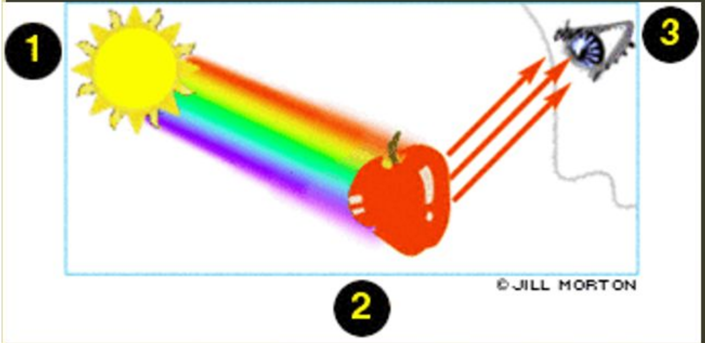
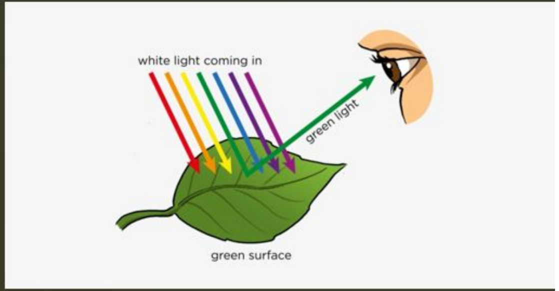
Table of Contents

- Background
- Method
- Results
- Conclusion & Significance
- Next steps

Background

How do we see the color of an object?

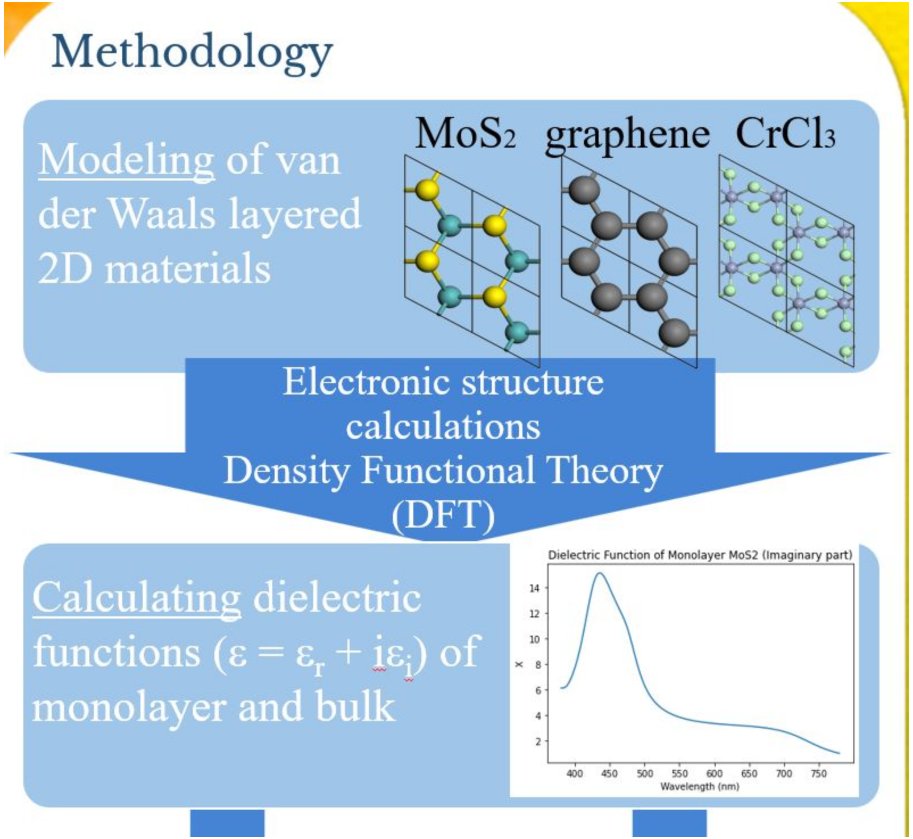




Three processes:

1. Incident white light
2. **Certain** color reflected by the object
3. Human eyes' perception

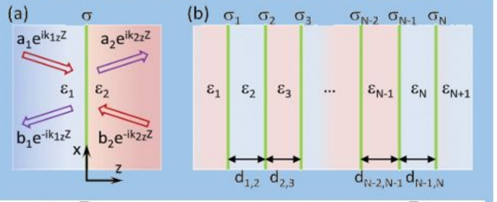
Brief Review of Method



Brief Review of Method

multi-layers

Extrapolate for multi-layers from transfer matrix method



$$\begin{bmatrix} b_1 \\ a_{N+1} \end{bmatrix} = \begin{bmatrix} M_{21}/M_{11} & (M_{11}M_{22} - M_{12}M_{21})/M_{11} \\ 1/M_{11} & -M_{12}/M_{11} \end{bmatrix} \begin{bmatrix} a_1 \\ b_{N+1} \end{bmatrix}$$

$$\mathcal{M} = D_{1\to 2}P(d_{1,2})D_{2\to 3}P(d_{2,3})\cdots P(d_{N-1,N})D_{N\to N+1}$$

$$D_{1\to 2} = \frac{1}{2}\begin{bmatrix} 1+\eta_s+\xi_s & 1-\eta_s+\xi_s \\ 1-\eta_s-\xi_s & 1+\eta_s-\xi_s \end{bmatrix} \quad P(\Delta z) = \begin{bmatrix} e^{-ik_z\Delta z} & 0 \\ 0 & e^{ik_z\Delta z} \end{bmatrix}$$

where **D** is the transmission matrix relating incident and reflected waves across the monolayer; **P** is the propagation matrix for the propagation of light in a homogenous medium; **M** is the transfer matrix relating incident and reflected waves across the multi-layer materials.

single layer/ bulk

Calculate reflectivity spectra using S Gupta et al.

$$T = \frac{4n_1n_2}{|n_1 + n_2 + \sigma_{2D}Z_{vac}|^2}$$

$$A = \frac{4n_1Re\{\sigma_{2D}\}Z_{vac}}{|n_1 + n_2 + \sigma_{2D}Z_{vac}|^2}$$

$$R = \left|\frac{n_2 - n_1 + \sigma_{2D}Z_{vac}}{n_1 + n_2 + \sigma_{2D}Z_{vac}}\right|^2$$

where **2D materials** are viewed as thin layers between two semi-infinite dielectric media slabs with normally incident and reflected waves across the interface; $\mathbf{n_{1,2}}$ are refractive indexes on the two sides of 2D materials (set to 1 as vacuum in our case); $\boldsymbol{\sigma_{2D}}$ is optical surface conductivity as a function of frequency and 2D wavevector; $\mathbf{Z_{vac}}$ is impedance of vacuum.

$$\sigma_{2D}(q=0,\omega) = i\omega\varepsilon_0(1-\varepsilon_{3D}(q=0,\omega))L$$

Brief Review of Method

CIEXYZ Color Space → sRGB

$$X = k \int_{380\text{ nm}}^{780\text{ nm}} d\lambda\, \bar{x}(\lambda) R(\lambda) S(\lambda)$$

$$Y = k \int_{380\text{ nm}}^{780\text{ nm}} d\lambda\, \bar{y}(\lambda) R(\lambda) S(\lambda)$$

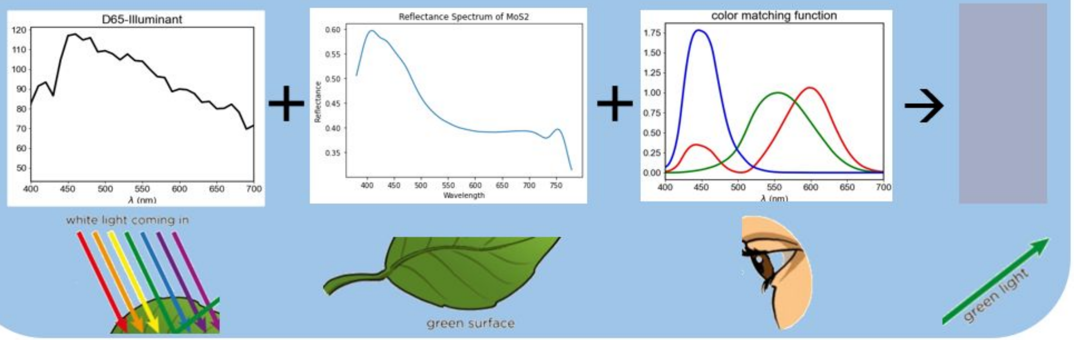
$$Z = k \int_{380\text{ nm}}^{780\text{ nm}} d\lambda\, \bar{z}(\lambda) R(\lambda) S(\lambda)$$

$$\begin{bmatrix} R_{linear} \\ G_{linear} \\ B_{linear} \end{bmatrix} = \begin{bmatrix} 3.2410 & -1.5374 & -0.4986 \\ -0.9692 & 1.8760 & 0.0416 \\ 0.0556 & -0.2040 & 1.0570 \end{bmatrix} \begin{bmatrix} X \\ Y \\ Z \end{bmatrix}$$

$$F(t) = \begin{cases} 12.92{*}t, & t \leq 0.00304 \\ 1.055{*}t^{1/2.4} - 0.055, & t > 0.00304 \end{cases}$$

$$RGB_{sRGB} = 255 * F(RGB_{linear})$$

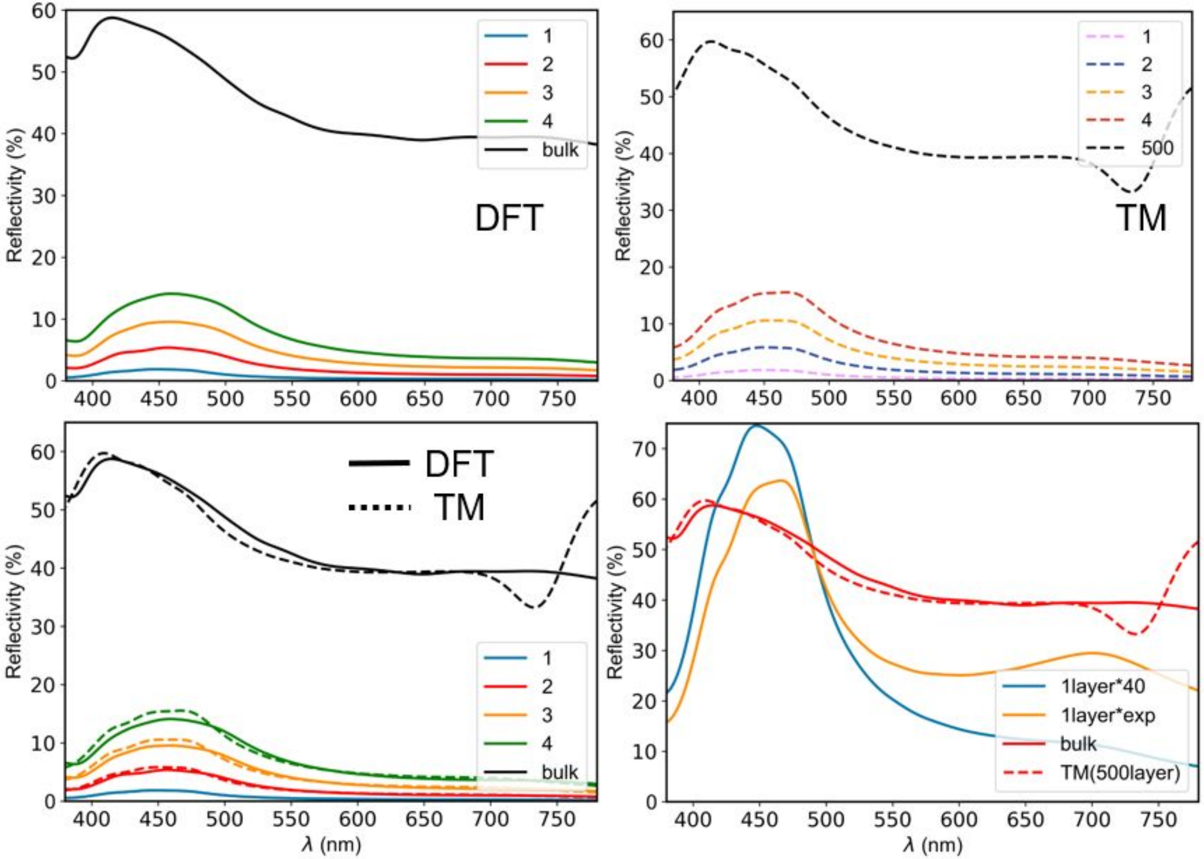
where **x(λ), y(λ) and z(λ)** are the color matching function; **R(λ)** is the reflectivity obtained by DFT/transfer matrix method; **S(λ)** is the spectral power distribution of one of the standard CIE illuminant (D65, D50…..).



Results: Table of Selected 2D Materials

Types	Materials
TMD	**MoS_2, WSe_2**
Graphene	**Graphene**
Chromium Trihalides CrX_3 (X=Cl, Br, I)	**$CrCl_3$**

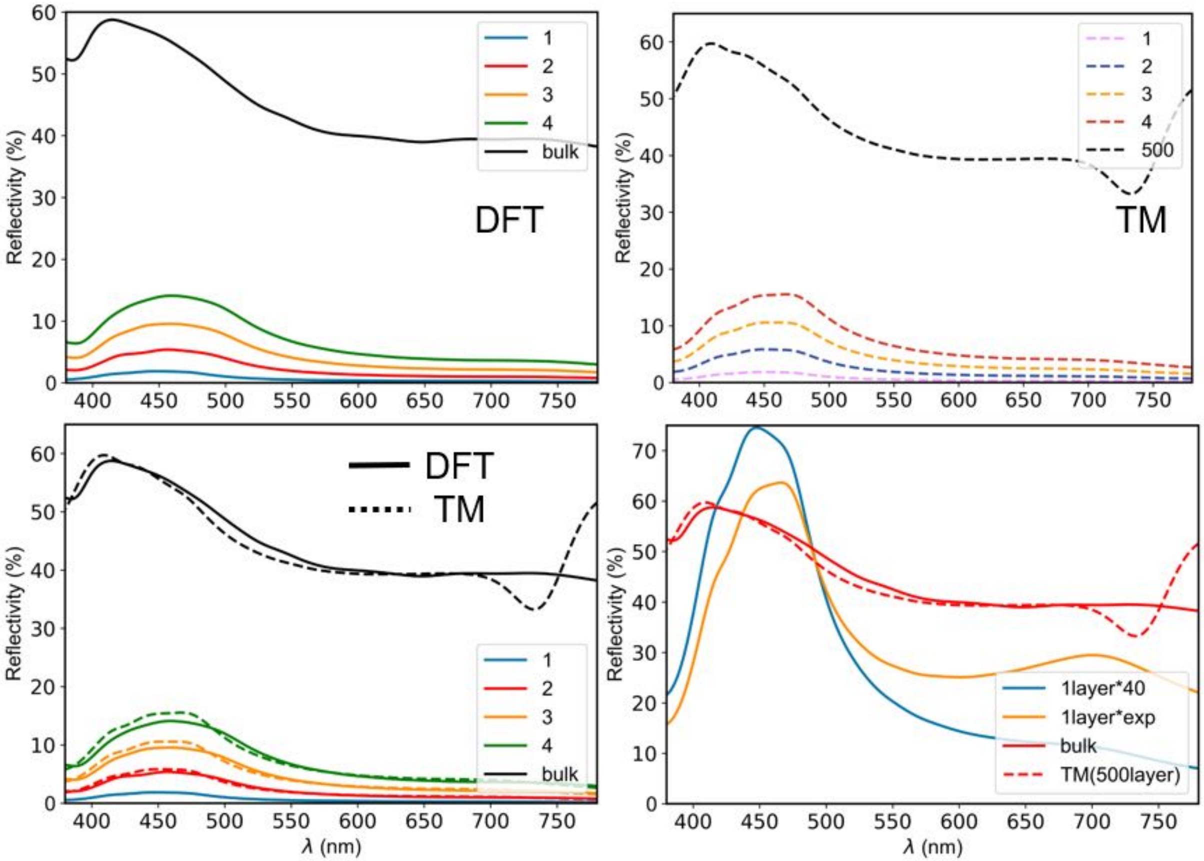
MoS2



AA stacking
Transfer matrix (TM) method:
Interlayer separation 6.81Å

credit. Qin-Kun Li

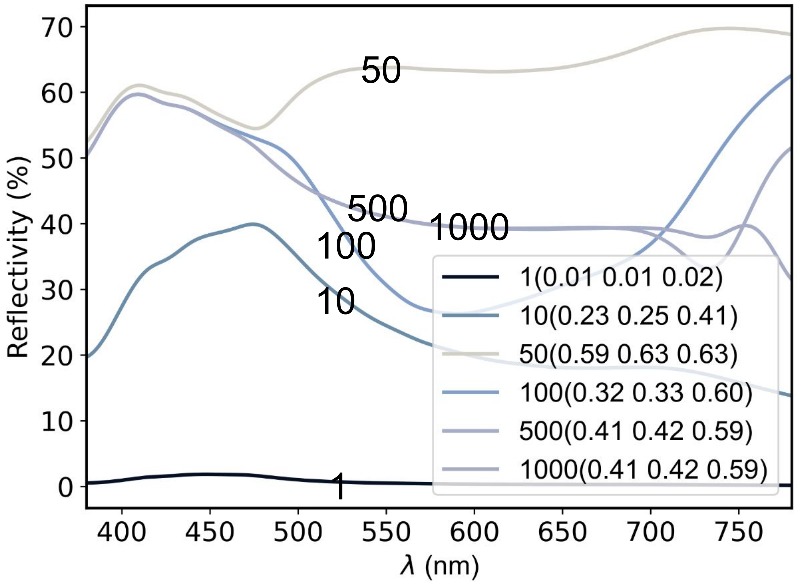
MoS2

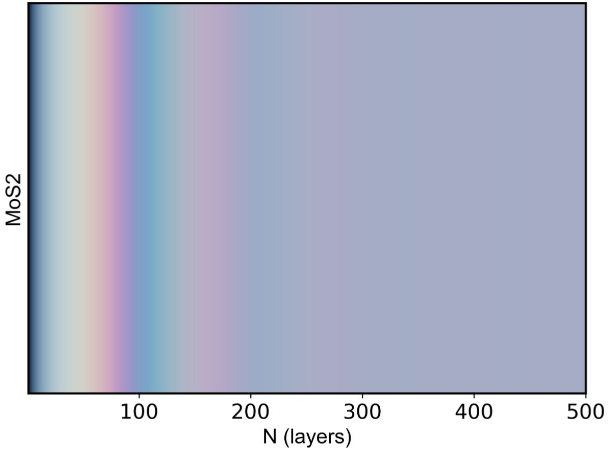


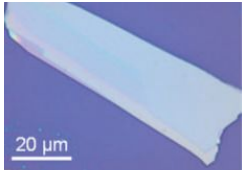
AA stacking
Transfer matrix (TM) method:
Interlayer separation 6.81Å

- TM produces good fit
  - deviation after 700 nm
- Peaks between 400-550 nm
  - expect blue/ purple
- Single layer and bulk: no linear or exponential relationship

MoS2



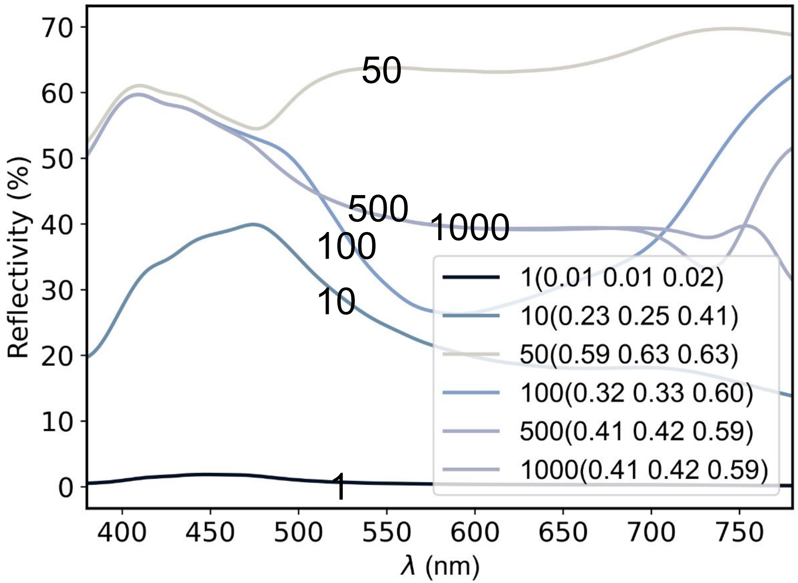


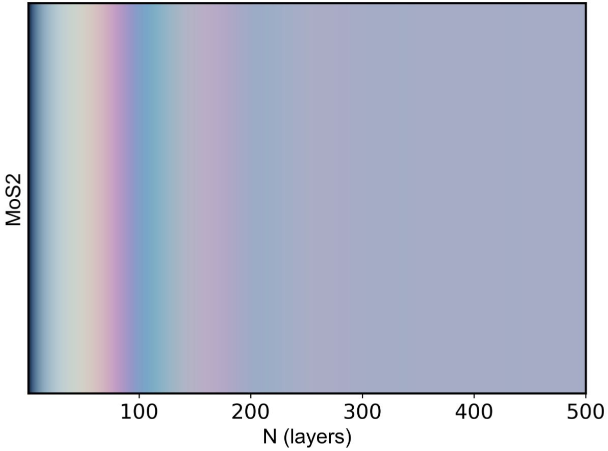


thick (>100 nm) MoS2 flake
APL Materials 2, 092509 (2014)

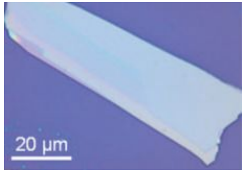
crystalline bulk MoS2 single crystal
grown by Sn flux method
CrystEngComm, 2015,17, 4026

MoS2





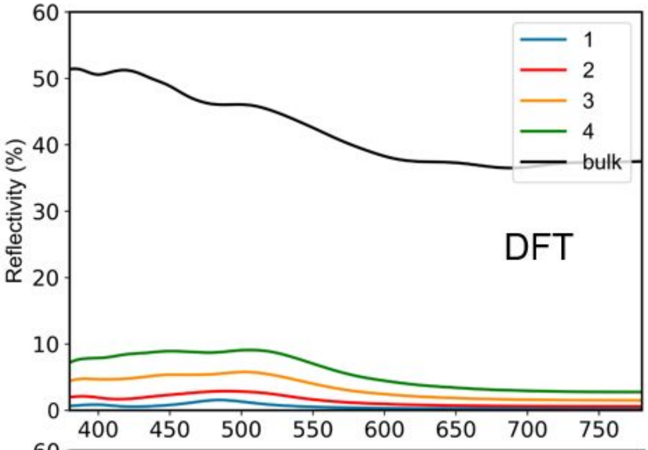
- Single layer is black
- 50 layers is creamy white
- Color is stable after ~400 layers
- **Pattern:**
  - pink/blue stripes fading periodically
  - “blending yogurt”

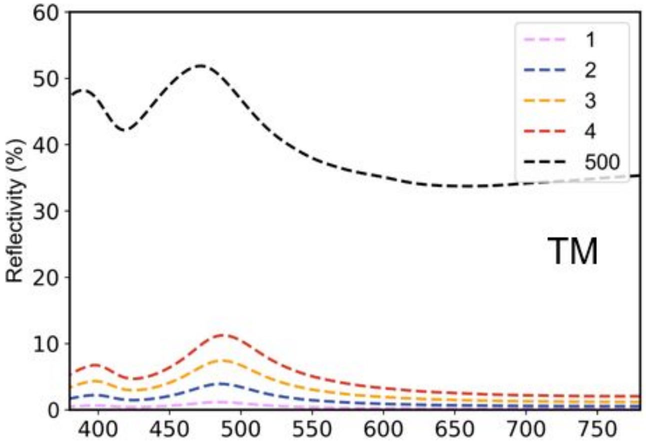


thick (>100 nm) MoS2 flake
APL Materials 2, 092509 (2014)

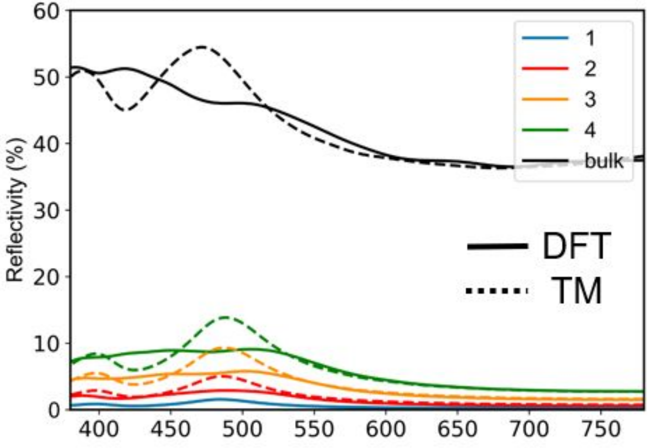
crystalline bulk MoS2 single crystal
grown by Sn flux method
CrystEngComm, 2015,17, 4026

WSe2



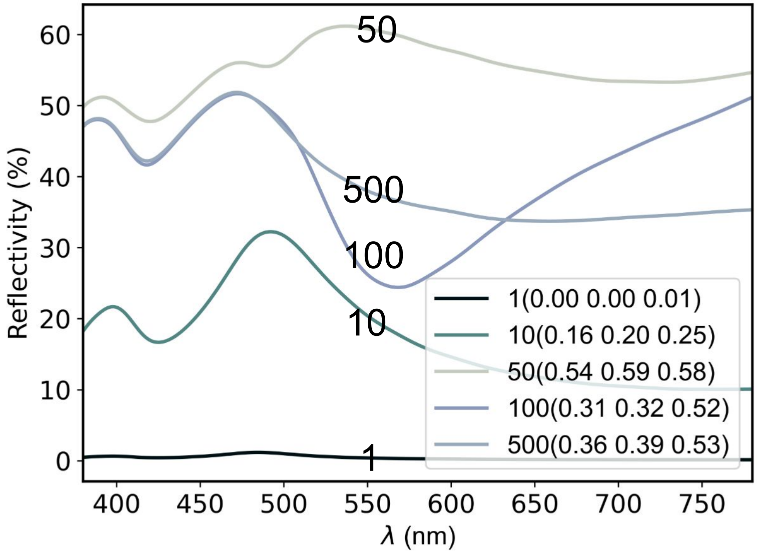


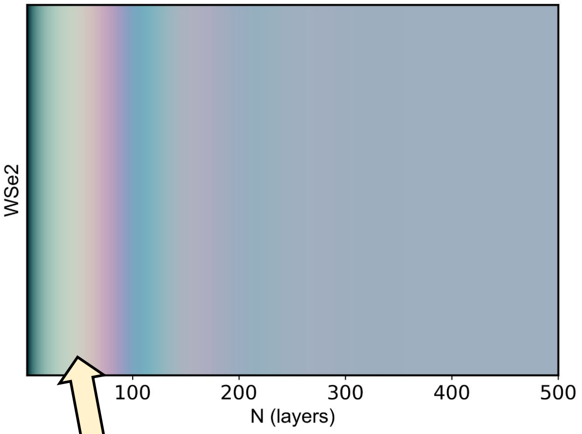
AA stacking
Transfer matrix (TM) method:
Interlayer separation 7.03Å



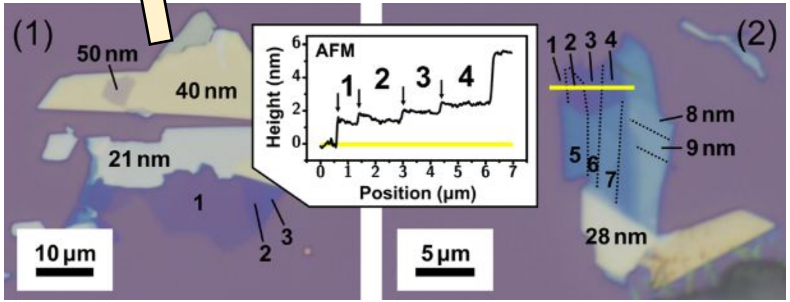
- DFT has plateau instead of peaks
- TMM produces wave-like curves
  - based on monolayer

WSe2



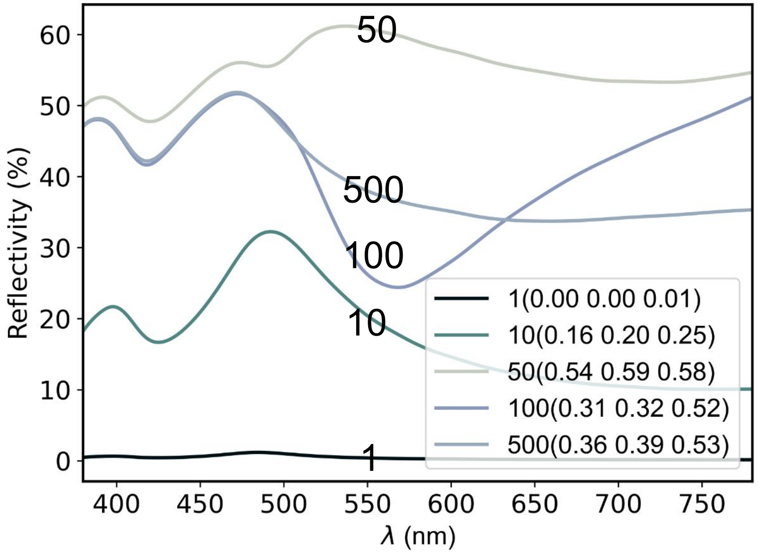


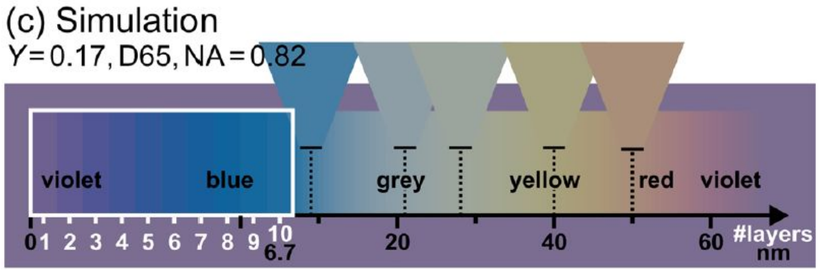
- Peaks in green & blue + purple
- Same pattern as MoS2
  - 50 layers creamy white
  - “blending yogurt”



J. Appl. Phys. 2015, 118, 145305

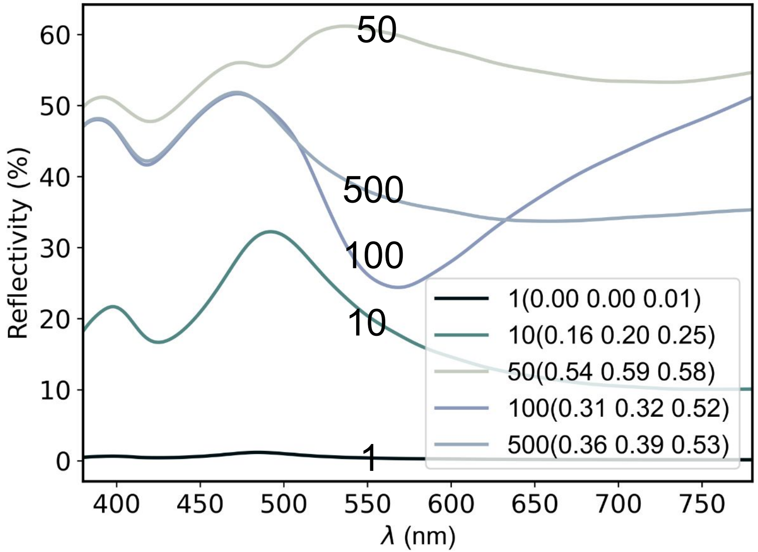
WSe2

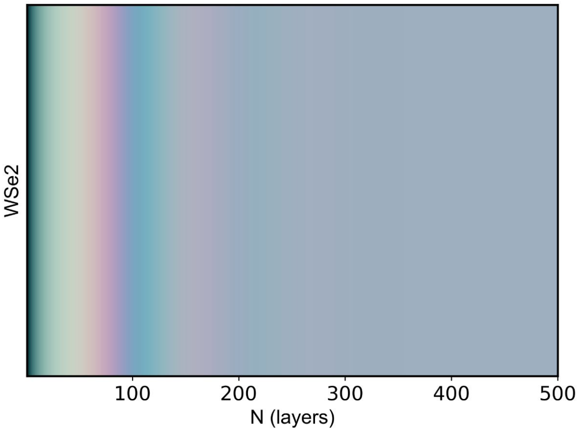


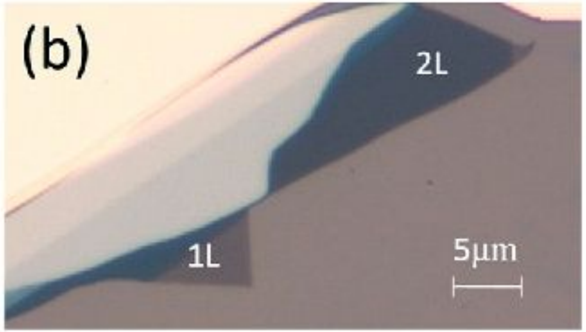


J. Appl. Phys. 2015, 118, 145305

WSe2

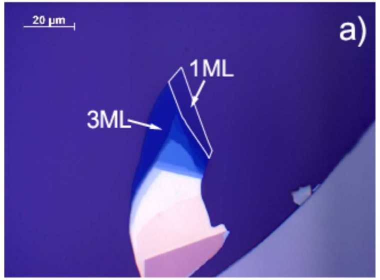






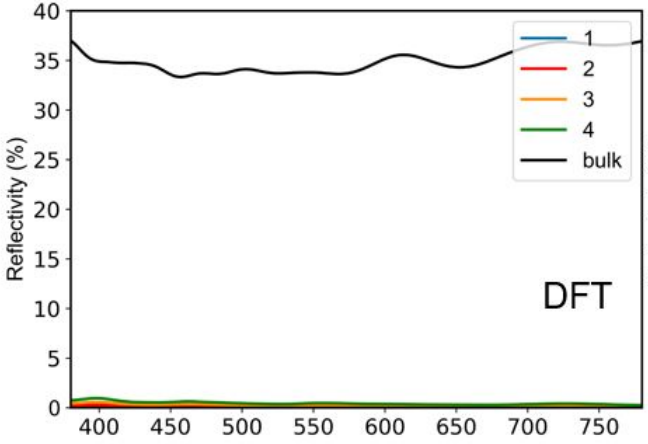
Optical microscopic image of pre-marked WSe2 **monolayer, bilayer, and thin bulk flakes**;

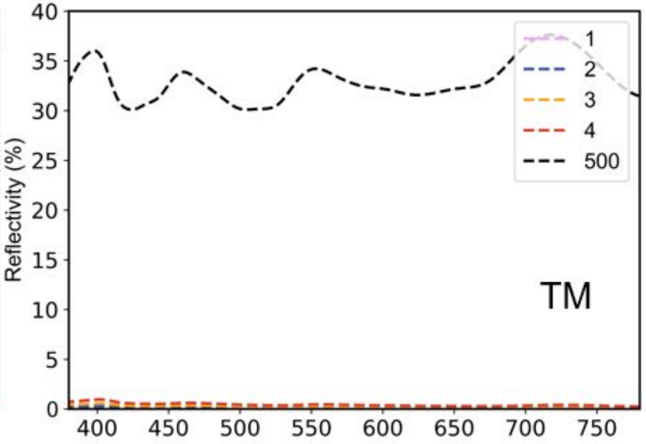
101901-3 Yan et al. Appl. Phys. Lett. 105, 101901 (2014)



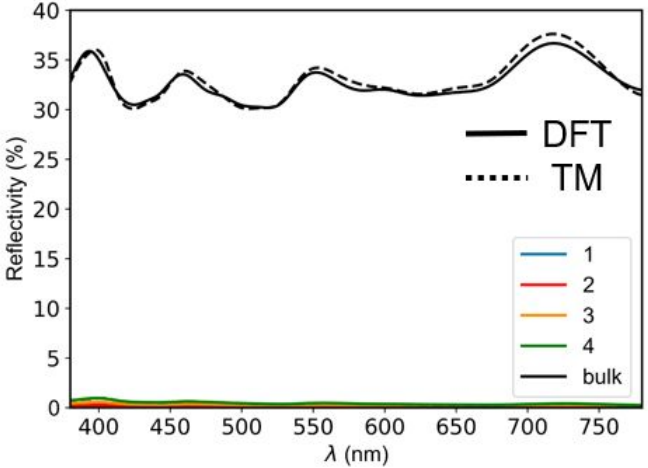
Temperature dependence of photoluminescence lifetime of atomically thin WSe2 layer

graphene



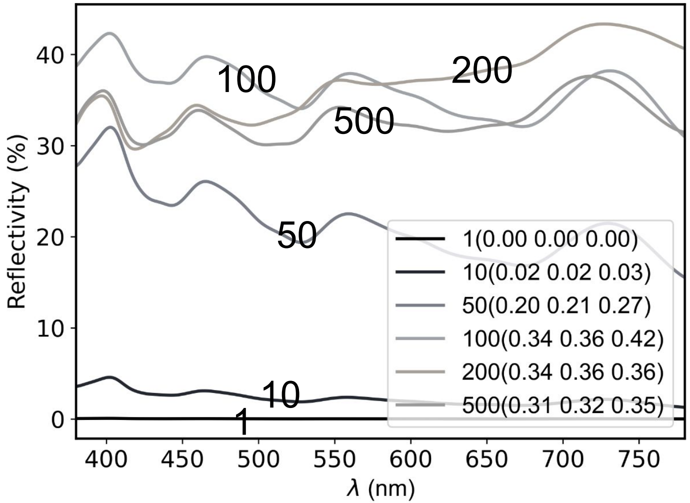


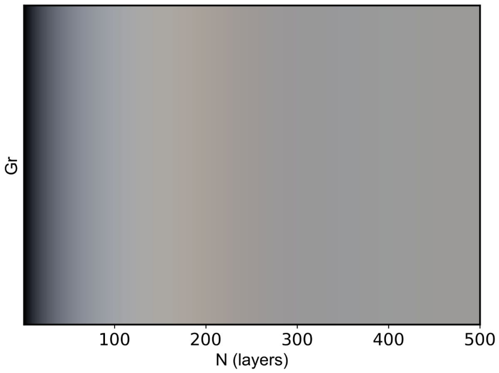
AA stacking
Transfer matrix (TM) method:
Interlayer separation 3.53Å

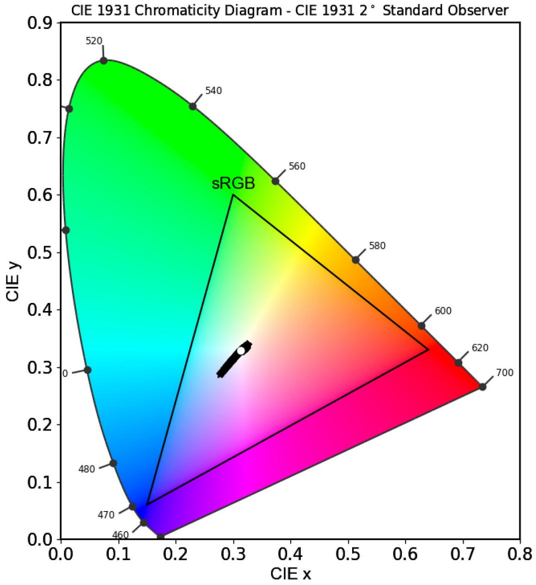


- Multiple peaks
- TM produces good fitting

graphene

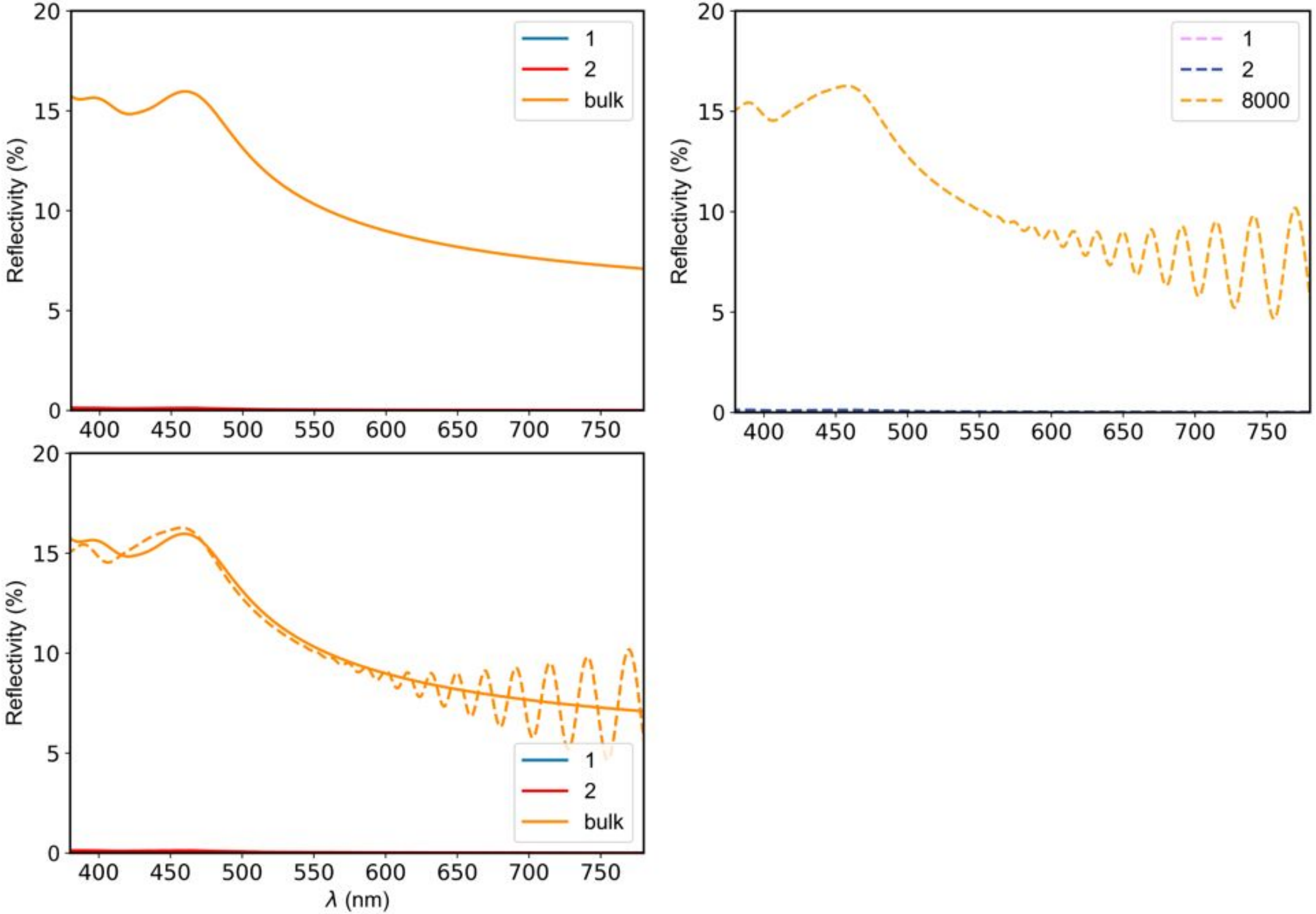






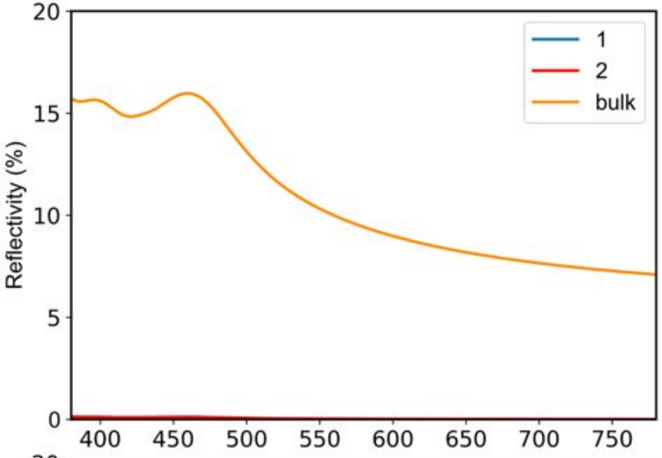
- Less "colorful" than other materials
  - low reflectivities & multiple peaks
  - due to simple structure and composition?

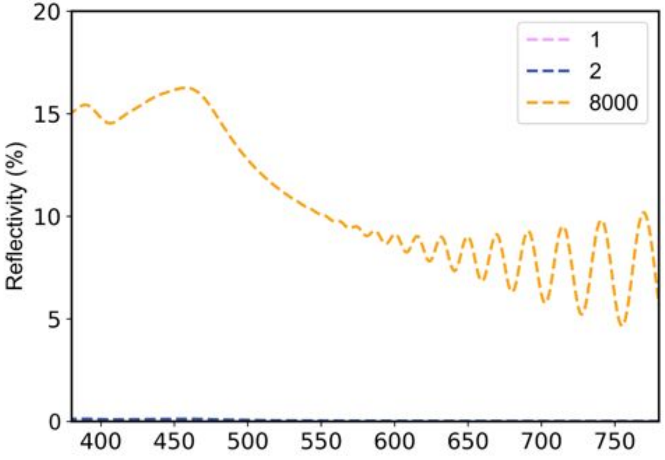
CrCl3



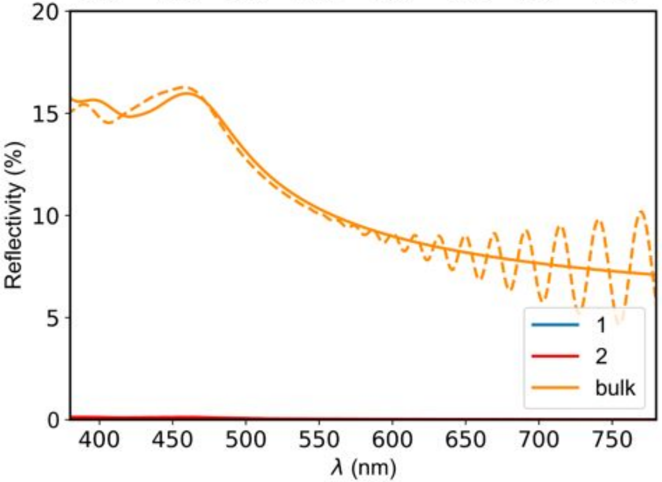
AA stacking
Transfer matrix (TM) method:
Interlayer separation 6.01Å

CrCl3



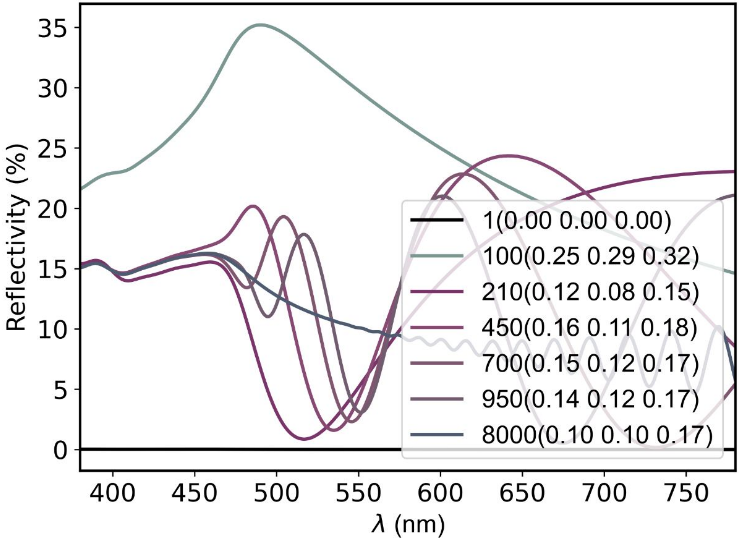


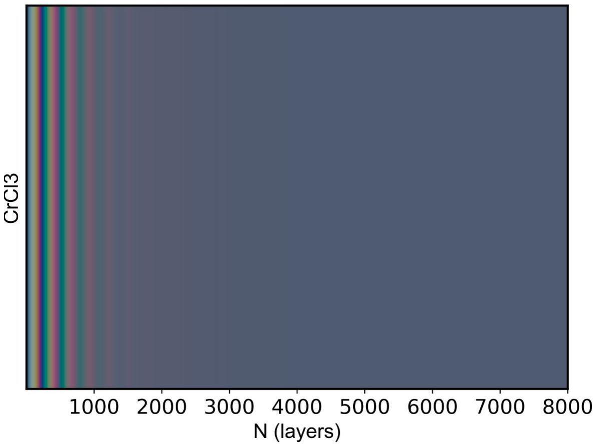
AA stacking
Transfer matrix (TM) method:
Interlayer separation 6.01Å

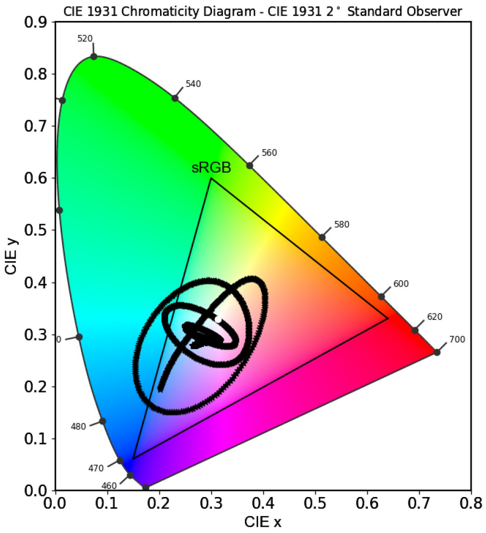


- Bulk has big drop
    - expect mixture of blue and purple
- TM produces wave-like fluctuations near the end

CrCl3

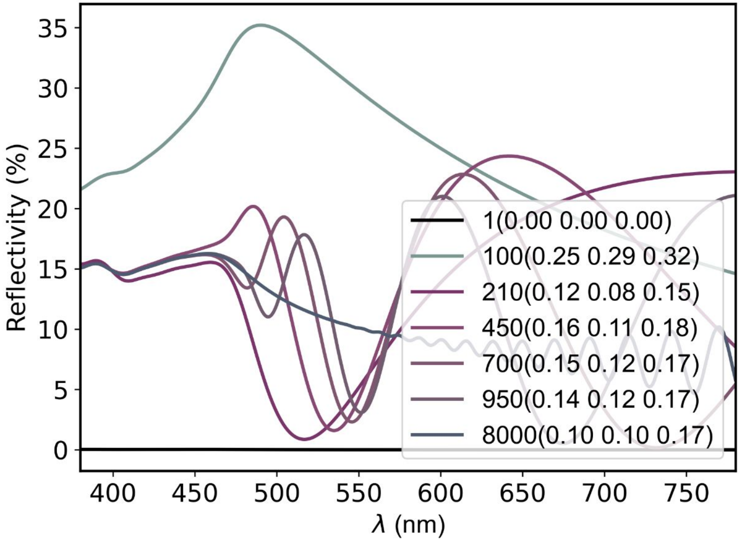


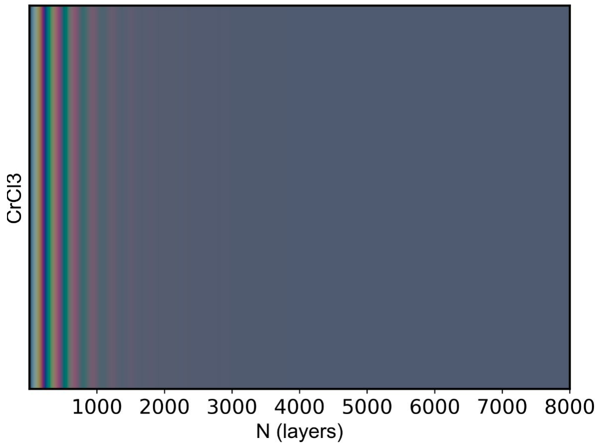


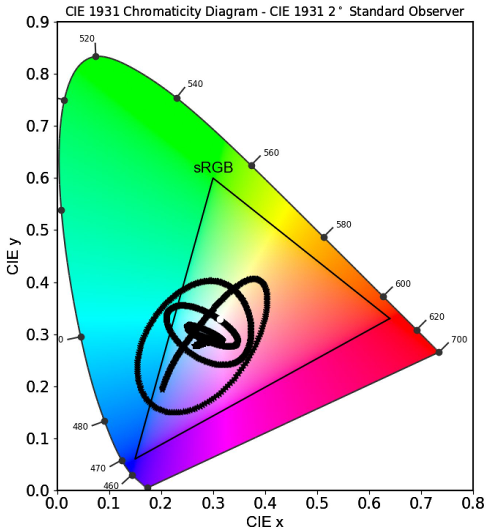


Bulk single crystal of CrCl3
Nat. Phys. 2019 15, 1255

CrCl3





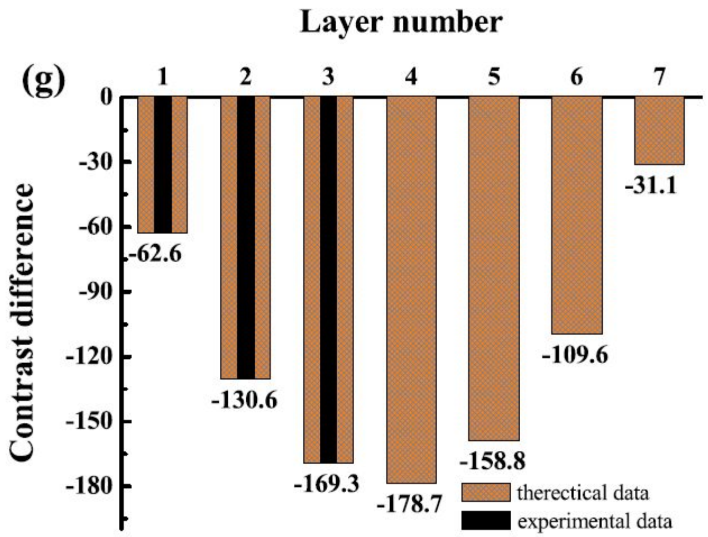


- Pink curves have two peaks
  - violet + red → magenta
- Constant color at ~3000 layers
  - takes longer than other materials

Bulk single crystal of CrCl3
Nat. Phys. 2019 15, 1255

Applications

1. Thickness identification for material characterization

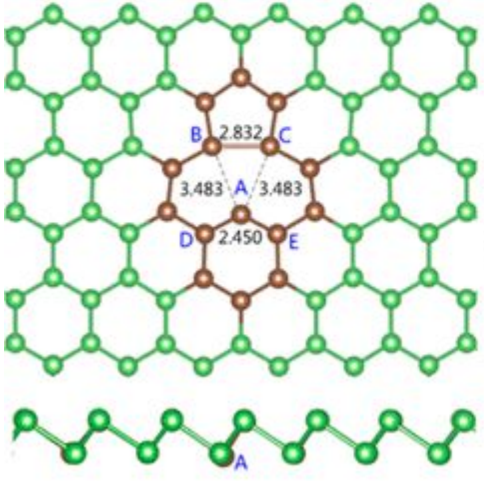


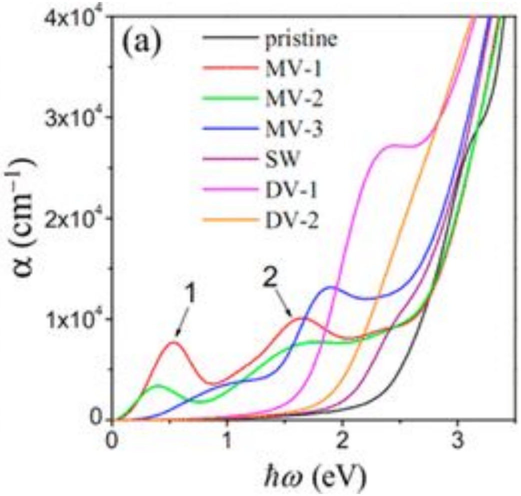
Optical thickness identification of few-layer MoS_2 deposited by chemical vapor deposition

Zusong Zhu *et al* 2019 *Mater. Res. Express* 6 045025

Applications

2. Defect identification





Optical Identification of Topological Defect Types in Monolayer Arsenene by First-Principles Calculation

J. Phys. Chem. C 2016, 120, 43, 24917–24924

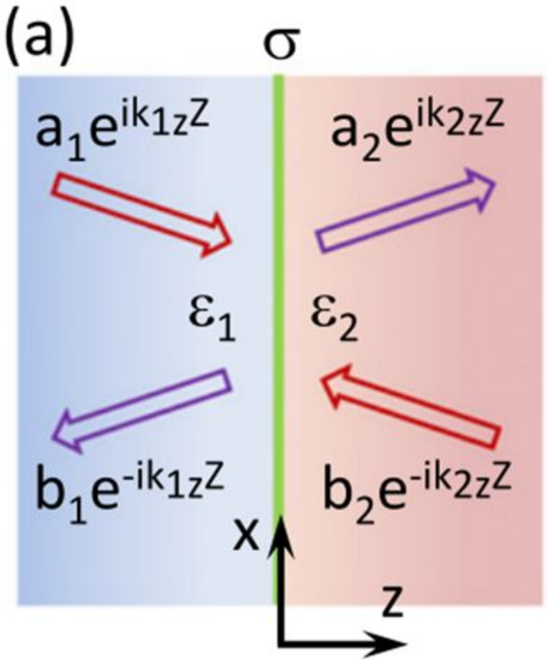
Feedback

- color not resulted from defects
- gold

Color Project Progress

Yang Yiyi
Zhu Zien
Wu Xinan

Explanation of Color Oscillation



$$\begin{bmatrix} a_1 \\ b_1 \end{bmatrix} = D_{1\to 2} \begin{bmatrix} a_2 \\ b_2 \end{bmatrix}$$

$$D_{1\to 2,m} = \frac{1}{2}\begin{bmatrix} 1+\eta_m+\xi_m & 1-\eta_m-\zeta_m\xi_m \\ 1-\eta_m+\zeta_m\xi_m & 1+\eta_m-\xi_m \end{bmatrix}$$

$$\tilde{r} = \frac{b_1}{a_1} = \frac{D_{21}}{D_{11}}, \tilde{t} = \frac{a_2}{a_1} = \frac{1}{D_{11}}$$

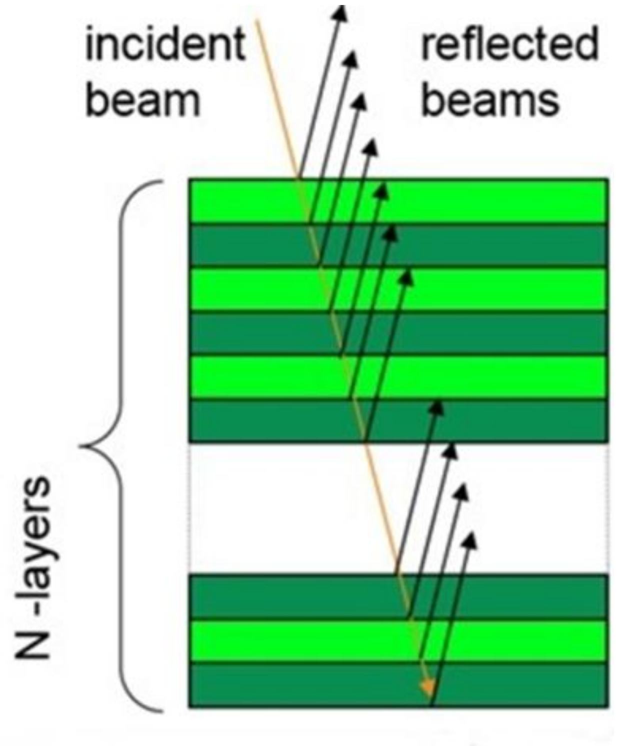
for p polarization:

$$\tilde{r}_p = \frac{1-\eta_p+\xi_p}{1+\eta_p+\xi_p} = \frac{\tilde{\sigma}_p}{2+\tilde{\sigma}_p Z_{vac}}, \ \tilde{t}_p = \frac{2}{1+\eta_p+\xi_p} = \frac{2}{2+\tilde{\sigma}_p Z_{vac}}$$

for p polarization:

$$\tilde{r}_s = \frac{1-\eta_s-\xi_s}{1+\eta_s+\xi_s} = \frac{-\tilde{\sigma}_s}{2+\tilde{\sigma}_s Z_{vac}}, \ \tilde{t}_s = \frac{2}{1+\eta_s+\xi_s} = \frac{2}{2+\tilde{\sigma}_s Z_{vac}}$$

Explanation of Color Oscillation



$$\tilde{r}_{total} = \tilde{r} + \tilde{r}\tilde{t}^{2}\Delta^{2} + \tilde{r}\tilde{t}^{4}\Delta^{4} + \cdots + \tilde{r}\tilde{t}^{2(N-1)}\Delta^{2(N-1)}$$

Let $\tilde{x} = \tilde{t}^{2}\Delta^{2} = \tilde{t}^{2}e^{i2kd}$

$$\tilde{r}_{total} = \tilde{r}(1 + \tilde{x} + \tilde{x}^{2} + \cdots + \tilde{x}^{N-1}) = \tilde{r}\frac{1-\tilde{x}^{N}}{1-\tilde{x}}$$

$$R_N = \tilde{r}_{total}{}^{*}\tilde{r}_{total} = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^{2} \cdot |1-\tilde{x}^{N}|^{2}$$

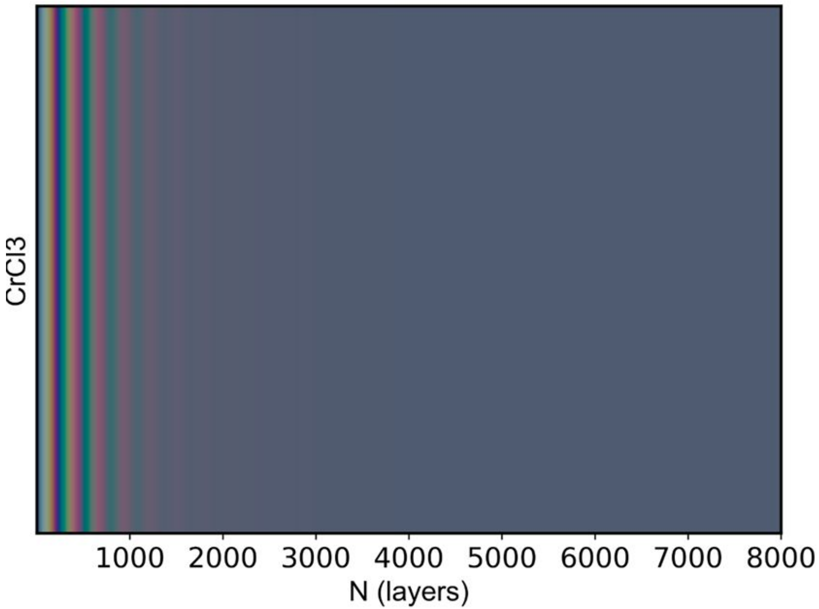
$$\tilde{x} = \tilde{t}^{2}e^{i2kd} = Te^{2i(kd+\theta)} \qquad T = \tilde{t}^{*}\tilde{t} \approx 1, \tilde{t} = \sqrt{T}\,e^{i\theta}$$

$$R_N = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^{2} \cdot (1 - 2\mathrm{Re}(\tilde{x}^{N}) + |\tilde{x}^{N}|^{2}) = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^{2} \cdot [1 - 2T^{N}\mathrm{Re}(e^{2iN(kd+\theta)}) + T^{2N}]$$

Explanation of Color Oscillation

$$R_N = \tilde{r}_{total}{}^* \tilde{r}_{total} = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot |1-\tilde{x}^N|^2$$

$$R_N = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot (1-2\mathrm{Re}(\tilde{x}^N)+|\tilde{x}^N|^2) = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot [1-2T^N \mathrm{Re}(e^{2iN(kd+\theta)})+T^{2N}]$$



when the number of layers increases by δN, and satisfy the following relation:

$$\delta N \cdot (kd+\theta) = m\pi$$

Then the periodicity factor will not change, which happens to be the most important part of the change. so we will see the same color as for the previous layers, but the overall brightness is reduced due to more absorption.

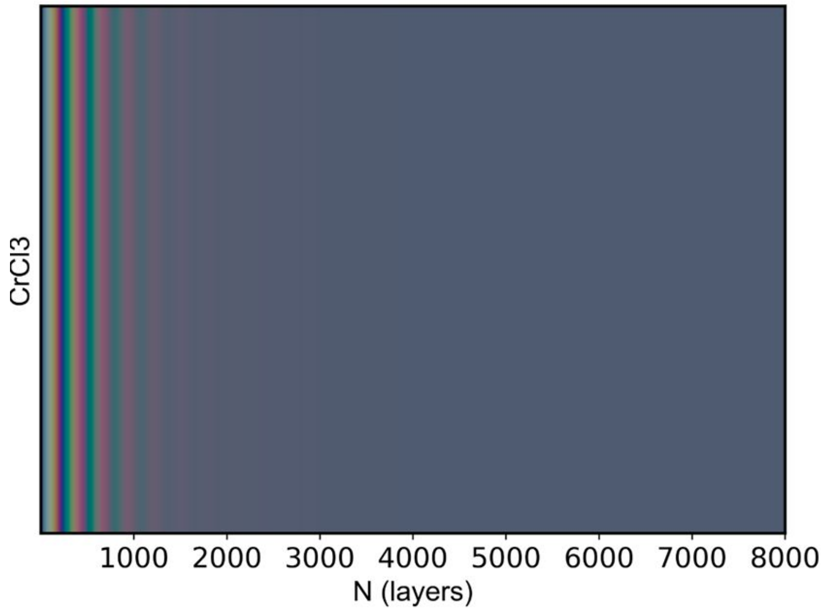
$$N \to \infty,\ \tilde{x}^N = T^N e^{2iN(kd+\theta)} \to \infty$$

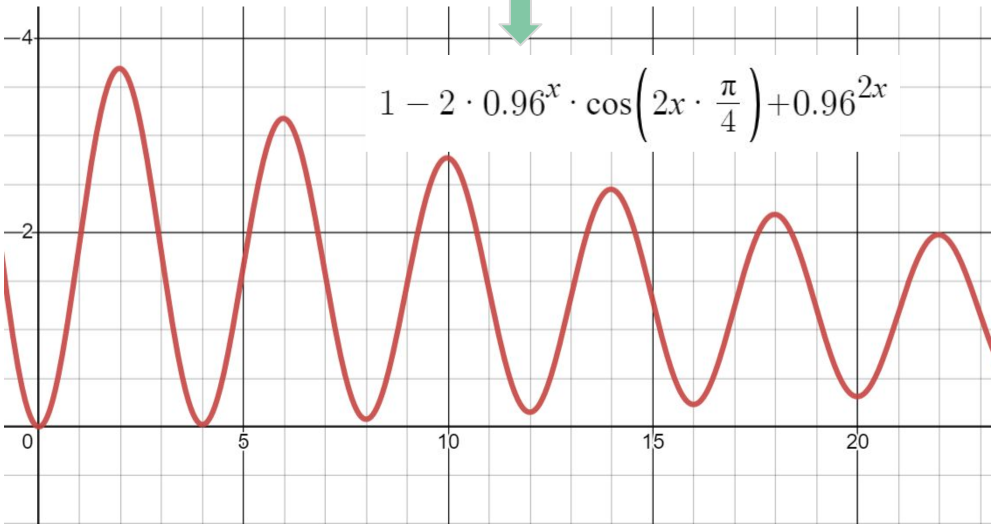
$$R_\infty(\lambda) = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2$$

Explanation of Color Oscillation

$$R_N = \tilde{r}_{total}{}^* \tilde{r}_{total} = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot |1-\tilde{x}^N|^2$$

$$R_N = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot (1 - 2\mathrm{Re}(\tilde{x}^N) + |\tilde{x}^N|^2) = \left|\frac{\tilde{r}}{1-\tilde{x}}\right|^2 \cdot [1 - 2T^N \mathrm{Re}(e^{2iN(kd+\theta)}) + T^{2N}]$$

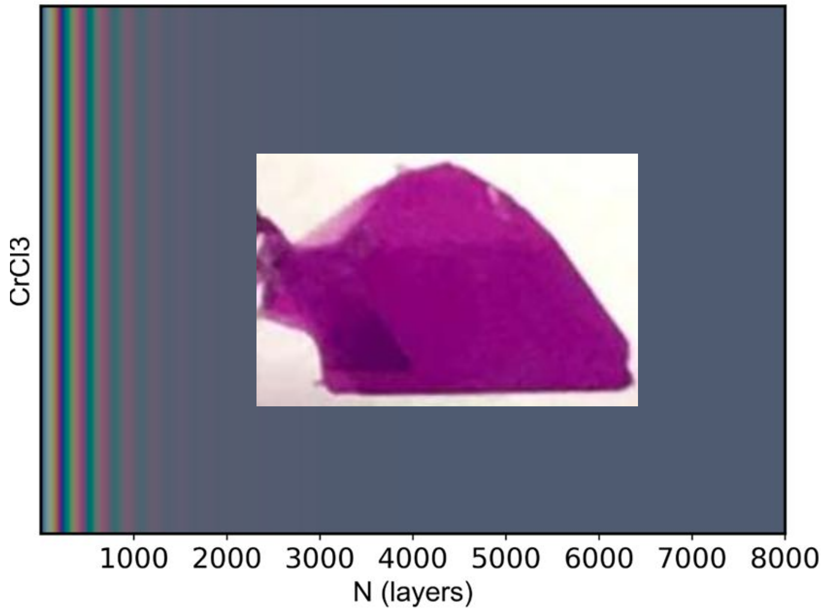




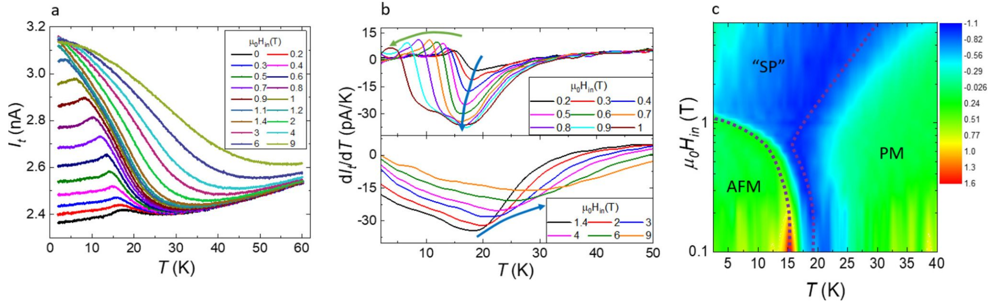
CrCl3 Color Mismatch

CrX3 has complex magnetic properties

$$\sigma_{2D}(q=0,\omega) = i\omega\varepsilon_0(1 - \varepsilon_{3D}(q=0,\omega))L$$

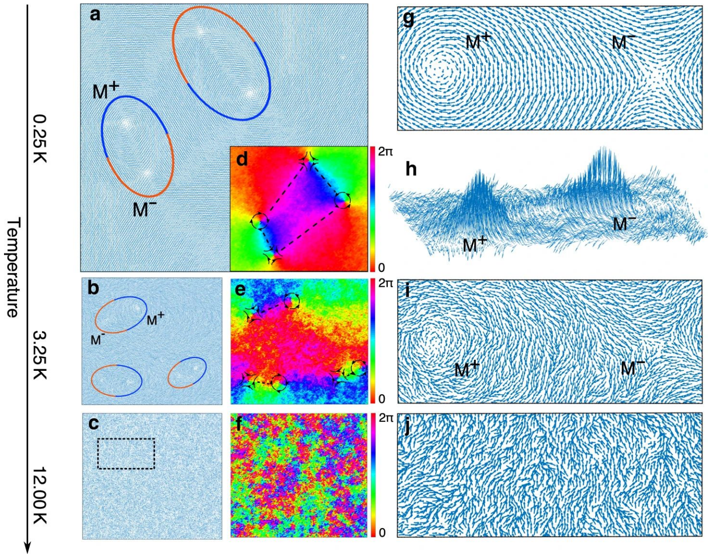


“The van der Waals magnetic insulator $CrCl_3$ shows layered antiferromagnetism down to the bilayer limit with the magnetic moments in the plane of the layers and little or no anisotropy within the plane.”



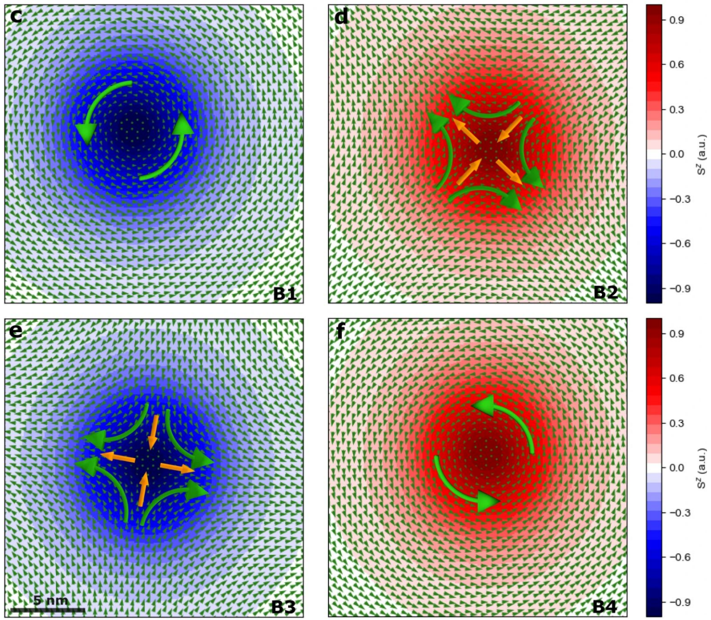
Nano Letters 2019 *19* (6), 3993-3998

CrCl3 Color Mismatch



"Our studies show that the overall isotropic in-plane magnetized 2D material such as ML $CrCl_3$ could provide the opportunity for generating meron-type topological defects without involving any other interactions."

Nat Commun 11, 4724 (2020)



"Their dynamics is determined by the interplay between the strong in-plane dipolar interactions and the weak out-of-plane magnetic anisotropy stabilising a vortex core within a radius of 8–10 nm."

Nat Commun 12, 185 (2021)

Outlook for future work

1) Comparison with other methods of measuring the number of layers by optical properties（Li, et al. (2017). Advanced Functional Materials.）
2) Consider the effect of substrate reflection
3) Comparison with results from membrane calculation software (like TFCalc)
4) Consider different types of light sources and matching functions(different races; microscope)
5) Different stacking methods for 2D materials