Investigating moiré interlayer excitons under the influence of atomic reconstructions

Nils-Erik Schütte, **Carl Emil Mørch Nielsen**, **Niclas Götting**, **Frederik Lohof**, **Gabriel Bester**, **and Christopher Gies**

Present address:
German Aerospace Center
Institute for Satellite Geodesy and Inertial sensing, and
University of Bremen
Institute for Theoretical Physics

18.03.2024

University of Bremen

QUANTUM TEAM

Universität Hamburg
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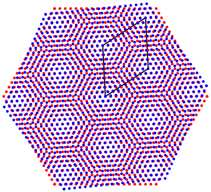
DFG Deutsche Forschungsgemeinschaft
German Research Foundation

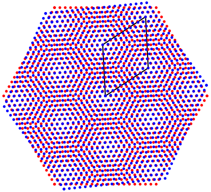
SPP 2244

Motivation



Twisted TMDs

Motivation



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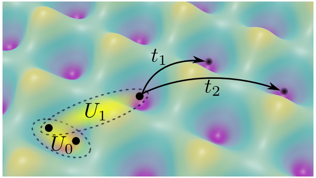
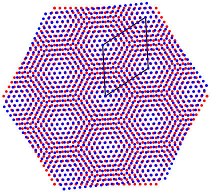


Götting *et al.*, Phys. Rev. B **105**, 165419 (2022)

Motivation



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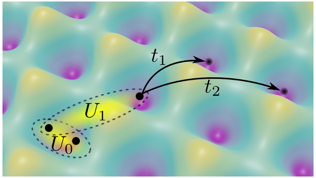

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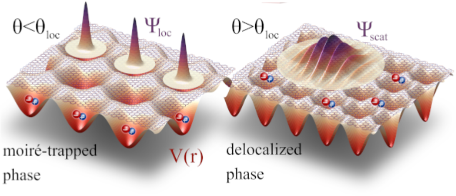
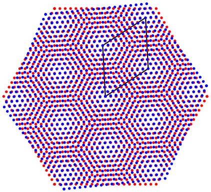
Quantum Phases



Brem *et al.* Nano Lett., 20, 12, 8534-8540 (2020)

Motivation



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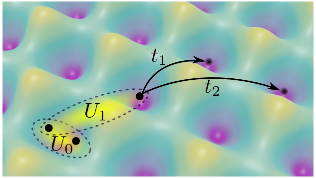

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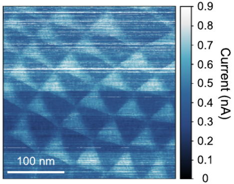
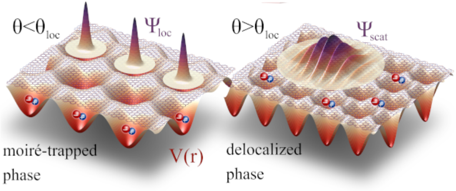


Rosenberger *et al.* ACS Nano, **14**, 4, 4550-4558 (2020)

Atomic Reconstructions

Quantum Phases



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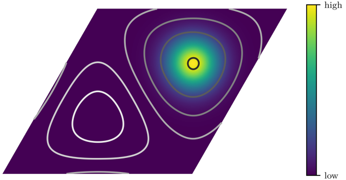
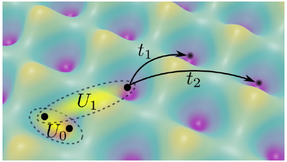
Outline

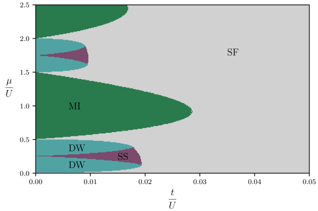
Interlayer Excitons



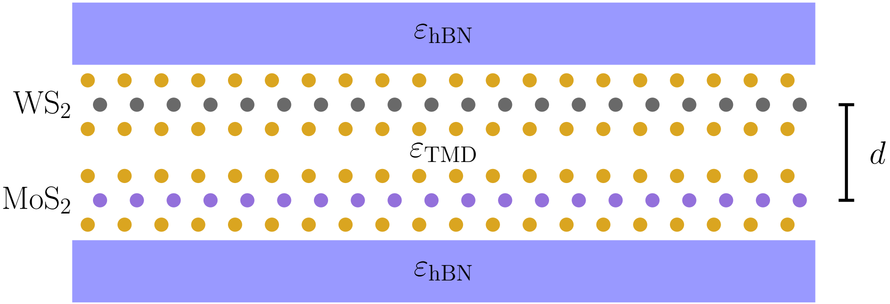
Mapping onto a Bose-Hubbard Model



Influence of Atomic Reconstructions on the Quantum Phases of IXs

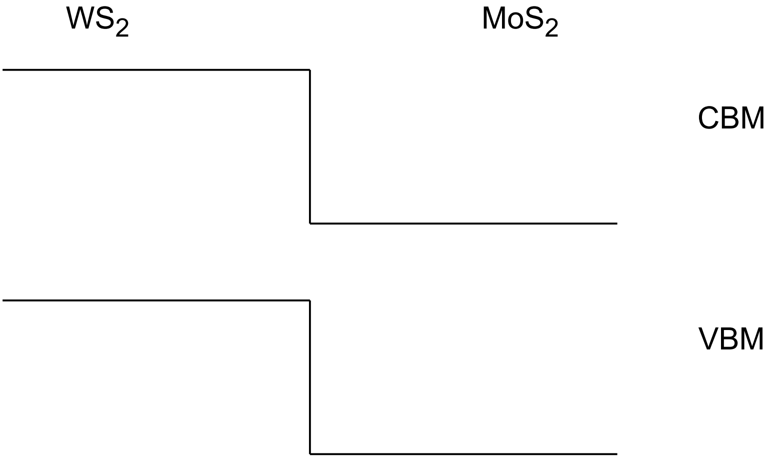


Stacking two TMDs

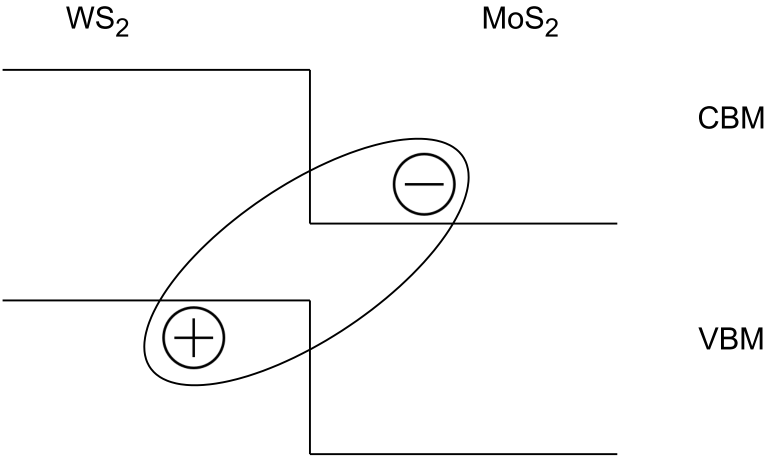


Stacking structure: R^X_X,
X: chalcogen atom

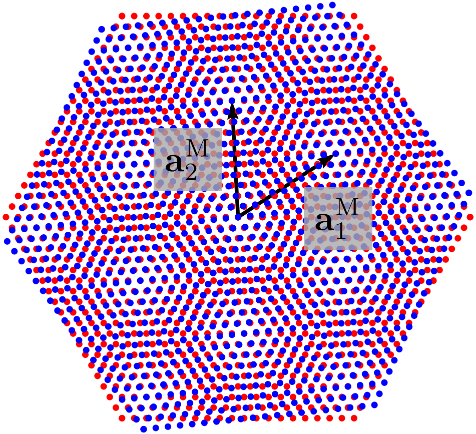
Type-II Band Alignment



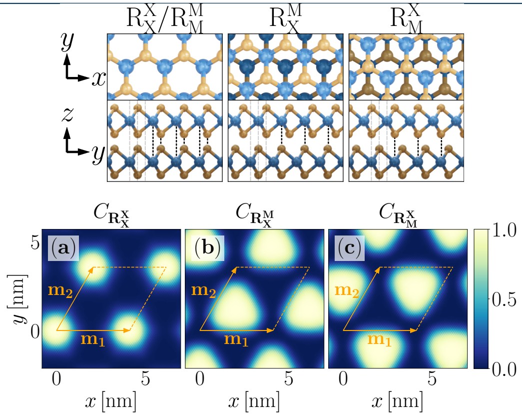
Exciton Formation



Introducing Twist Angle



Atomic Reconstructions



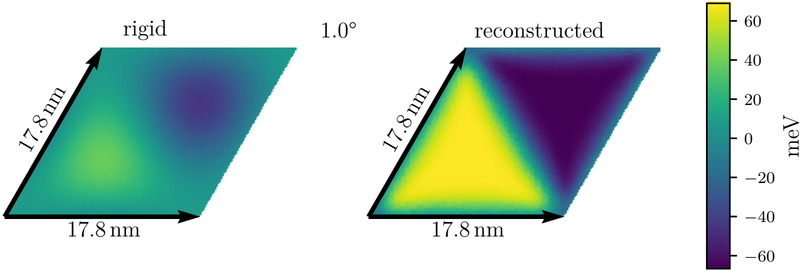
AG Bester, Universität Hamburg

Nielsen *et al.*, Phys. Rev. B 108, 045402 (2023)

Moiré Potential

- band gap as effective potential for excitons:

$$V^{\mathrm{M}}(\boldsymbol{r}) = \sum_{\boldsymbol{G}} V_{\boldsymbol{G}} \mathrm{e}^{\mathrm{i}\boldsymbol{G}\boldsymbol{r}}$$



Wu *et al.*, Phys. Rev. Lett. 118, 1474019 (2017)
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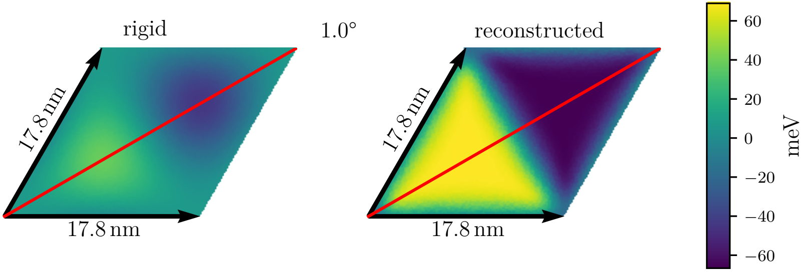

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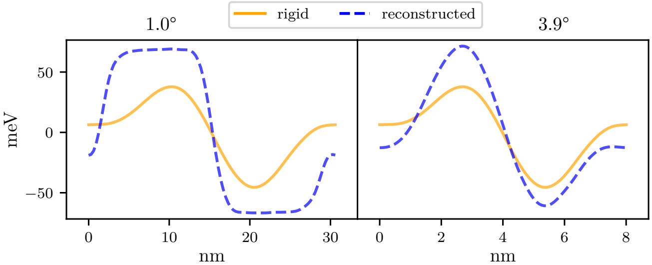


Wu *et al.*, Phys. Rev. Lett. 118, 1474019 (2017)
Nielsen *et al.*, Phys. Rev. B 108, 045402 (2023)

IX Hamiltonian

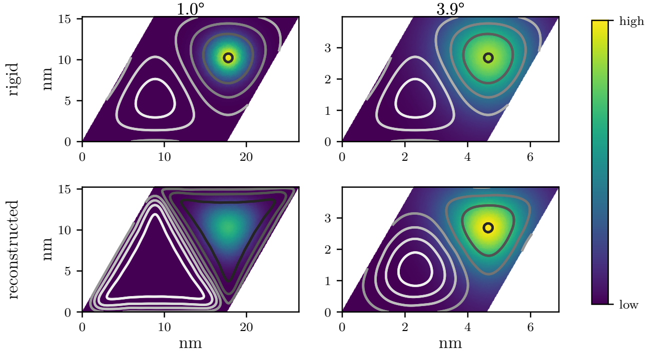
$$H = -\frac{\hbar^2}{2M}\Delta + V^{\mathrm{M}}(\boldsymbol{r})$$

- approximate excitons as bosons
- effective single-particle Hamiltonian for center of mass motion
- solve H in Bloch basis
- Wannier functions from Bloch functions via Fourier like transform

Wu *et al.*, Phys. Rev. B 97, 035306 (2018)

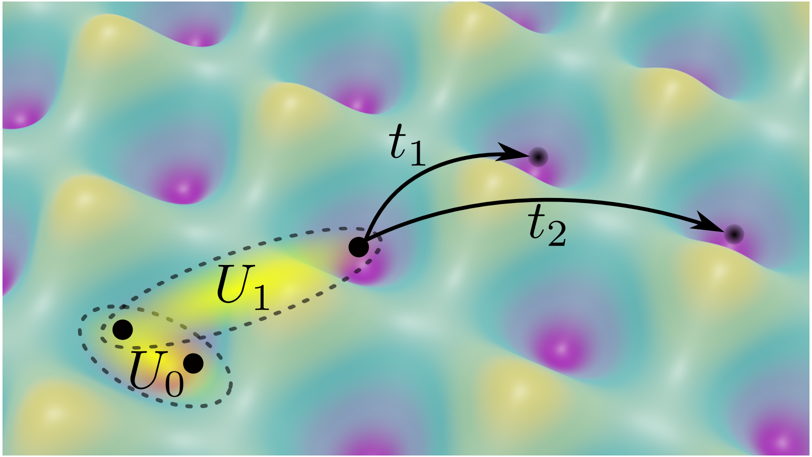
IX Wave Functions

$$H = -\frac{\hbar^2}{2M}\Delta + V^{\mathrm{M}}(\boldsymbol{r})$$



Wu *et al.*, Phys. Rev. B 97, 035306 (2018)

Bose-Hubbard Model



Götting *et al.*, Phys. Rev. B **105**, 165419 (2022)

Bose-Hubbard Model

$$H = -t \sum_{\langle ij \rangle} \hat{b}_i^\dagger \hat{b}_j + \frac{U}{2} \sum_i \hat{n}_i(\hat{n}_i - 1) - \mu \sum_i \hat{n}_i + \frac{V}{2} \sum_{\langle ij \rangle} \hat{n}_i \hat{n}_j$$

May, Diploma thesis, `http://users.physik.fu-berlin.de/~pelster/Theses/may.pdf`

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Mean-field approximation:

$$\varphi_i = \langle \hat{b}_i \rangle$$
$$\varrho_i = \langle \hat{n}_i \rangle$$

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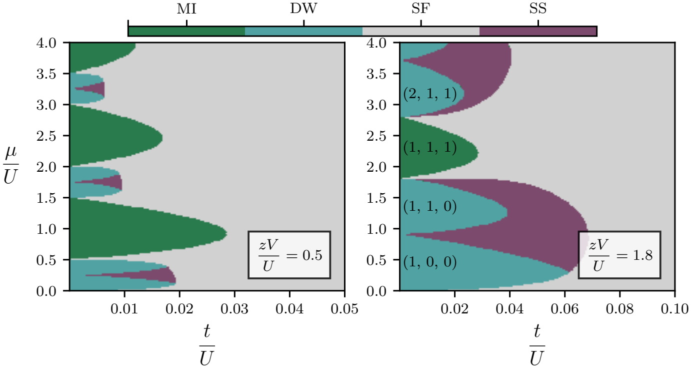


May, Diploma thesis, `http://users.physik.fu-berlin.de/~pelster/Theses/may.pdf`

Hubbard Parameters

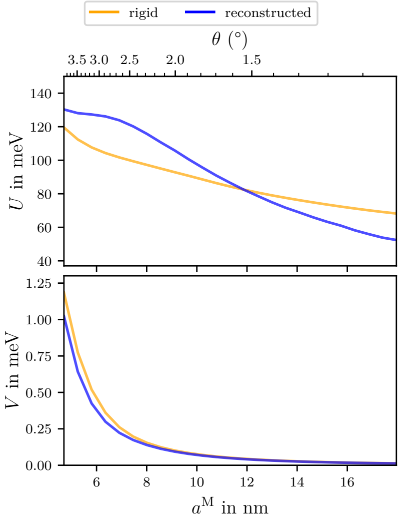
Hopping parameter:

$$t_n = \frac{1}{N} \sum_{\boldsymbol{Q}} \mathrm{e}^{\mathrm{i}(\boldsymbol{R_0} - \boldsymbol{R}_n)\boldsymbol{Q}} E_{\boldsymbol{Q}}$$

Hubbard U parameter:

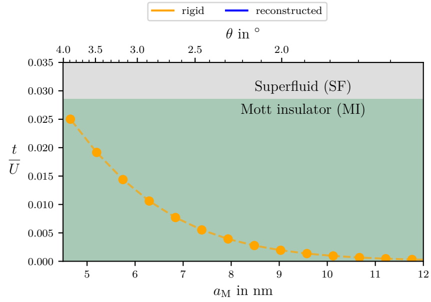
$$U_n = \int \int_{\mathbb{R}^2} |\omega_{\boldsymbol{R}_0}(\boldsymbol{r})|^2 |\omega_{\boldsymbol{R}_n}(\boldsymbol{r}')|^2 \tilde{U}(\boldsymbol{r} - \boldsymbol{r}') \mathrm{d}^2 r \mathrm{d}^2 r'$$

hBN is used as sub- and superstrate

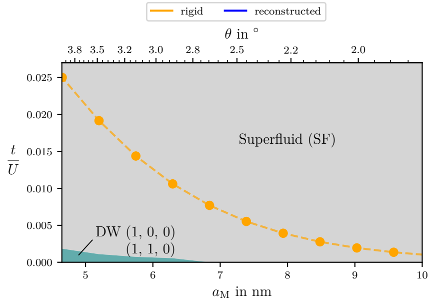


Quantum Phases of IXs

- maximal one boson per lattice site
- material system: MoS_2/WS_2
- relaxed: phases are pushed deeper into MI region



filling: $\varrho = 1$



filling: $\varrho = \frac{1}{3}, \frac{2}{3}$

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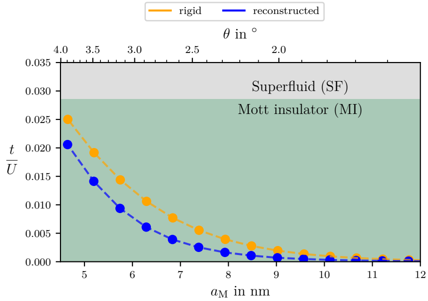

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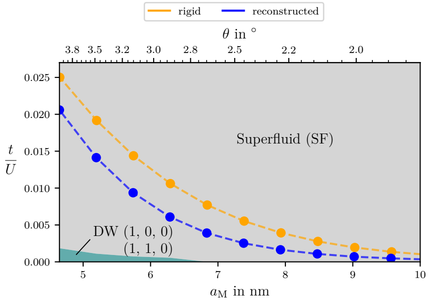

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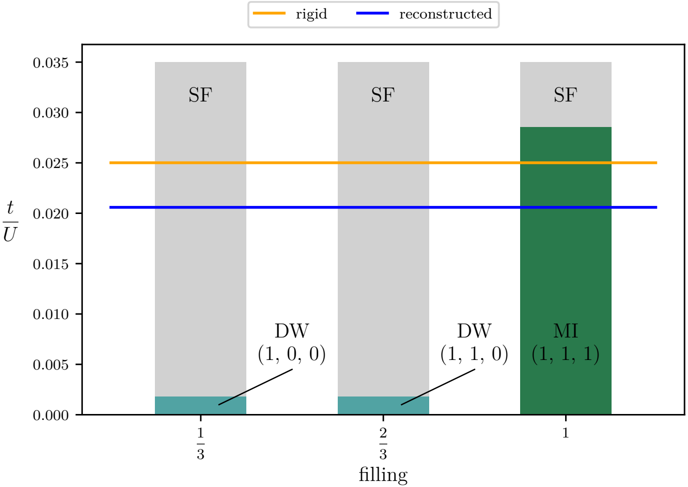
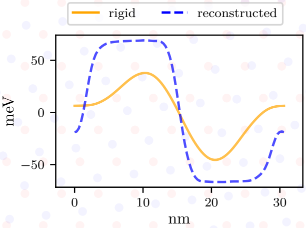
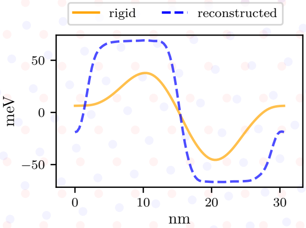
Quantum Phases of IXs

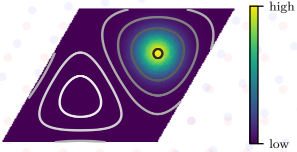


Conclusion

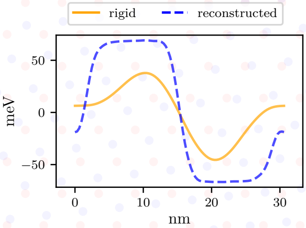


Conclusion

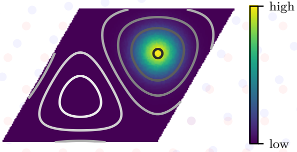




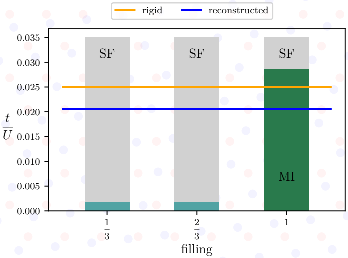
Conclusion



rigid
reconstructed
50
0
−50
meV
0
10
20
30
nm

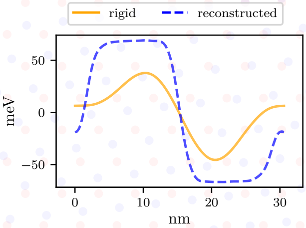


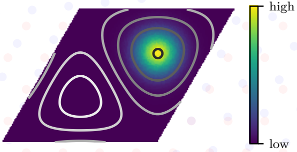
high
low

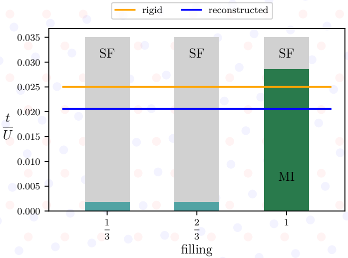


rigid
reconstructed
0.035
0.030
0.025
0.020
0.015
0.010
0.005
0.000
t/U
SF
SF
SF
MI
1/3
2/3
1
filling

Conclusion







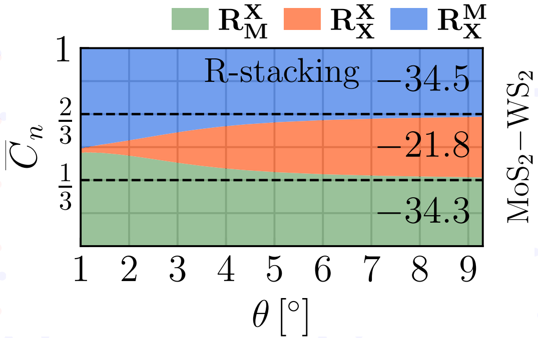
Thank you for listening!

Outlook

- cluster mean-field [1] or Monte-Carlo methods [2]
- finite-temperature theory by using the thermal average [1]
- include description of exciton-polaritons [3]

[1] Malakar *et al.* Phys. Rev. B 102, 184515 (2020)
[2] Bogner *et al.* Eur. Phys. J. B 92, 111 (2019)
[3] Byrnes *et al.* Phys. Rev. B 81, 205312 (2010)

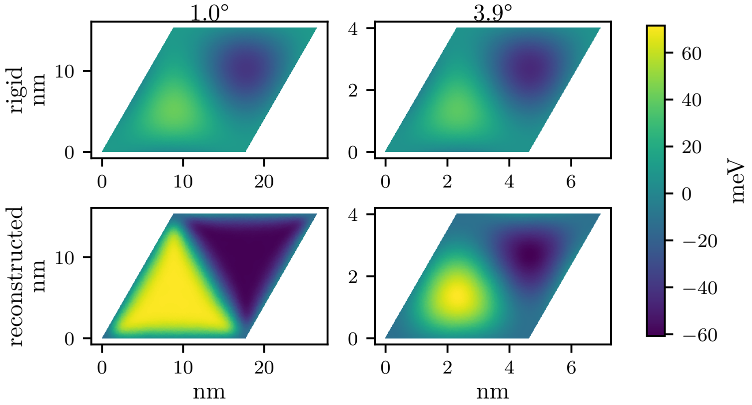
Atomic Reconstructions



Band gap as a function of in-plane distance between adjacent metal atoms and interlayer distance:

$$E_g = E_g(\Delta r_m, d(\Delta r_m))$$

Moiré Potential



IX Hamiltonian

Effective single particle Hamiltonian for IXs:

$$H = -\frac{\hbar^2}{2M}\Delta_{\boldsymbol{r}} + V^{\mathrm{M}}(\boldsymbol{r})$$

Bloch functions:

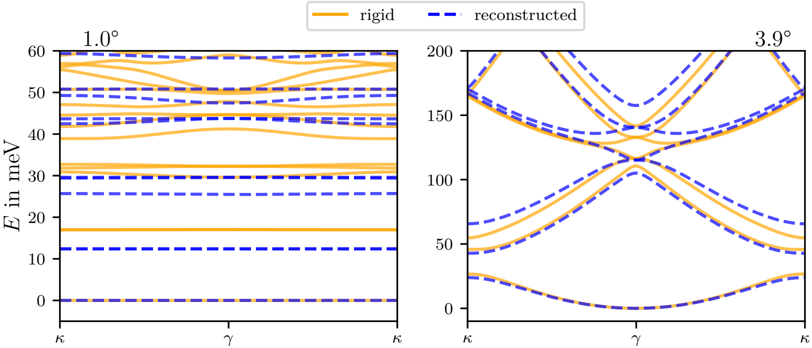
$$\chi_{\boldsymbol{Q}}^{(\alpha)} = \frac{1}{\sqrt{V}}\sum_{\boldsymbol{G}^{\mathrm{M}}} c_{\boldsymbol{Q}-\boldsymbol{G}^{\mathrm{M}}}^{(\alpha)} \mathrm{e}^{\mathrm{i}(\boldsymbol{Q}-\boldsymbol{G}^{\mathrm{M}})\boldsymbol{r}}$$

Wannier functions as IX wave functions:

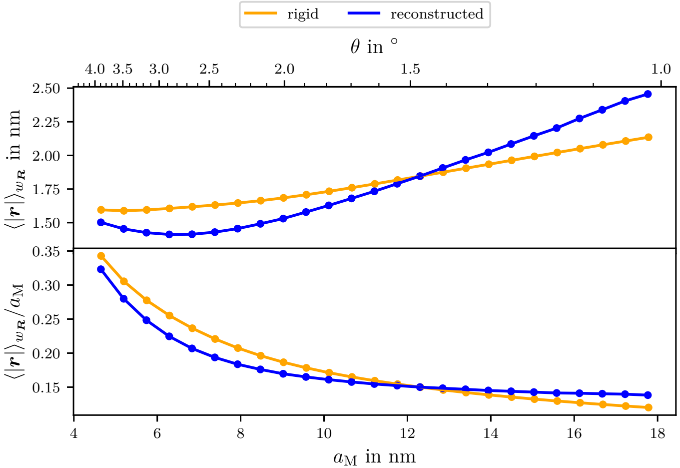
$$\omega_{\boldsymbol{R}}(\boldsymbol{r}) = \frac{1}{\sqrt{N}}\sum_{\boldsymbol{Q}} \mathrm{e}^{-\mathrm{i}\boldsymbol{Q}\boldsymbol{R}}\chi_{\boldsymbol{Q}}(\boldsymbol{r})$$

Band Structure

$$\hat{H} = -\frac{\hbar^2}{2M}\Delta + V^{\mathrm{M}}(\boldsymbol{r})$$



Quantum Phases of IXs



Bose-Hubbard Model

Extended Bose-Hubbard Hamiltonian:

$$H = -t \sum_{\langle ij \rangle} \hat{b}_i^\dagger \hat{b}_j + \frac{U}{2} \sum_i \hat{n}_i(\hat{n}_i - 1) - \mu \sum_i \hat{n}_i + \frac{V}{2} \sum_{\langle ij \rangle} \hat{n}_i \hat{n}_j$$

Mean-field approximation:

$$\varphi_i = \left\langle \Psi \middle| \hat{b}_i \middle| \Psi \right\rangle$$

$$\varrho_i = \langle \Psi \,|\, \hat{n}_i \,|\, \Psi \rangle$$

$$\begin{aligned} \hat{b}_i^\dagger \hat{b}_j &= (\varphi_i^* + \delta \hat{b}_i^\dagger)(\varphi_j + \delta \hat{b}_j) \\ &= \varphi_i^* \varphi_j + \varphi_i^* \delta \hat{b}_j + \varphi_j \delta \hat{b}_i^\dagger + \delta \hat{b}_i^\dagger \delta \hat{b}_j \end{aligned}$$

Bose-Hubbard Model

Extended Bose-Hubbard Hamiltonian:

$$H^{\mathrm{MF}}_{\mathrm{UC}} = -\, t \sum_X (-\varphi^*_X \overline{\varphi}_X + \overline{\varphi}^*_X \hat{b}_X + \overline{\varphi}_X \hat{b}^\dagger_X) + \frac{U}{2} \sum_X \hat{n}_X(\hat{n}_X - 1) - \mu \sum_X \hat{n}_X$$
$$+\, V \sum_X \bar{\varrho}_X \left(\hat{n}_X - \frac{1}{2}\varrho_X\right)$$

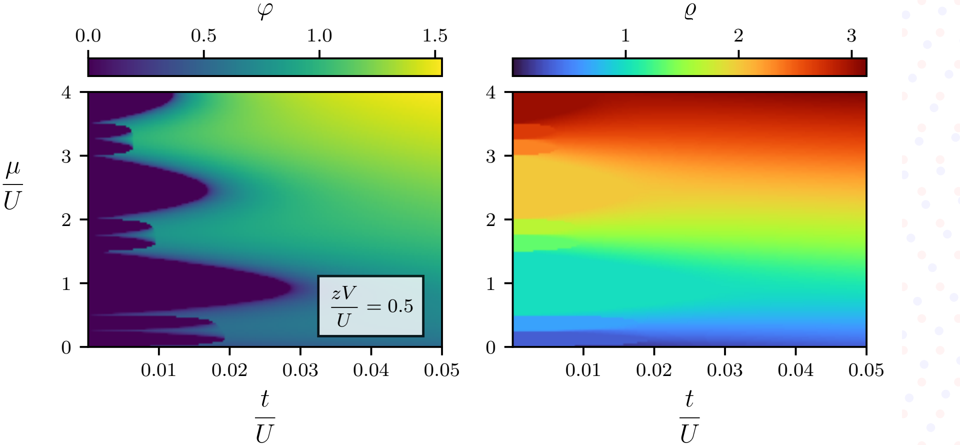
Hopping parameters:

$$t_n = \frac{1}{N} \sum_{\boldsymbol{Q}} \mathrm{e}^{\mathrm{i}(\boldsymbol{R}-\boldsymbol{R}')\boldsymbol{Q}} E_{\boldsymbol{Q}}$$

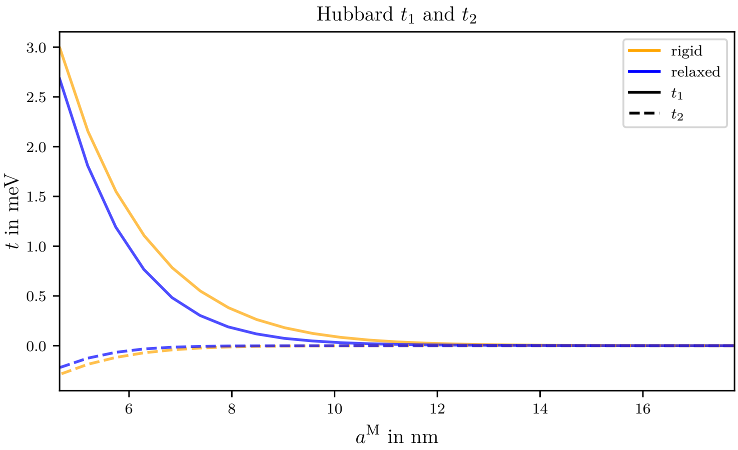
Interaction parameters:

$$U_n = \int\!\!\int_{\mathbb{R}^2} |\omega_{\boldsymbol{R}_0}(\boldsymbol{r})|^2 |\omega_{\boldsymbol{R}_n}(\boldsymbol{r}')|^2\, \tilde{U}(\boldsymbol{r} - \boldsymbol{r}')\, \mathrm{d}^2 r\, \mathrm{d}^2 r'$$

Bose-Hubbard Model



Hopping Parameter



Dipole-Dipole Interaction

Dipole-dipole repulsive interaction:

$$\tilde{U}(r) = \frac{e^2}{2\pi\varepsilon_\mathrm{r}\varepsilon_0}\left(\frac{1}{r} - \frac{1}{\sqrt{r^2+d^2}}\right)$$

Transformation to momentum space:

$$\tilde{U}(q) = \frac{e^2}{\varepsilon_0\varepsilon_\mathrm{r}}\frac{1}{q}\left(1-\mathrm{e}^{-dq}\right)$$

$$\varepsilon_\mathrm{r}(q) \quad \rightarrow \quad \varepsilon_\mathrm{TMD}\frac{1-\tilde{\varepsilon}_1\alpha-\tilde{\varepsilon}_2\alpha+\tilde{\varepsilon}_1\tilde{\varepsilon}_2}{1+\tilde{\varepsilon}_1\alpha+\tilde{\varepsilon}_2\alpha+\tilde{\varepsilon}_1\tilde{\varepsilon}_2}$$

$$\tilde{\varepsilon}_1 = \frac{1-\varepsilon_\mathrm{hBN}}{1+\varepsilon_\mathrm{hBN}} \quad , \quad \tilde{\varepsilon}_2 = \frac{\varepsilon_\mathrm{TMD}-1}{\varepsilon_\mathrm{TMD}+1} \quad , \quad \alpha = \mathrm{e}^{-qh_\mathrm{TMD}}$$

Dipole-Dipole Interaction

Fromfactor accounting the finite width of the bilayer[1]

$$F(q) = \frac{2}{\pi} \arctan\left(\frac{\pi}{q h_{\mathrm{eff}}}\right)$$

$$\tilde{U}(\boldsymbol{r}) = \frac{1}{2\pi}\frac{e^2}{\varepsilon_0}\frac{1}{\varepsilon_{\mathrm{TMD}}}\int_0^\infty F(q)\left(1-\mathrm{e}^{-dq}\right)\frac{1+\tilde{\varepsilon}_1\alpha+\tilde{\varepsilon}_2\alpha+\tilde{\varepsilon}_1\tilde{\varepsilon}_2}{1-\tilde{\varepsilon}_1\alpha-\tilde{\varepsilon}_2\alpha+\tilde{\varepsilon}_1\tilde{\varepsilon}_2} J_0(qr)\,\mathrm{d}q$$

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