Ab initio Study of 2D Materials as Photocathode Capping Layers and Potential Photocathodes

Photocathode Physics for Photoinjectors

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Joint work with Johannes Kevin Nangoi, Tomás Arias

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- **Capping Layers as 2D Materials**
- Theory of Impact of Capping Layers on Quantum Efficiency (QE) and Mean-Transverse Energy (MTE)
- Results: Graphene, hBN, 1H-$NbSe_2$

Why Scattering States are Important

2D Materials: Photocathode capping layers

Photocathode surfaces are prone to damage due to various processes including:

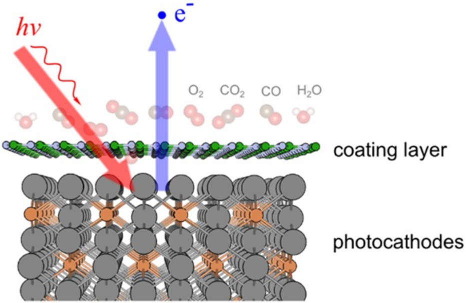

Figure: DOI: 10.1038/s41699-018-0062-6

Why Scattering States are Important

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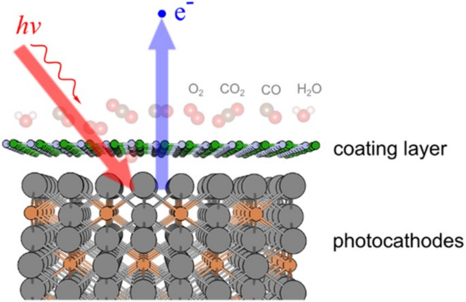

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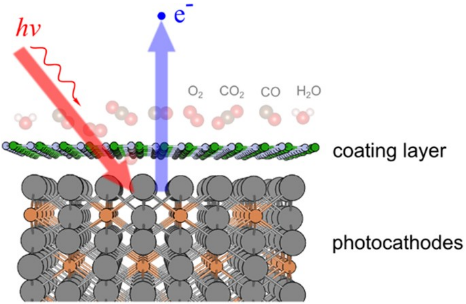

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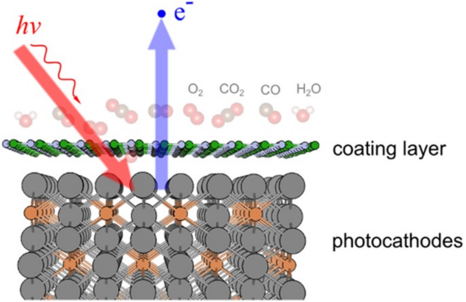

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Why Scattering States are Important

2D Materials: Photocathode capping layers

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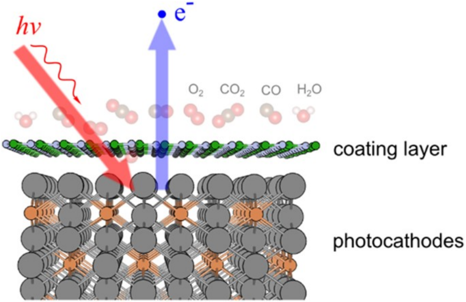
- general chemical contaminants

These particular processes can be lessened by introducing *capping layers*.



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Why Scattering States are Important

Graphene Capping Layer

Liu et al. showed that graphene is a good capping layer for copper photocathodes.

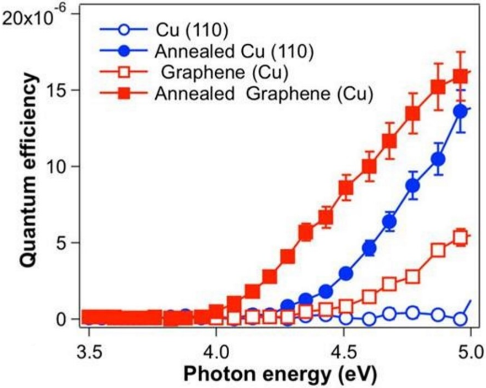

Figure: Adapted from Liu et al., DOI: 10.1063/1.4974738

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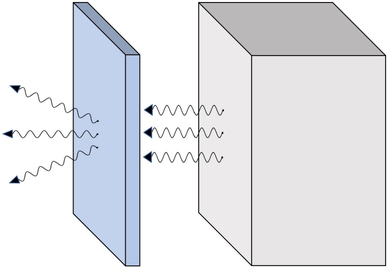
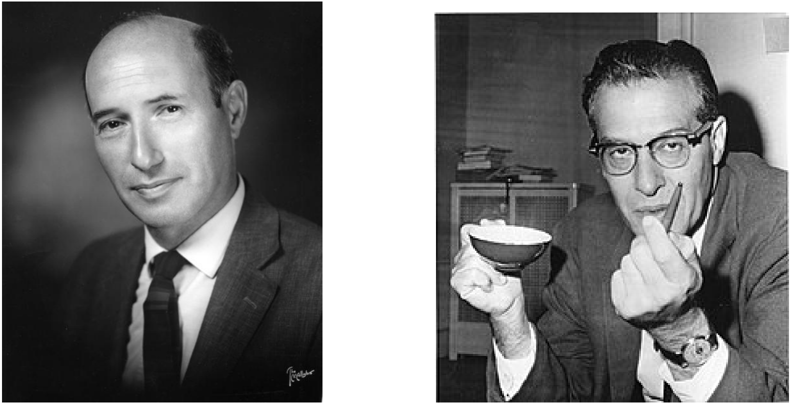
Why Scattering States are Important

Electron Transparency and Reflectivity

- Mean-transverse energy (MTE) and quantum efficiency (QE) are modulated by capping layers
- We need electron **transparency** and **reflectivity**
- Transmission and reflection can be taken directly from **scattering states**



Go Retro! Solving for Scattering States



Go Retro! Solving for Scattering States

Review: Scattering Theory

If we introduce a perturbing potential $V(\mathbf{r})$ to an unperturbed Hamiltonian H, then the new eigenstate is:

$$\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} + \int d^3r' \; G_{\mathbf{k}}(\mathbf{r}-\mathbf{r}')V(\mathbf{r}')\psi_{\mathbf{k}}(\mathbf{r}') \; (\textbf{Lippmann-Schwinger})$$

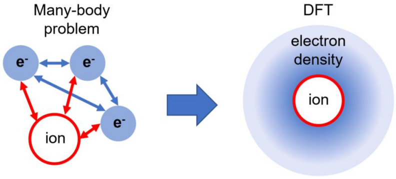
$$G_{\mathbf{k}}(\mathbf{r}) = -\frac{m}{2\pi\hbar^2}\frac{e^{ikr}}{r} \; (\textbf{Green's Function})$$

In practice, $V(\mathbf{r})$ is taken from density-functional theory (DFT)

Density Functional Theory (DFT)

1/2

Solve the many-body problem by minimizing the ground-state energy functional (of electron density).



Kohn-Sham Auxilliary System

$$-\frac{\nabla^2}{2}\psi_i(\mathbf{r}) + V^{SC}(\mathbf{r})\psi_i(\mathbf{r}) = \epsilon_i\psi_i(\mathbf{r})$$

Many-body Problem → Single-particle QM

Density Functional Theory (DFT)

2/2

Typical DFT software packages solve the Kohn-Sham states with periodic boundary conditions.

Pros:

- Plane-wave basis set → Fast Fourier Transforms

Cons:

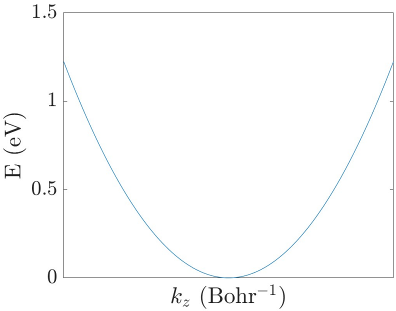
- **(2D materials) Difficulties with scattering states**

Fixing the PBC Issue

Trying to use PBCs

Graphene Energy Spectrum

$(k_x = k_y = 0)$



This is what we want

Graphene Band Structure

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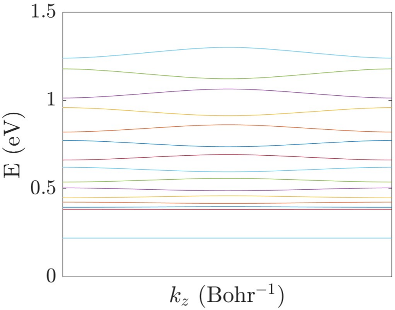
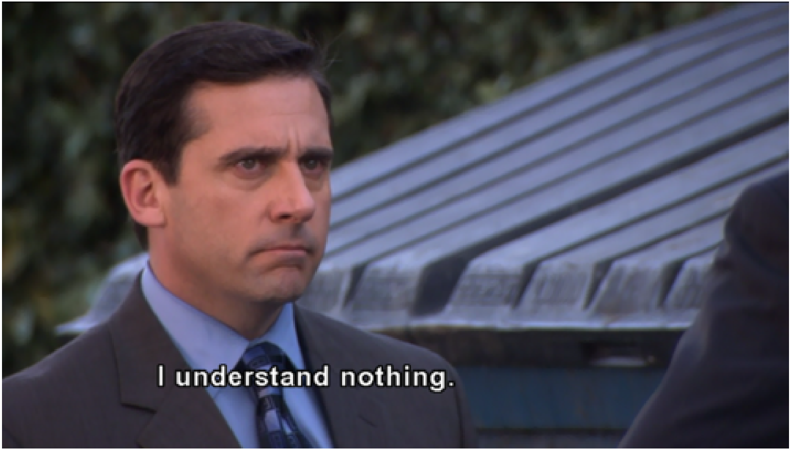


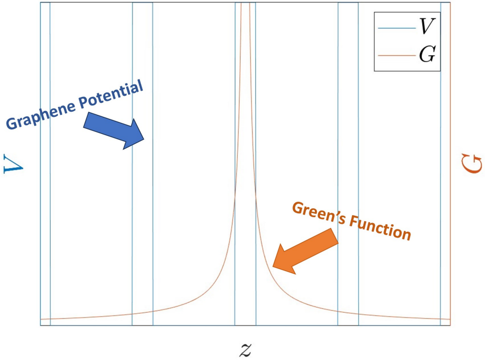
This is what we get

BUT WHY ISN'T IT COMPATIBLE???



A Modified Green's Function

- The Green's Function is proportional to $1/r$ → cannot neglect interactions with periodic images!



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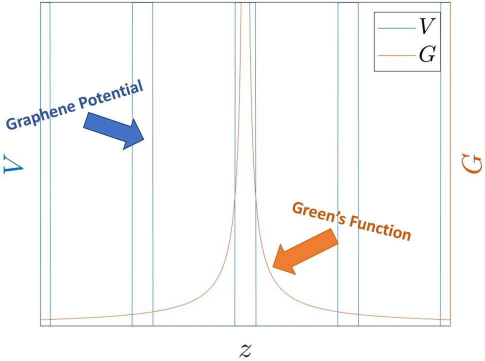
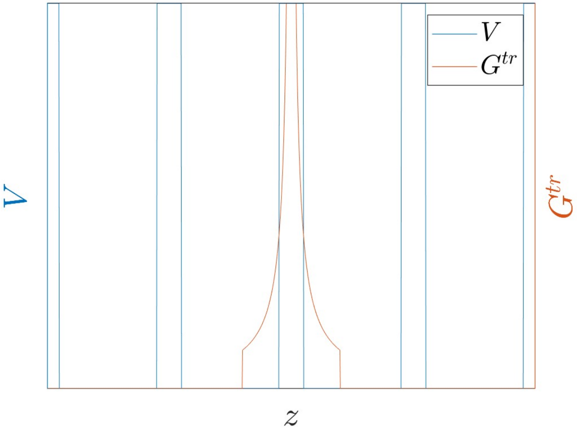


- **Problem:** How do we cut out interactions with periodic images?

A Modified Green's Function

Answer: Cut off the Green's Function past a fixed distance!

$$G_{\mathbf{k}}(\mathbf{r}) \longrightarrow G_{\mathbf{k}}^{tr}(\mathbf{r}) = G_{\mathbf{k}}(\mathbf{r})\Theta(L - |z|)$$



The Technique

Simple method!

1. Replace G with G^{tr}
2. Extract the self-consistent potential (V^{SC}) from DFT
3. Insert these ingredients into the LS Equation and solve!

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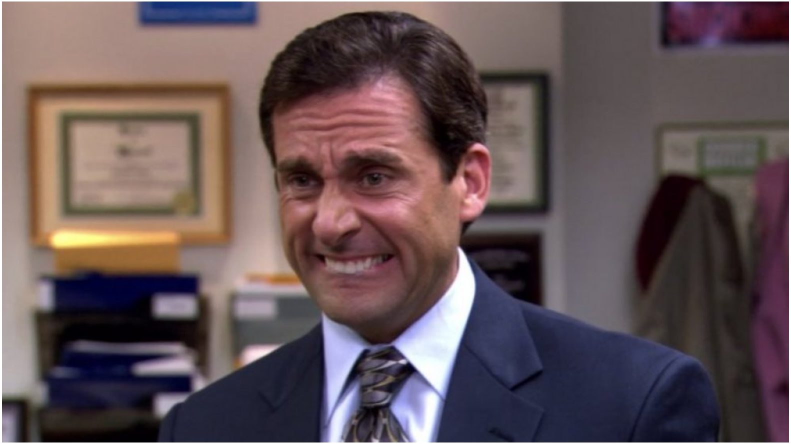
Let's go a bit deeper!

The Technique



The Technique

The Method in More Detail

Rearranging the Lippmann-Schwinger Equation with algebra (in bra-ket notation), we have:

$$(1 - \widehat{V}^{SC}\,\widehat{G}_{\mathbf{k}}^{tr})(\widehat{V}^{SC}\,|\psi_{\mathbf{k}}\rangle) = (\widehat{V}^{SC}\,|\phi_{\mathbf{k}}\rangle)$$

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Looks like $Ax = b$**!**

- Solve for $x = \widehat{V}^{SC}\,|\psi_{\mathbf{k}}\rangle$ using a suitable numerical solver
- Substitute back into the original equation: $|\psi_{\mathbf{k}}\rangle = |\phi_{\mathbf{k}}\rangle + \widehat{G}^{tr}_{\mathbf{k}}(\widehat{V}^{SC}\,|\psi_{\mathbf{k}}\rangle)$

Using the Method

With the solution, we can:

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With the solution, we can:

- Find scattering solutions at arbitrary excess energies
- Find transmission (electron transparency) and reflection (electron reflectivity) coefficients
- Create photoemission plots

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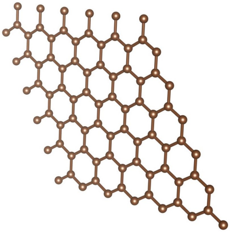
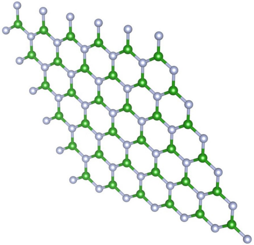
Results: Three Different Materials

Graphene



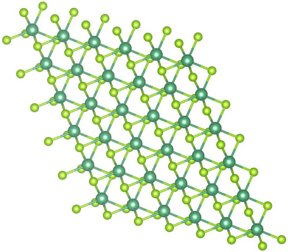
- Prototypical 2D material
- Experimental data exists

Hexagonal Boron Nitride



- Same unit cell as graphene
- Slightly bigger bond length
- Material of recent interest

1H-Niobium Diselenide



- 3-atom thick material
- Heavier atoms
- Better against ion back-bombardment

Results: Testing Graphene

Experimental Results

Liu et al. showed that graphene is a good capping layer for copper photocathodes.

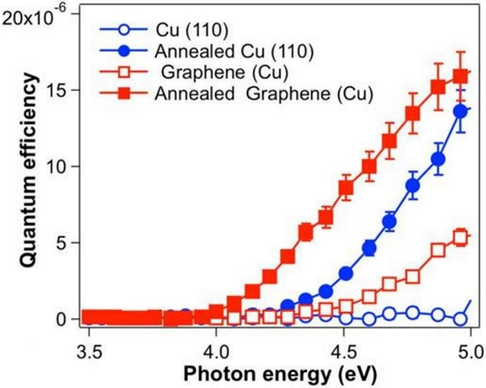
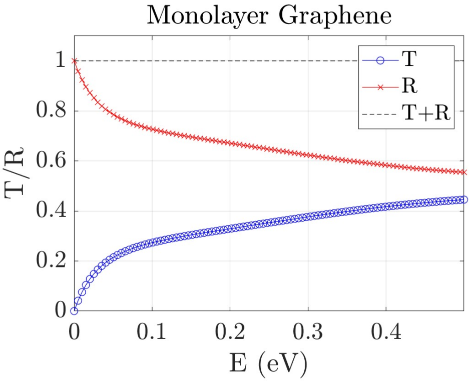


Figure: Adapted from Liu et al., DOI: 10.1063/1.4974738

Results: Graphene

Electron Transmission and Reflection

We first study the **electron transparency** at normal incidence



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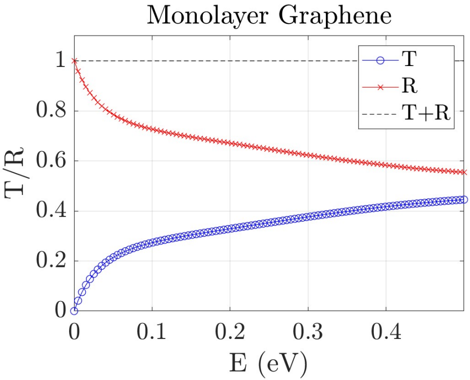

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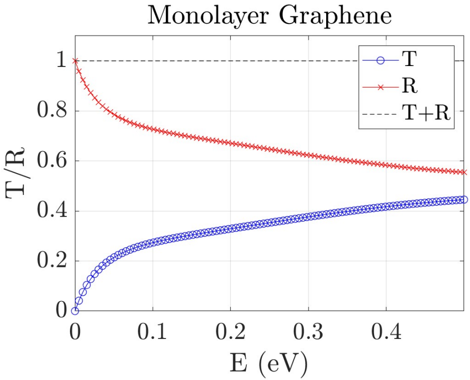
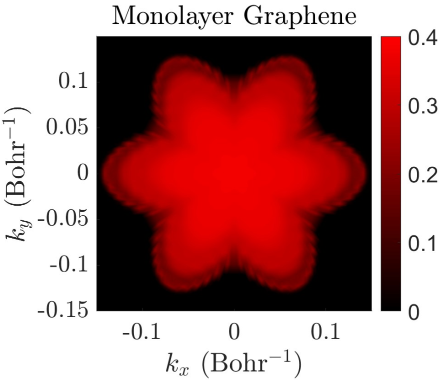


- A modest electron transparency predicted
- The electron transparency is (possibly) high enough to offset the reflection from oxidation layers

Results: Graphene

Angular Dependency for Electron Transmission

The **angular distribution of electron transparency** for graphene at 0.3 eV:



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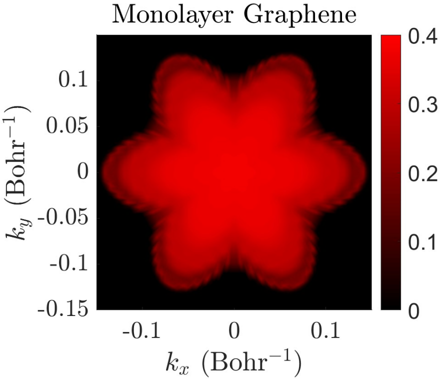

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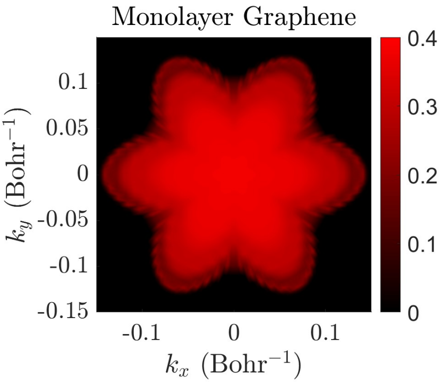

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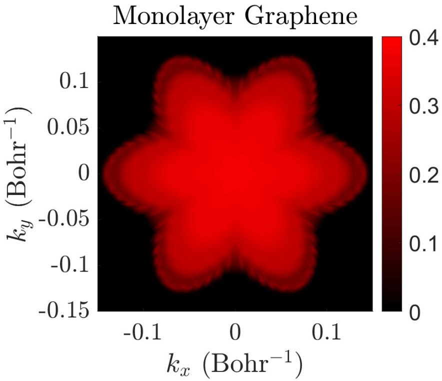

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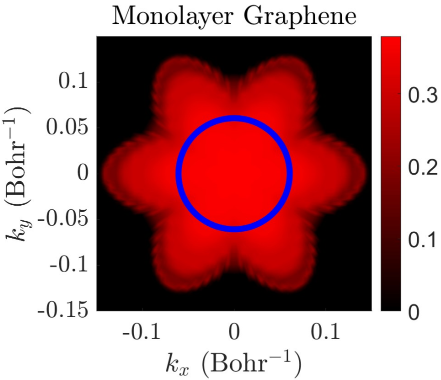

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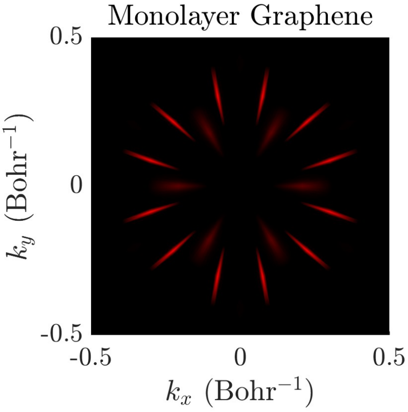
Using the Truncated Green's Function Technique

Photoemission with 12 eV photons (4.67 eV max excess energy):



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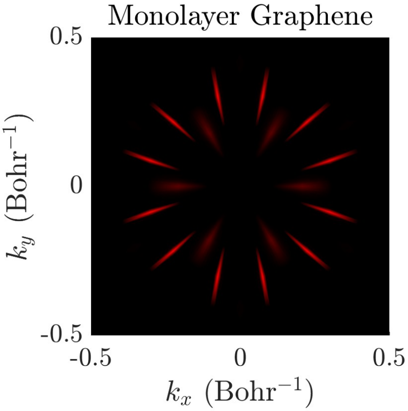

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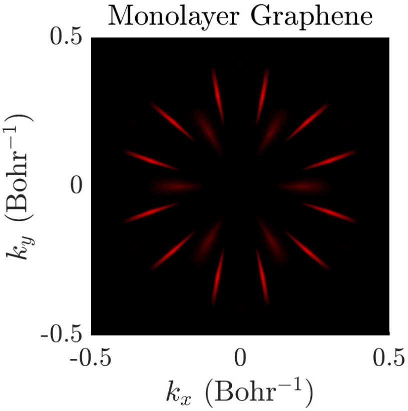

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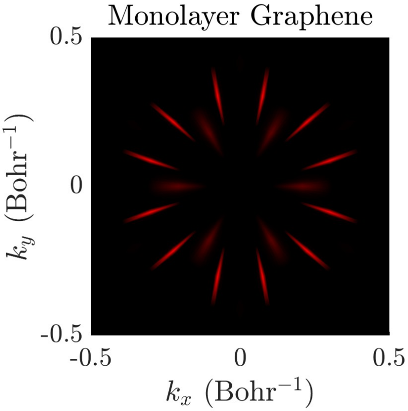

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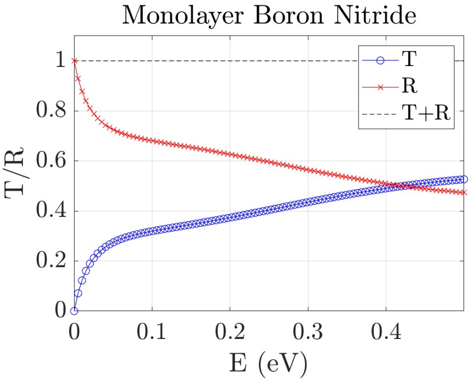
- six-fold rotational symmetry

Results: hBN

Electron Transmission and Reflection



Results: hBN

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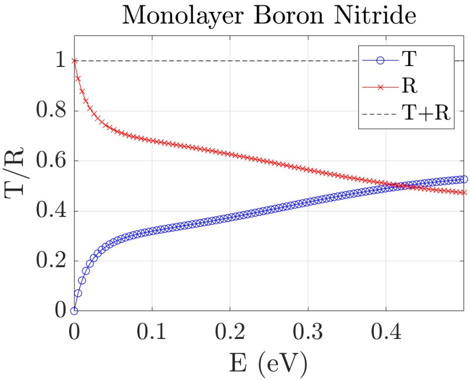
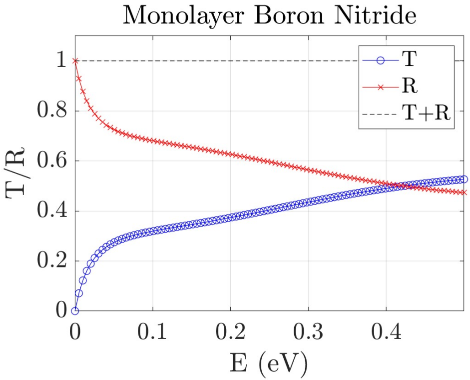


- Transmission and reflection have similar shape to graphene

Results: hBN

Electron Transmission and Reflection



- Transmission and reflection have similar shape to graphene
- Transmission is marginally better

Results: hBN

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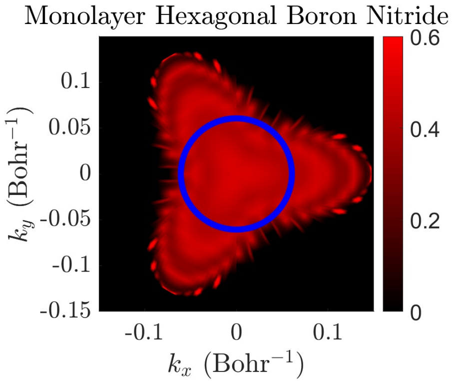
Electron transmission at 0.3 eV excess energy:



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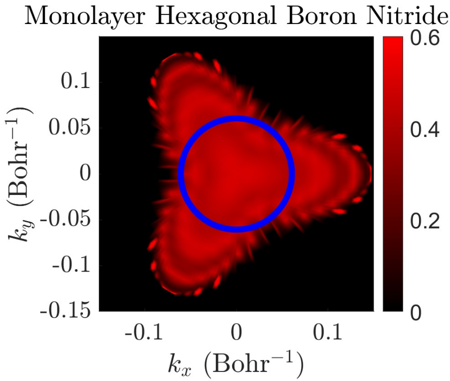

- Three-fold rotational symmetry

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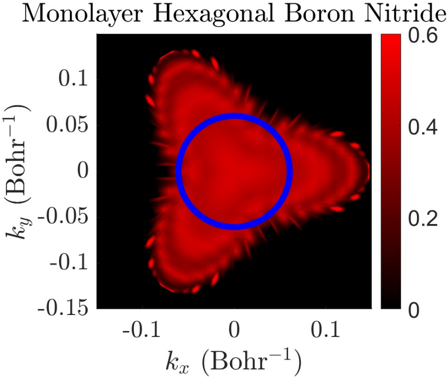

- Three-fold rotational symmetry
- Nearly constant transmission

Results: hBN

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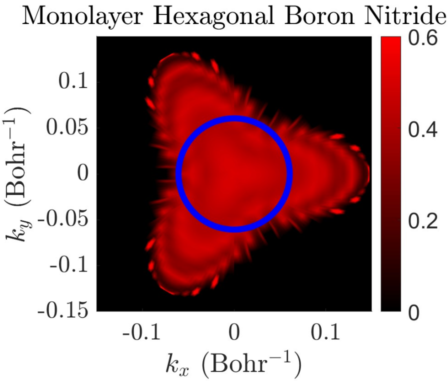

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- Transmit ≤ 50 meV transverse energy (blue) electrons without much distortion!

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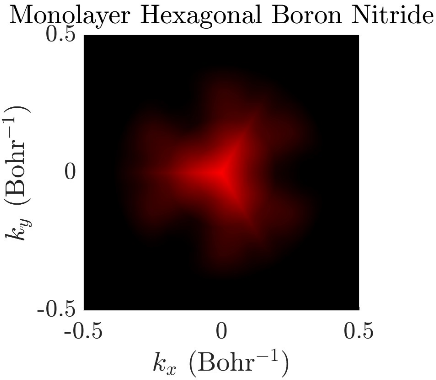
(Direct) Photoemission

Photoemission with 10 eV photons (4.1 eV max excess energy):



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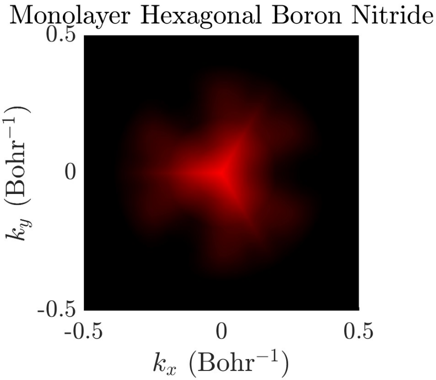

- $MTE \approx 0.373\ eV$

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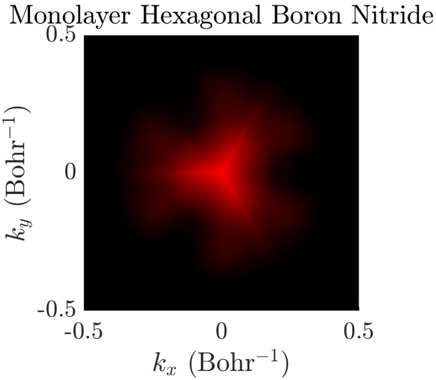

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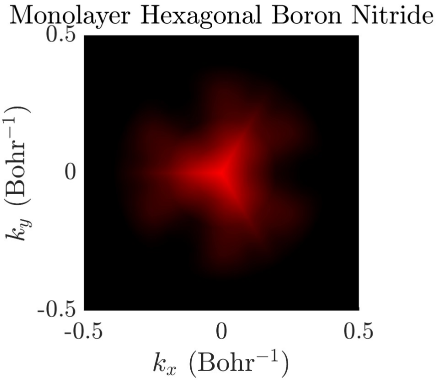
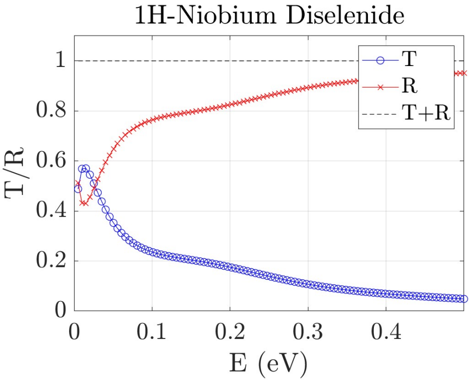


- $MTE \approx 0.373\ eV$
- Three-fold rotational symmetry
- Possibly a good photoemitter! Narrow beam produced!

Results: 1H-NbSe$_2$

Electron Transmission and Reflection



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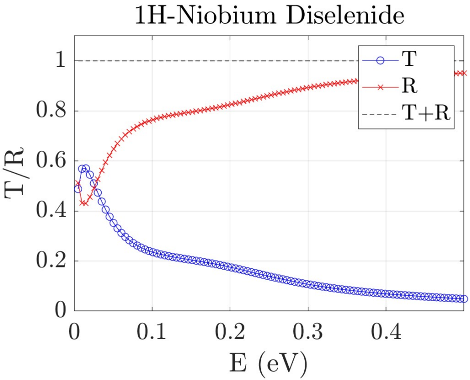

- Three-atom thick material → better protection against ion back-bombardment

Results: 1H-NbSe$_2$

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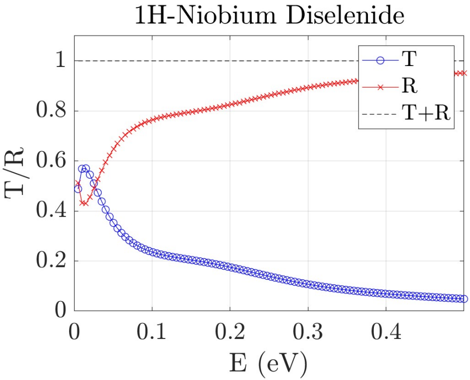

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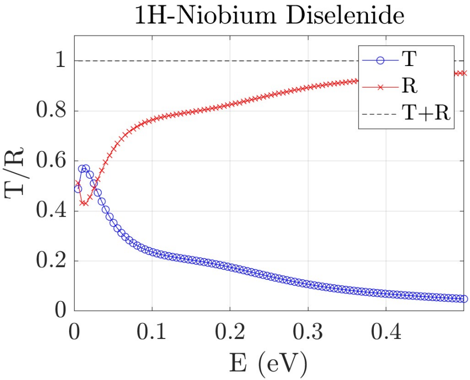
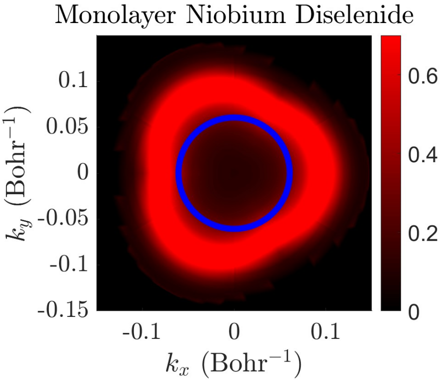


- Three-atom thick material → better protection against ion back-bombardment
- Transmission decays!
- BUT! Better transmission for $E \lessapprox 0.1$ eV!

Results: 1H-NbSe$_2$

Angular Dependency for Electron Transmission

Electron transmission at 0.3 eV excess energy



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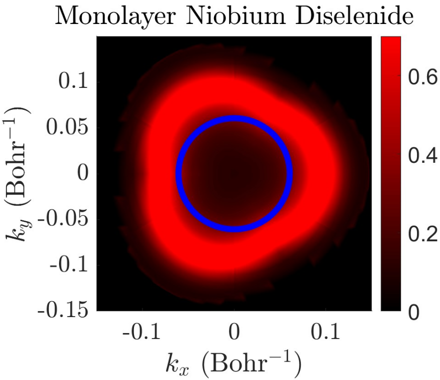

- The transmission is localized about a ring

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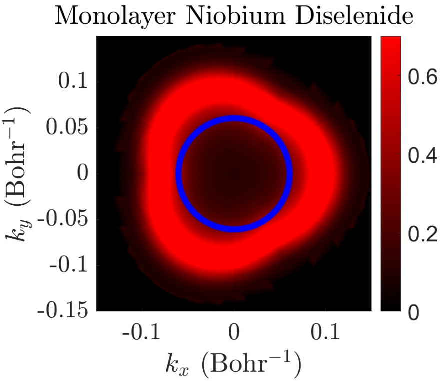

- The transmission is localized about a ring
- Three-fold rotational symmetry

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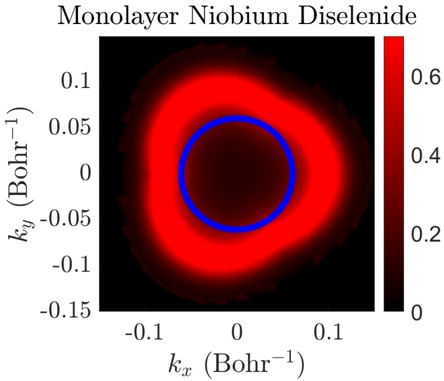
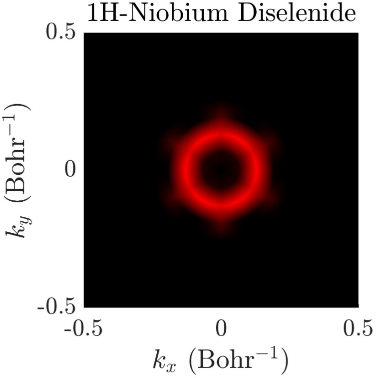


- The transmission is localized about a ring
- Three-fold rotational symmetry
- Not ideal! Highly distorted beam for ≤ 50 meV beam (blue).

Results: 1H-$NbSe_2$

(Direct) Photoemission

Photoemission with 6 eV photons (1.1 eV max excess energy):



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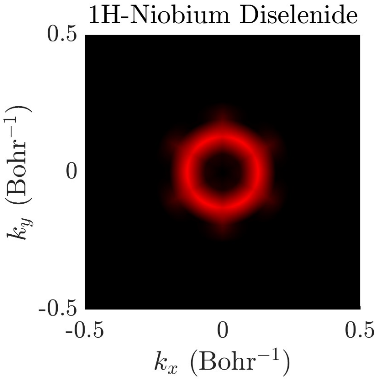

- $MTE \approx 0.239\ eV$

Results: 1H-$NbSe_2$

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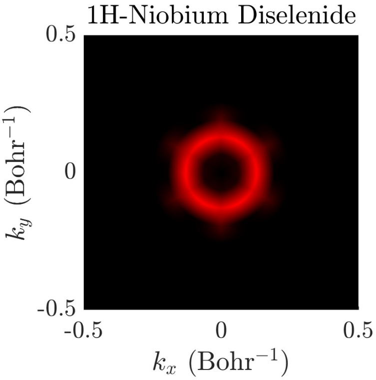

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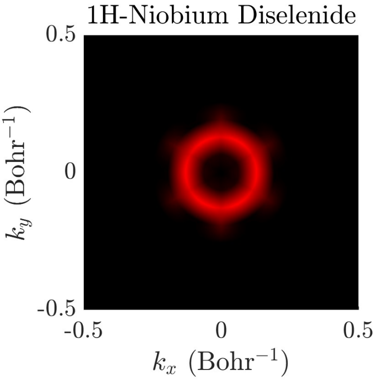

- $MTE \approx 0.239\ eV$
- Six-fold rotational symmetry
- Ideal! Want low MTE beams!

Results: Summary of Energy Distribution

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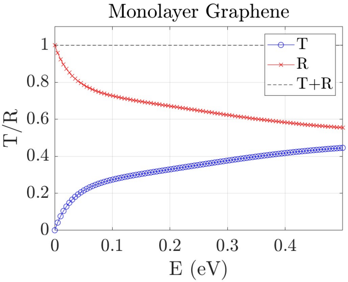

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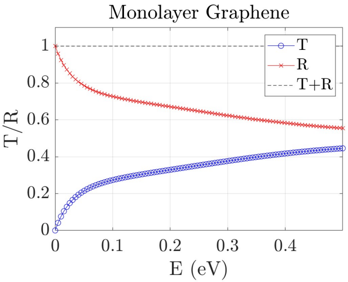

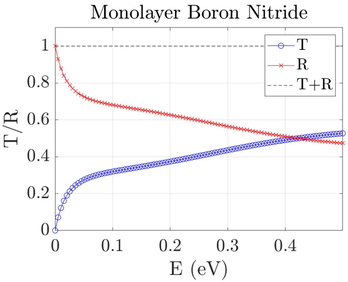

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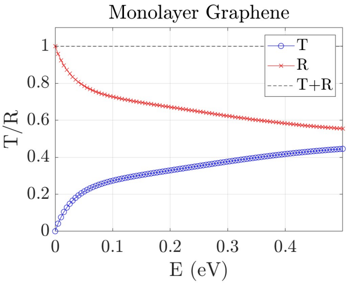

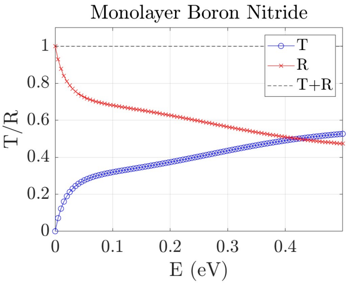

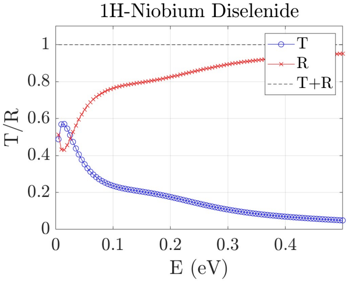

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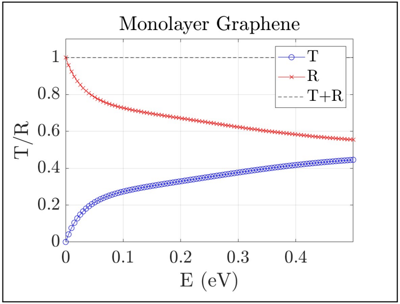
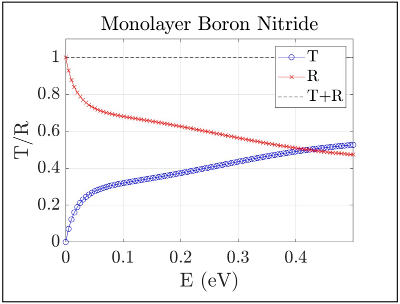
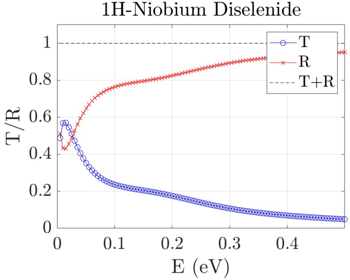

Monolayer Graphene


Monolayer Boron Nitride


1H-Niobium Diselenide

Results: Summary of Transverse Momenta Distribution

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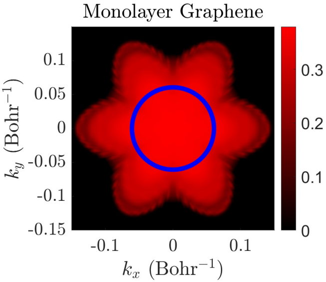

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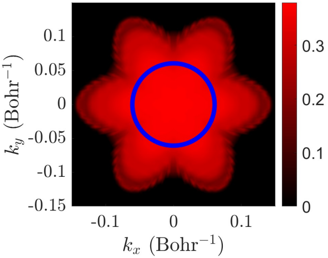

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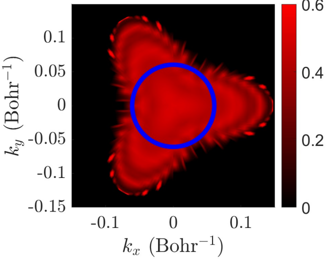

Monolayer Hexagonal Boron Nitride

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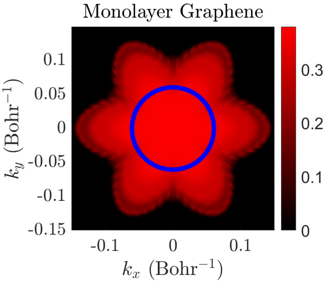
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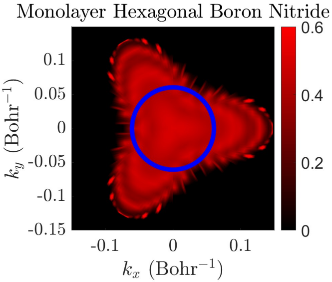
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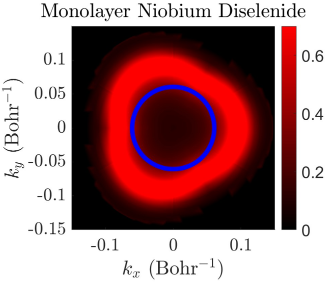
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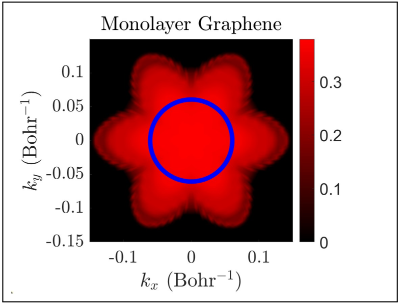
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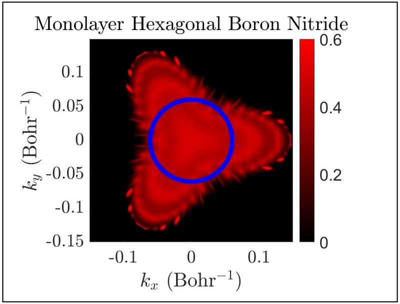
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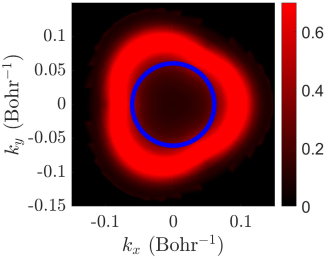

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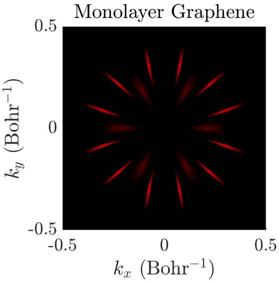

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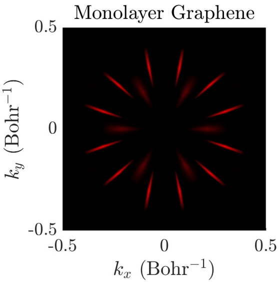

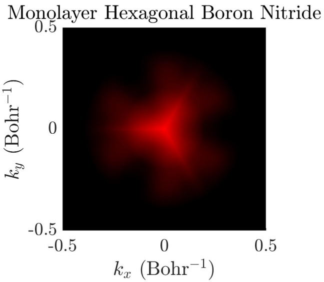

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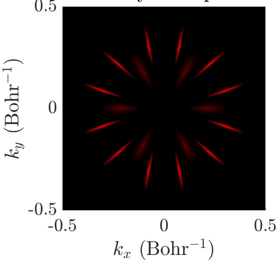
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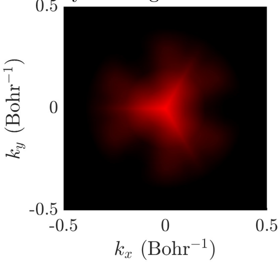
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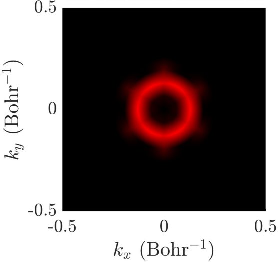
1H-Niobium Diselenide

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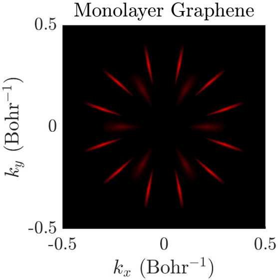
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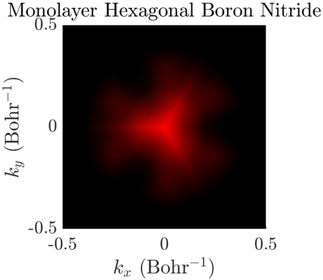
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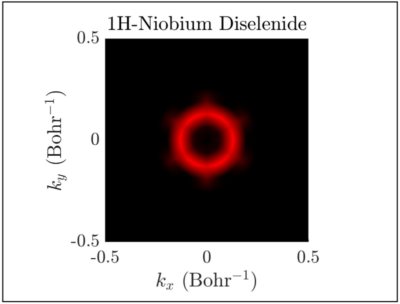
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Conclusions

Material	**Energy**	**Angular**	**Photocathode**
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hBN	☺ ($\gtrapprox 0.1$ eV)	☺	☹
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- Any suggestions?

Thank you for listening!

Tyler Wu

tcw66@cornell.edu