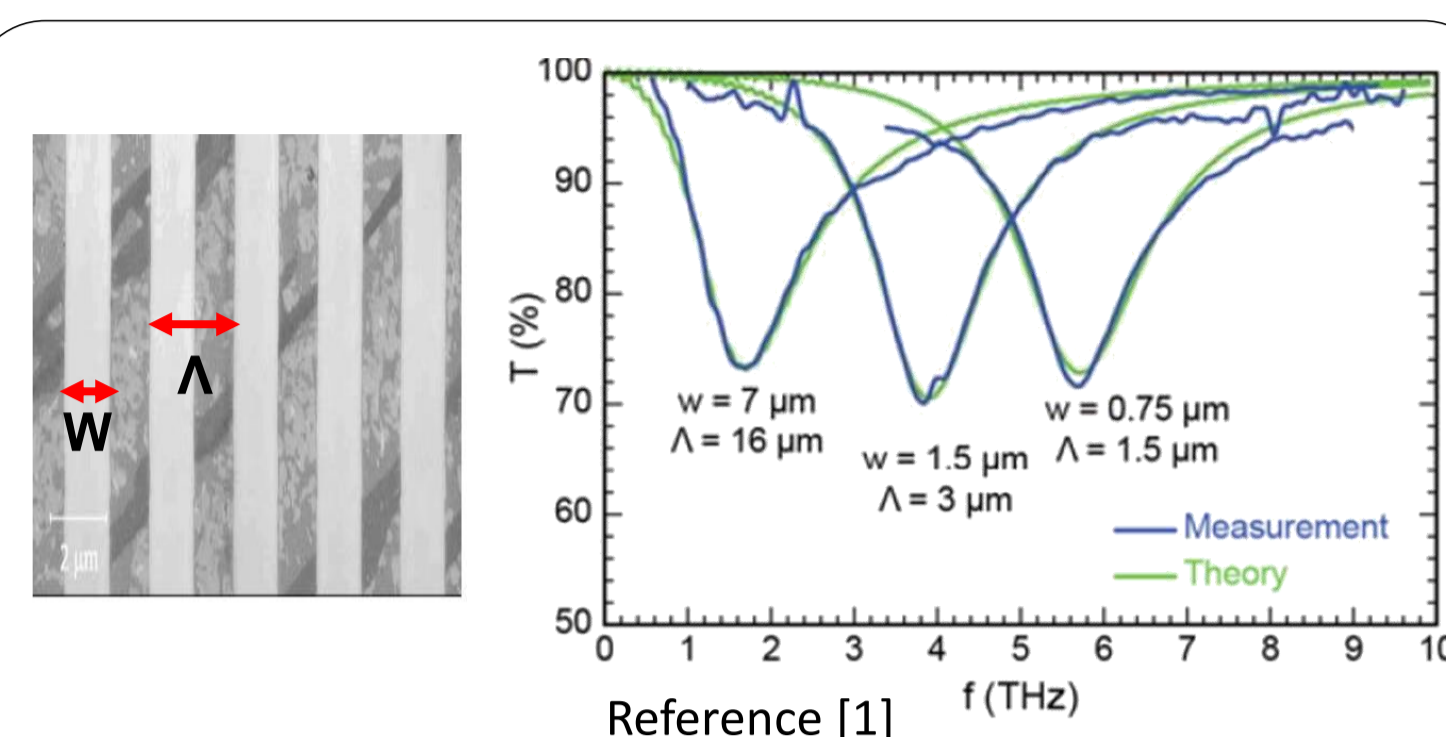
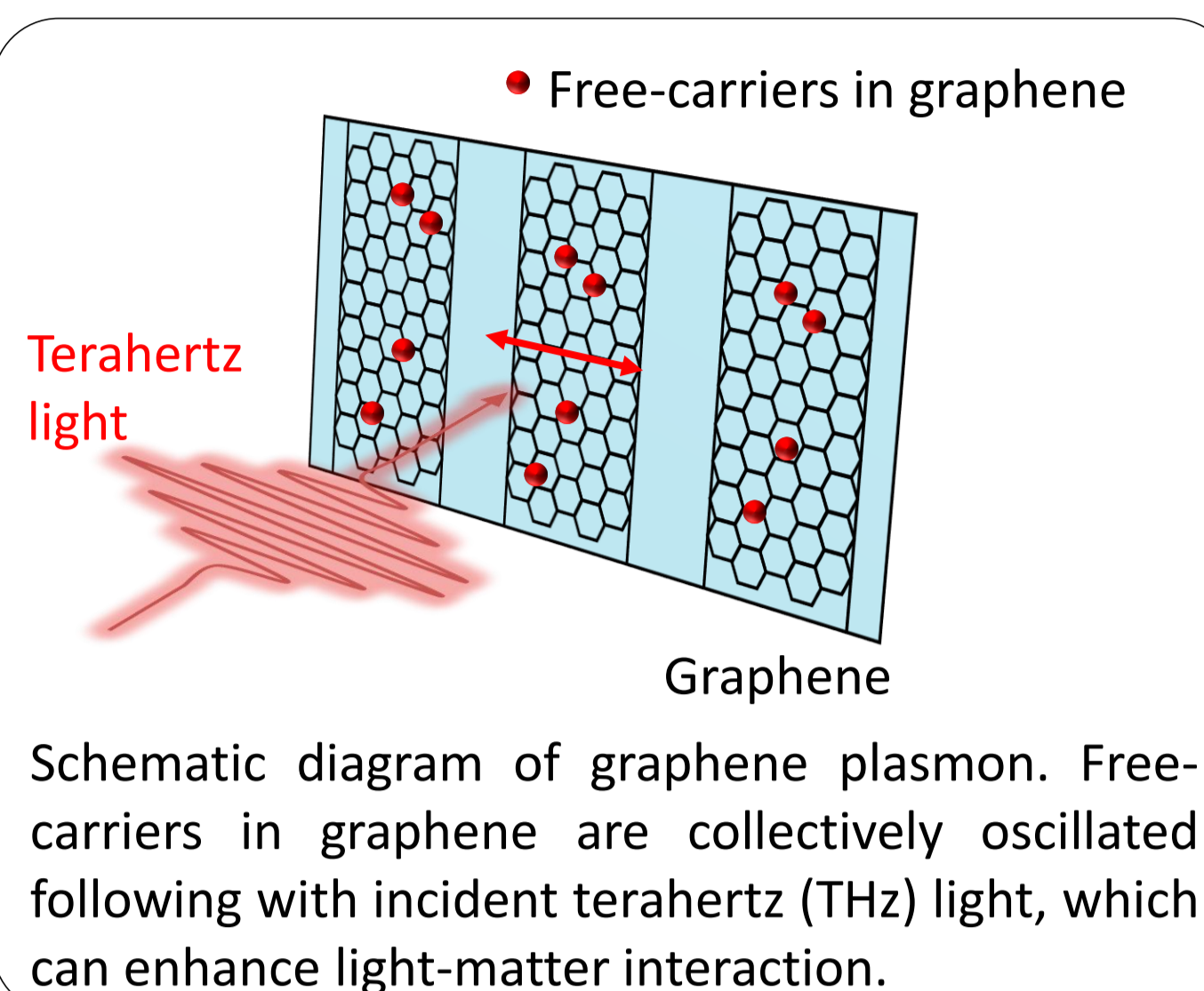


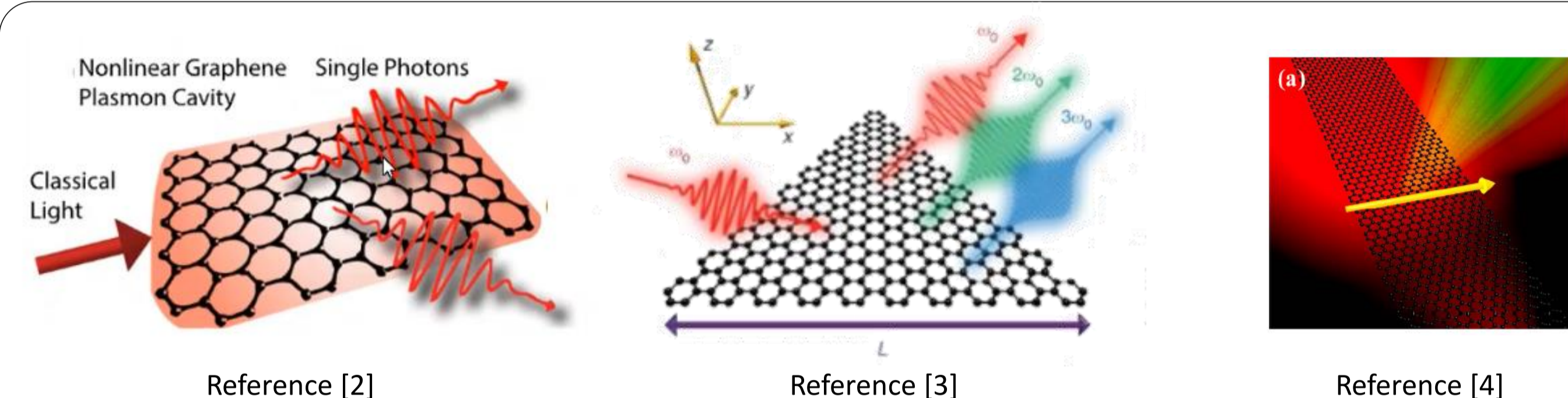
Motivation

Significant enhanced light-matter interaction in graphene using plasmons.



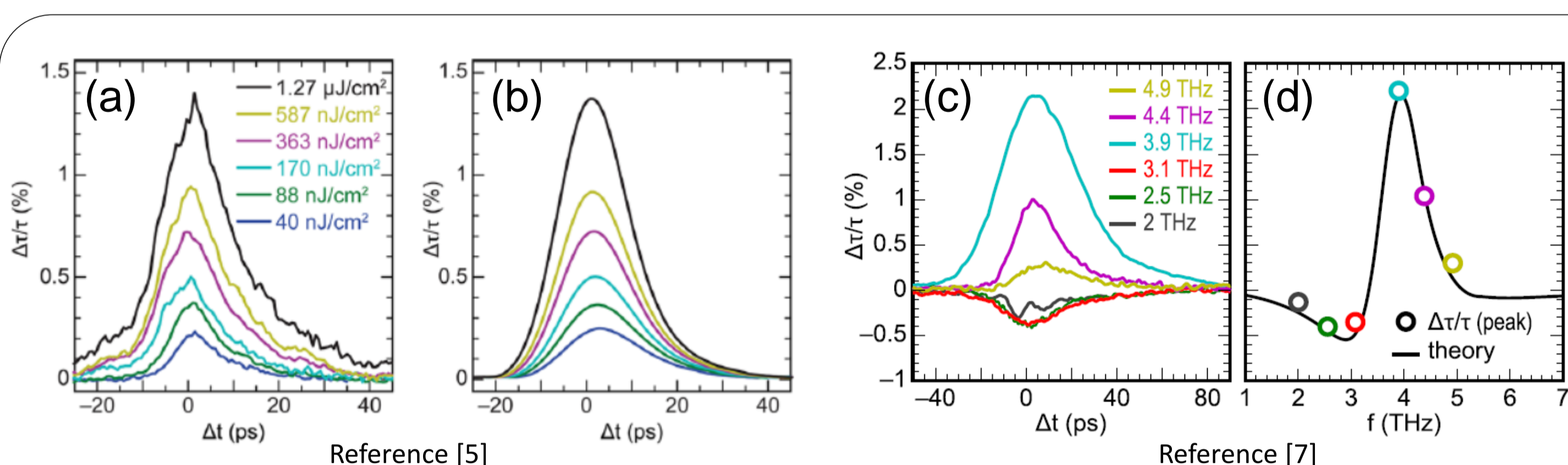
1. Significant enhanced absorption was reported, which can be achieved by means of the plasmonic devices.
2. Plasmon frequency can be tailored by changing the width of the graphene ribbons.

Theoretical predictions for the enhancement of nonlinear properties



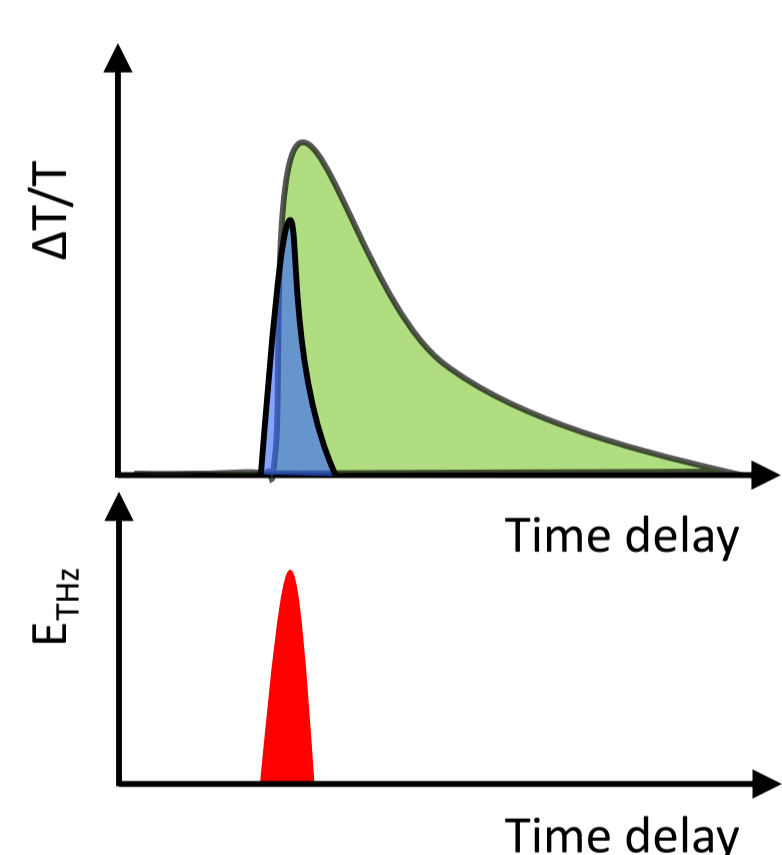
Using plasmon in graphene, several theoretical predictions have suggested that nonlinear properties can be enhanced as well.

Experimental verifications for the enhancement of nonlinear properties



1. Strong-pump induced transmission was observed (Fig. (a)), which matches well with a two-temperature model (Fig. (b)).
2. Red shift of the plasmon frequency was predicted [5].
3. To confirm the red shift of plasmon frequency, pump-induced transmission was obtained at various frequencies, change of sign of pump-induced transmission was observed (Fig. (c) and (d)) [6, 7].
4. These experimental studies suggest that **thermal nonlinearity** plays a pivotal role for nonlinear plasmonics in graphene.

Ultrafast response time of plasmonic nonlinearity



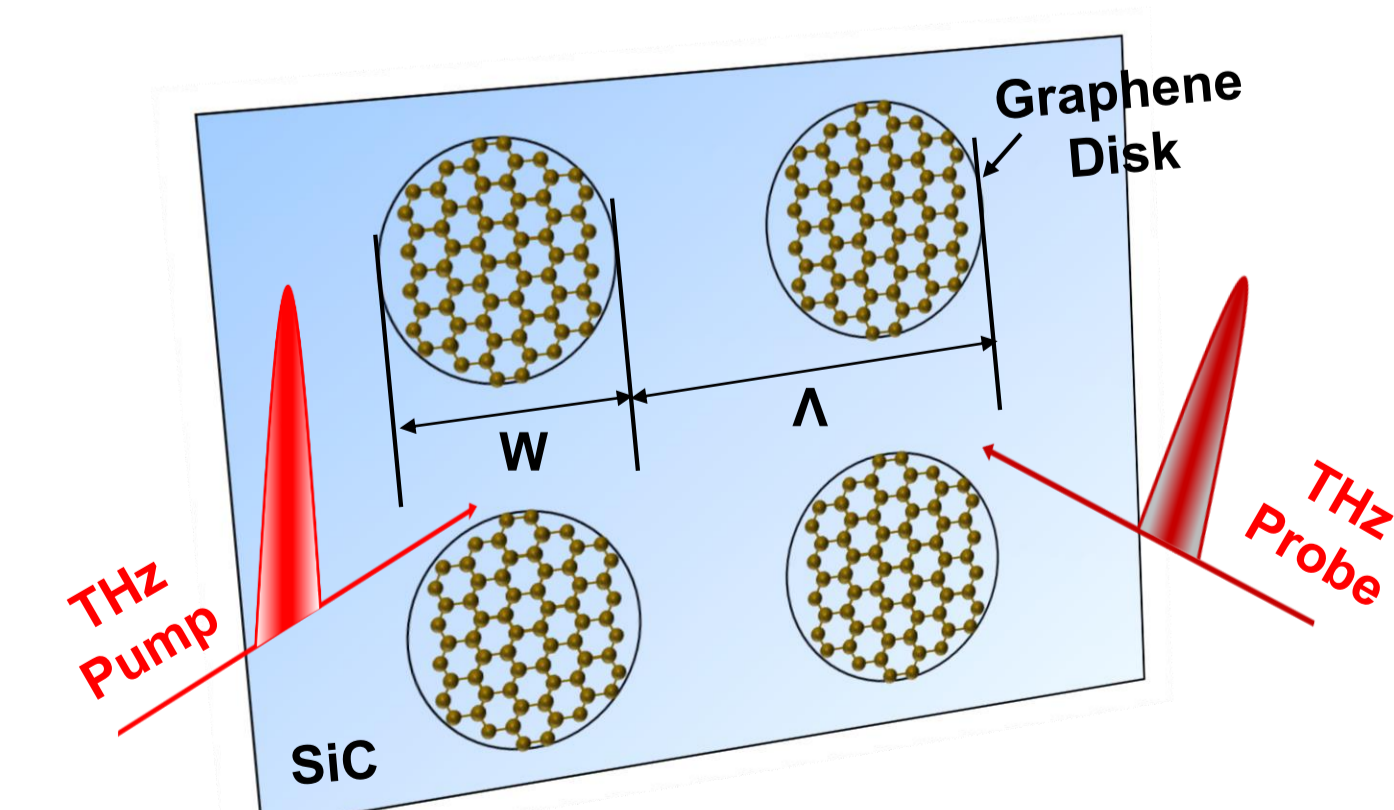
1. Thermal nonlinear effect depends on the electron temperature, leading to slow response (green curve in left figure).
2. Ultrafast response is expected for plasmonic nonlinearity (blue curve in left figure) [8, 9].

→ lack of experimental studies that characterized plasmonic nonlinearity!

in focus of this study: observation of plasmonic nonlinearity

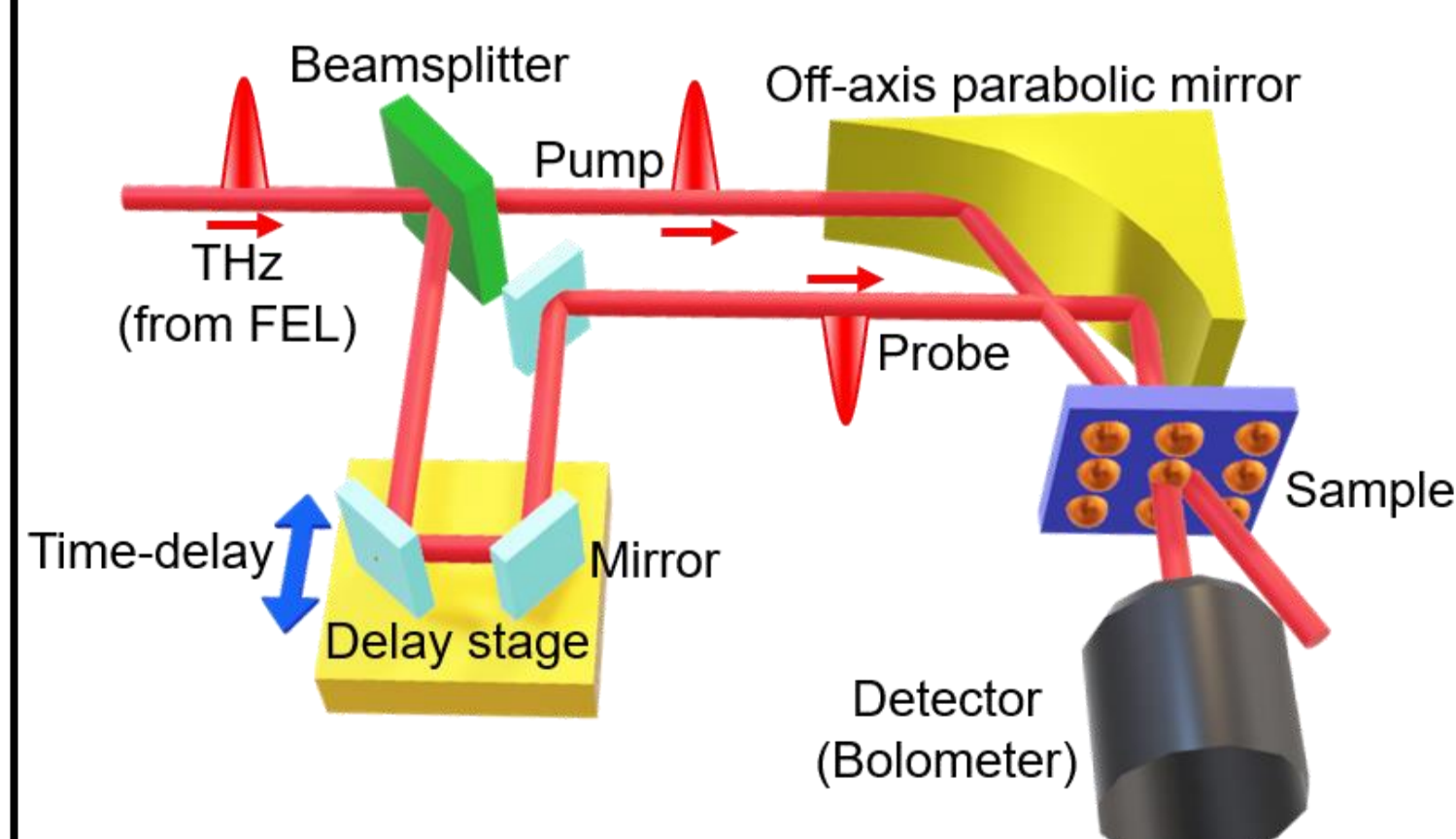
Sample and experimental setup

Sample (Graphene disks) geometry



Graphene disks were fabricated from hydrogen intercalation of epitaxial graphene on SiC.
W: Width of graphene disks (1280 nm)
Lambda: Period of graphene disks (1.85 micrometers)

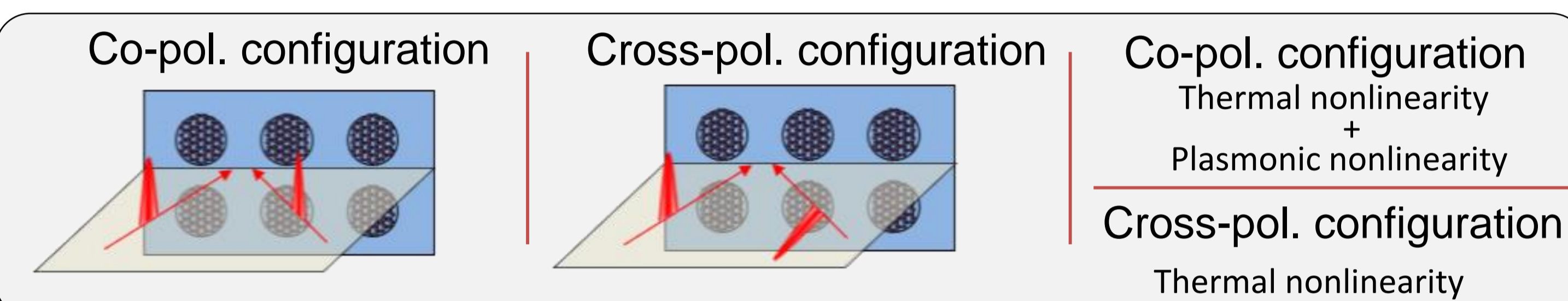
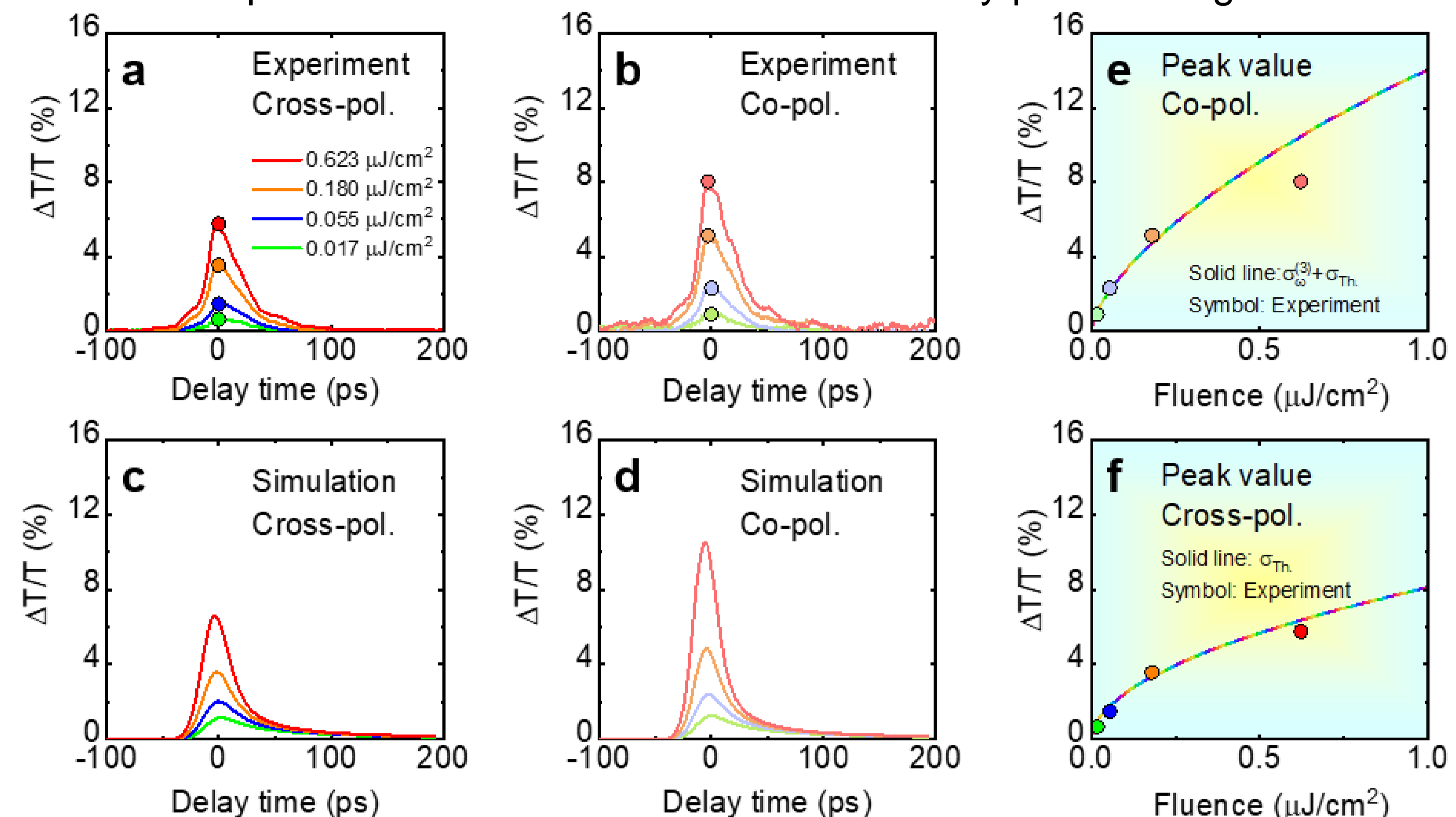
Experimental setup



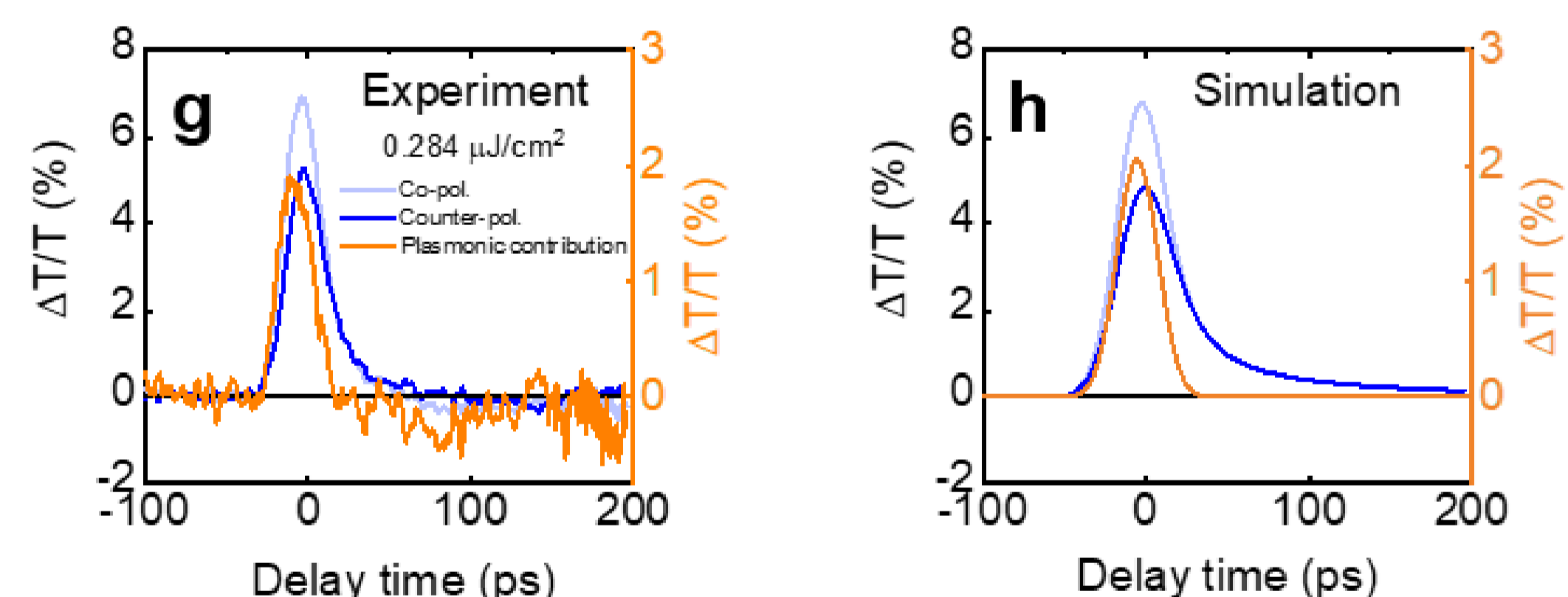
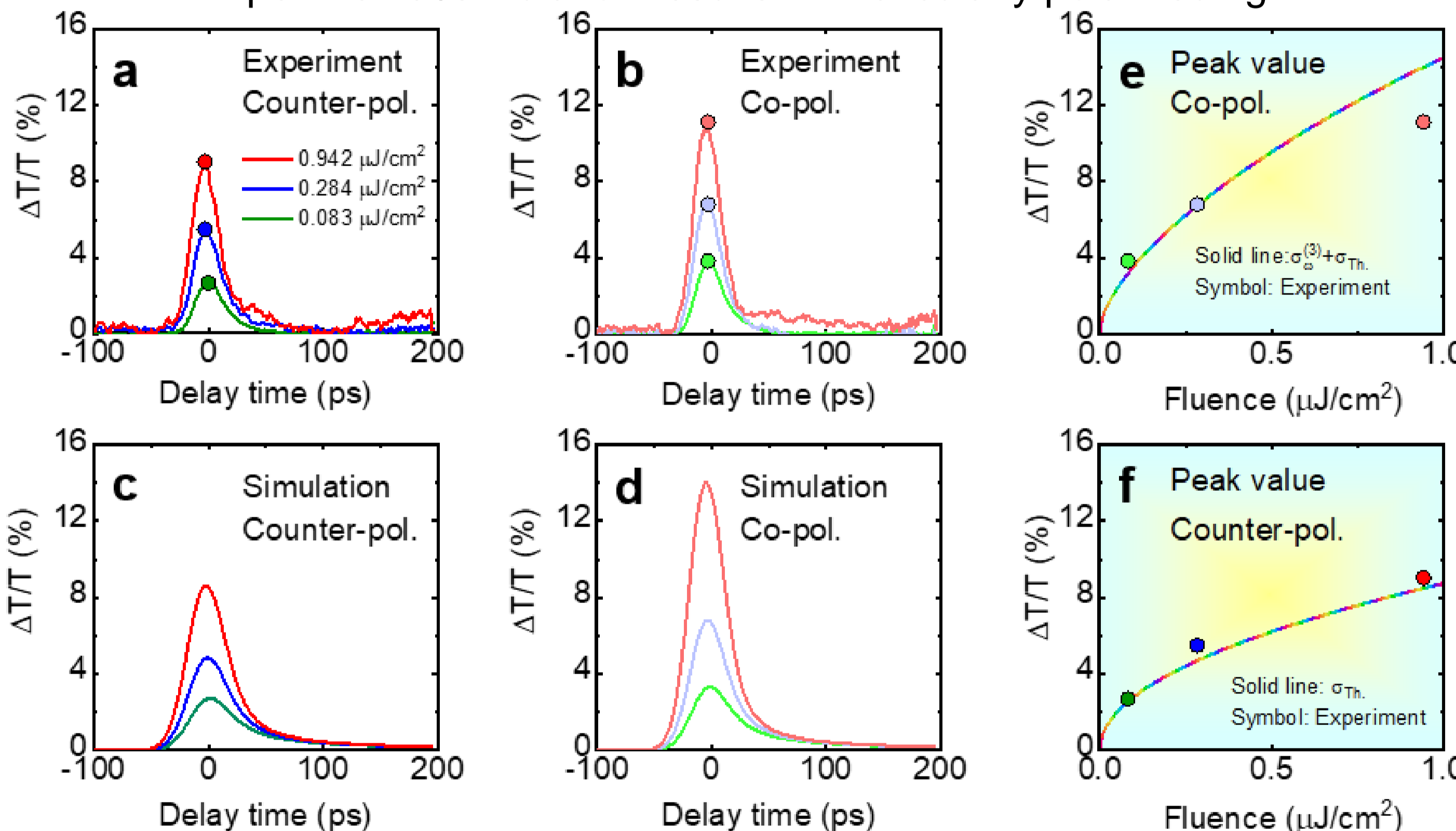
THz source: Helmholtz-Zentrum Dresden-Rossendorf
 Center wavelength: 3.5 THz
 Repetition rate: 13 MHz

Experimental/simulation results

Experimental/simulation results with linearly polarized-light



Experimental/simulation results with circularly polarized-light



1. pump-induced increase of transmission in both polarization configurations
2. more enhanced increase of transmission is observed in co-polarization configuration
3. → clear experimental evidence showing plasmonic nonlinearity beyond thermal effects
4. fast response time of plasmonic nonlinearity

Simulation model

Thermal nonlinearity

$$\sigma_{\omega}^{(1)} = \frac{w}{\Lambda} \frac{iD(T_e)}{\pi} \frac{1}{(\omega_{\text{eff}}(T_e) + i\Gamma(T_e))}$$

$D(T_e)$: Temperature-dependent Drude weight.
 $\Gamma(T_e)$: Temperature-dependent scattering rate.
 $\omega_{\text{eff}}(T_e) = (\omega^2 - \omega_{\text{p0}}^2(T_e))/\omega$ (ω_{p0} : Resonance frequency of plasmon).

Plasmonic nonlinearity (Kerr effect)

$$\sigma_{\omega}^{(3)} = \frac{w}{\Lambda} \frac{i9e^6 v_F^2}{4\pi\hbar^4 D(T_e)} \frac{1}{(\omega_{\text{eff}}(T_e) + i\Gamma(T_e))(-\omega_{\text{eff}}(T_e) + i\Gamma(T_e))(2\omega_{\text{eff}}(T_e) + i\Gamma(T_e))}$$

v_F : Fermi-velocity of graphene. (10⁸ cm/s)

Electronic temperature T_e (balanced equation)

$$\alpha_s T_e \frac{dT_e}{dt} + \beta_s (T_e^3 - T_L^3) = \mathfrak{I}(\omega_0, T_e) I(t)$$

α_s : Specific heat of electron in graphene.
 β_s : Super-collision cooling coefficient.
 T_L : Lattice temperature.
 $\mathfrak{I}(\omega_0, T_e) I(t)$: Increase of temperature via absorption.

Total conductivity and transmission (equivalent circuit model)

$$\sigma_{\text{tot}} = Q_1 \sigma_{\omega}^{(1)} + Q_2 \sigma_{\omega}^{(3)} [E \cdot E]$$

$$T = \frac{4\sqrt{\epsilon_1 \epsilon_2}}{Z_0^2 \left| \frac{\sqrt{\epsilon_1} + \sqrt{\epsilon_2}}{Z_0} + \sigma_{\text{tot}} \right|^2}$$

Z_0 : Impedance of free space (377 Ω).
 ϵ_1 : Dielectric constant for vacuum (1).
 ϵ_2 : Dielectric constant for SiC (10).

References

- [1]: K. M. Daniels *et al.*, 2D mater. **4**, 025034 (2017).
- [2]: M. Gullans *et al.*, Phys. Rev. Lett. **111**, 247401 (2013).
- [3]: J. D. Cox *et al.*, Nat. Comm. **5**, 5725 (2014).
- [4]: J. D. Cox *et al.*, ACS Nano **10**, 1995 (2016).
- [5]: M. M. Jadidi *et al.*, Nano Lett. **16**, 2734 (2016).
- [6]: M. Chin *et al.*, J. Phys. Photonics **3**, 01LT01 (2021).
- [7]: M. M. Jadidi *et al.*, ACS Photonics **6**, 302 (2019).
- [8]: J. D. Cox *et al.*, ACS Nano **10**, 1955 (2016).
- [9]: M. Kauranen *et al.*, Nat. Photonics **6**, 737 (2012).