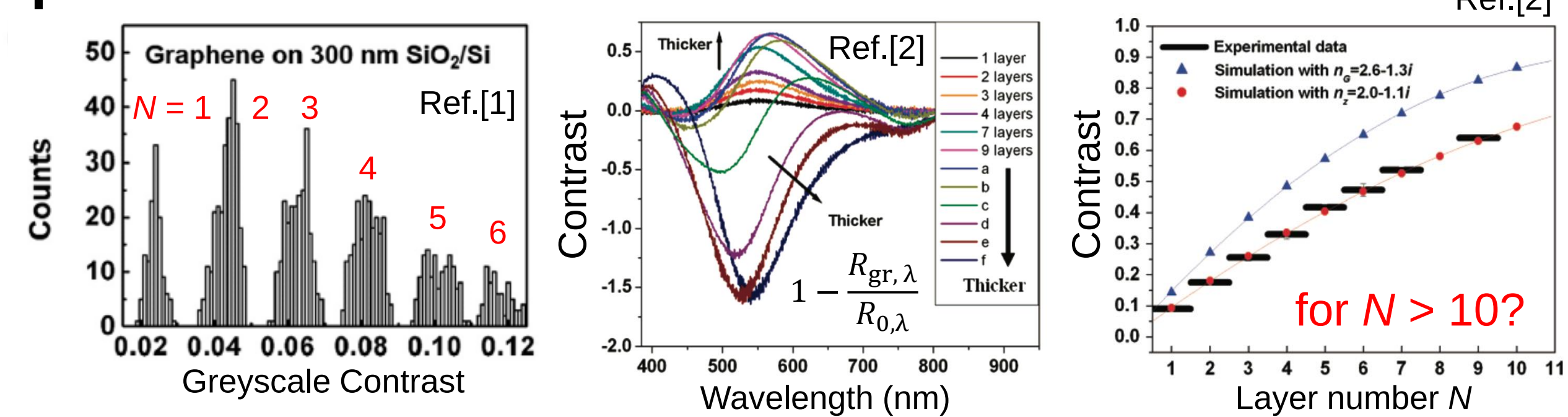


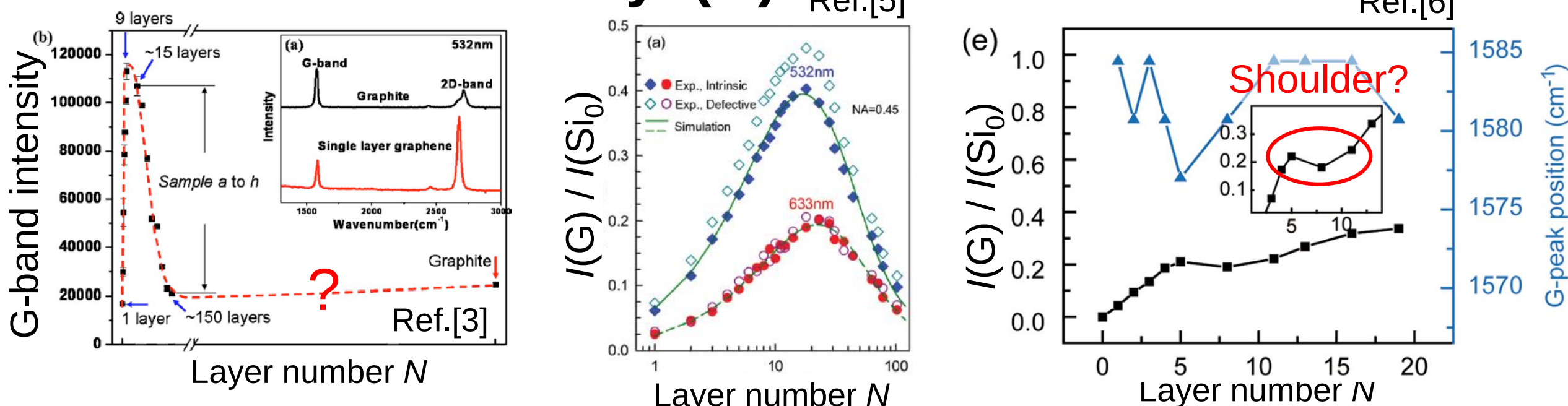
Introduction

Optical contrast C and Reflectance R



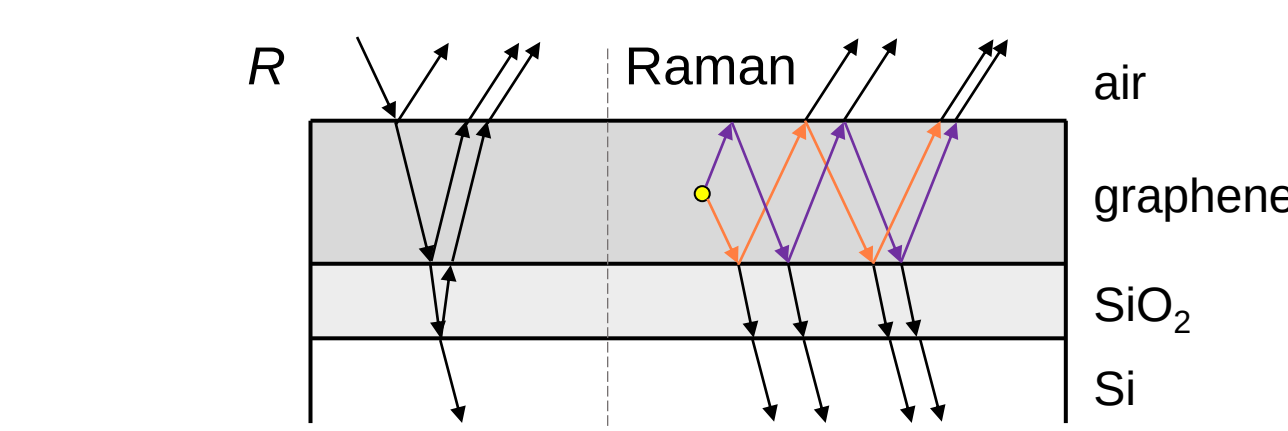
- RGB and grayscale contrasts and reflectance R are well quantized up to 6/10 layers [1]
 - Reflectance R has complex N -dependence for $N > 10$, but it has not been studied systematically [2]
- ⇒ what happens for $N > 10$?

Raman G-band intensity $I(G)$



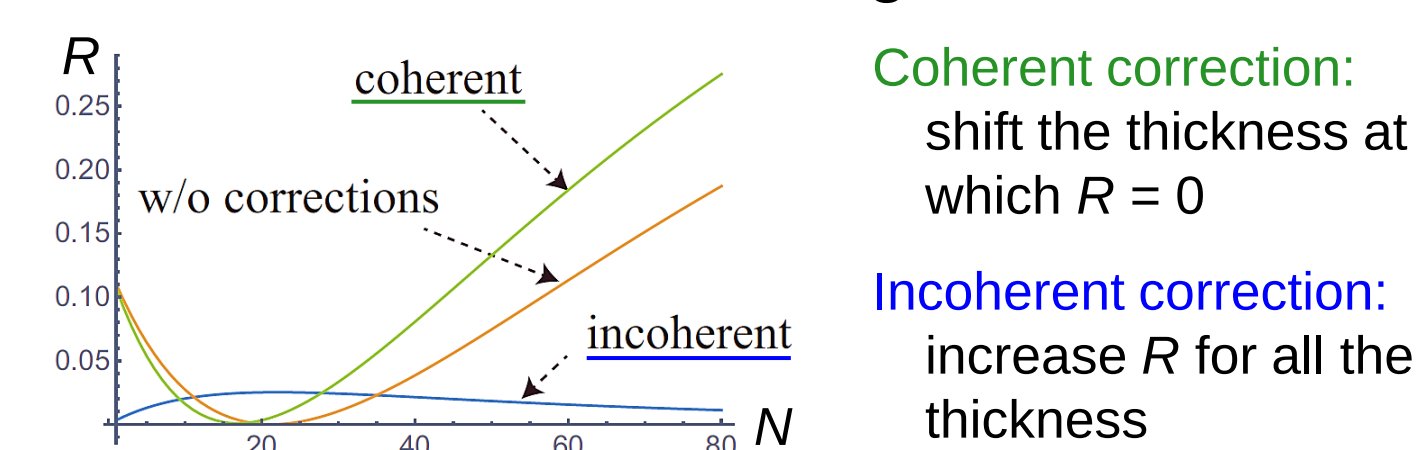
- $I(G)$ shows reentrant behavior with N , and then approaches the value of bulk graphite [3-6]
- ⇒ how $I(G)$ approaches that of bulk graphite?

Reflectance R of incident light
Multiple reflection / Interference of scattered light



Motivation

- C and R for **thicker graphene**?
- Interference of Raman scattered light?
- Does Raman scattered light affect the R ?



Reflectance and Raman scattering are studied for the same multi-layer graphene on SiO_2/Si up to $t \sim 100$ nm ($N \sim 300$ layers)

Experimental

Multi-layer graphene

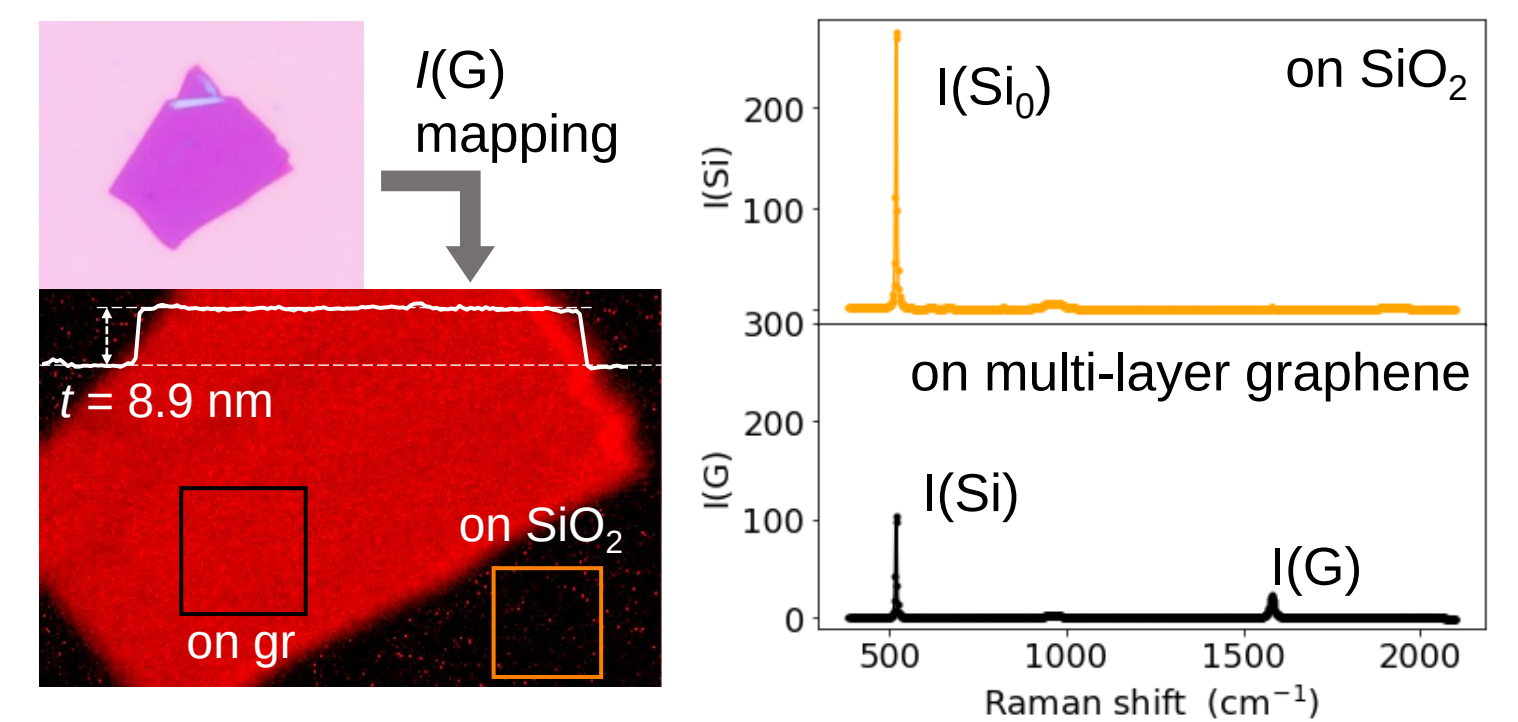
- Exfoliated graphene on SiO_2 ($t = 274 \pm 1$ nm)/Si
- Graphene thickness is measured by AFM
- SiO_2 thickness is determined from reflectance measurement [9]

RGB and Reflectance

- Optical images are taken under the same brightness and exposure time
- R is measured by a reflectometer [9] with a 50x objective lens (spot size of 5 μm)

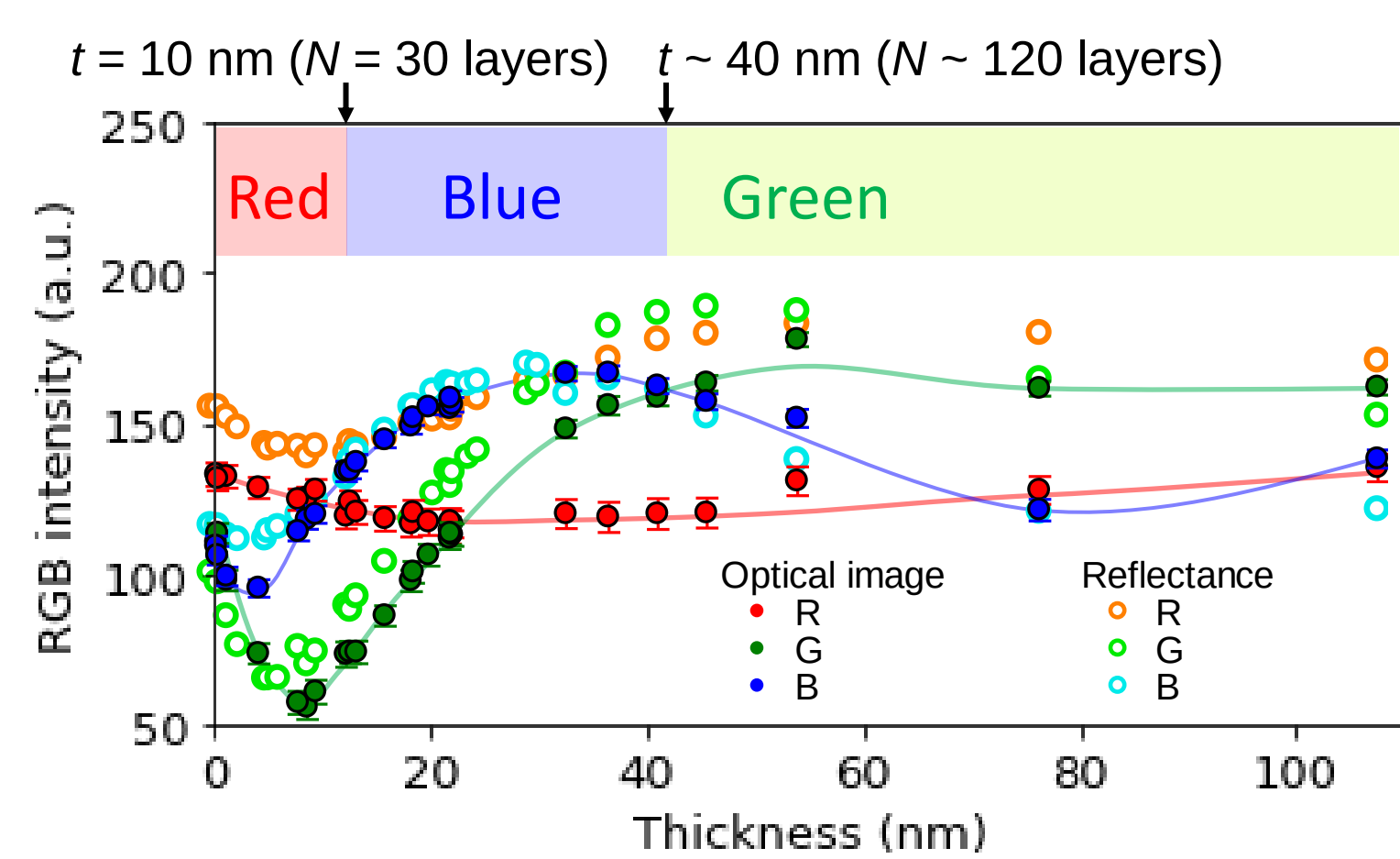
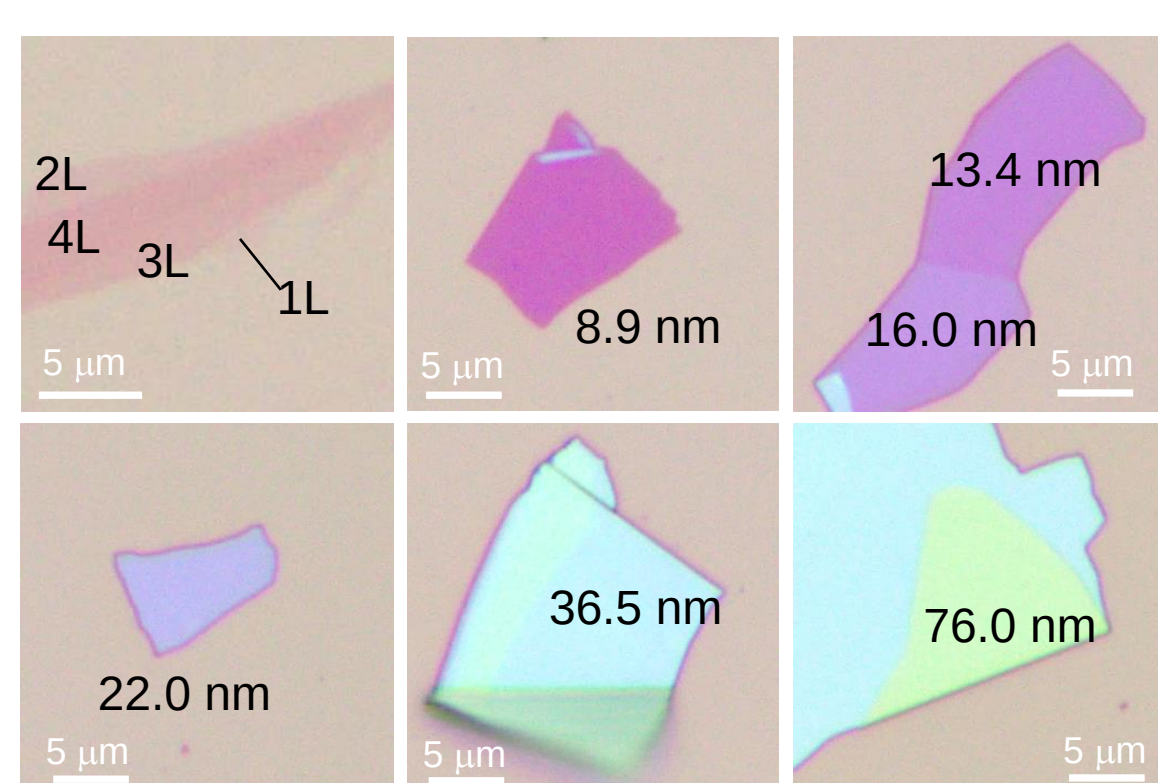
Raman spectroscopy

- Excitation light: 532nm laser
- Each signal intensity $I(*)$ is defined as the peak height, and normalized by $I(\text{Si}_0)$

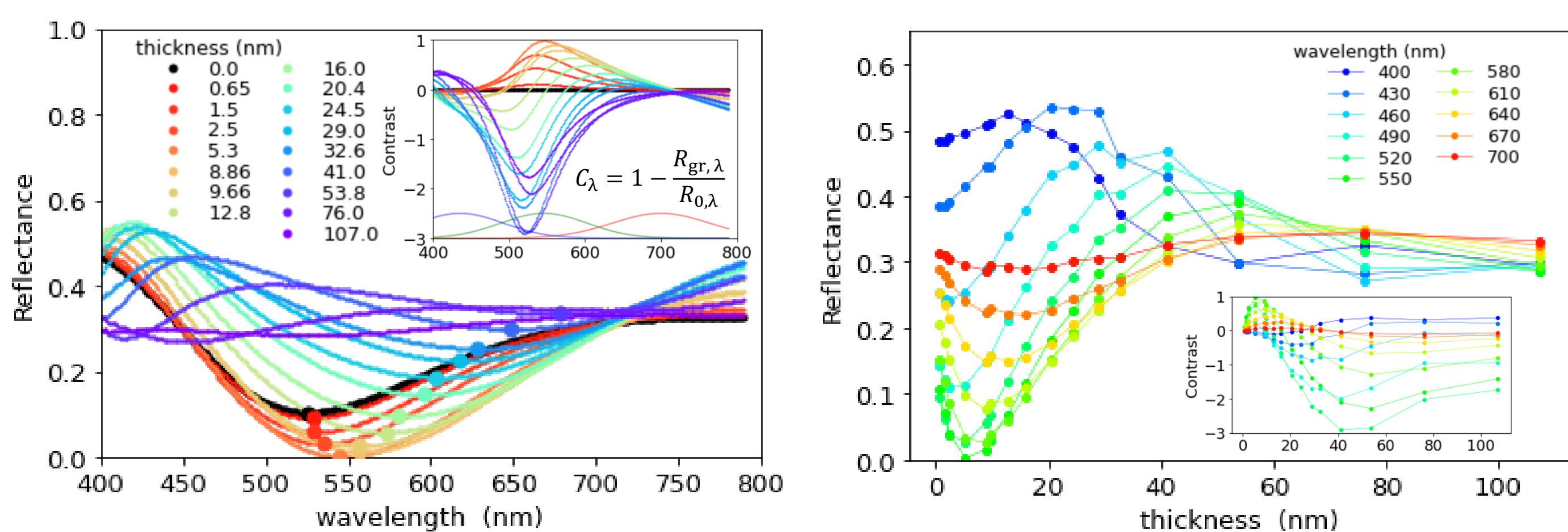


Reflectance

RGB intensity



Reflectance R



- R has periodic λ -dependence for thin graphene, and R becomes almost λ -independent for thick graphene
- Zero-Reflectance** at $t \sim 5.3$ nm and $\lambda \sim 540$ nm → *graphene is "invisible"*
- C around $\lambda \sim 550$ nm changes most drastically with t
- RGB values obtained from optical images are well reproduced by R with simple gaussian distribution of RGB up to $t = 107$ nm

Model calculation

Fresnel's equation

$$R = \left[\frac{r_1 e^{i(\phi_1 + \phi_2)} + r_2 e^{-i(\phi_1 - \phi_2)} + r_3 e^{-i(\phi_1 + \phi_2)} + r_1 r_2 r_3 e^{i(\phi_1 - \phi_2)}}{e^{i(\phi_1 + \phi_2)} + r_1 r_2 e^{-i(\phi_1 - \phi_2)} + r_1 r_3 e^{-i(\phi_1 + \phi_2)} + r_2 r_3 e^{i(\phi_1 - \phi_2)}} \right]^2$$

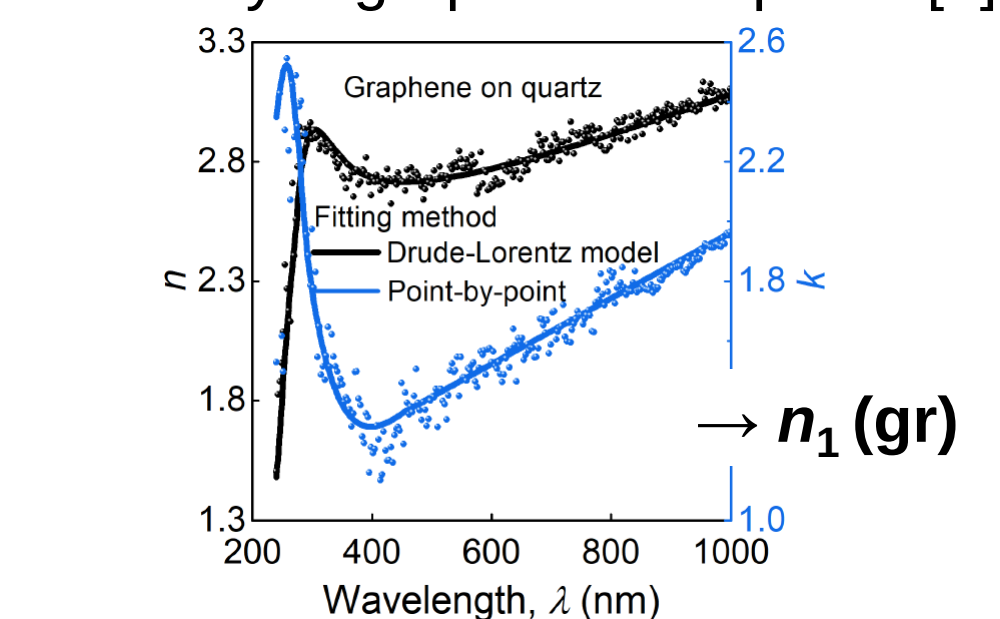
$$r_1 = \frac{n_0 - n_1}{n_0 + n_1}, \quad r_2 = \frac{n_1 - n_2}{n_1 + n_2}, \quad r_3 = \frac{n_2 - n_3}{n_2 + n_3}$$

$$\phi_1 = \frac{2\pi n_1 d_1}{\lambda}, \quad \phi_2 = \frac{2\pi n_2 d_2}{\lambda}$$

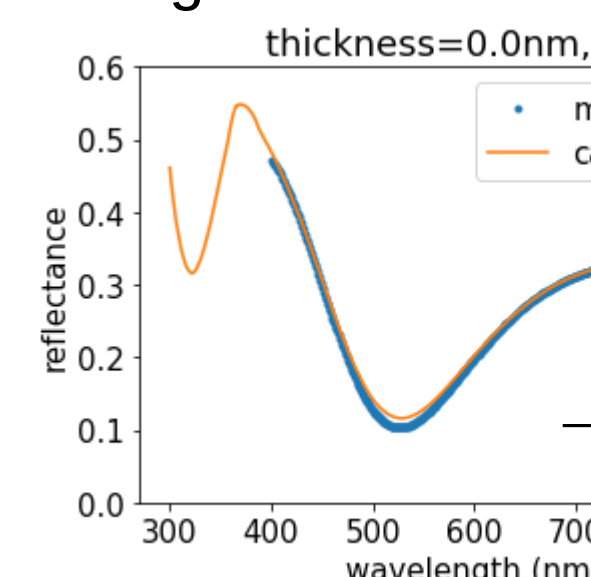
n_i : refractive index
 d_i : thickness

d_1 (gr) n_0 (air) = 1
 n_1 (gr): from Ref.[7]
 d_2 (SiO_2) = 274 nm
 n_2 (SiO_2) = 1.43
 n_3 (Si): from Ref.[8]

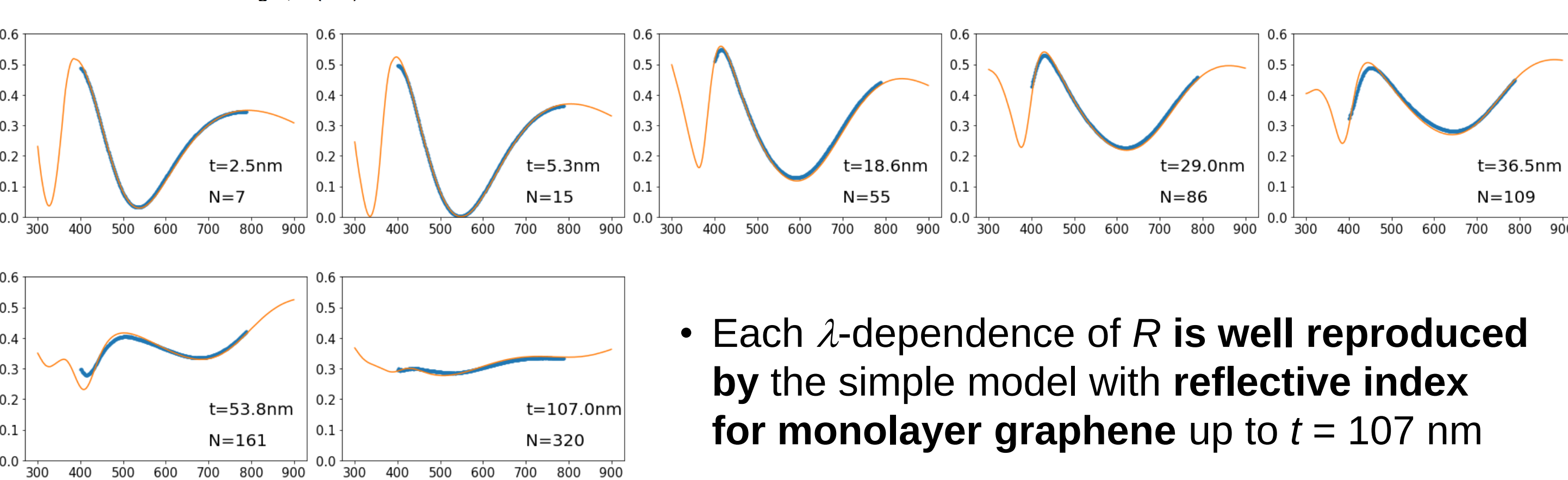
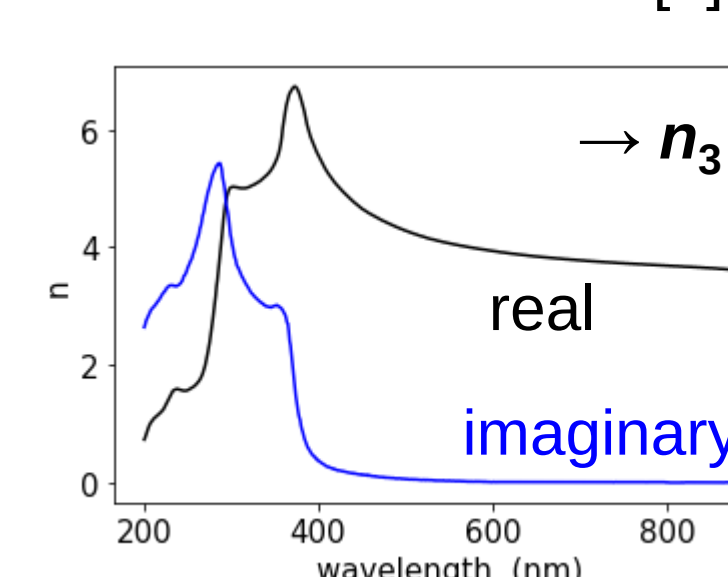
Monolayer graphene on quartz [7]



Fitting of $N = 0$ data



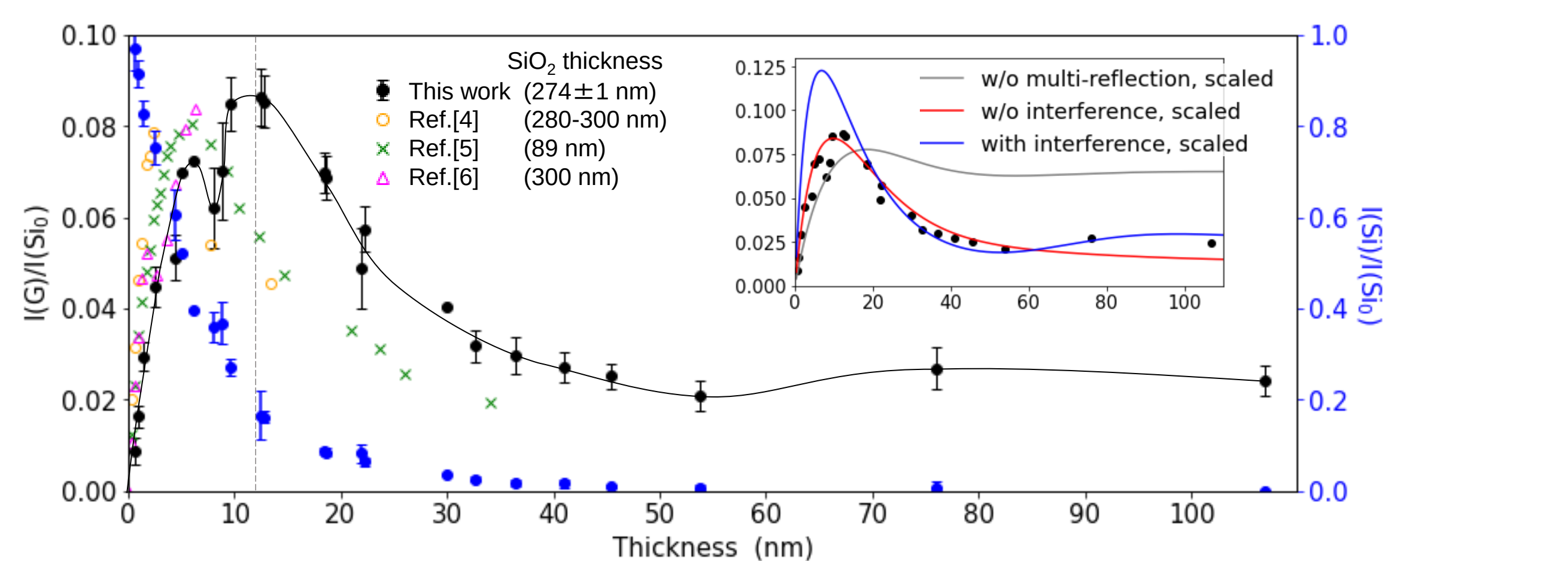
Refractive index of Si [8]



- Each λ -dependence of R is well reproduced by the simple model with reflective index for monolayer graphene up to $t = 107$ nm

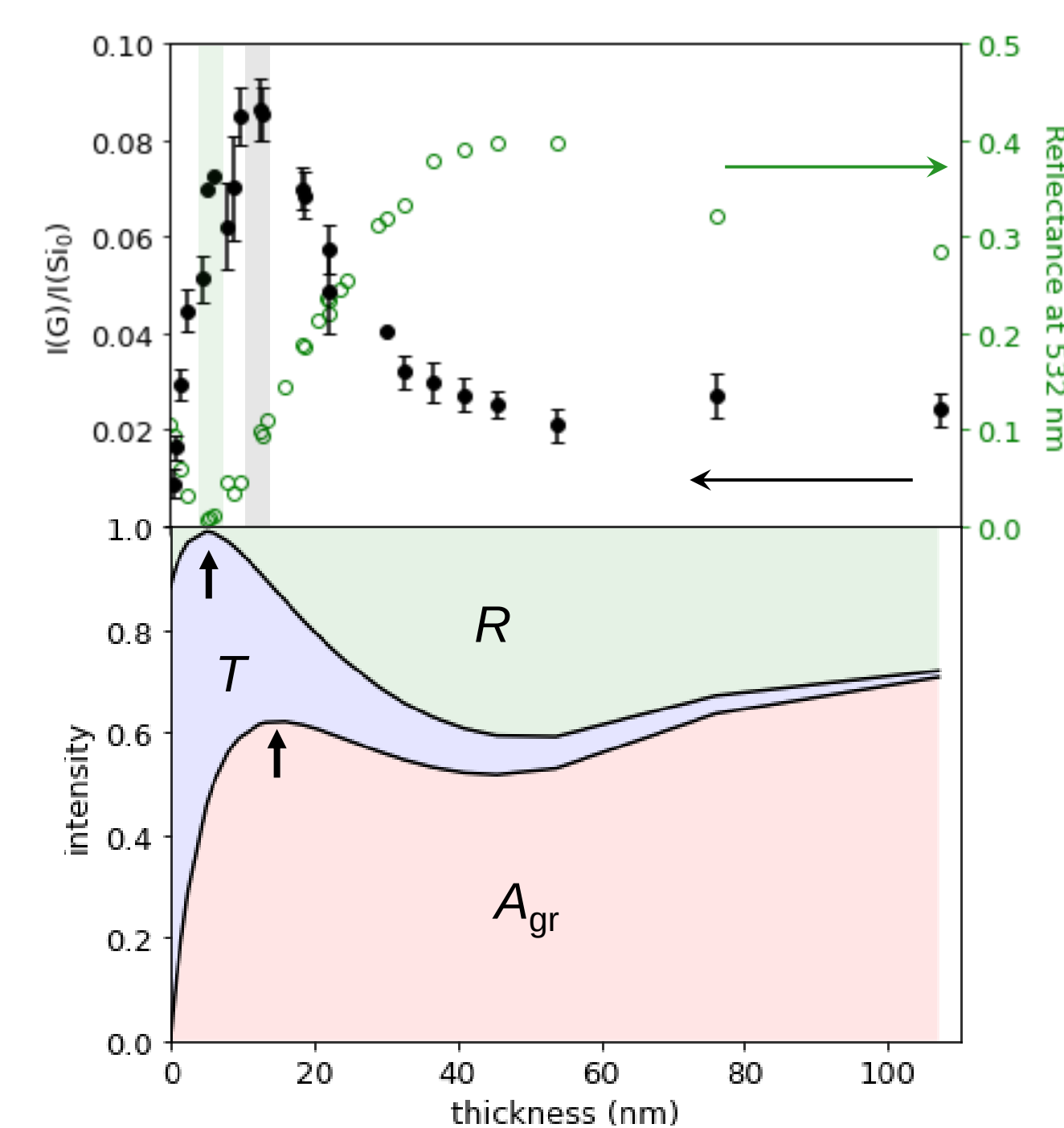
Raman Spectroscopy

G-band intensity $I(G)$



- $I(G)/I(\text{Si}_0)$ has a peak at $t_{\text{peak}} \sim 12$ nm
Each study [4,5,6] shows different t_{peak} , possibly due to the tiny difference of the SiO_2 thickness
- Saturate but **finite periodic t -dependence** at $t > 30$ nm
Comparison with calculated $I(G)$ (intensity is scaled) revealed that not only the multiple reflection but also the **interference of scattered light are important**
- $I(G)/I(\text{Si}_0)$ decays "almost" exponentially with t
The behavior is not a simple exponential due to the multiple reflection of scattered light

Relation between $I(G)$ and R



- $I(G)/I(\text{Si}_0)$ becomes large when the incident light (532 nm) is less reflected (namely, the light is "trapped" inside multi-layer graphene)
- Peak of $I(G)/I(\text{Si}_0)$ does not coincide with the minima of R
← The thickness, where the light absorption by graphene (A_{gr}) takes its maxima, is thicker than the thickness at R minima, because some amount of light transmits to SiO_2 and is absorbed by Si

$$I = R + A_{\text{gr}} + T$$

I : Incident laser intensity (= 1)
 R : Reflectance
 A_{gr} : absorption by graphene
 T : absorption by Si

Summary

- RGB values by optical images are well reproduced by R up to 107 nm.
- R spectrum are well reproduced by Fresnel's equation with refractive index for monolayer graphene up to 107 nm.
- Not only multiple reflection but also interference of light emitted by Raman scattering is important to understand $I(G)$ especially at $t > 50$ nm.
- $I(G)$ is large where R is small, since the incident light is effectively "trapped" inside multi-layer graphene.

Reference

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- [2] Z. H. Ni *et al.*, Nano Lett. **7**, 2758 (2007).
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- [4] D. Yoon *et al.*, J. Kor. Phys. Soc **55**, 1299 (2009).
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- [6] Y-S. No *et al.*, Sci. Rep. **8**, 571 (2018).
- [7] M. A. El-Sayed *et al.*, Nanomaterials **11**, 1230 (2021).
- [8] D. E. Aspnes and A. A. Studna, Phys. Rev. B **27**, 985 (1983).
- [9] R measurement was performed in Takeda clean room, supported by "Nanotechnology Platform Japan" of the MEXT Grant No.JPMXP09F21UT0045