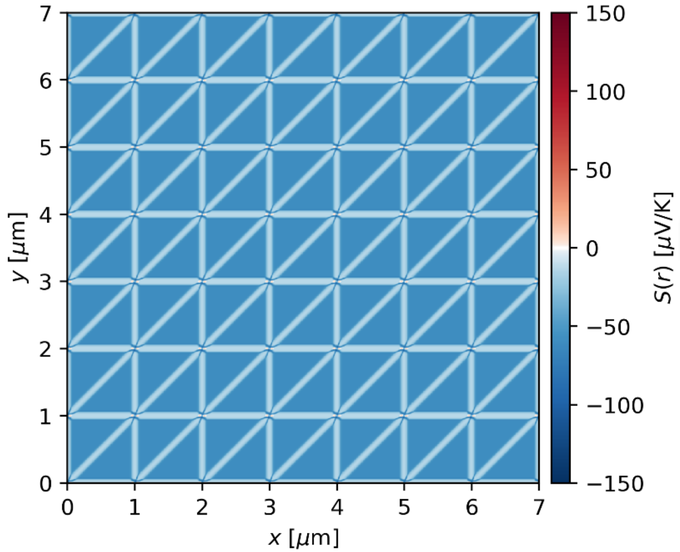
Seebeck $S(\vec{d}(\vec{r}))$



Seebeck coefficient



Photoresponsivity

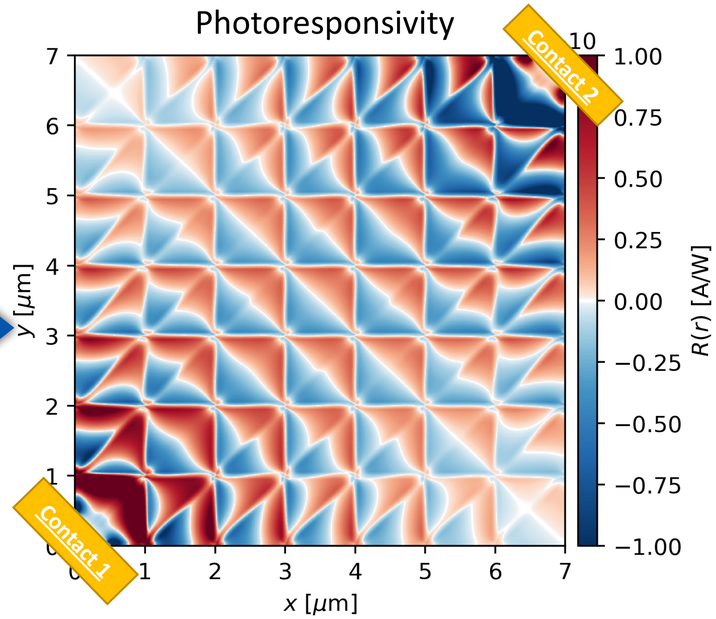
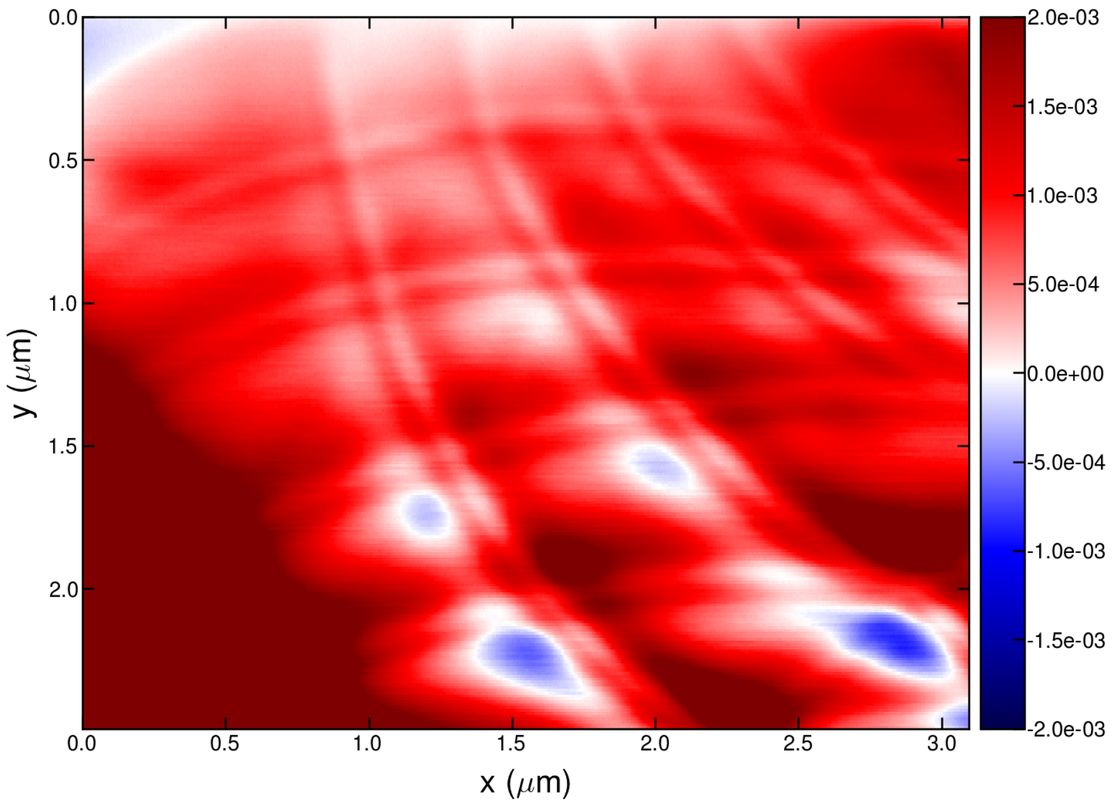
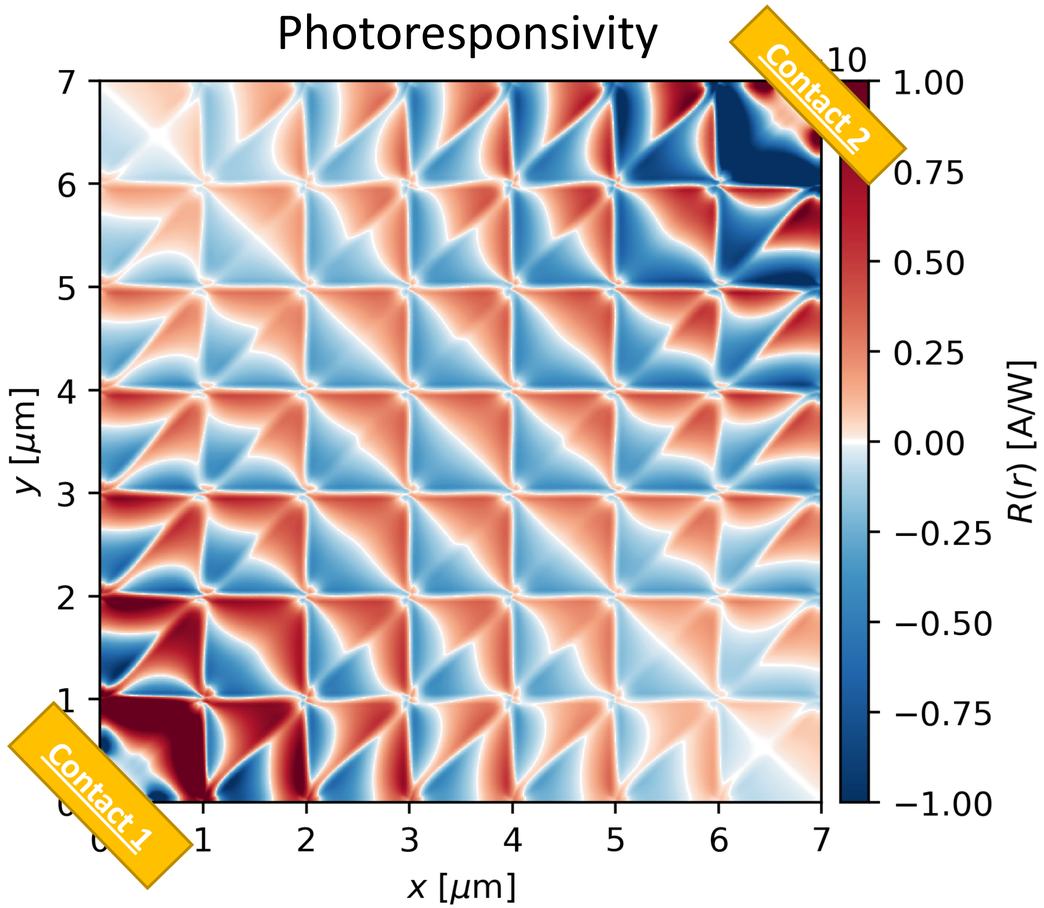


Photo-thermoelectric model

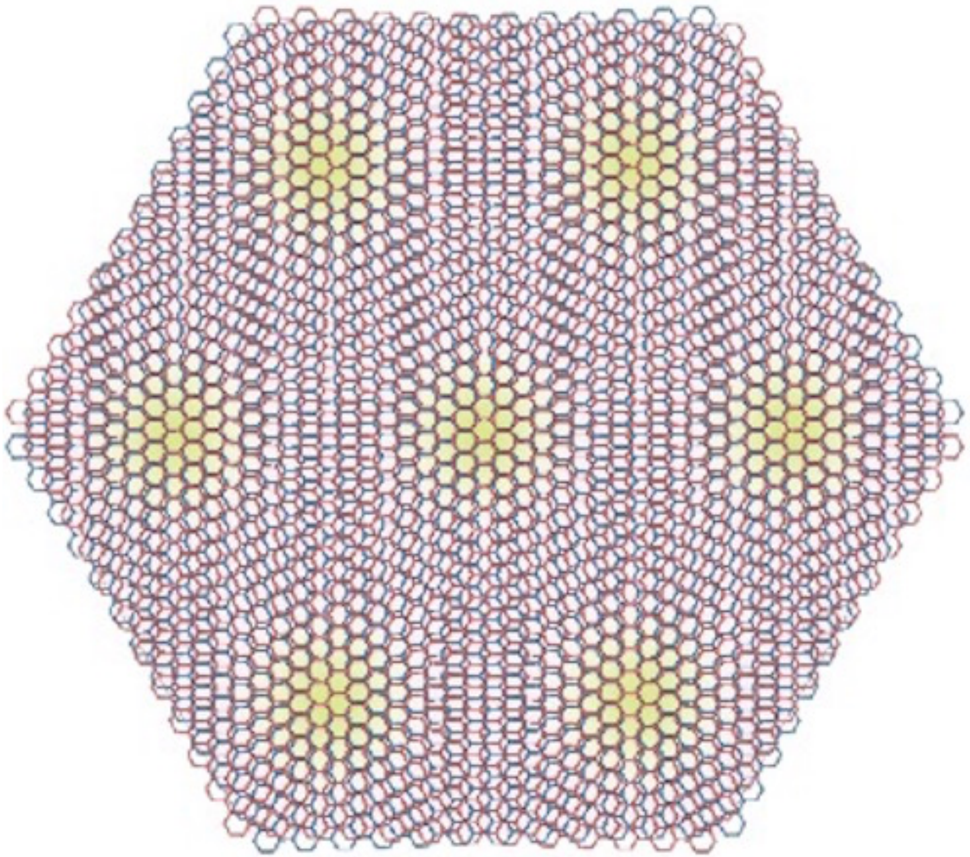




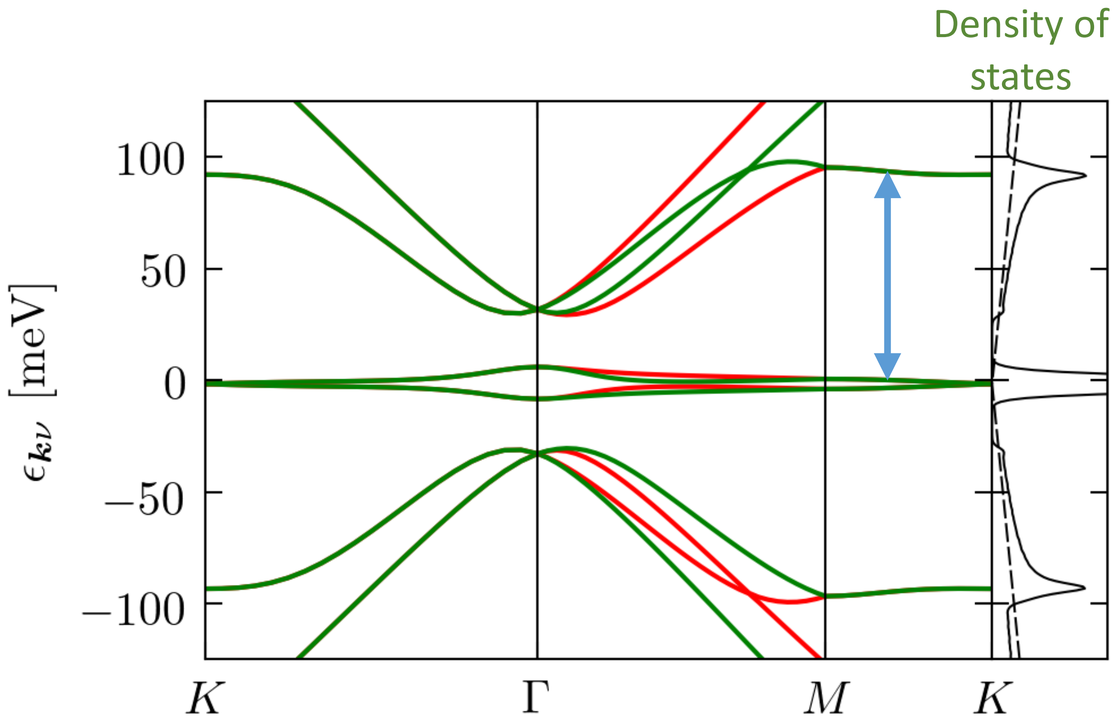
Hesp et al., Arxiv 2011.05060. Nature Communications 2021

Optical transitions of magic angle graphene

Twist angle ~1.3°
Moire period ~15nm



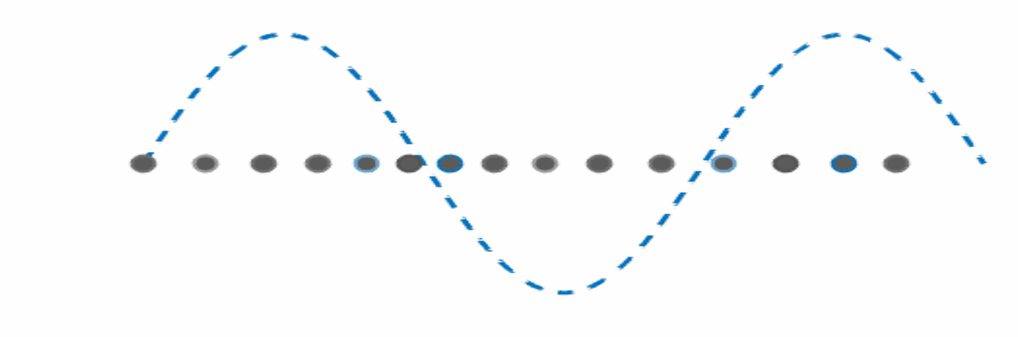
Accessible with infrared light



~100meV

P. Novelli, I. Torre, F.H.L. Koppens, F. Taddei, and M. Polini, Phys. Rev.)

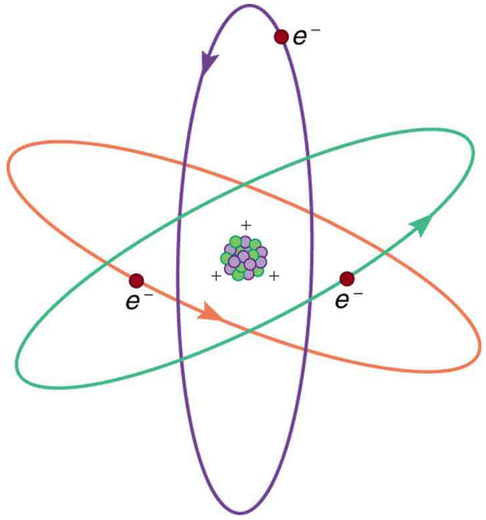
Intraband plasmons: Free electrons
(e.g. metals, doped graphene, etc.)



Optical Energy range 0.01 - 3eV
(Visible, Infraed, Terahertz)

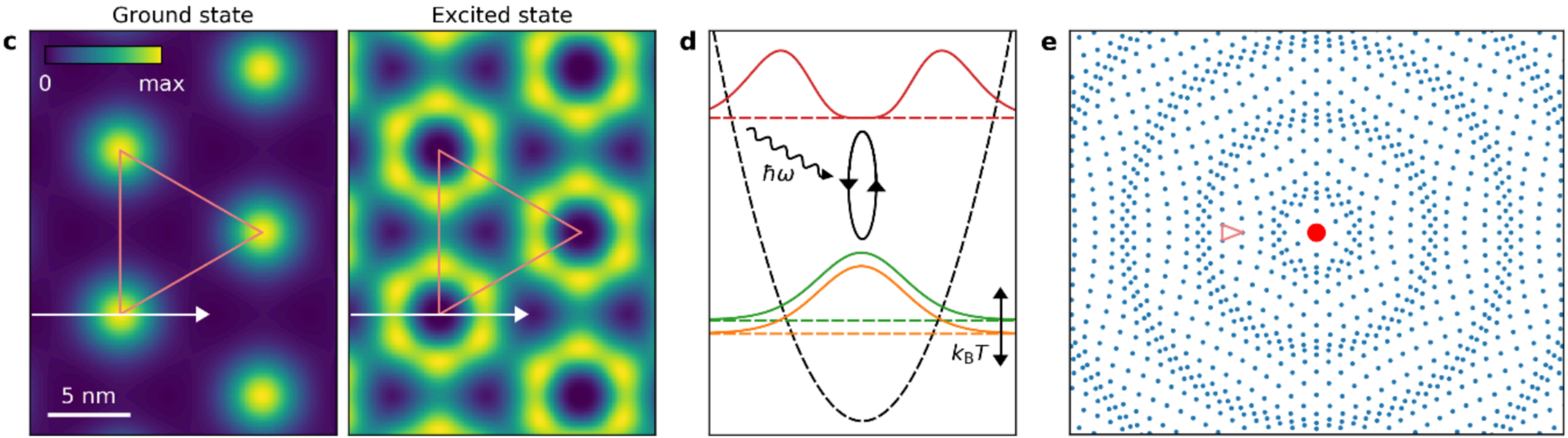
Wavelength ~ PhotonWavelength / 200

Interband plasmons: Bound electrons



Higher energy range 3-15 eV
(UV)

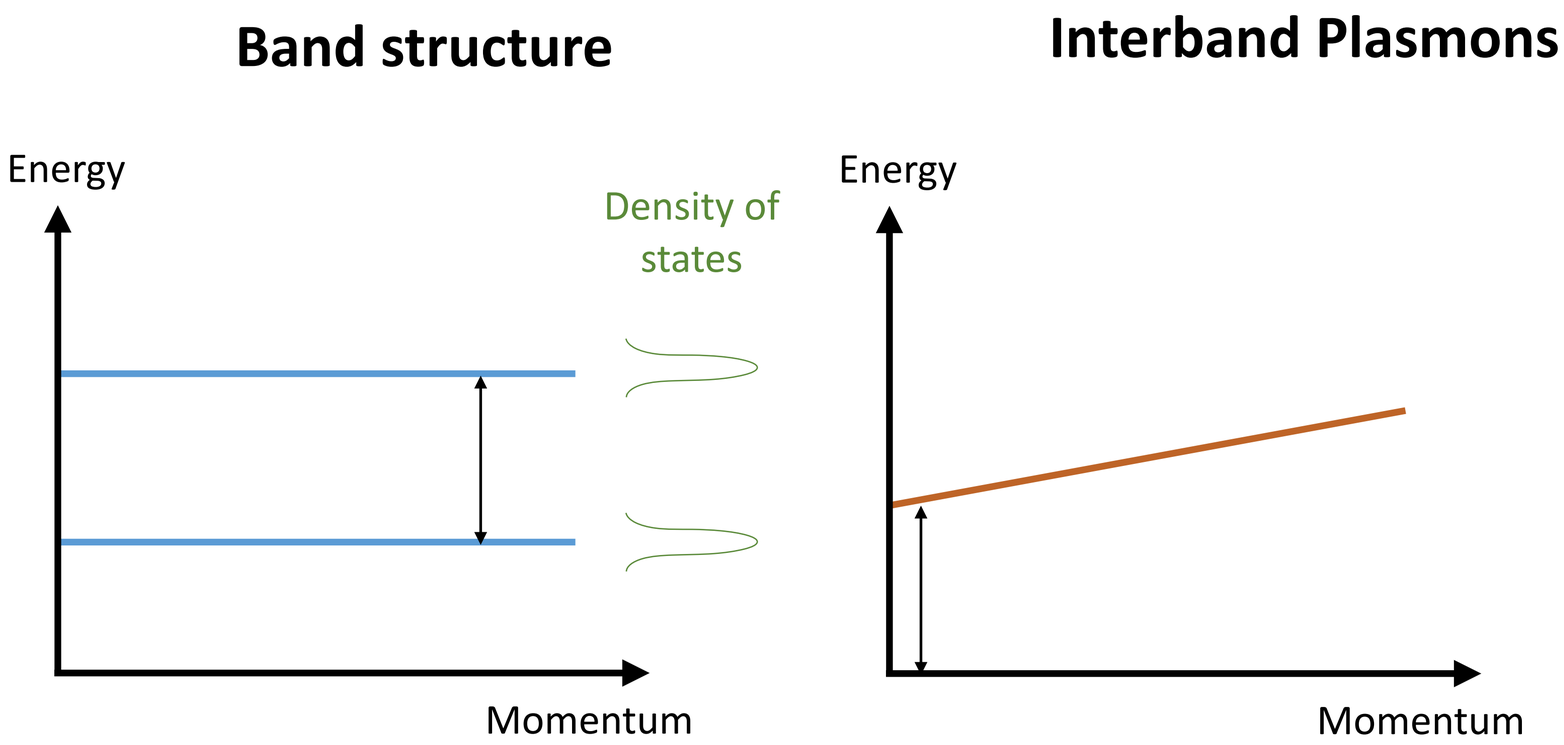
Interband Plasmons in Twisted Graphene



Hesp et al., Arxiv 1910.07893

Theory on interbank plasmons in twisted graphene: Stauber et al, Nano Letters 2016
Optical properties of twisted graphene: Moon, Koshino, PRB 2013

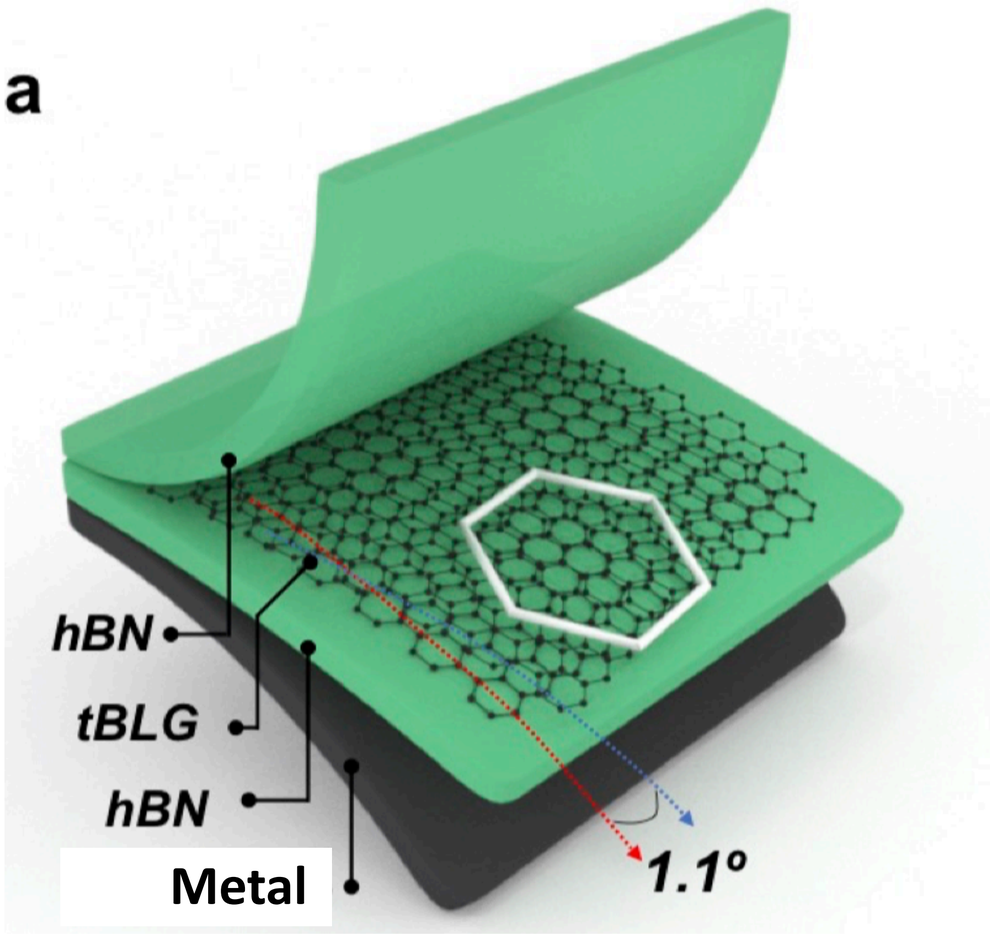
Analogy: quantum Hall bulk magnetoplasmons

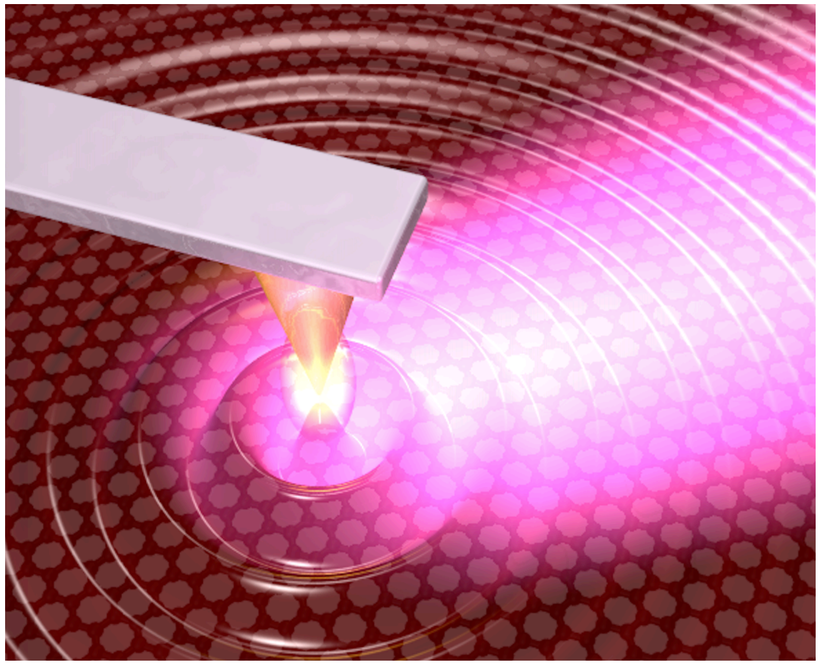


See e.g. Fetter et al., PRB 2015, and many others

s-SNOM on magic angle graphene (undoped!!)

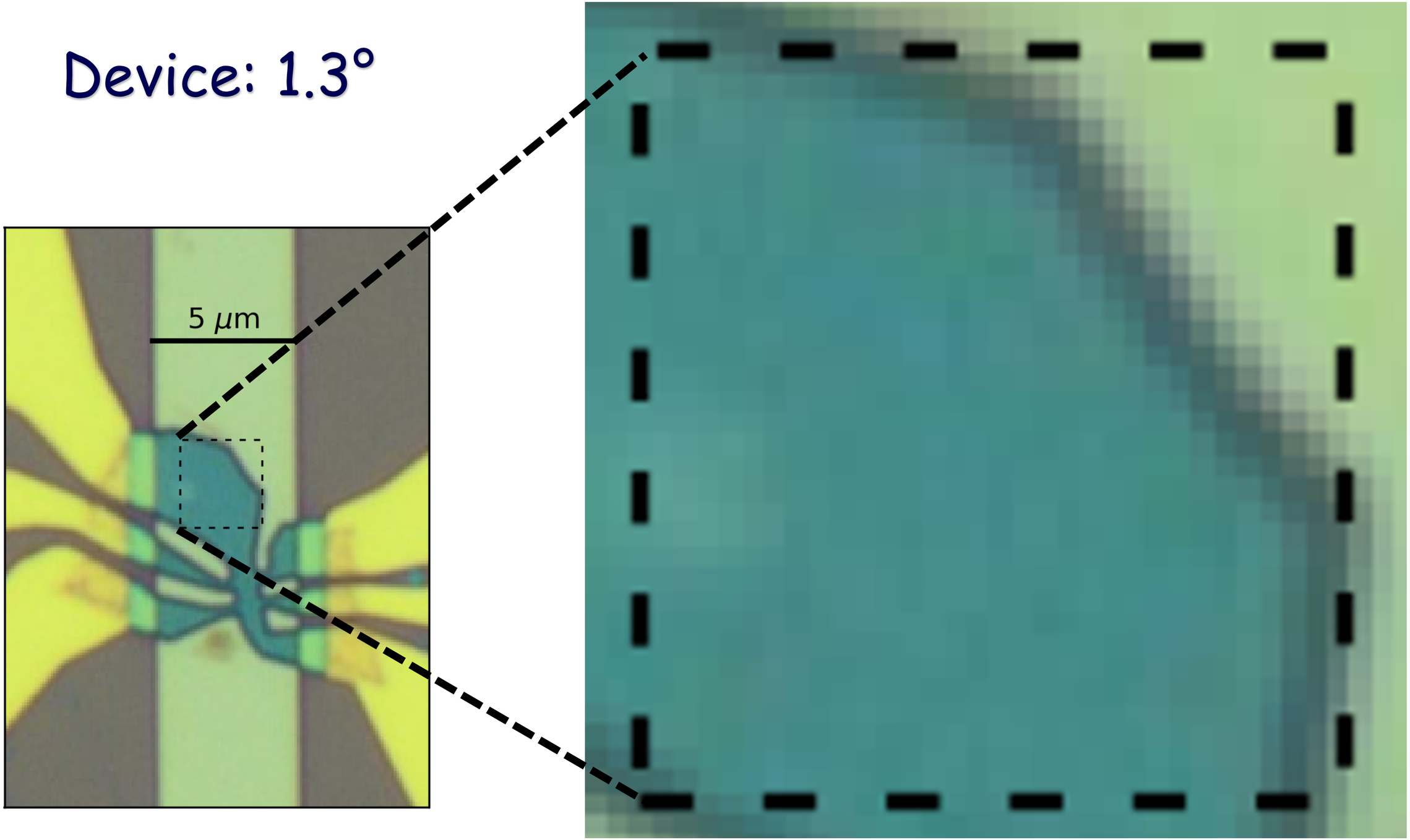
Excitation ~0.2eV (~6μm)
Hot spot under tip ~20nm





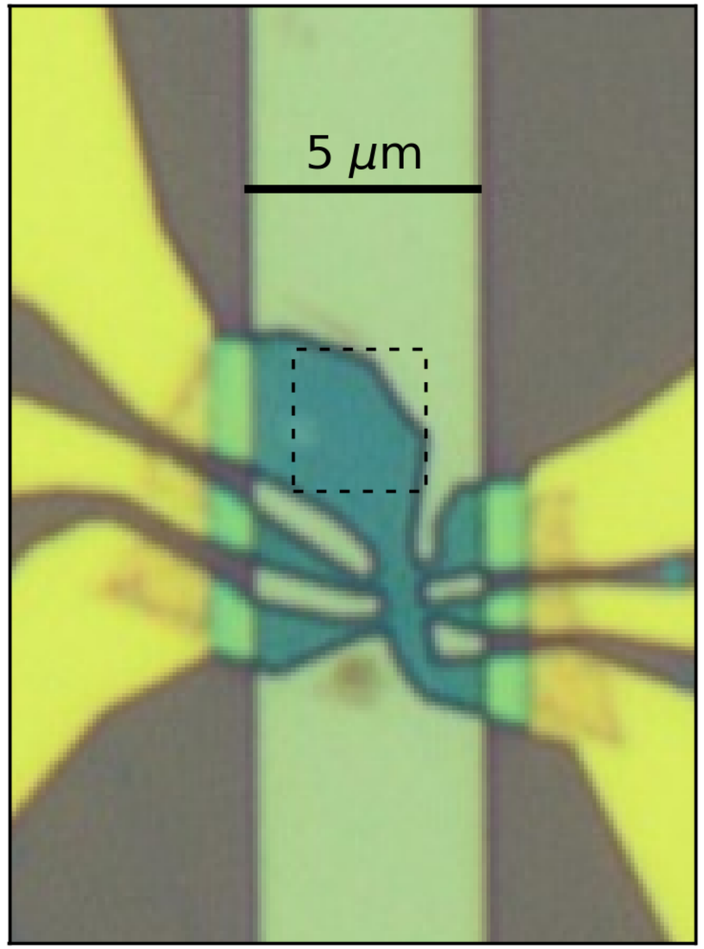
Hesp et al., Nature Physics 2021

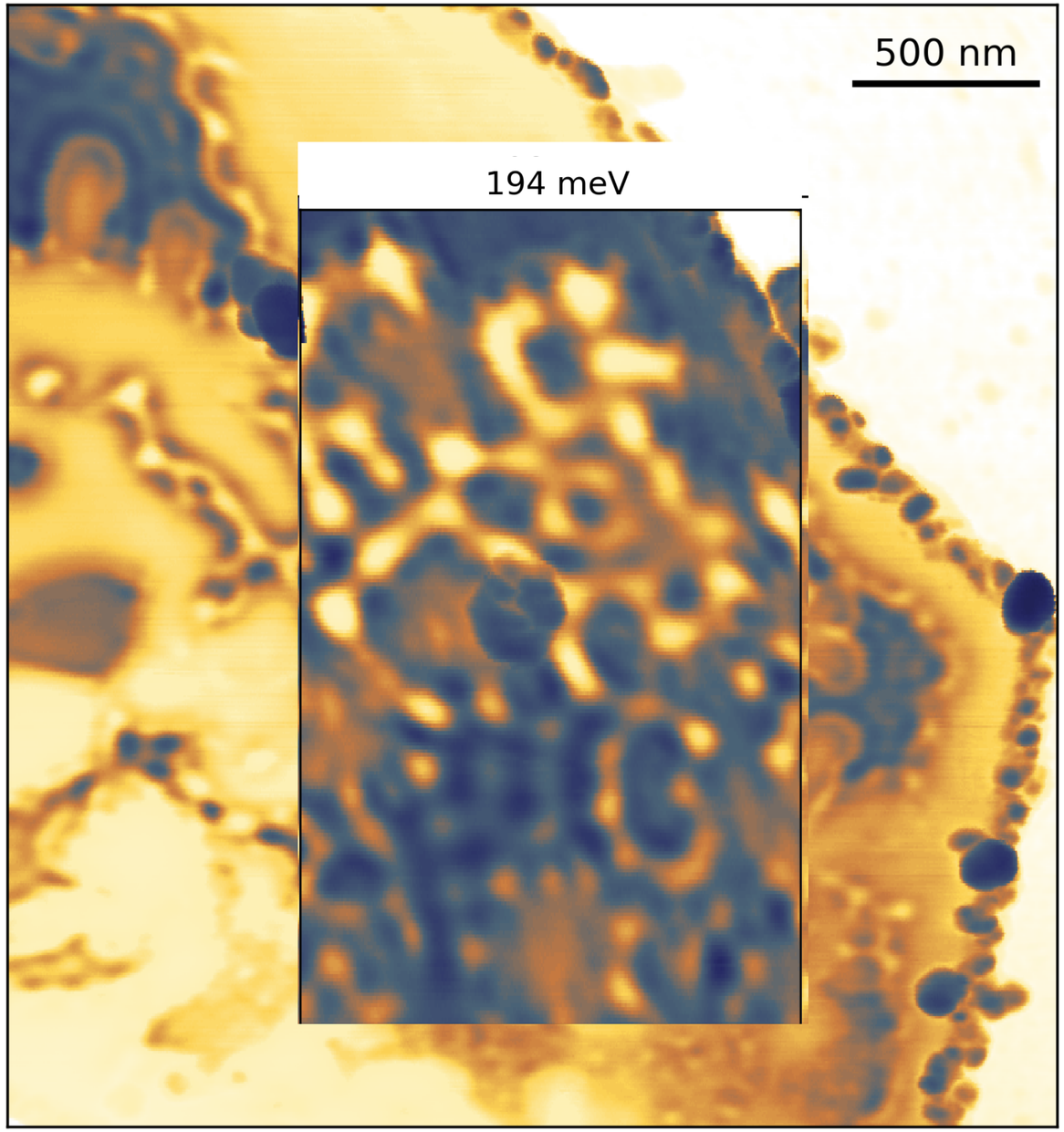
Device: 1.3°



Hesp et al., Nature Physics 2021

Collective excitations: interband plasmons

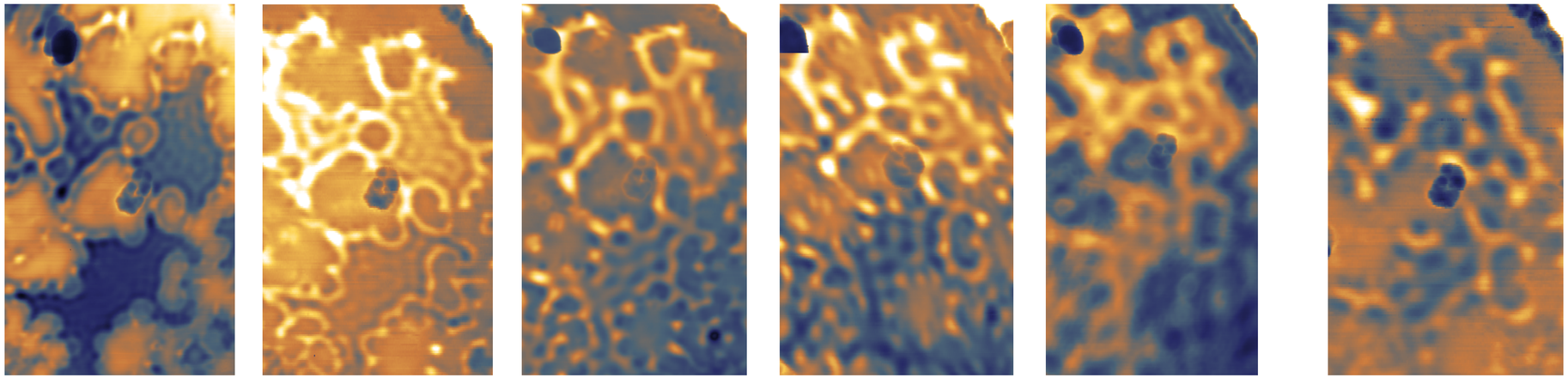


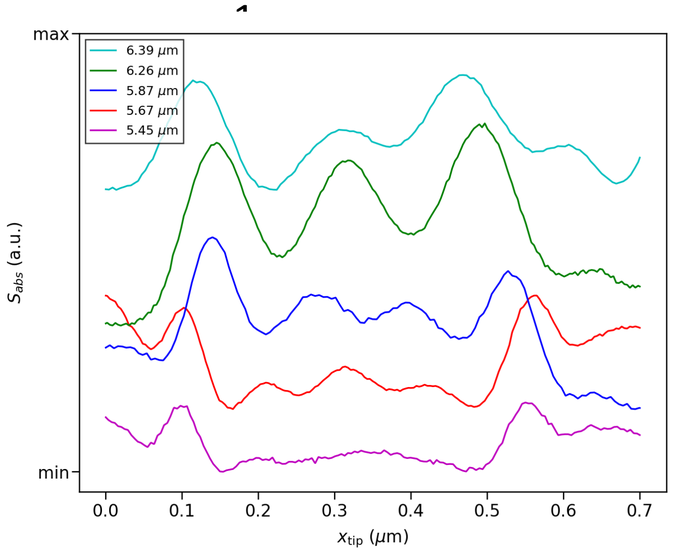


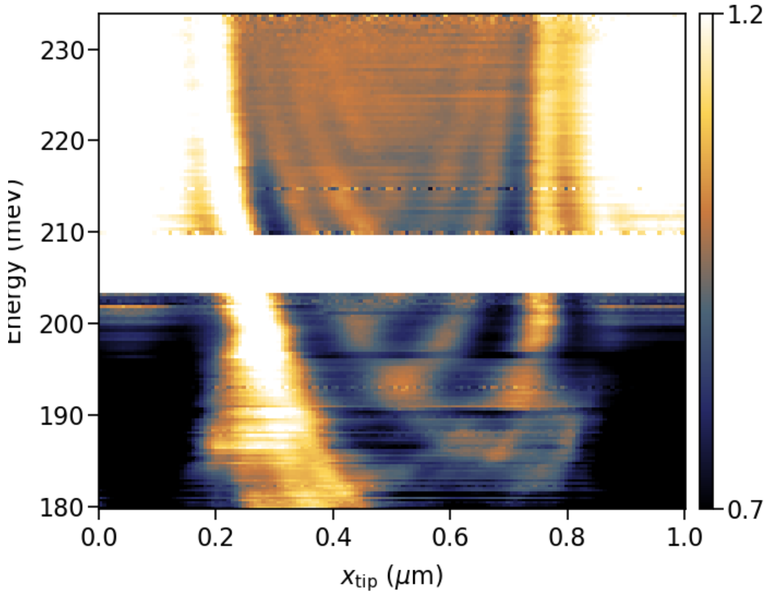
Hesp et al., Nature Physics 2021

Changing wavelength

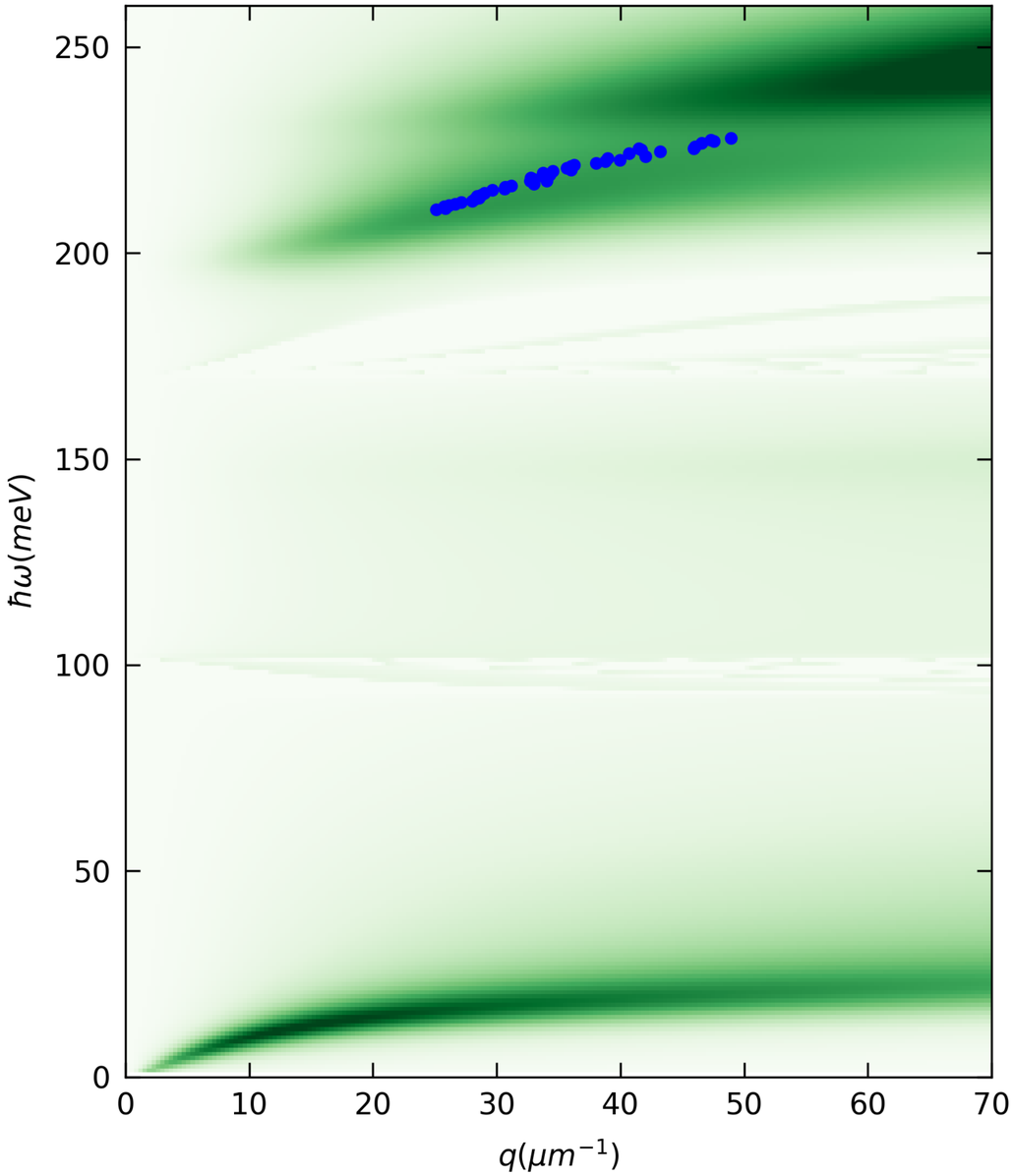
5.67um 5.87um 6.26um 6.39um 6.54um 10.6um







Plasmon dispersion



Theory: P. Novelli, I. Torre, F.H.L. Koppens, F. Taddei, and M. Polini, Phys. Rev. B 2019

Determine optical conductivity of TBG

- Optical conductivity ($\epsilon(\omega, q) = 0$)

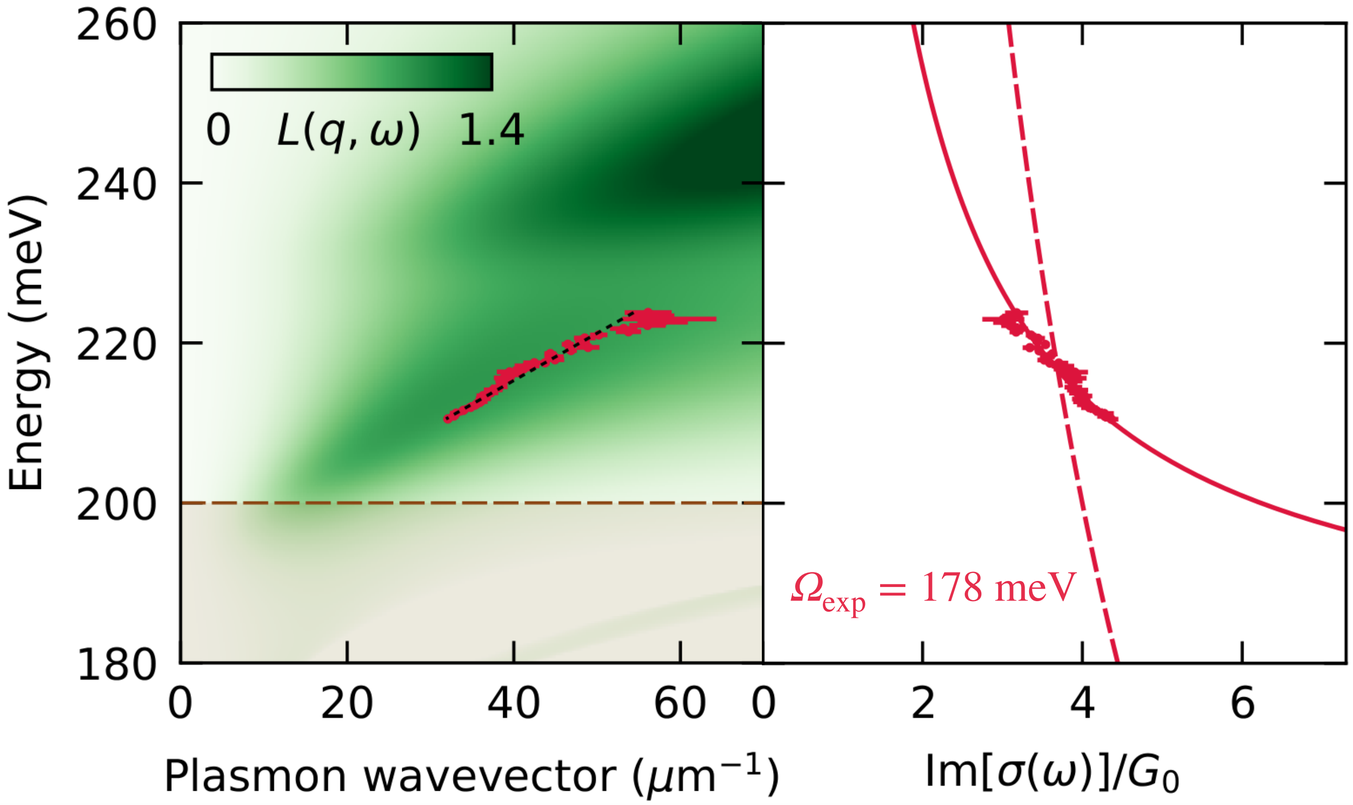
  $\sigma(\omega) = i\omega/(q^2 V_{q,\omega})$

- Drude response

  $\sigma_2(\omega) = G_0 W_0/\hbar\omega$

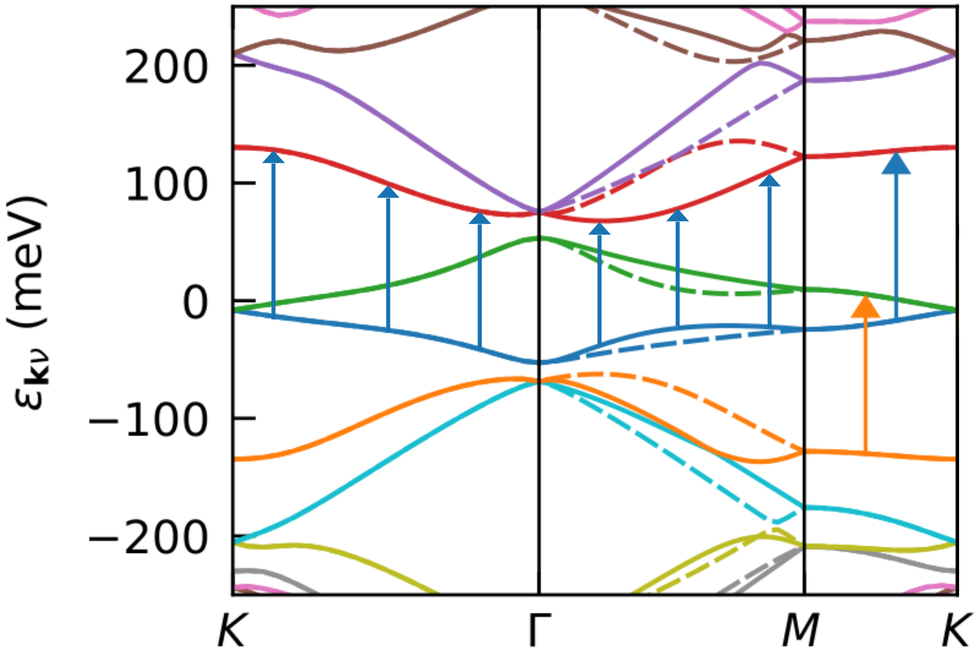
- Interband resonance

  $\sigma_2(\omega) = G_0 W_{\text{exp}} \dfrac{\hbar\omega}{\hbar^2\omega^2 - \hbar^2\Omega_{\text{exp}}^2}$



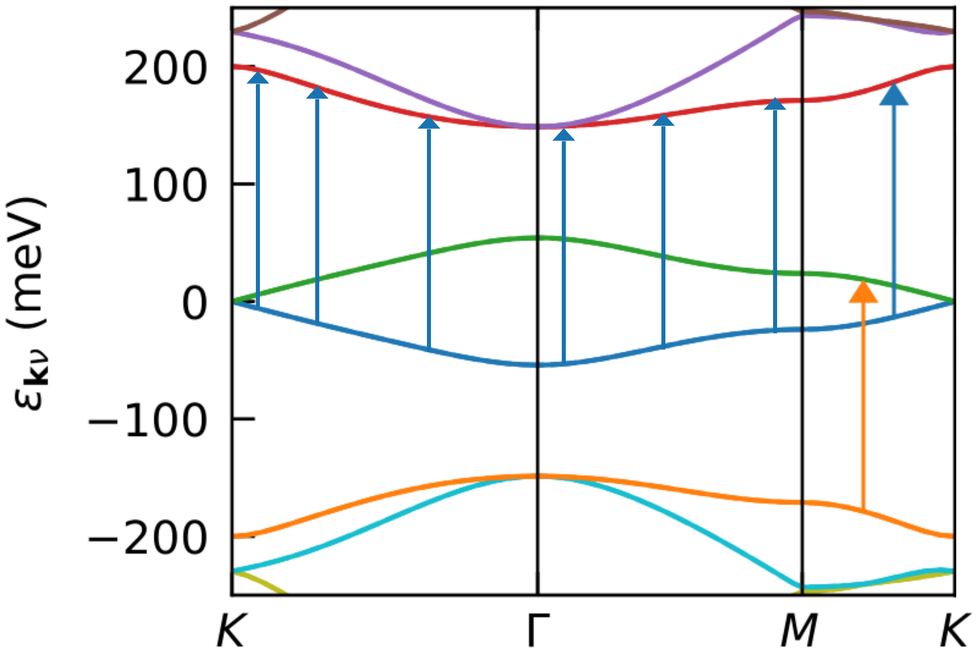
Reduced tunneling enhances band nesting

Typical tunneling parameters



$u_{\mathrm{AA}} = 80$ meV
$u_{\mathrm{AB}} = 100$ meV

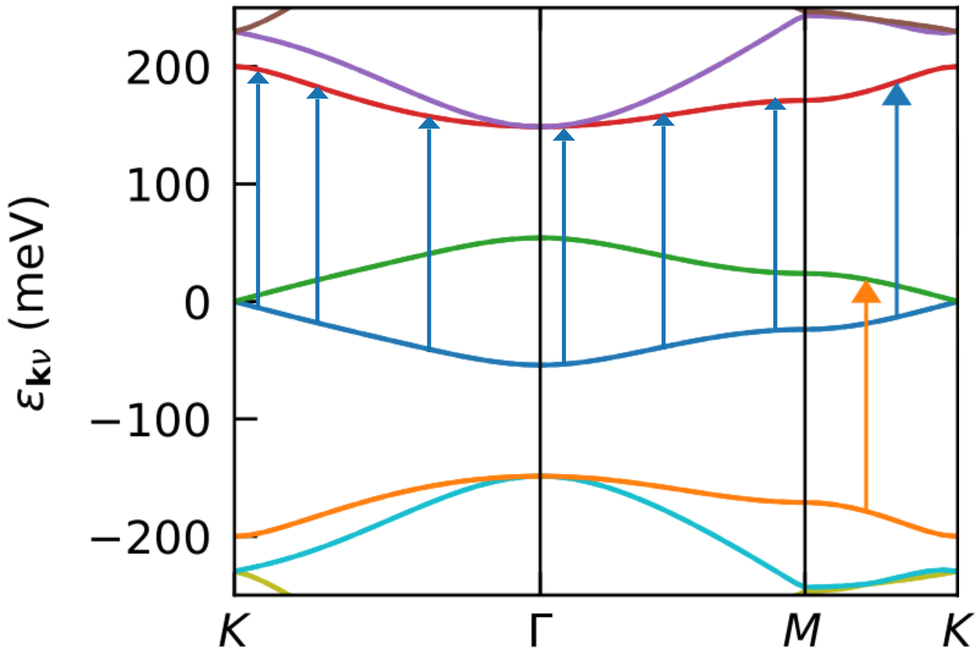
Reduced tunneling strength in AA-regions

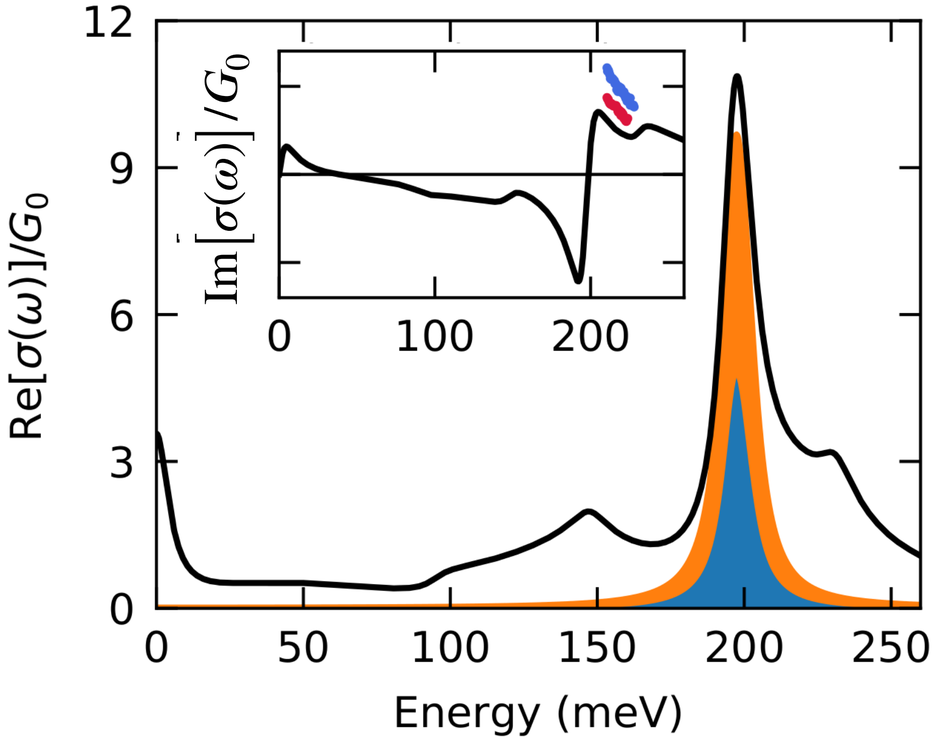


$u_{\mathrm{AA}} = 0$ meV
$u_{\mathrm{AB}} = 100$ meV

Enhanced interband resonance

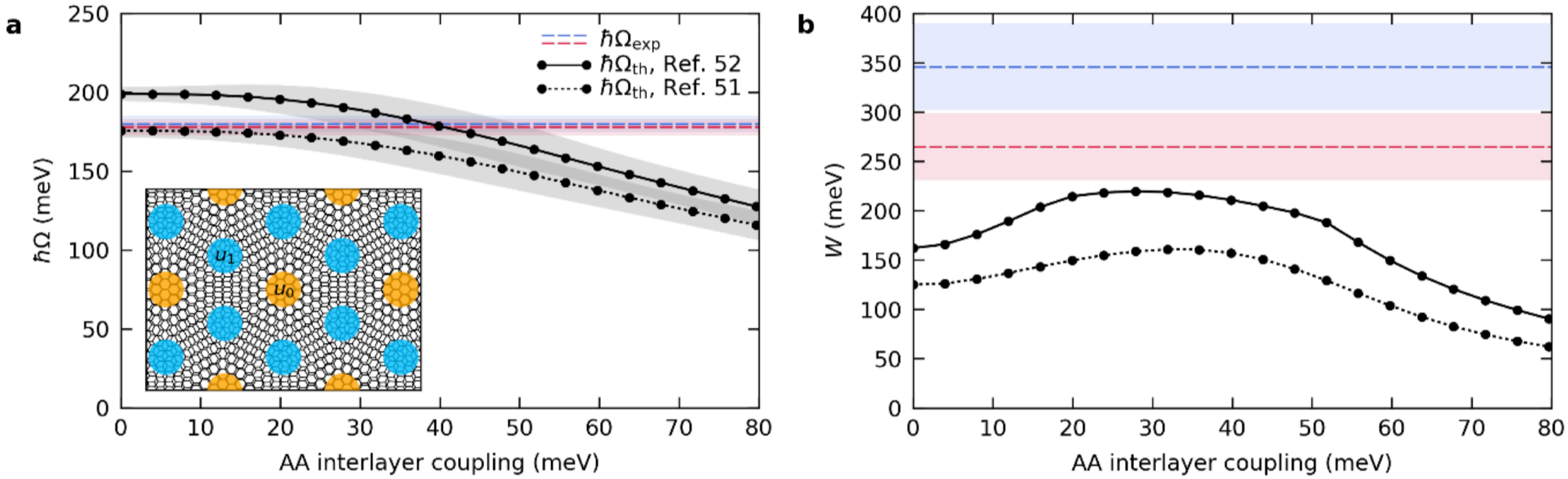
Reduced tunneling strength in AA-regions





Resonance frequency

Spectral weight



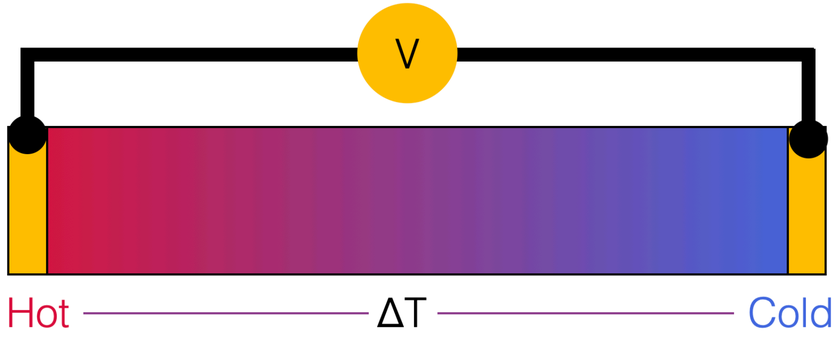
Our data show u_0 ~40meV~0.5u_1—> strong corrugations or e-e interactions

Photothermoelectric effect in TBG

Approach 2. Using AFM tip to heat up electrons locally.

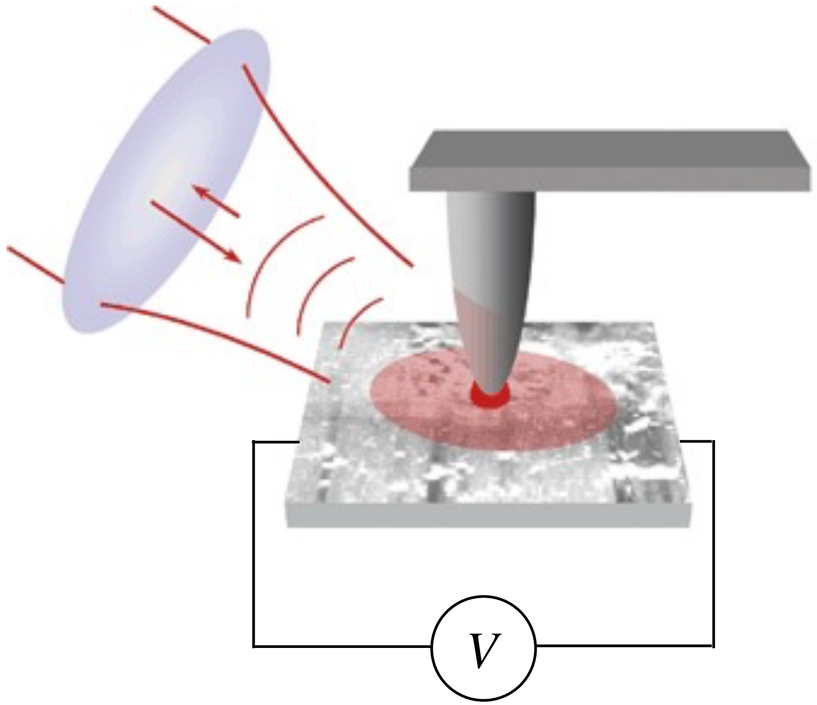
Irradiated tip acts as a local hot spot right above a planar sample.

Conventional thermoelectric effect:

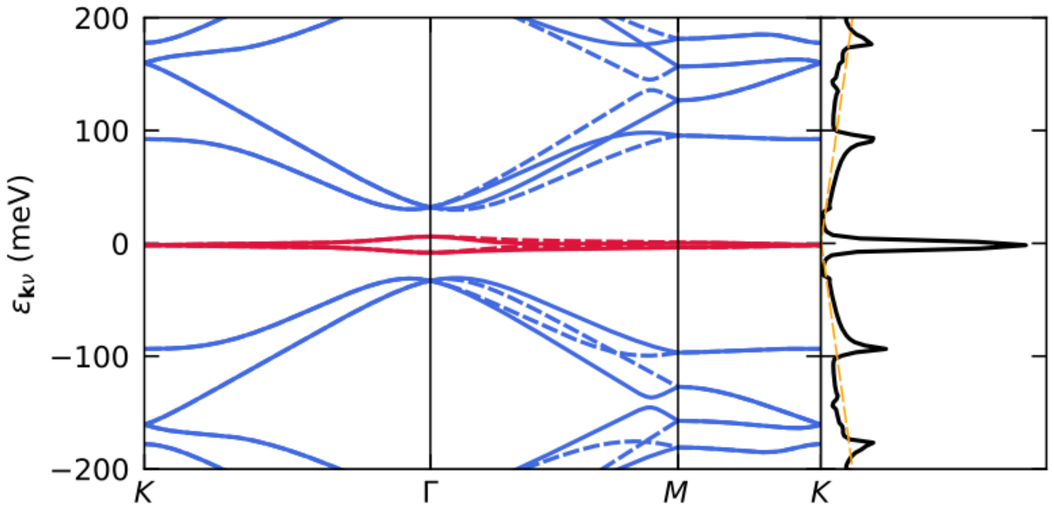


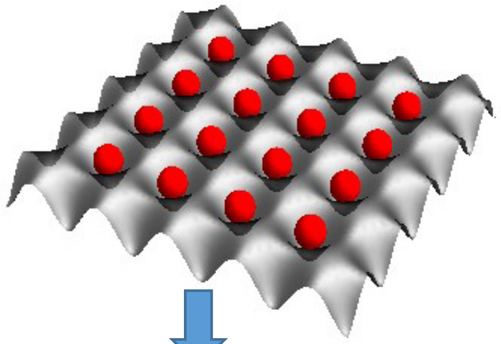
$$V = S\Delta T$$

Light "heats up" electrons:

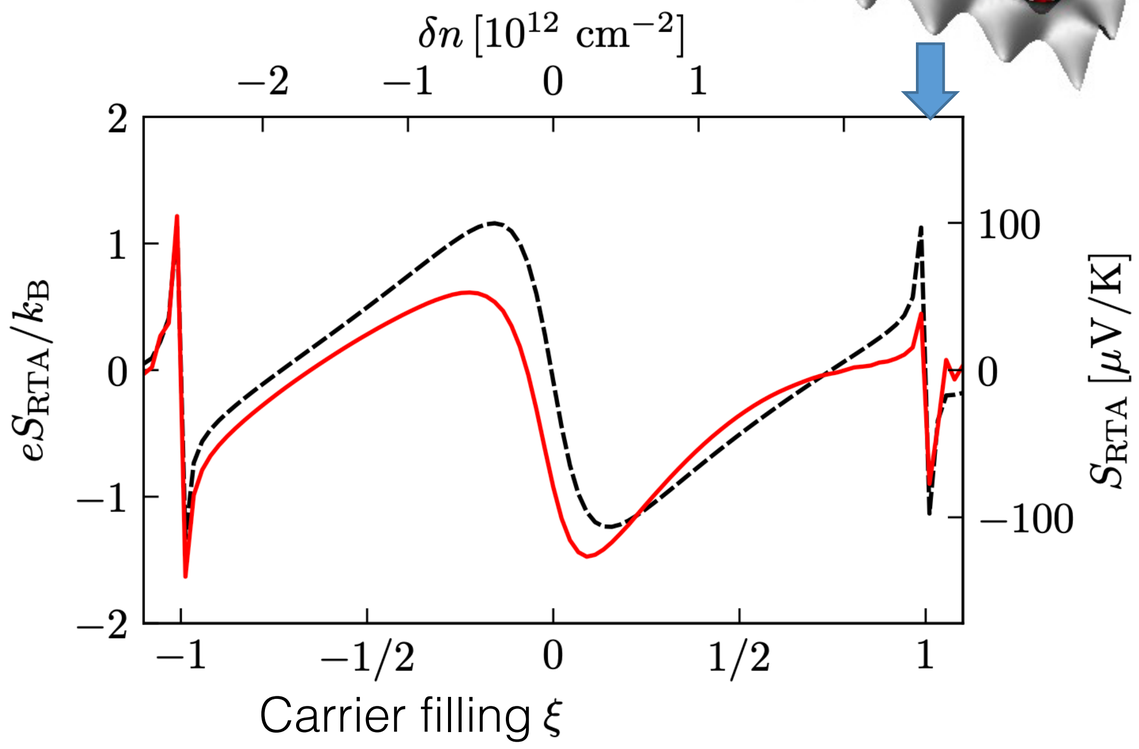


Bandstructure and Seebeck coefficient
Magic angle graphene: Θ=1.05°



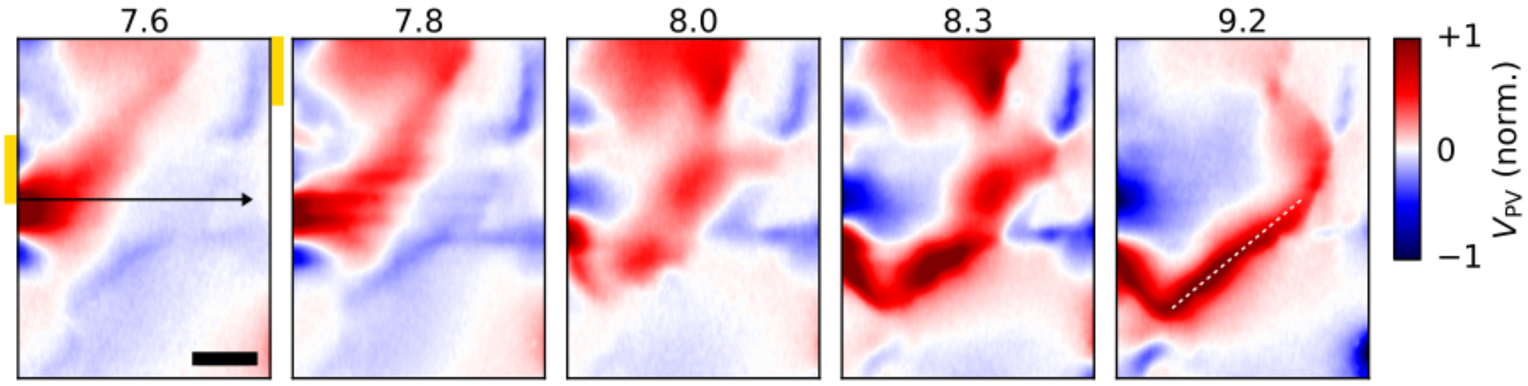


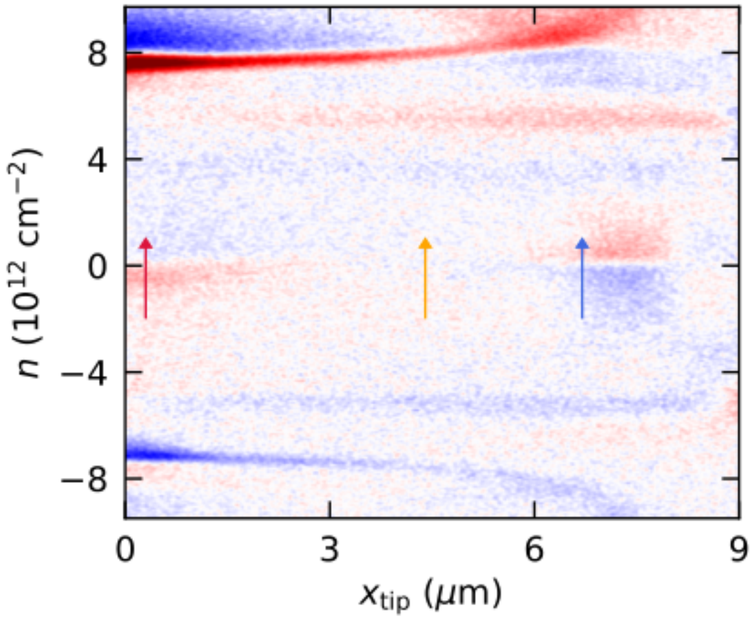
Seebeck coefficient



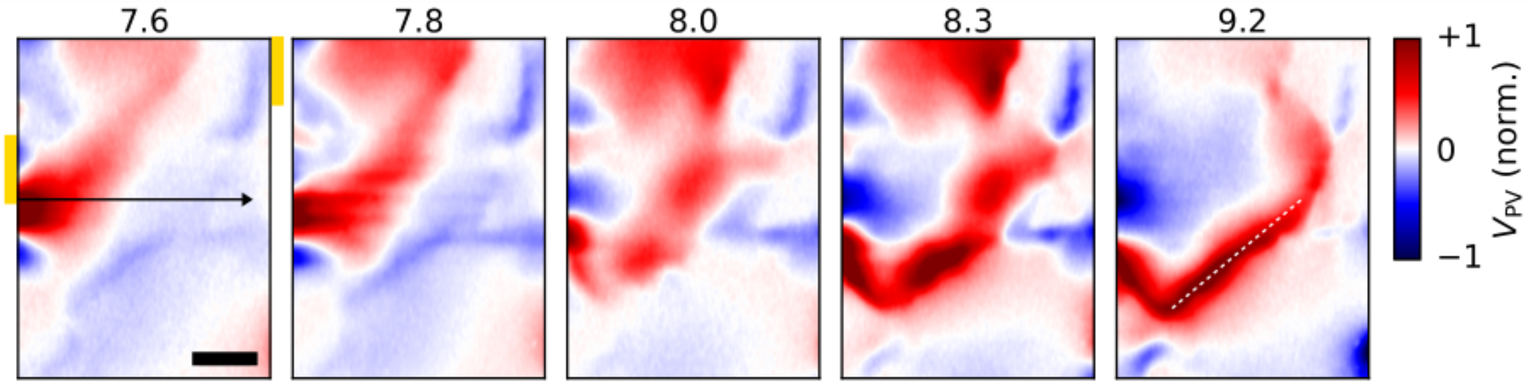
Theory: P. Novelli, I. Torre, F.H.L. Koppens, F. Taddei, and M. Polini, PRB 2020

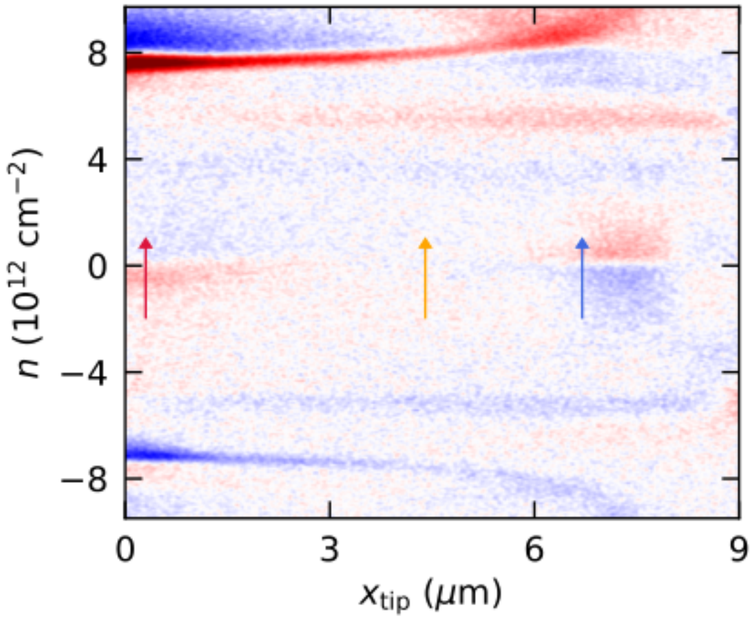
Cryogenic photocurrent nanoscopy

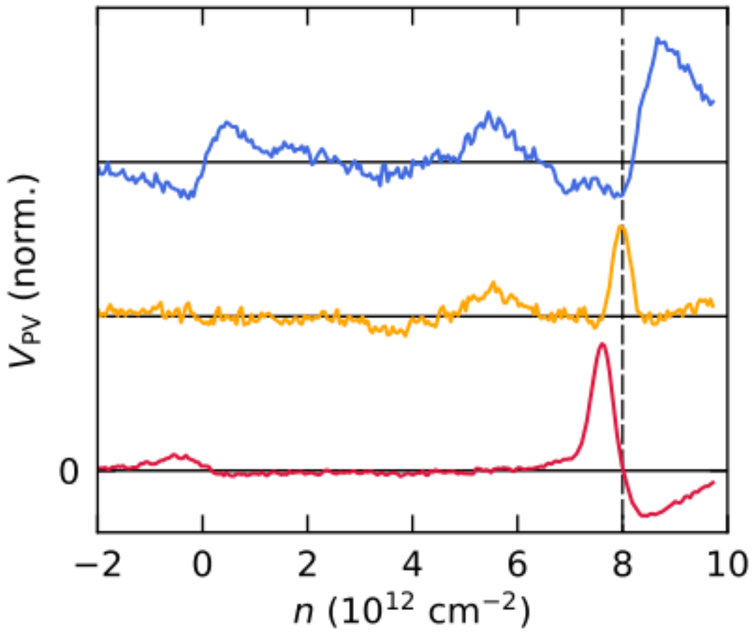




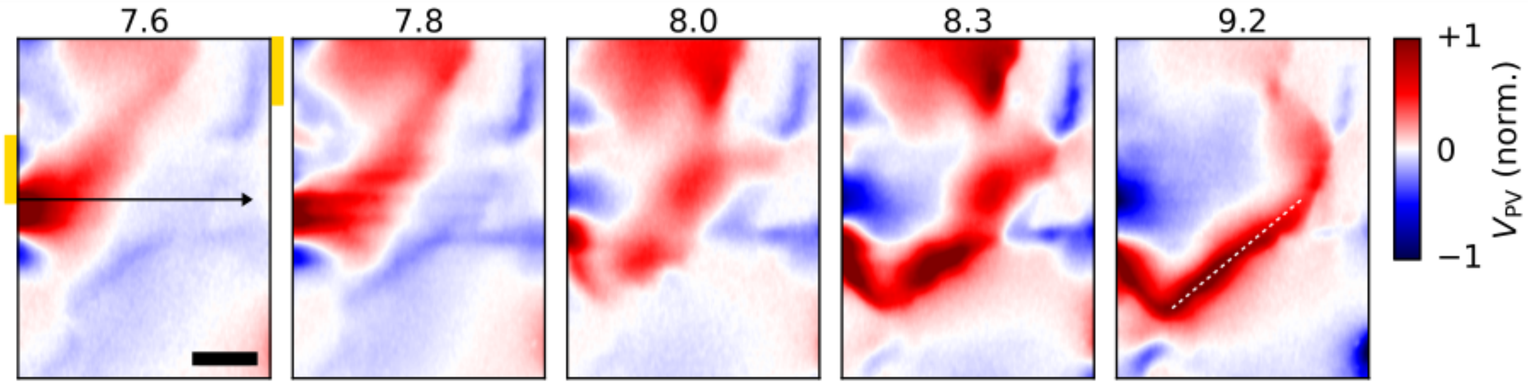
Cryogenic photocurrent nanoscopy

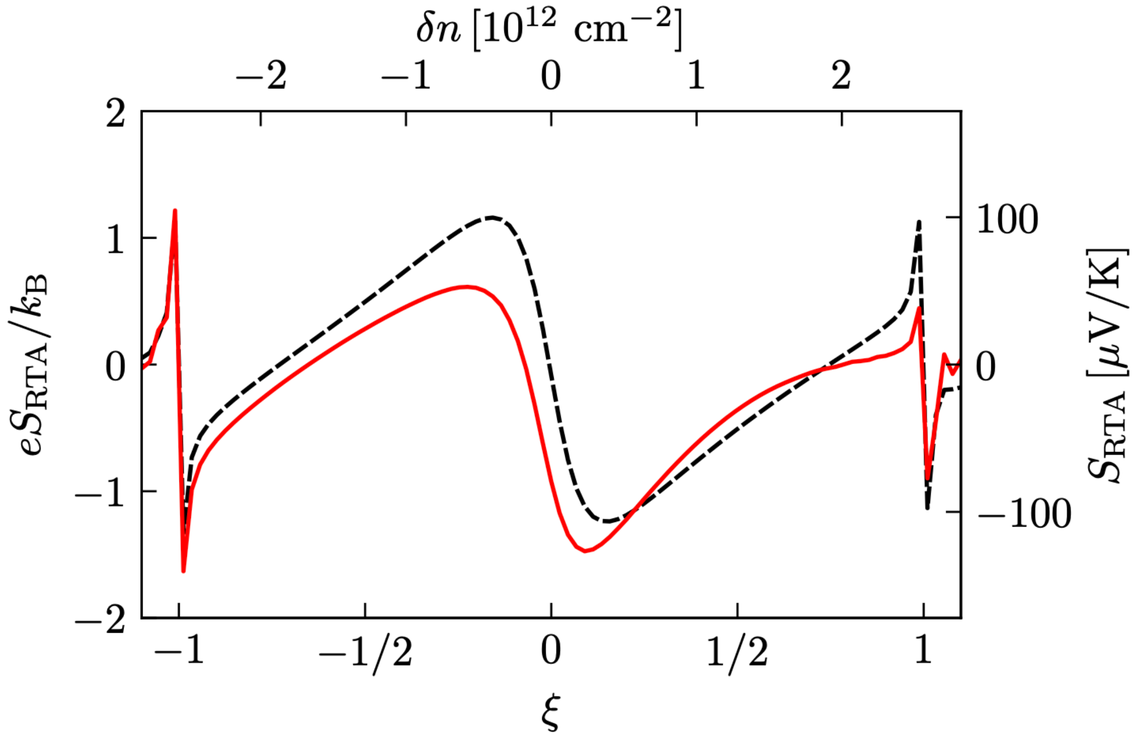


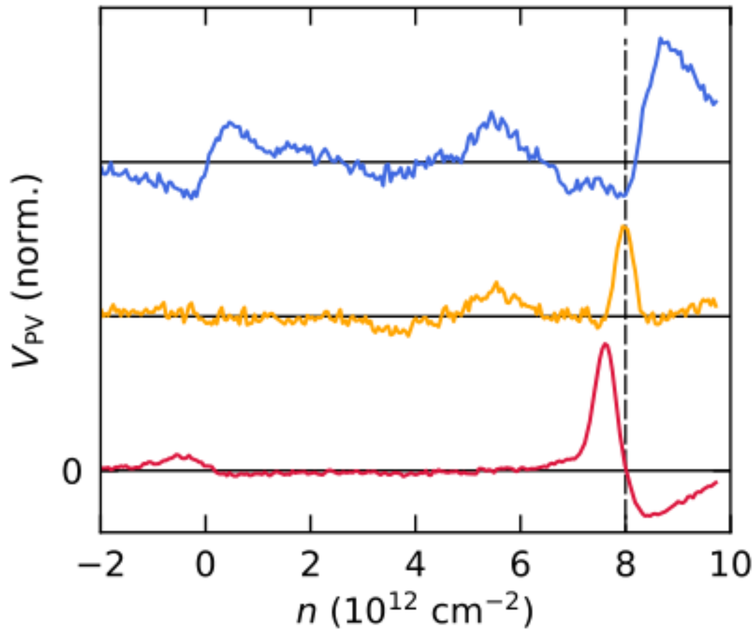




Cryogenic photocurrent nanoscopy

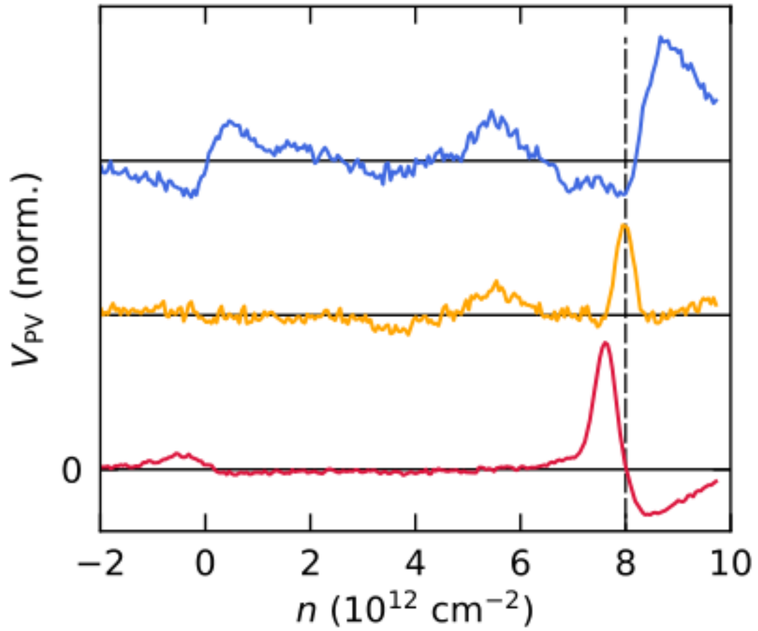




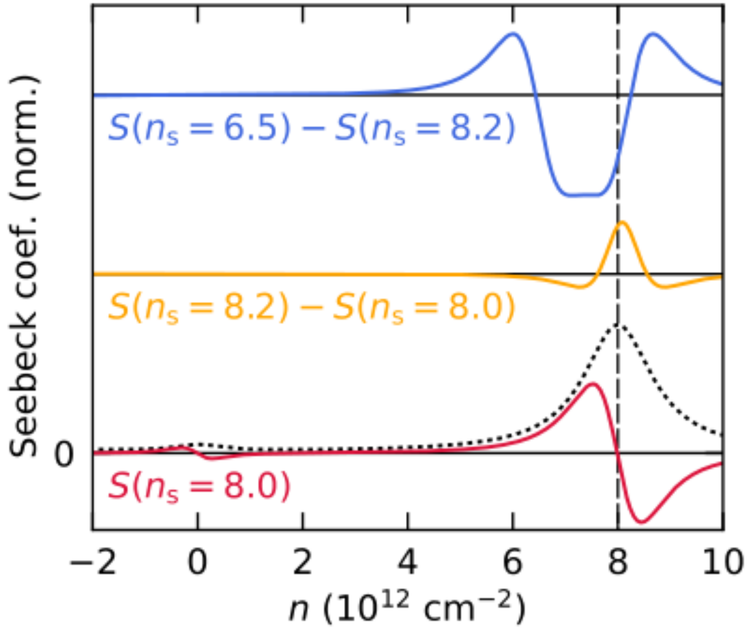


Probing twist angle variations with photovoltage nanoscopy

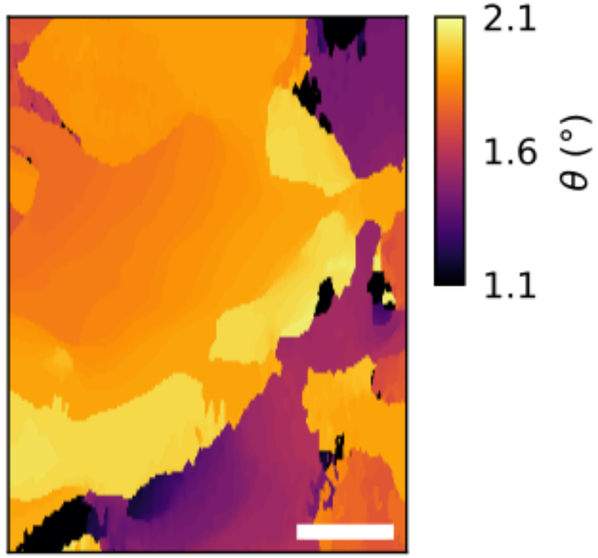
Experiment



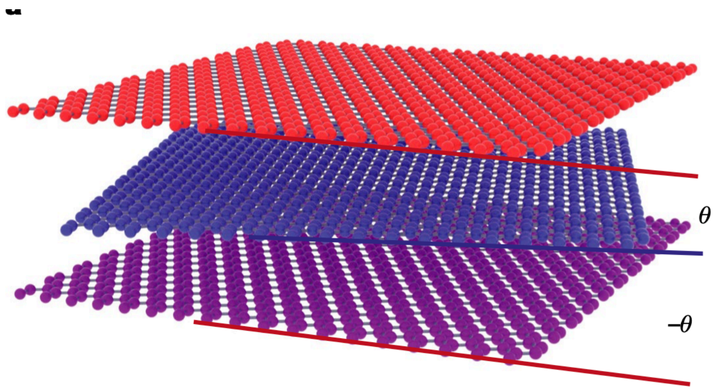
Simplified model

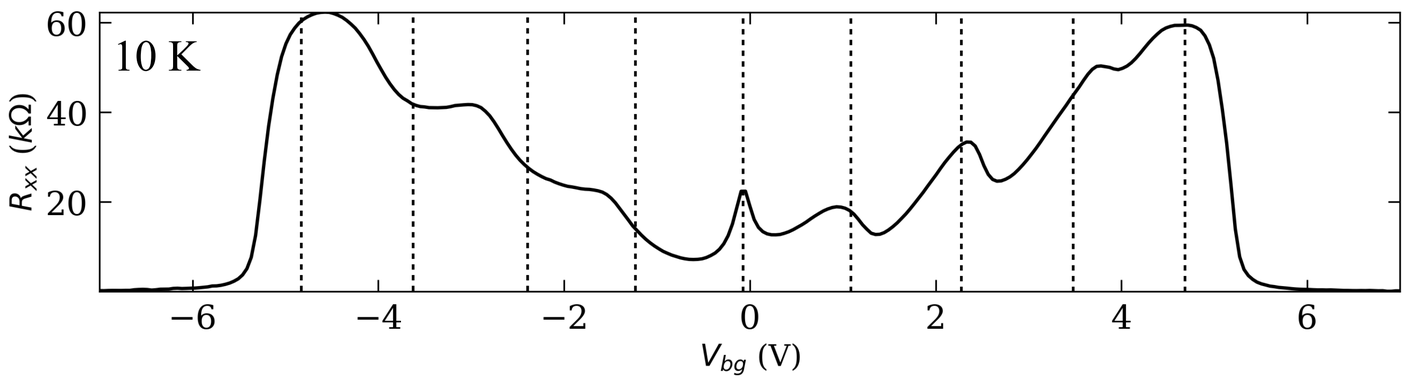


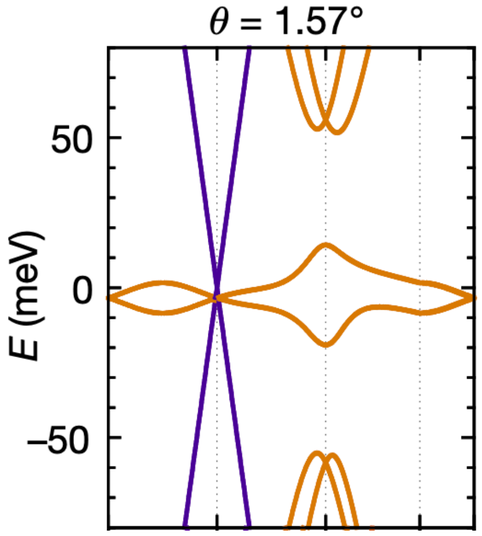
Deduced twist angle map



Local photovoltage studies in magic angle twisted trilayer graphene

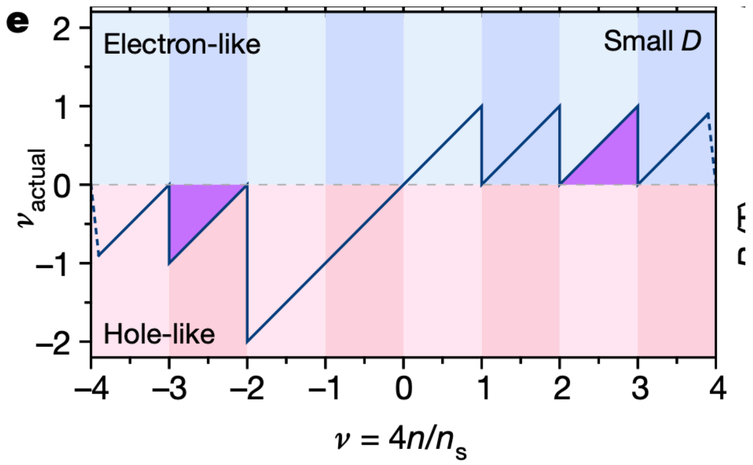




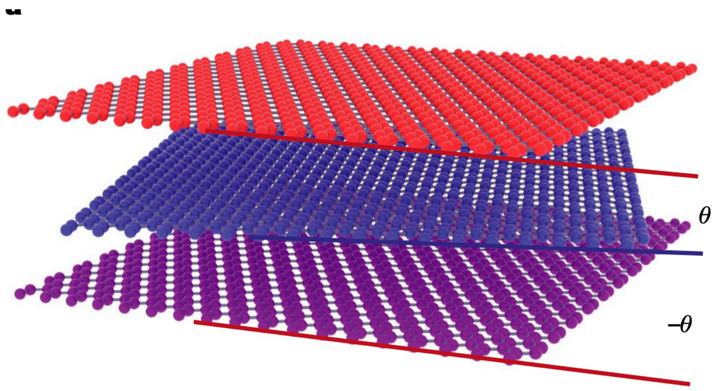


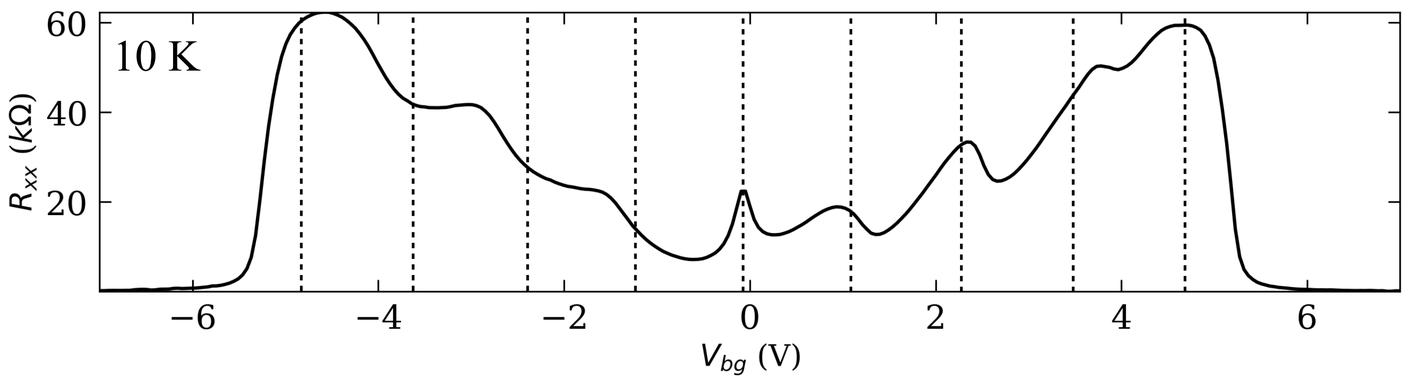
Article

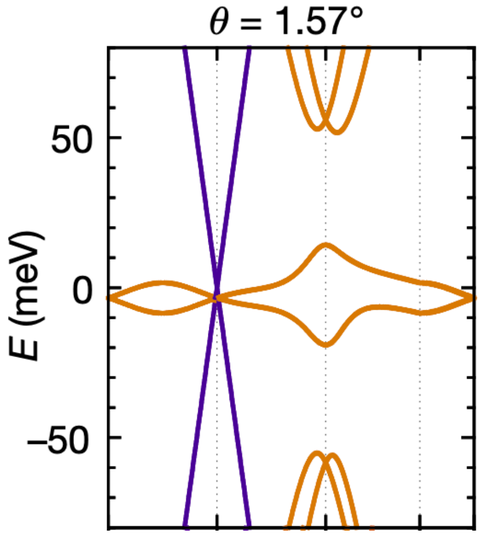
Tunable strongly coupled superconductivity in magic-angle twisted trilayer graphene

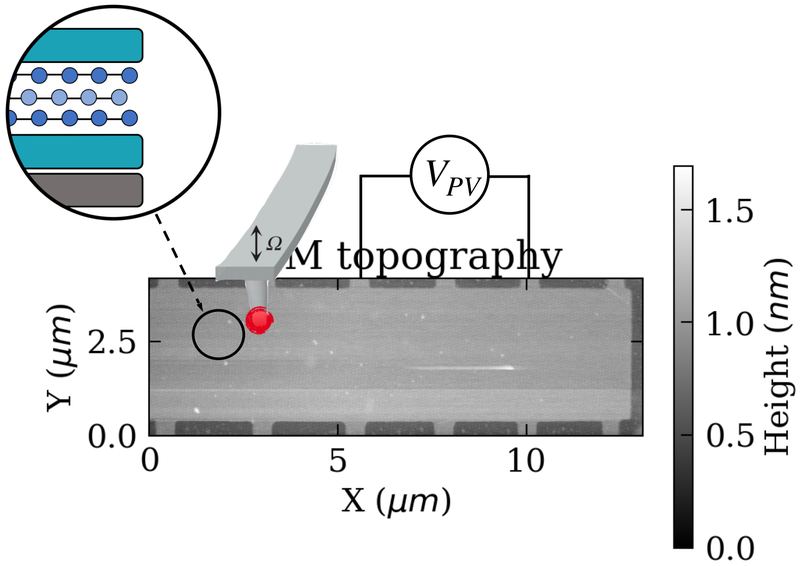


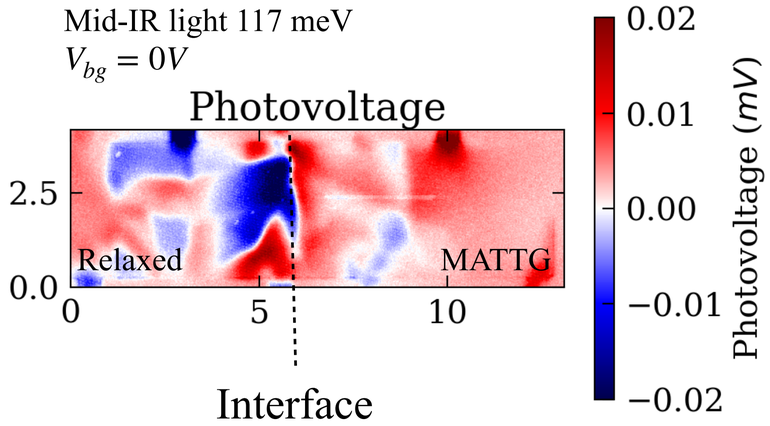
Local photovoltage studies in magic angle twisted trilayer graphene



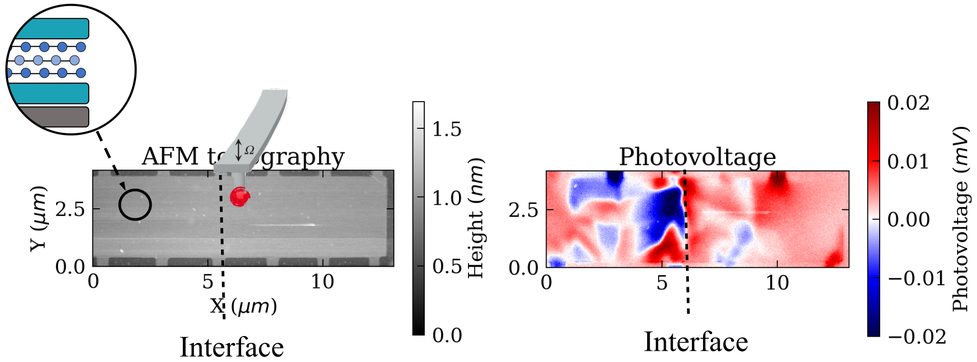


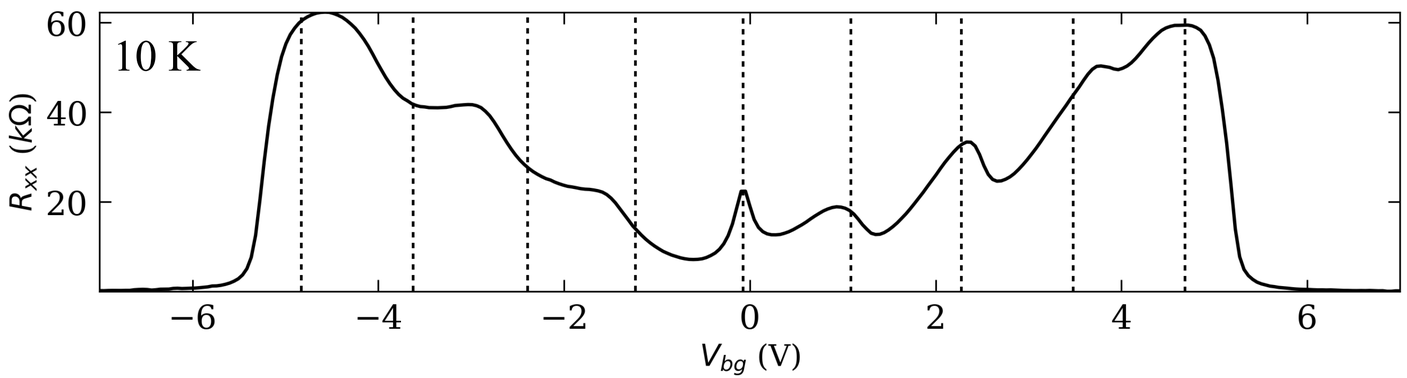






Local thermodynamic probe for strongly correlated electrons



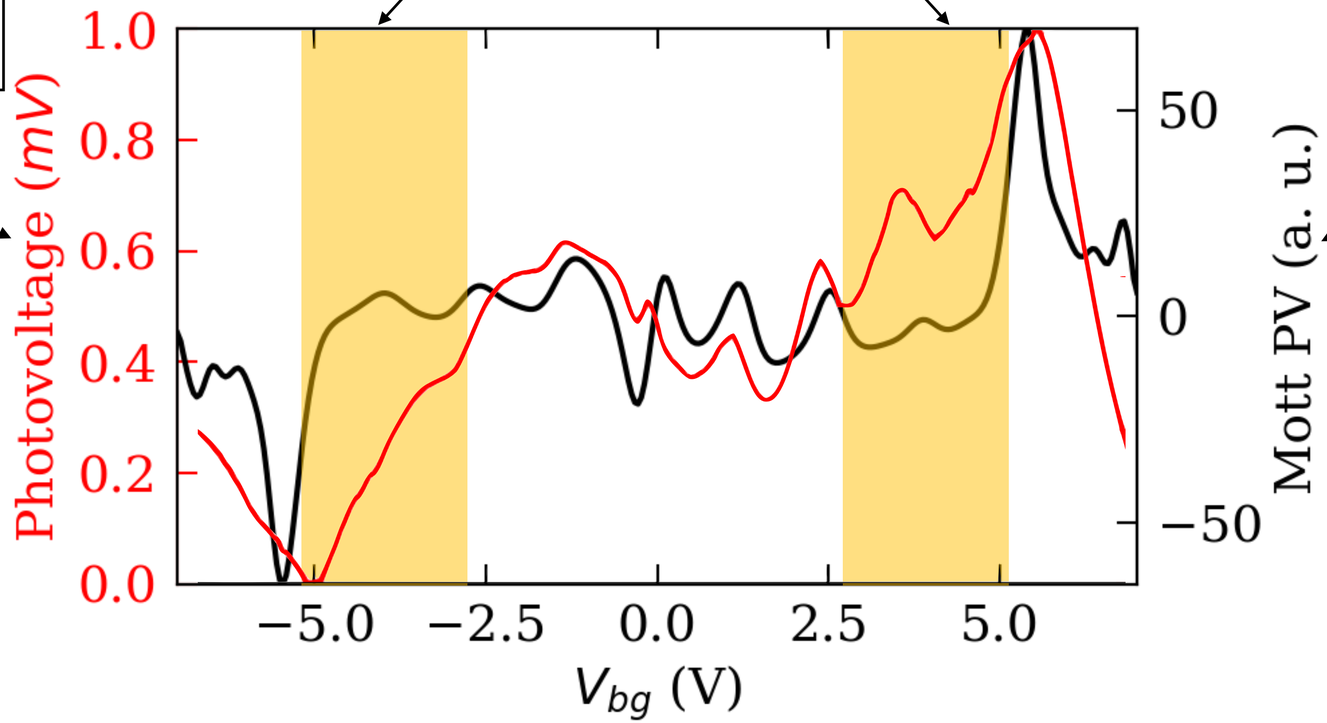


Comparison with weakly interacting electron gas:

Obtained from local photovoltage measurements

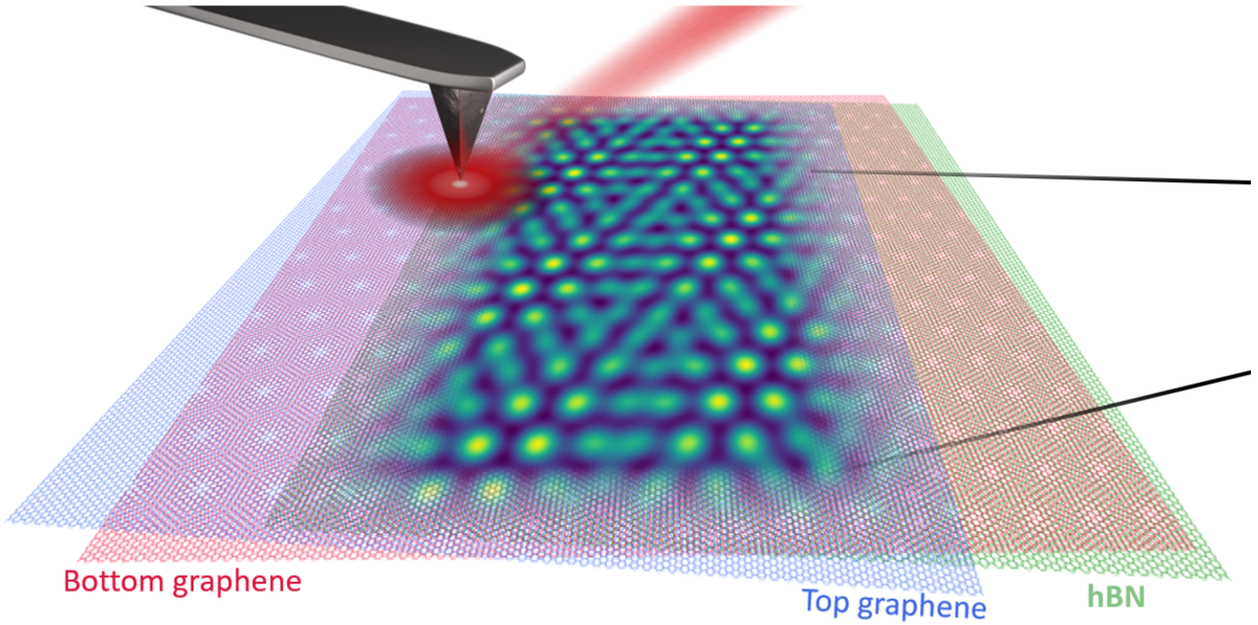
Excess of entropy close to fully filled and fully emptied flat bands

Calculated from 4-probe conductivity data

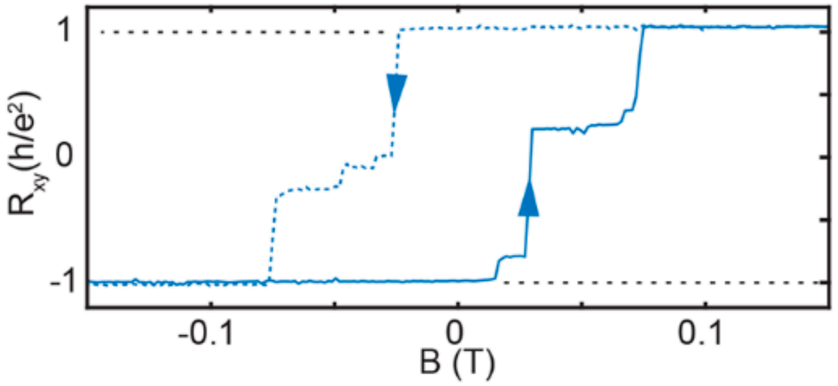


Can we do more? Resolving supermoiré using local photovoltage measurements

Imaging broken inversion symmetry in MATBG.

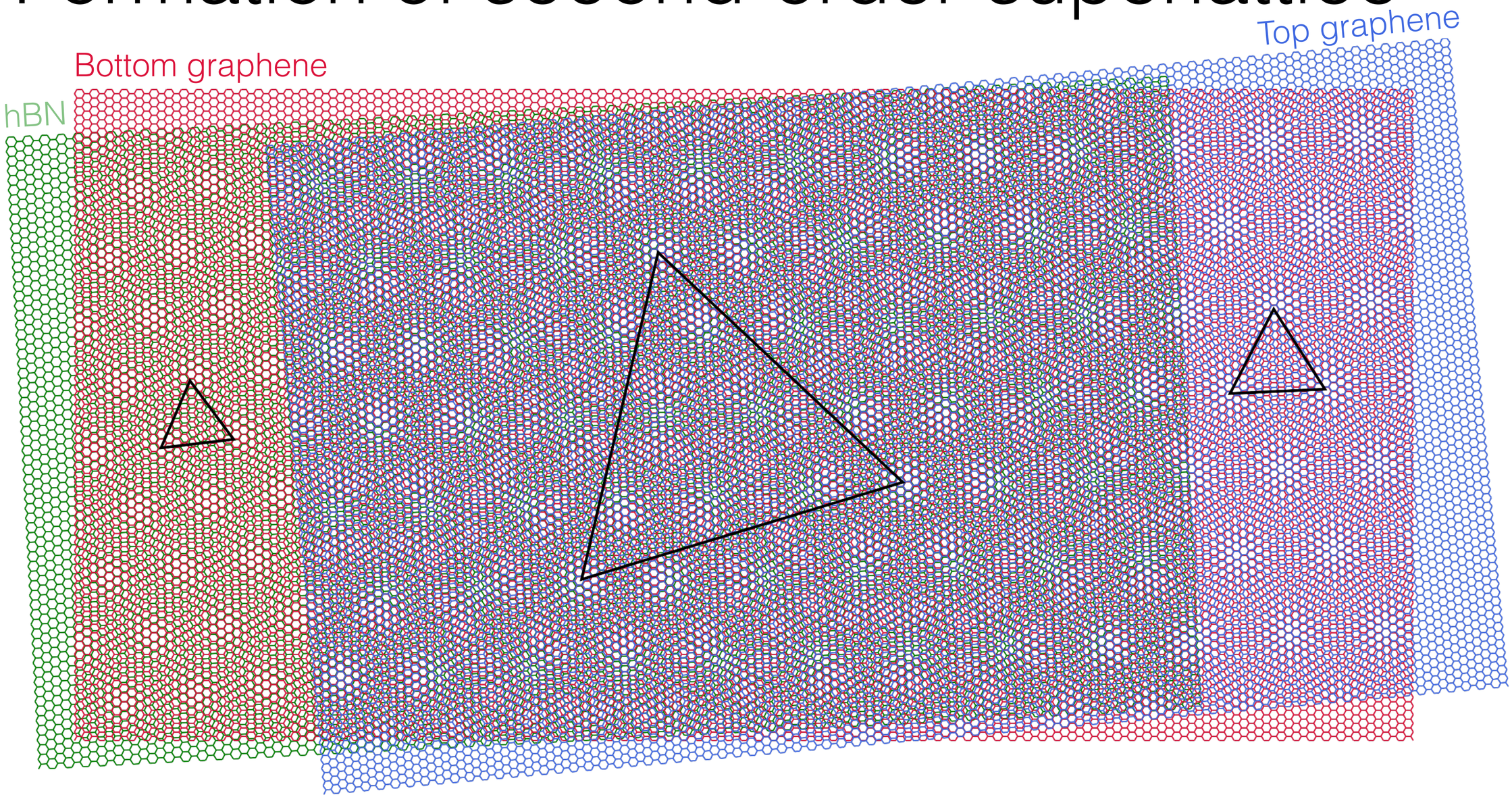


Chern insulators at ν=3



Serlin et. al., *Science* **367**, 6480 (2020)

Formation of second-order superlattice

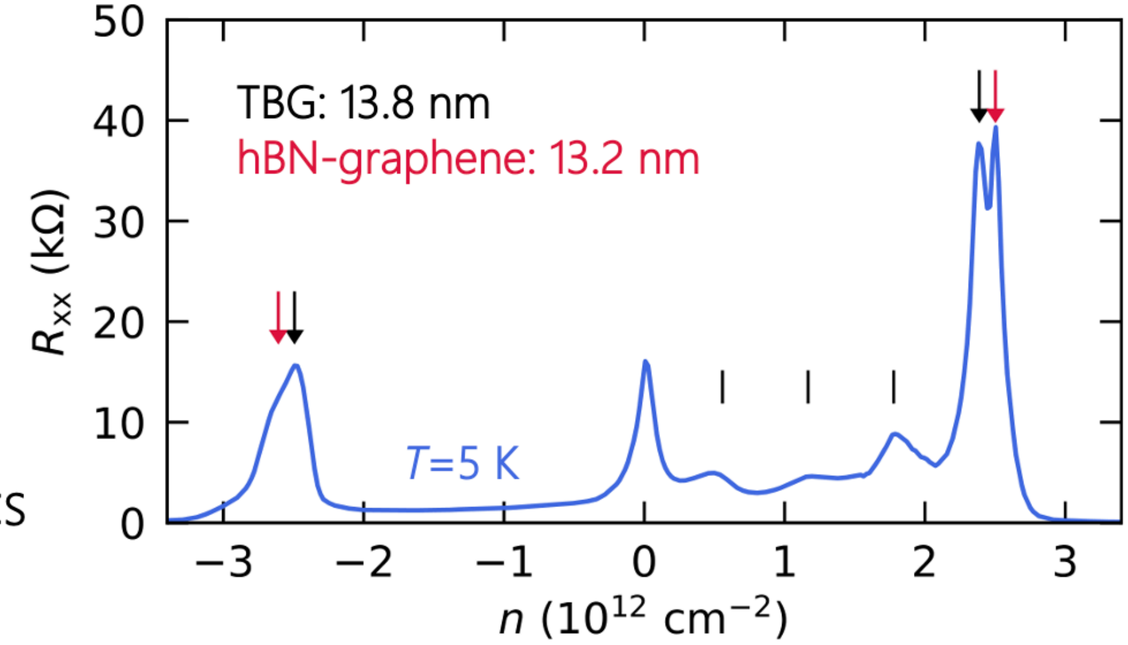


Resistance measurements suggests coexisting moiré lattices

- Peak in resistance at 4 e/unit cell
- Read superlattice periodicity:

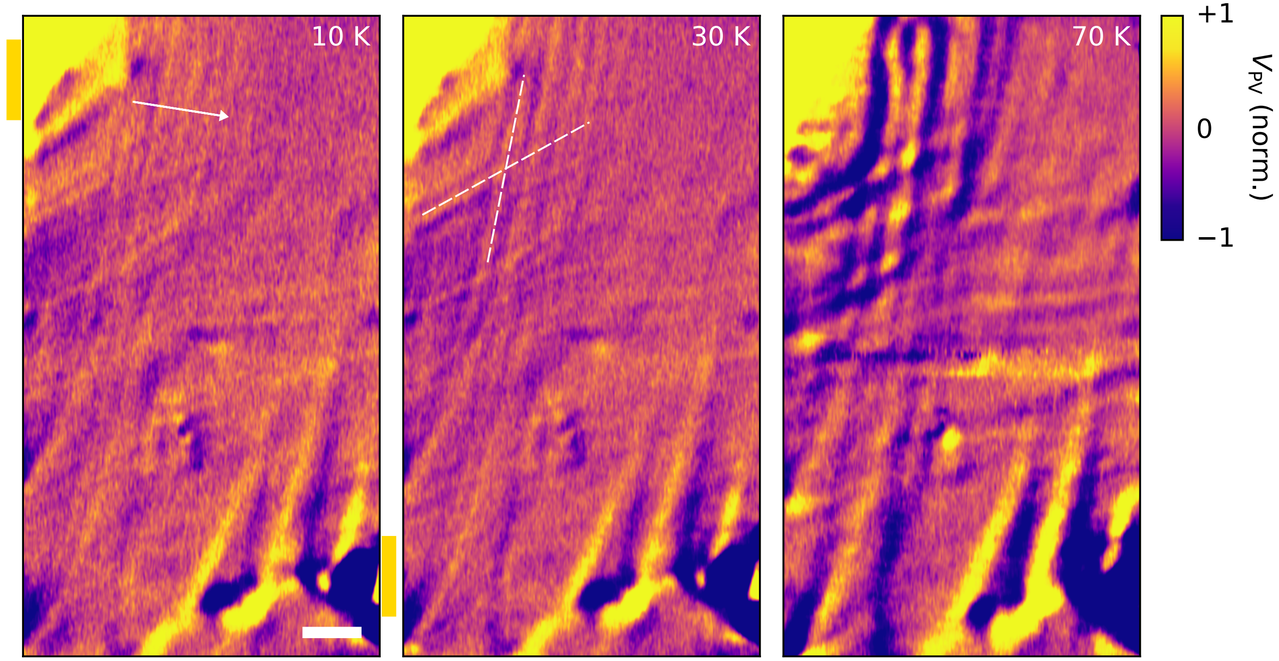
$$n_s = \frac{8}{\sqrt{3}\lambda_M^2}$$

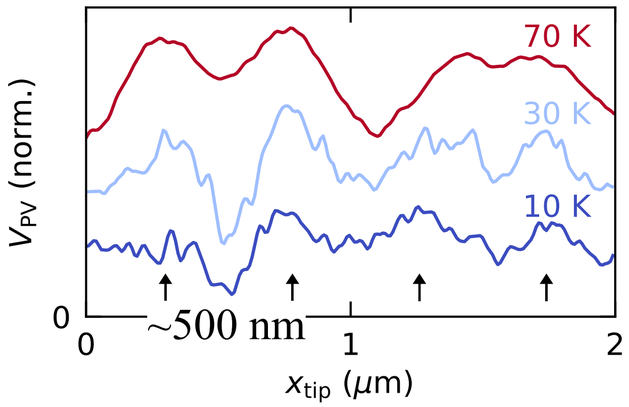
- Additional peaks: magic-angle physics

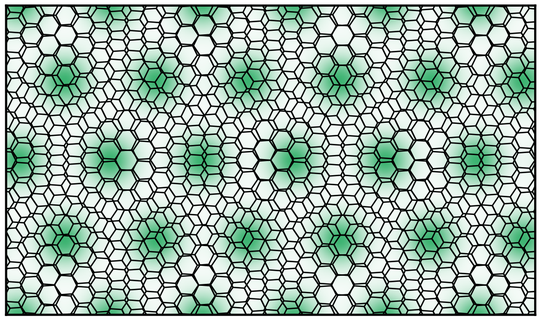


Photocurrent nanoscopy on TBG aligned on hBN

We only observe two sets of fringes.

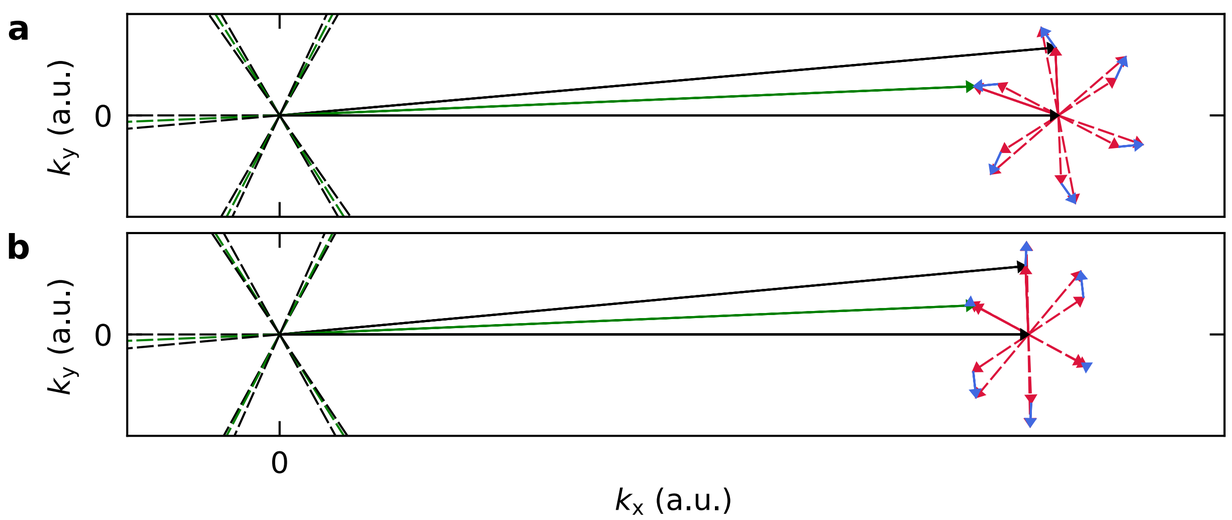


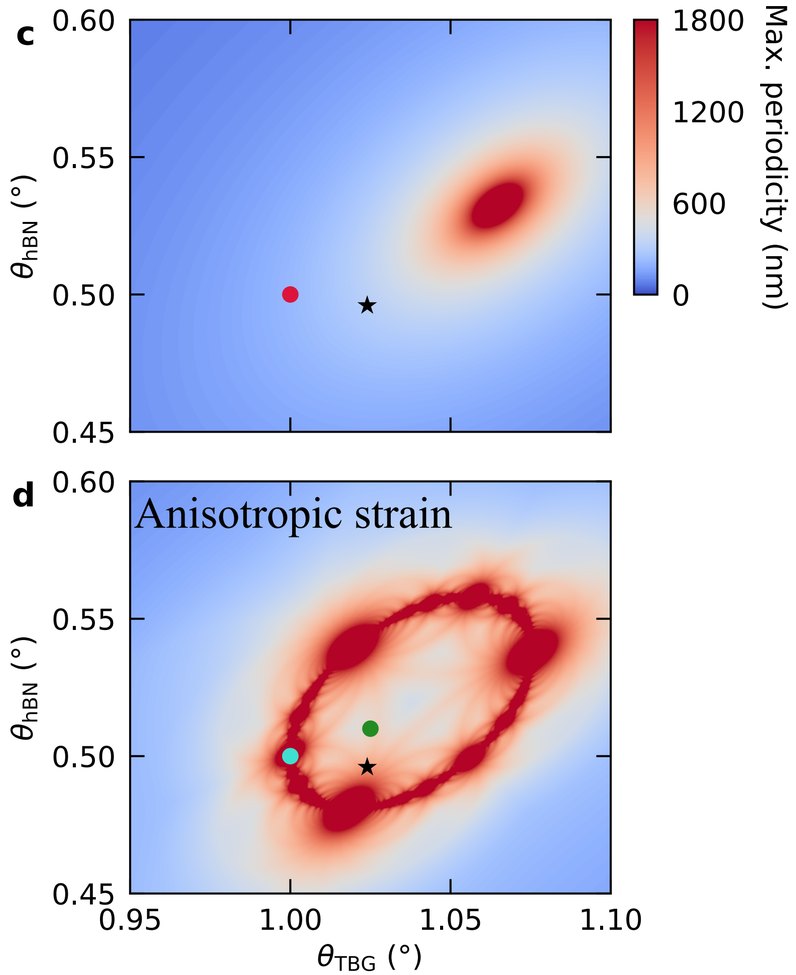


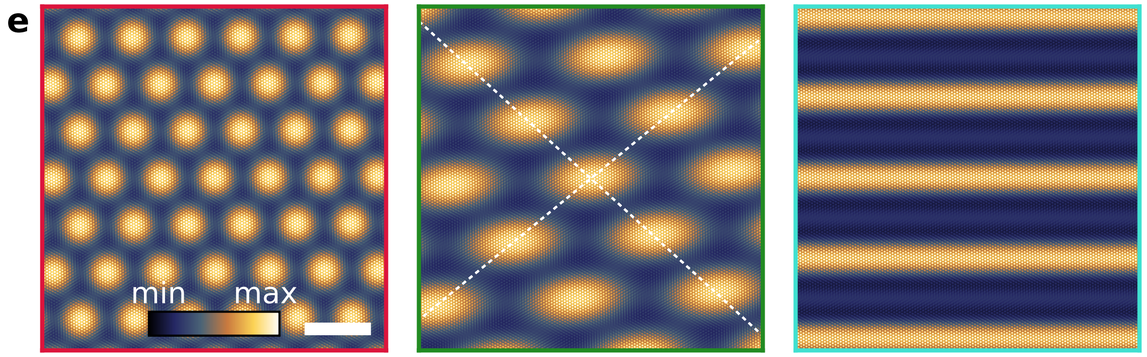


Hesp, PS, et. al., *In preparation*

Implications of strains on the supermoiré potential



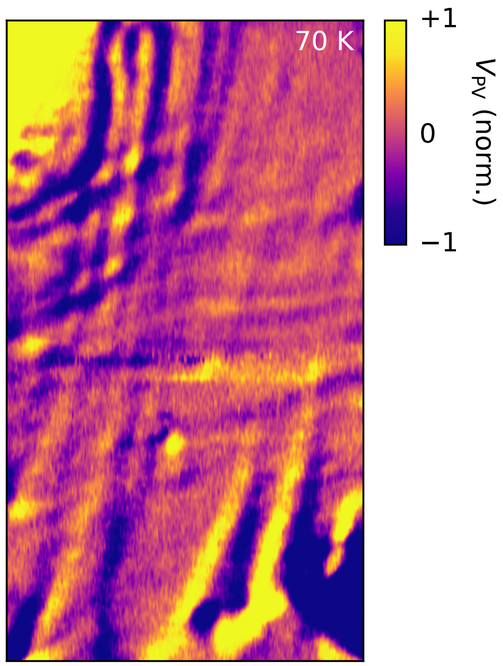


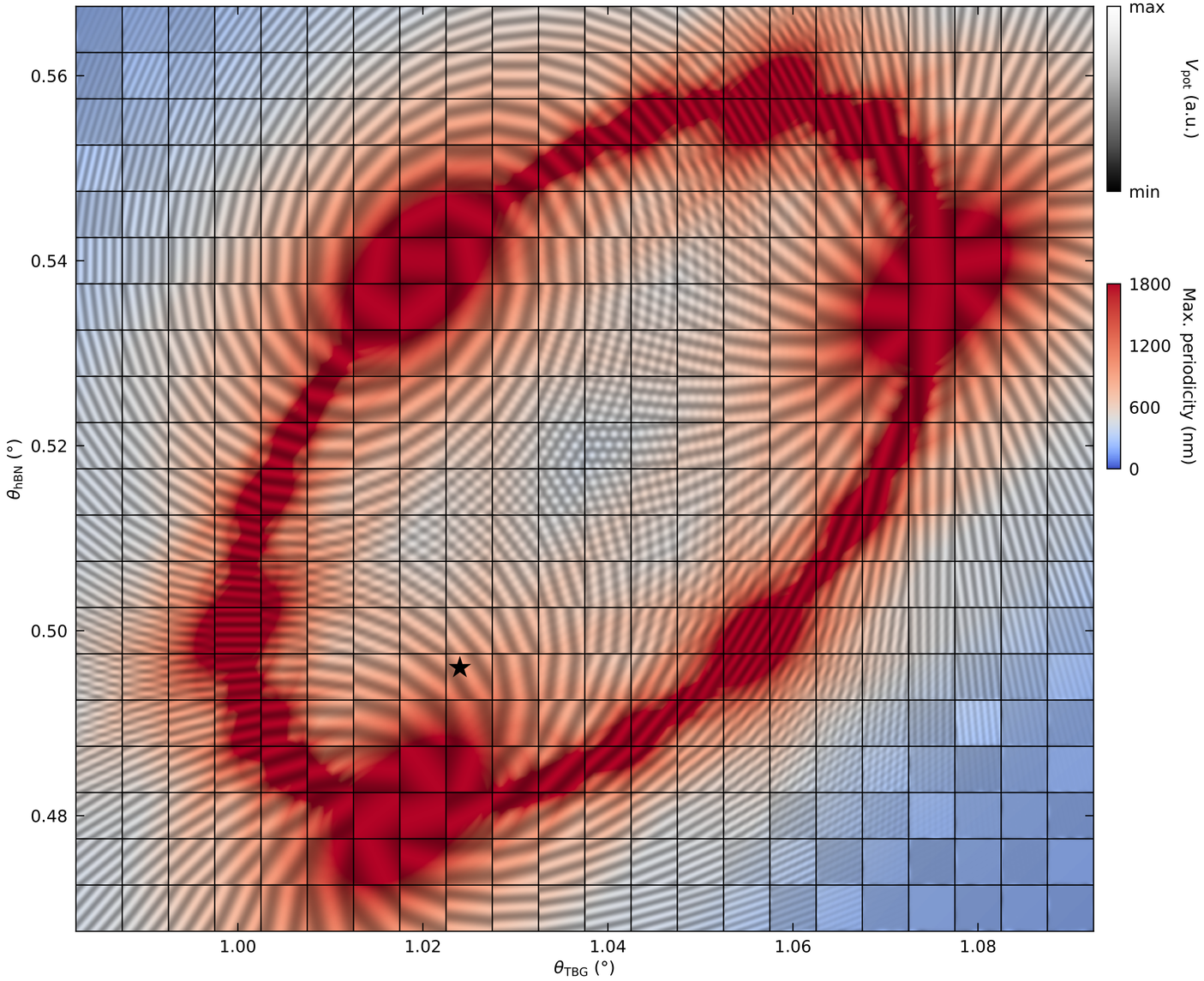


Calculated binding energy

Hesp, PS, et. al., *In preparation*

Strain engineering based on the MATBG/hBN superlattices

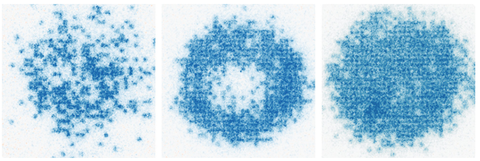


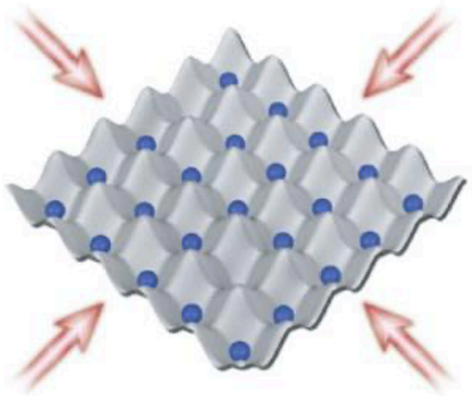


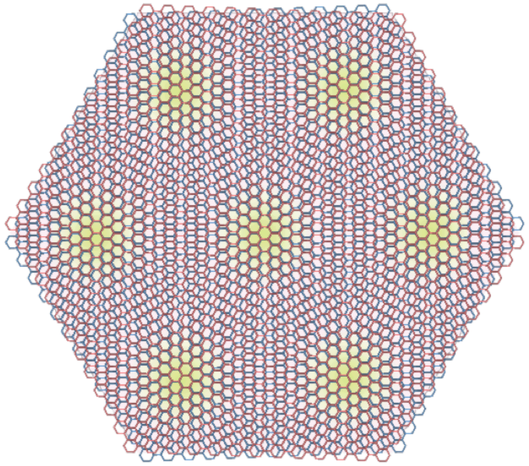
Outlook

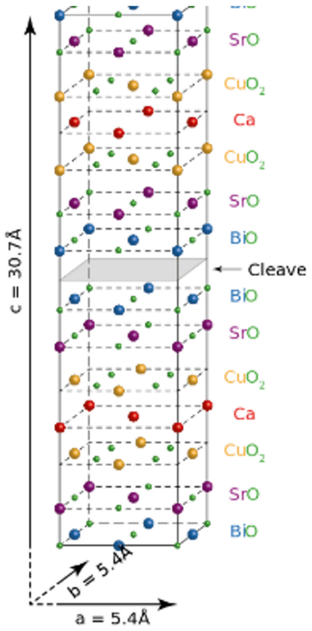
Nanoscale imaging of a solid-state quantum matter simulator

Cold Atoms Optical Lattices	Moiré Quantum Matter	Quantum Materials
Length scale ~ **1 micron**	Moiré length ~ **10nm**	Lattice scale ~ **few Å**









$Bi_2Sr_2Ca_nCu_{n+1}O_{2n+6-d}$

Optical (photocurrent) nanoscopy for THz frequencies:
Probe superconductivity
Real-sapce imaging of electronic correlations
Role of plasmons on correlations

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Prof. Pablo Jarillo-Herrero
Daniel Rodan
Yuan Cao

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