

Challenges to Create Graphene-Based Terahertz/Infrared Lasers

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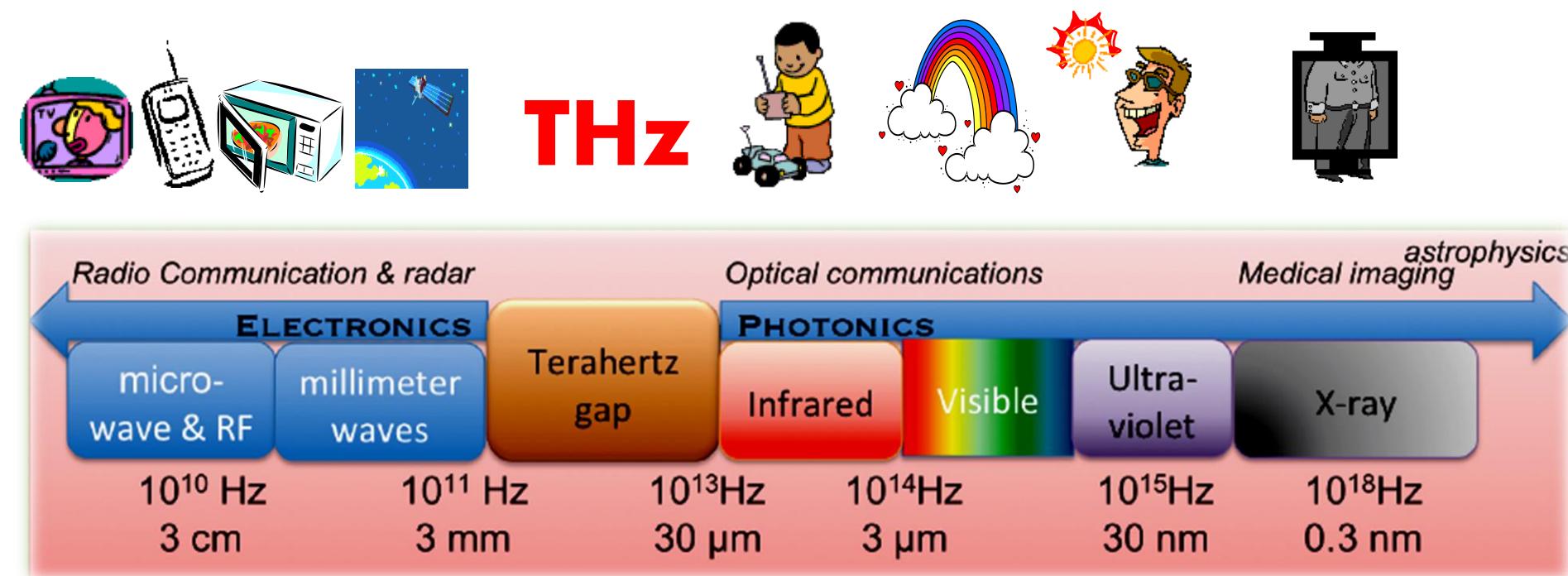
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Outline

- **Introduction and motivation**
- **Ultrafast carrier relaxation dynamics and THz/IR gain in optically/electrically pumped graphene**
- **Carrier heating & cooling effect in optical & injection pumping**
- **Graphene current-injection lasers**
- **Graphene active plasmonics for giant gain**
- **Summary**

Where is “Terahertz”?



Promising Applications for Terahertz ICTs

(Courtesy of Terahertz Technology Trend Investigation Committee, MIC, Japan)

Imaging

Prevention of terrorism and crime by detection of explosive and dangerous materials



THz Tech.



Biometrics



Detection of concealed articles by transparent imaging

Spectroscopy

Detection of drugs and infectious disease bacillus



Banned drug detection in envelopes

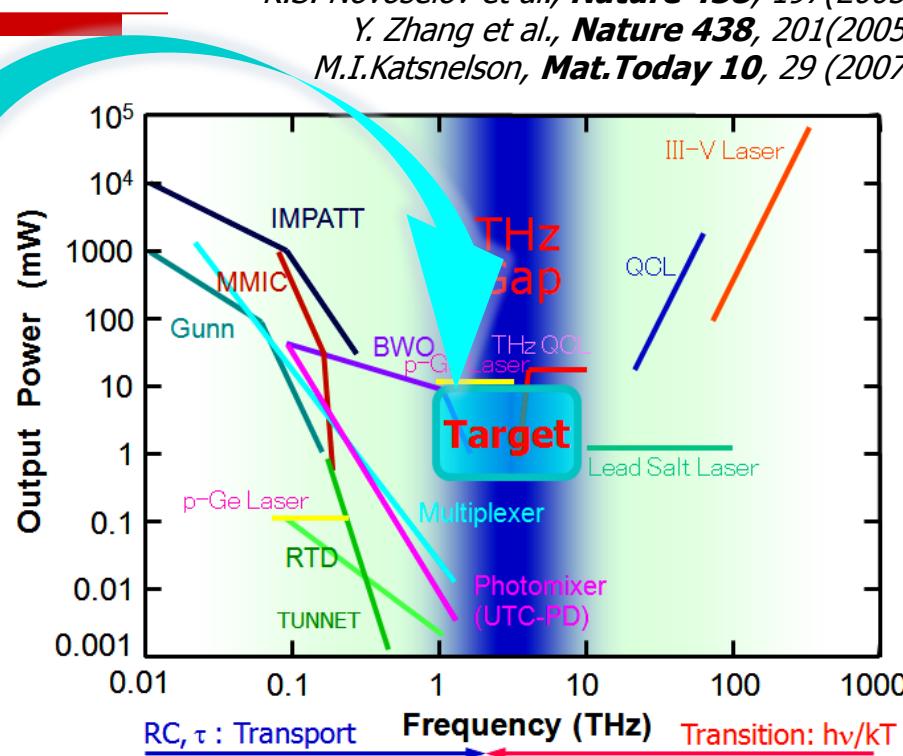
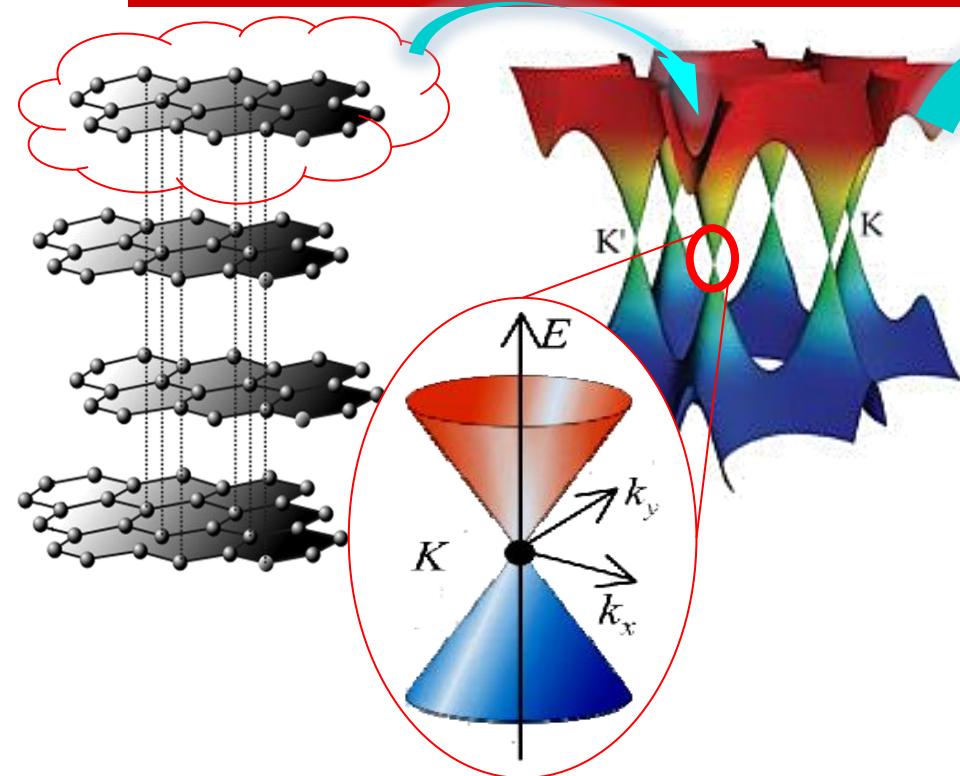
Telecom.

Ubiquitous ICT



Graphene can Bridge over the THz Gap!

P. R. Wallace, *PR 71*, 622 (1947).
K.S. Novoselov et al., *Science 306*, 666 (2004).
K.S. Novoselov et al., *Nature 438*, 197(2005).
Y. Zhang et al., *Nature 438*, 201(2005).
M.I. Katsnelson, *Mat.Today 10*, 29 (2007).

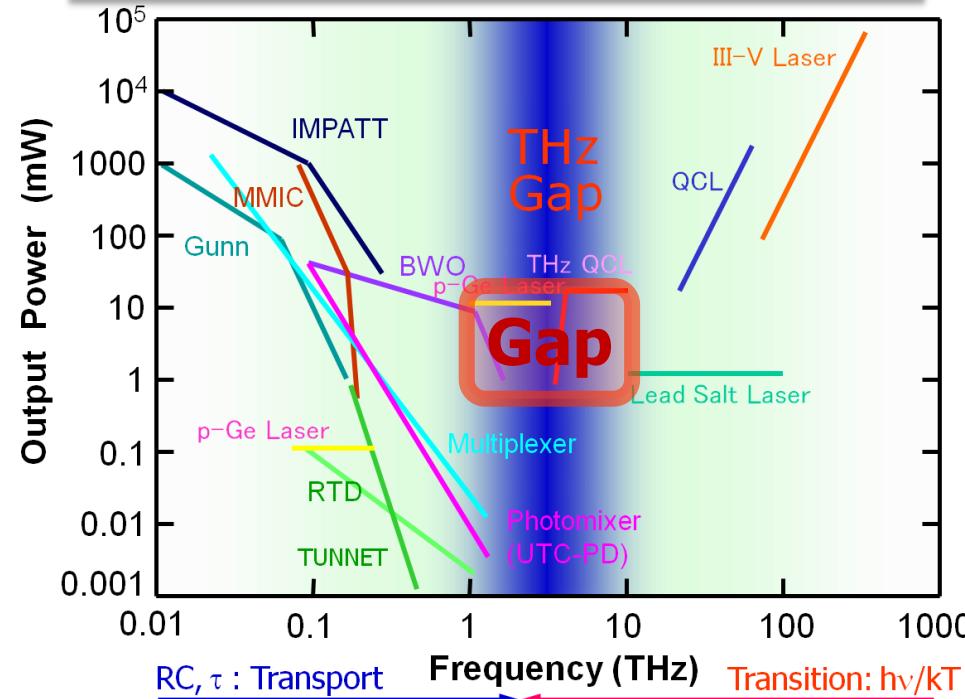


- Mono- or few layers of sp^2 bonded carbon atoms in a honeycomb lattice.
- Massless Dirac Fermions obey linear dispersion relation at K & K' points.
- High carrier mobility $\mu > 200,000 \text{ cm}^2/\text{Vs}$ at RT. (*cf.* InGaAs: $\mu \sim 12,000 \text{ cm}^2/\text{Vs}$)

Due to its unique transport properties, graphene is suitable for implementation in photonic devices.

Idea for Graphene THz/IR Lasers

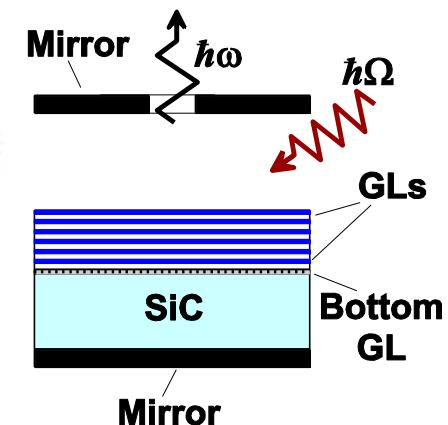
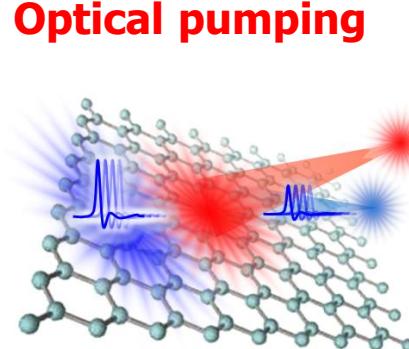
Current status of THz sources



(Courtesy of Terahertz Technology Trend Investigation Committee, MIC, Japan)

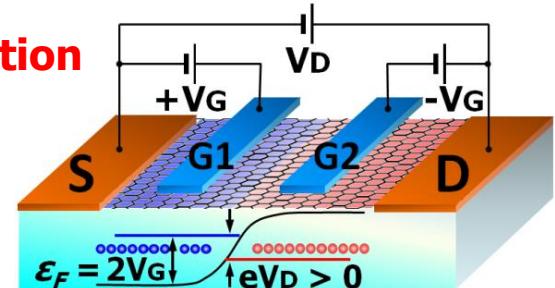
Proposal of graphene THz lasers

Optical pumping



V.Ryzhii, M.Ryzhii, T.Otsuji, *JAP* **101**, 083114 (2007).

Current injection



- QCLs only work at cryogenic temperatures.
- Need powerful, compact, room-temperature operating THz sources for imaging and communications.

M. Ryzhii and V. Ryzhii, *JJAP* **46**, L151 (2007).
V. Ryzhii, M. Ryzhii, V. Mitin, T. Otsuji, *JAP* **110**, 094503 (2011).

Negative dynamic conductivity of graphene with optical pumping

V. Ryzhii^{a)} and M. Ryzhii

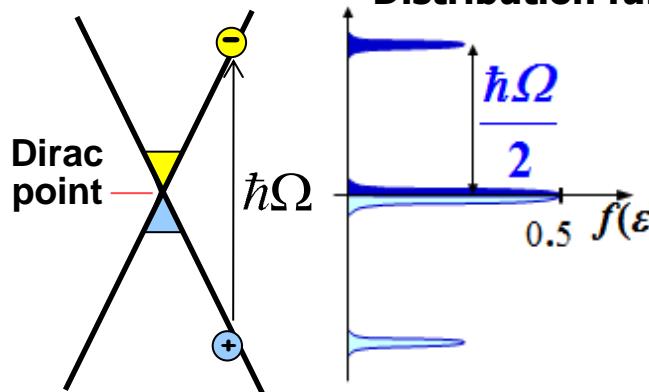
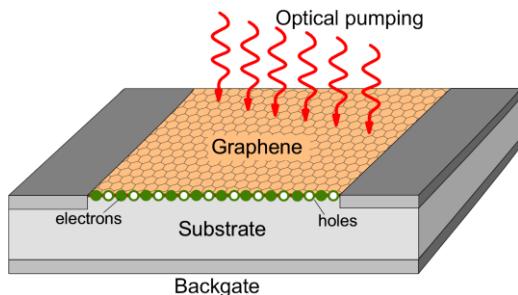
Computer Solid State Physics Laboratory, University of Aizu, Aizu-Wakamatsu 965-8580, Japan

T. Otsuji

Research Institute of Electrical Communication, Tohoku University, Sendai 980-8577, Japan

(Received 30 January 2007; accepted 8 February 2007; published online 26 April 2007)

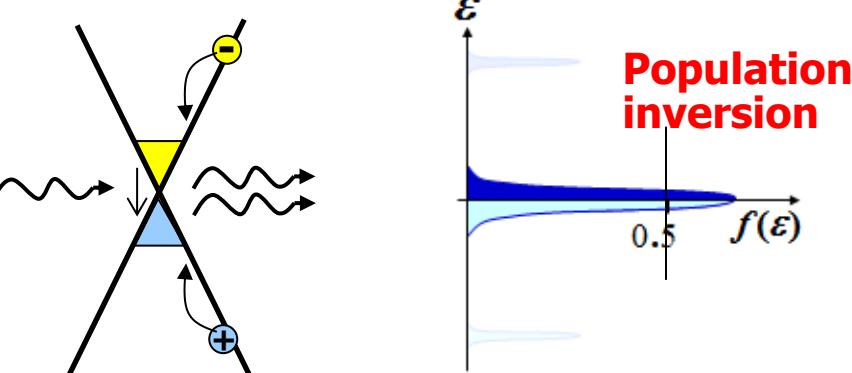
Optical pumping of intrinsic graphene



V. Ryzhii, M. Ryzhii, and T. Otsuji, JAP 101, 083114 (2007).

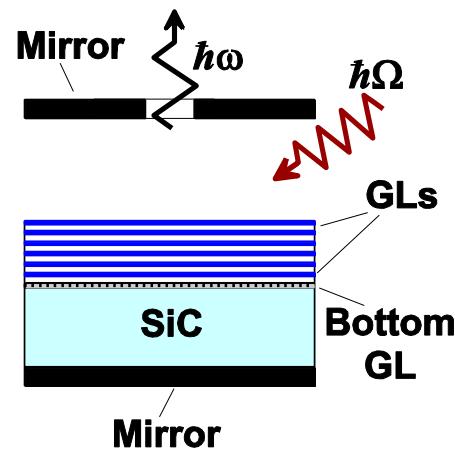
V. Ryzhii, et al., JAP 106, 084507 (2009).

Population inversion in THz (low-energy) region



$$1-10 \text{ THz} = 4-40 \text{ meV}$$

Cavity structure



Optical pumping

Relaxation by OPs

Accumulation of carriers near the Dirac point

Population inversion

$$1 - f_e - f_h = 1 - 2f < 0$$

Gain for THz waves + resonant cavity

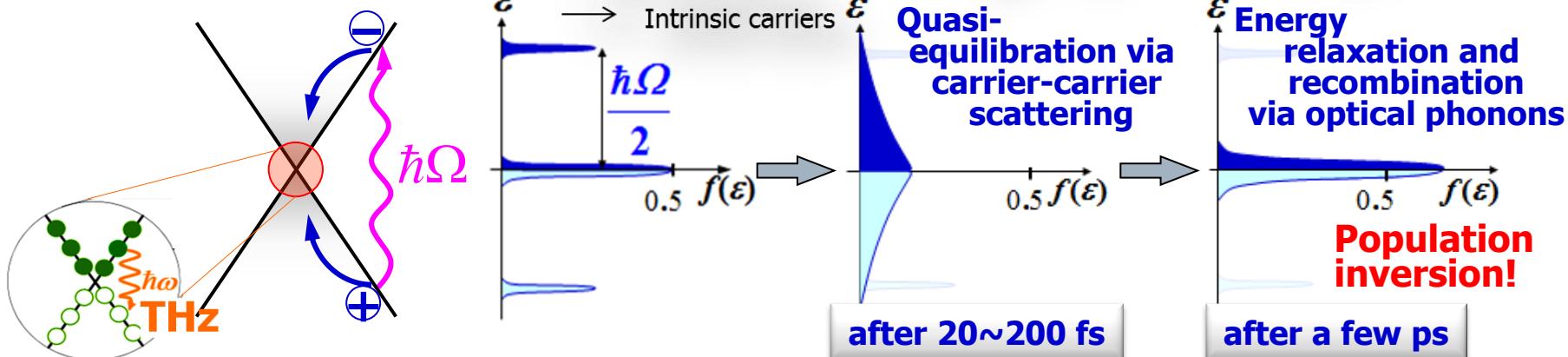
THz laser!

Carrier Relaxation Dynamics after Optical Pumping and Population Inversion at RT

Major carrier scatterings

- Carrier-carrier (CC) → Quasi-equilibration (20~200 fs)
- Intraband optical phonon (OP) → Energy relaxation (100 fs ~ a few ps)
- Interband OP → Energy relaxation & Recombination (1~10 ps)
- Auger-type → Recombination & Impact ionization (10 fs ~1 ps)

Distribution function



D. Sun et al., *PRL* **101**, 157402 (2008).

P.A. George et al., *Nano Lett.* **8**, 4248 (2008).

J. Dawlaty et al., *APL* **92**, 042116 (2008).

M. Breusing et al., *PRL* **102**, 086809 (2009).

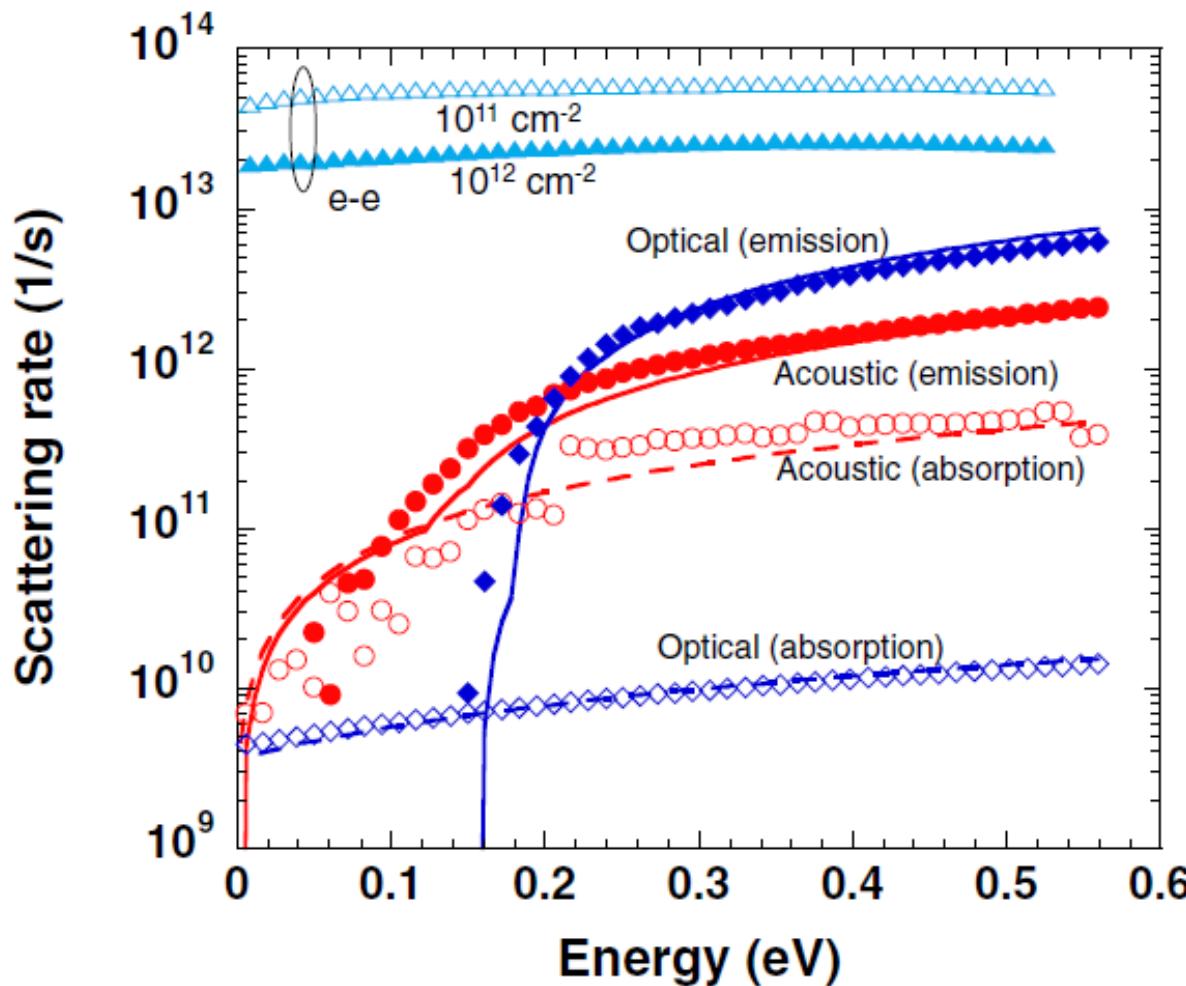
T. Winzer and E. Malic, *PRB* **85**, 241404(R) (2012).

Scattering Rates for Intrinsic Graphene Obtained by MC Simulation

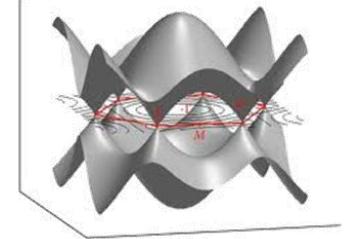
E. Sano, **JJAP** **50**, 090205 (2011).

X. Li et al., arXiv:1005.2631v1 (2010).

V. Perebeinos and Ph. Avouris, **PRB** **81**, 195442 (2010).



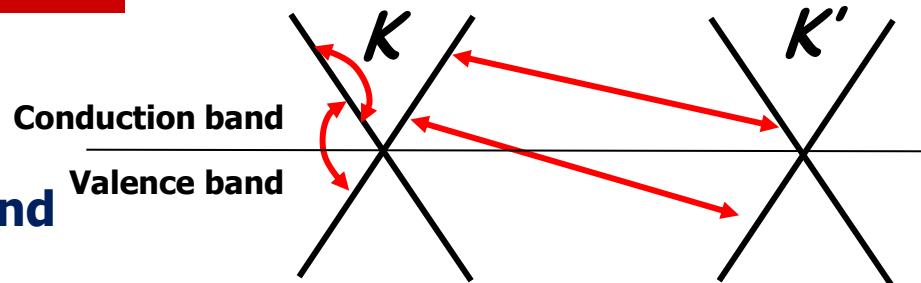
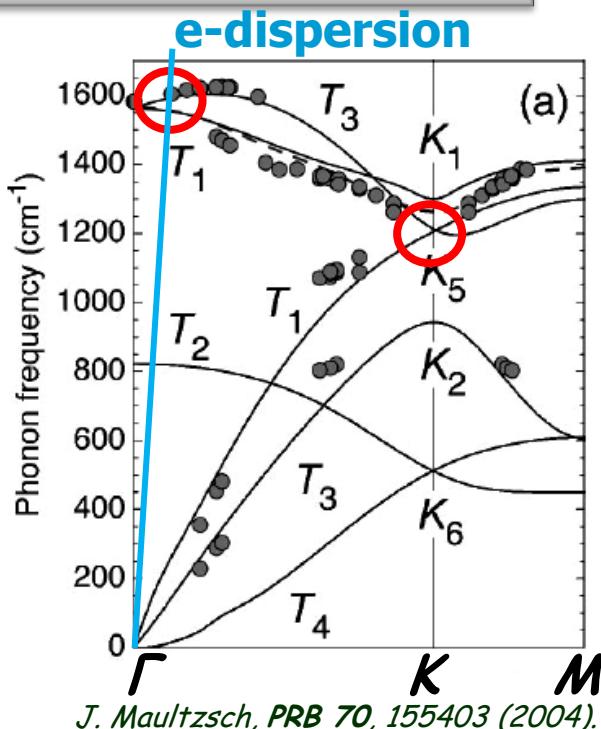
Optical Phonons in Graphene



Carriers interact with:

- Optical Phonons at Γ (Γ -LO&TO)
 - Intravalley & Intraband/Interband
- Optical Phonons at K (K-TO)
 - Intervalley & Intraband/Interband

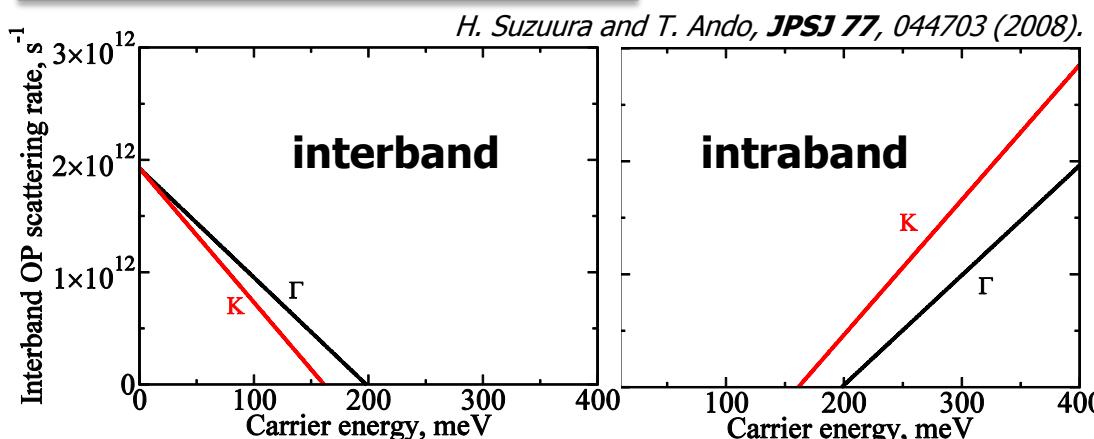
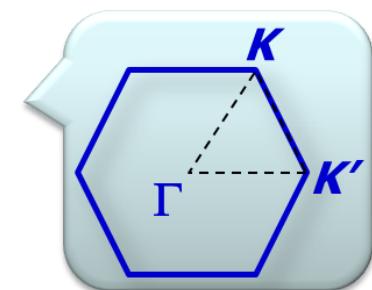
Phonon dispersion



Scattering rates

$$W_{A,\text{inter/tra}}^{(\pm)}(l\mathbf{k}, l\mathbf{k}') = \frac{2\pi}{\hbar} |\langle l\mathbf{k}' | H_{A,\text{inter/tra}} | l\mathbf{k} \rangle|^2 \cdot \delta(k - k' \pm \omega / v_F)$$

$$\frac{1}{\tau_{A,\text{inter/tra}}^{(\pm)}} = \sum_{\mathbf{k}'} W_{A,\text{inter/tra}}^{(\pm)}(l\mathbf{k}, l\mathbf{k}')$$



■ Time scale of OP emission: 300 fs \sim 3 ps

Theoretical Study of Graphene Under Pulse Excitation

A. Satou, T. Otsuji, and V. Ryzhii, **JJAP** **50**, 070116 (2011).
 H. Suzuura and T. Ando, **J. Phys. Soc. Jpn.** **77** 044703 (2008).

Rate equations for relaxation of quasi-Fermi level and carrier temperature

$$\frac{d\Sigma}{dt} = \frac{1}{\pi^2} \sum_{i=\Gamma,K} \int d\mathbf{k} \left[(1 - f_{h\omega_i - \nu_w \hbar k})(1 - f_{\nu_w \hbar k}) / \tau_{iO,\text{inter}}^{(+)} - f_{\nu_w \hbar k} f_{h\omega_i - \nu_w \hbar k} / \tau_{iO,\text{inter}}^{(-)} \right]$$

$$\frac{dE}{dt} = \frac{1}{\pi^2} \sum_{i=\Gamma,K} \int d\mathbf{k} \nu_w \hbar k \left[(1 - f_{h\omega_i - \nu_w \hbar k})(1 - f_{\nu_w \hbar k}) / \tau_{iO,\text{inter}}^{(+)} - f_{\nu_w \hbar k} f_{h\omega_i - \nu_w \hbar k} / \tau_{iO,\text{inter}}^{(-)} \right]$$

$$+ \frac{1}{\pi^2} \sum_{i=\Gamma,K} \int d\mathbf{k} h\omega_i \left[f_{\nu_w \hbar k} (1 - f_{\nu_w \hbar k + h\omega_i}) / \tau_{iO,\text{intra}}^{(+)} - f_{\nu_w \hbar k} (1 - f_{\nu_w \hbar k - h\omega_i}) / \tau_{iO,\text{intra}}^{(-)} \right]$$

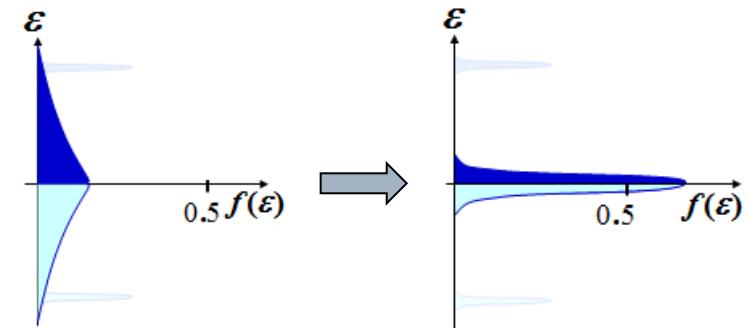
Quasi-Fermi distribution caused by CC scattering

$$f_{\nu_w \hbar k} = \frac{1}{\exp[(\nu_w \hbar k - \varepsilon_F)/k_B T_c] + 1}$$

with $\varepsilon_F = \varepsilon_F(t)$, $T_c = T_c(t)$

$\tau_{iO,\text{inter}}, \tau_{iO,\text{intra}}^{(\pm)}$

Relaxation time for interband and intraband OP ("+" for absorption and "-" for emission; i= K, Γ)



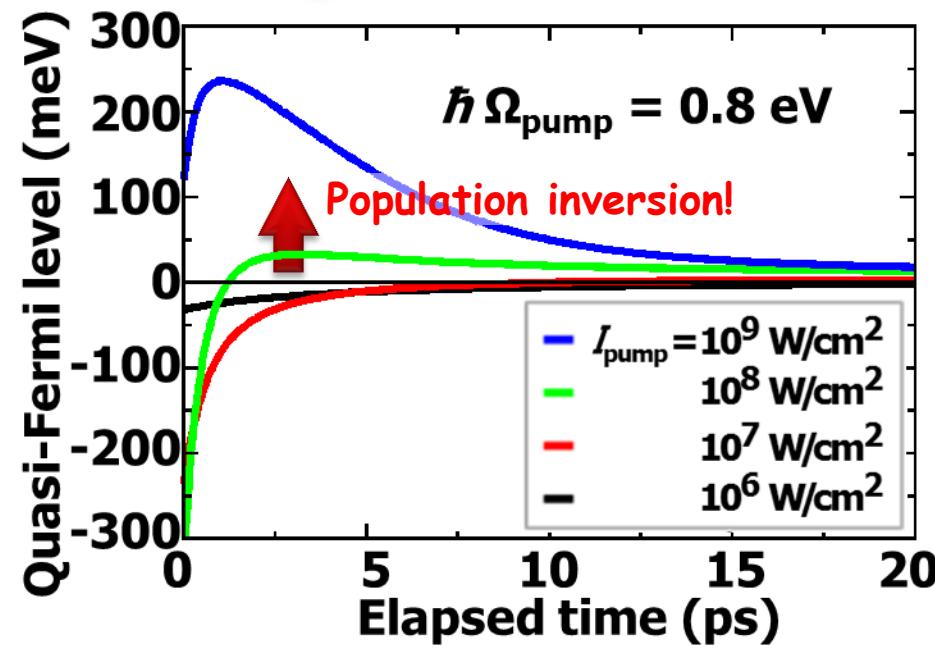
Relaxation of Quasi-Fermi Level and Carrier Temperature

A. Satou, T. Otsuji, and V. Ryzhii, **JJAP 50**, 070116 (2011).

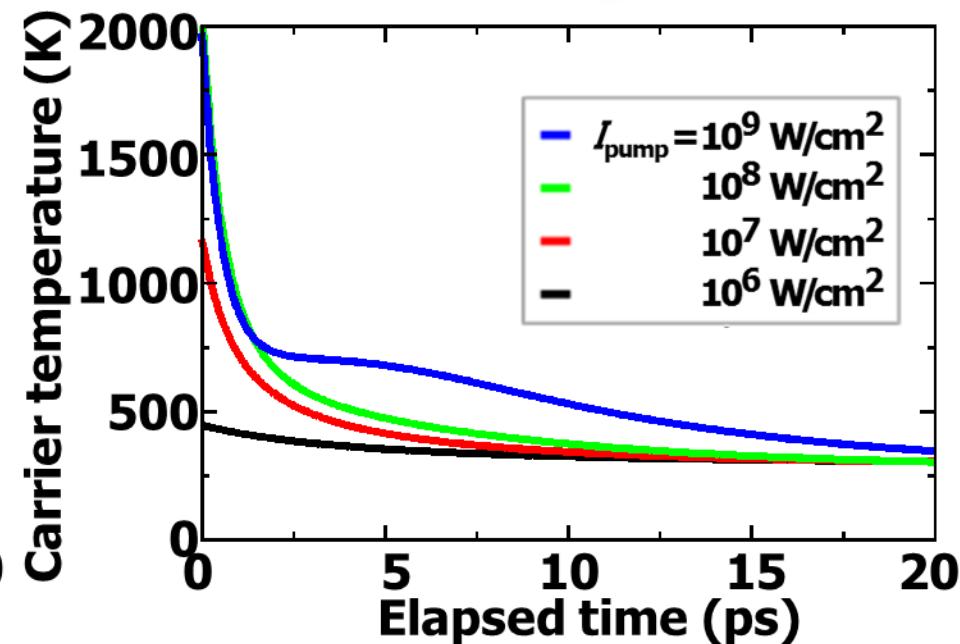
Δt Pulse width = 80 fs

I_{pump} Peak intensity

Quasi-Fermi level



Carrier temperature

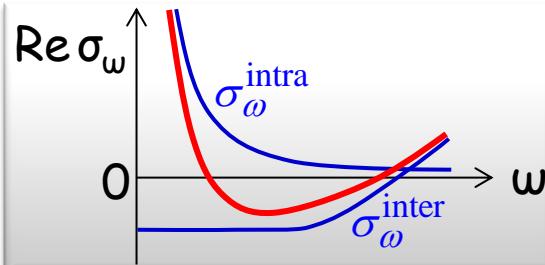


■ Population inversion occurs with a threshold of pumping intensity!

Time-Dependent Dynamic Conductivity

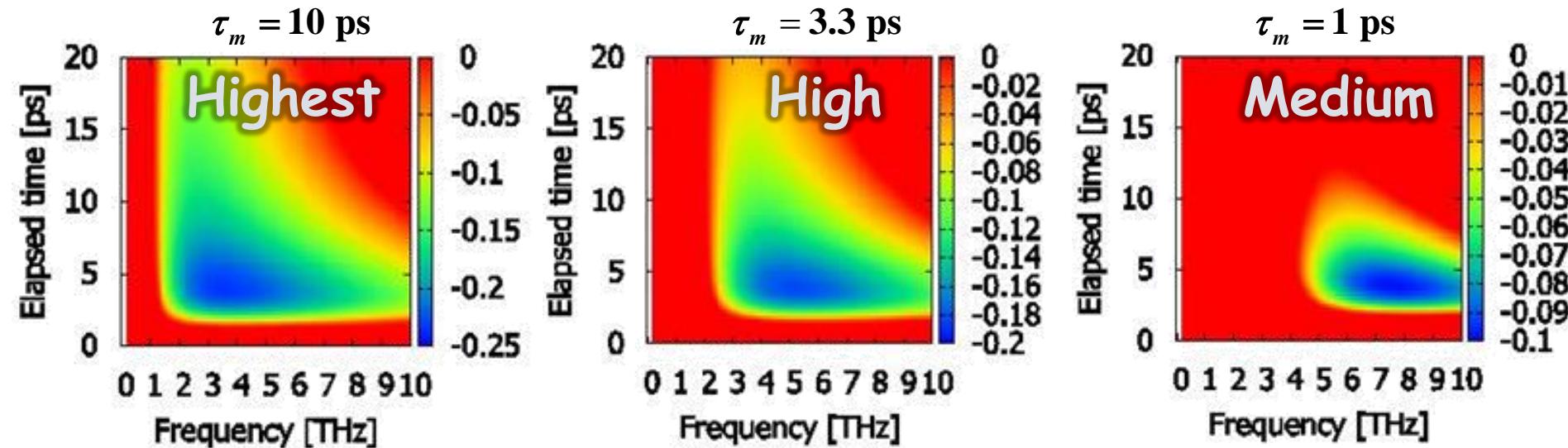
T. Otsuji et al., *J. Phys. D* **45**, 303001 (2012).

Dynamic conductivity



$$\begin{aligned} \text{Re } \sigma_{\omega} &= \text{Re } \sigma_{\omega}^{\text{inter}} + \text{Re } \sigma_{\omega}^{\text{intra}} \\ &\approx \frac{e^2}{4\hbar} \left(1 - 2f_{\hbar\omega}\right) + \frac{(\ln 2 + \varepsilon_F / 2k_B T)e^2}{\pi\hbar} \frac{k_B T \tau}{\hbar(1 + \omega^2 \tau^2)} \\ &\approx \frac{e^2}{4\hbar} \tanh\left(\frac{\hbar\omega - 2\varepsilon_F}{4k_B T}\right) + \frac{(\ln 2 + \varepsilon_F / 2k_B T)e^2}{\pi\hbar} \frac{k_B T \tau}{\hbar(1 + \omega^2 \tau^2)} \end{aligned}$$

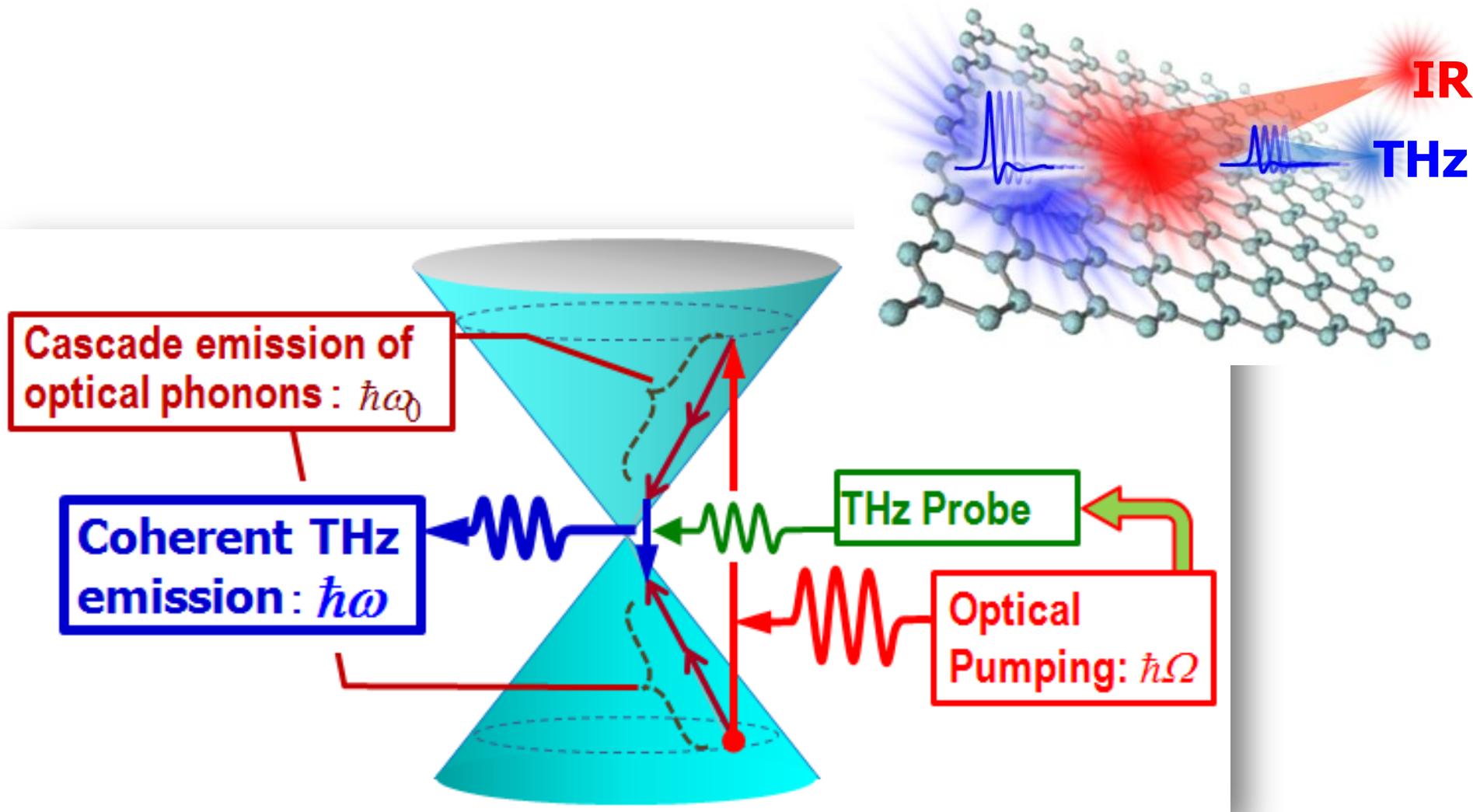
(intra) = Drude absorption



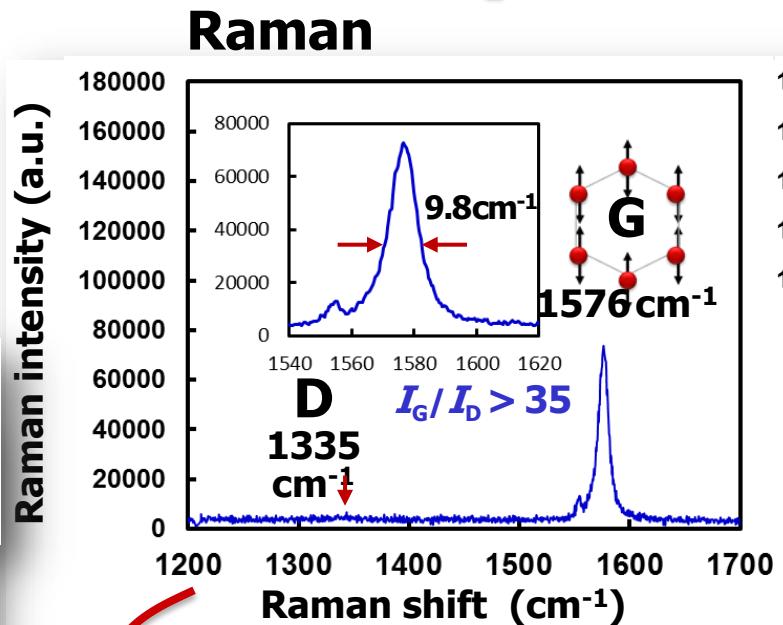
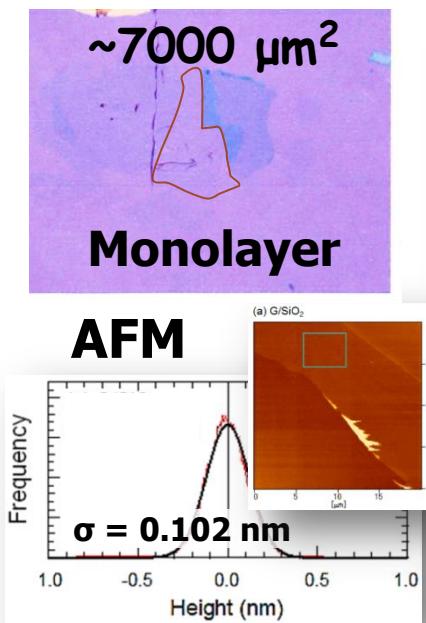
■ Longer relaxation time, larger and broader NDC

$I_{\text{pump}} = 10^8 \text{ W/cm}^2$
80 fs FWHM

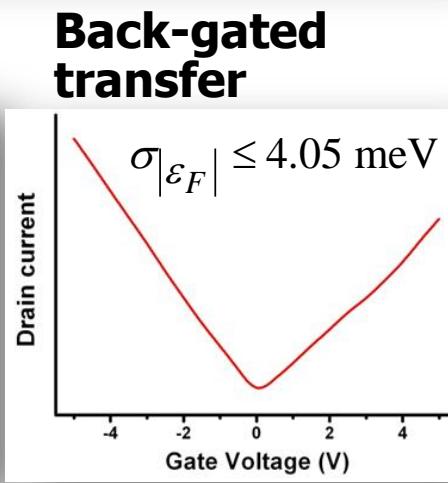
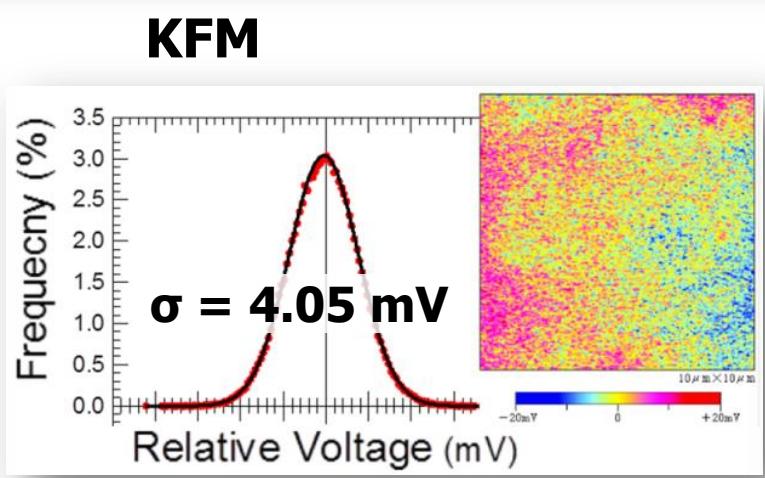
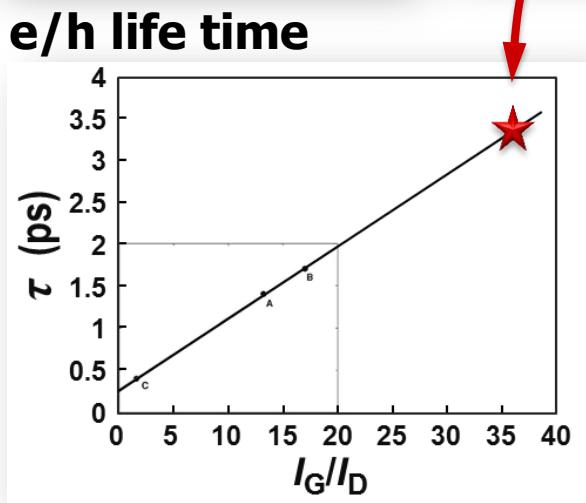
Proof-of-Concept Experiment: Optical Pumping/THz Probing Graphene



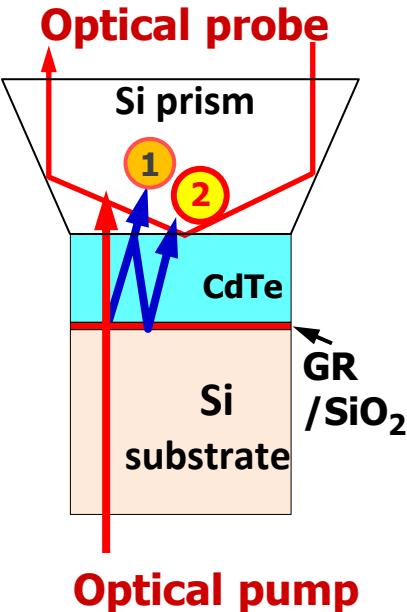
Exfoliated Monolayer Graphene/SiO₂/Si



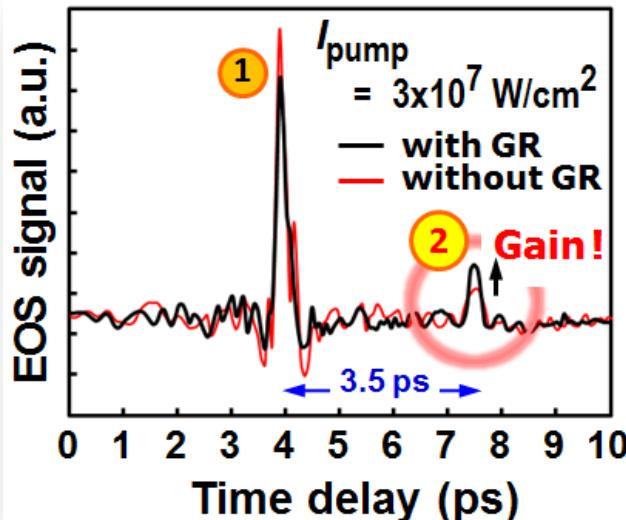
T. Otsuji et al., J. Phys. D 45, 303001 (2012).



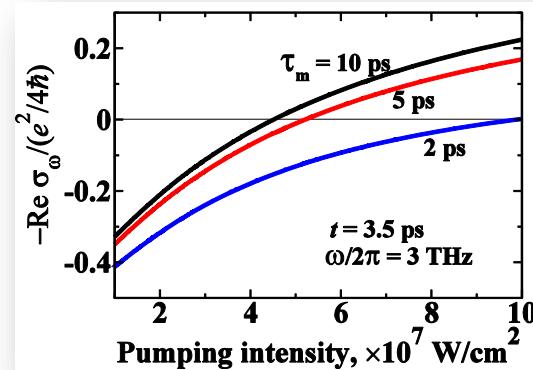
Observation of Threshold Behavior, Proving Stimulated THz Emission & Gain



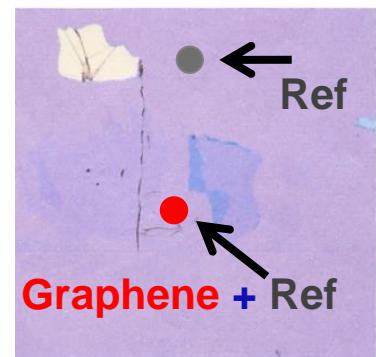
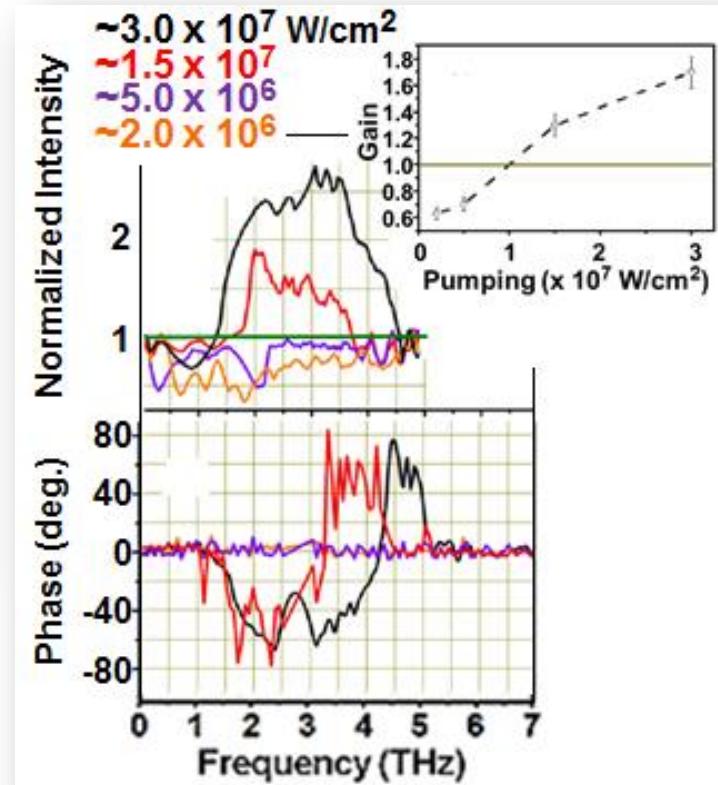
Experimental result



Theoretical result



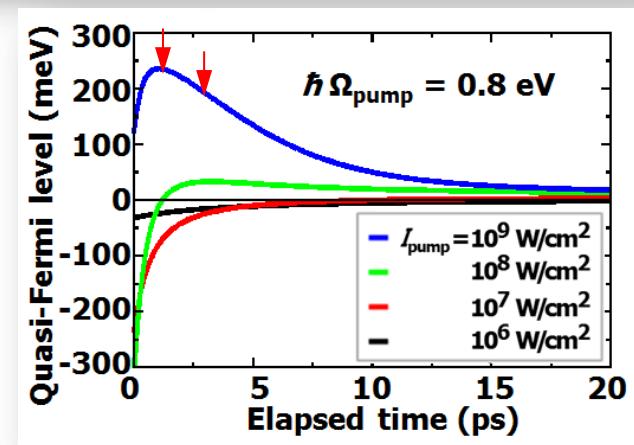
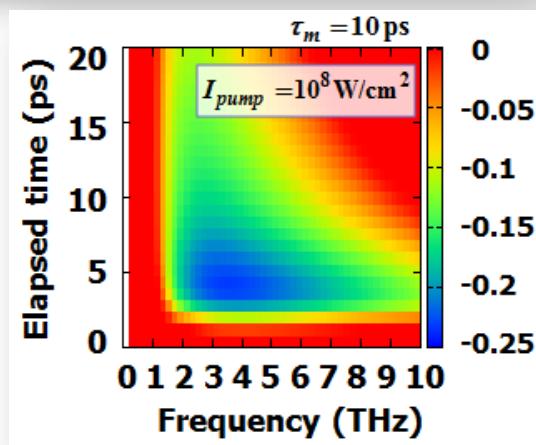
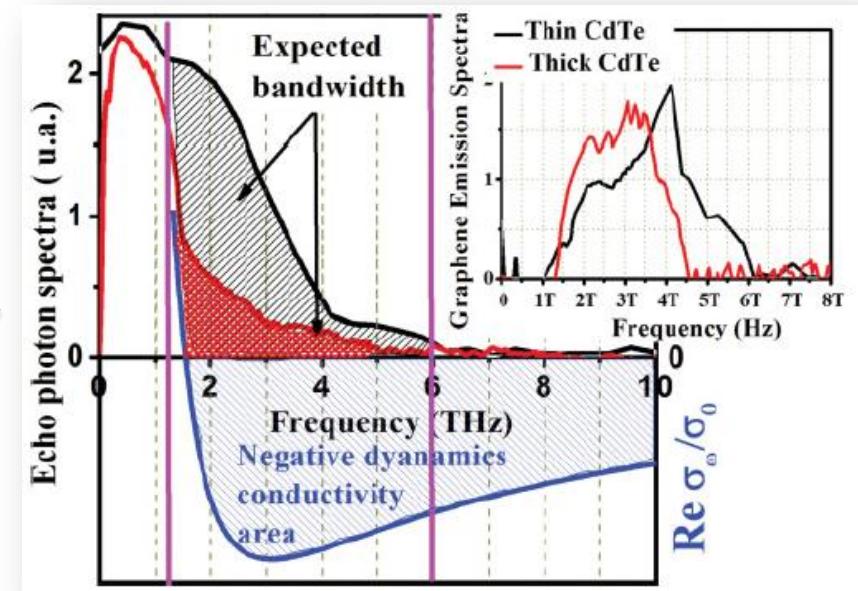
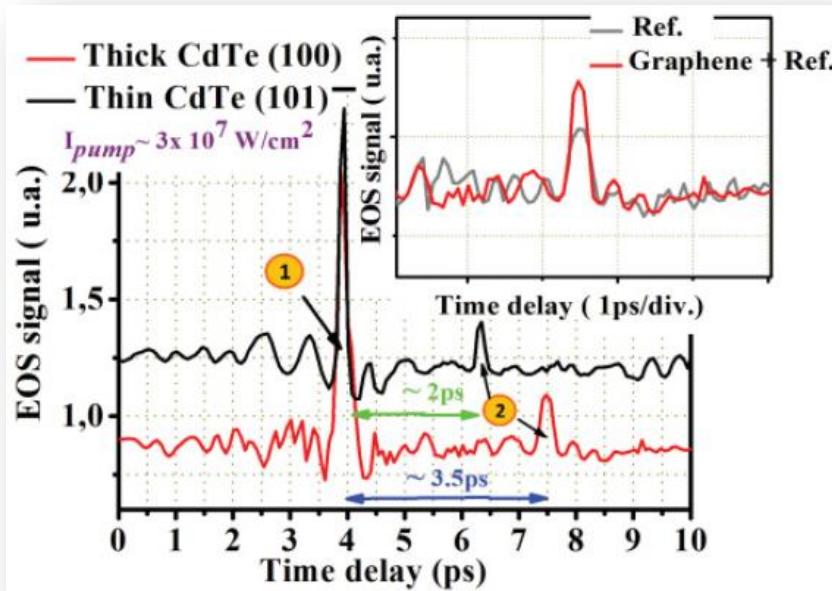
S. Boubanga Tombet et al., PRB 85, 035443 (2012).



- Threshold behavior
- Normal dispersion
- ☞ Proving the THz gain!

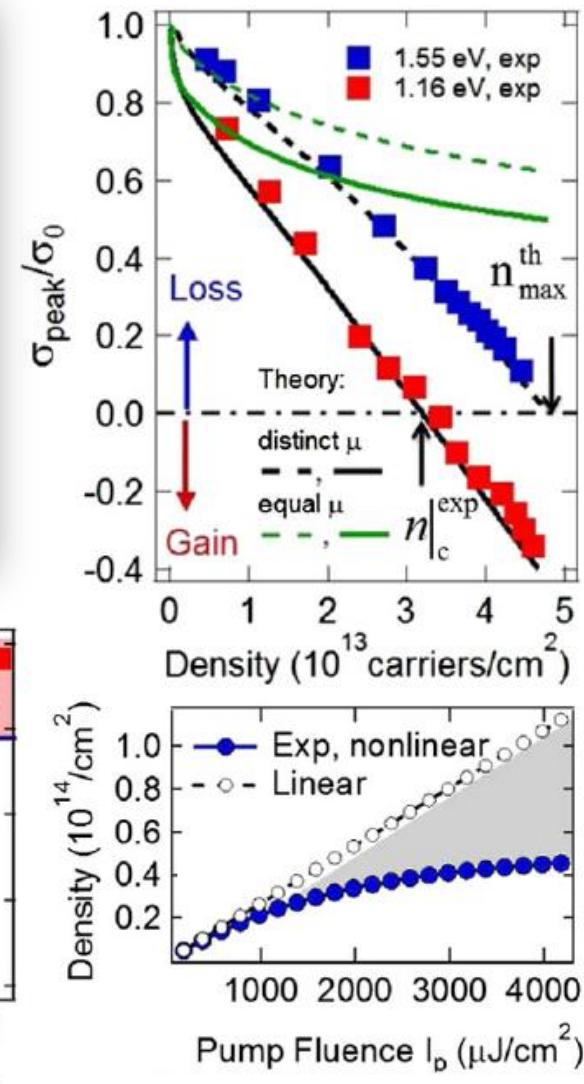
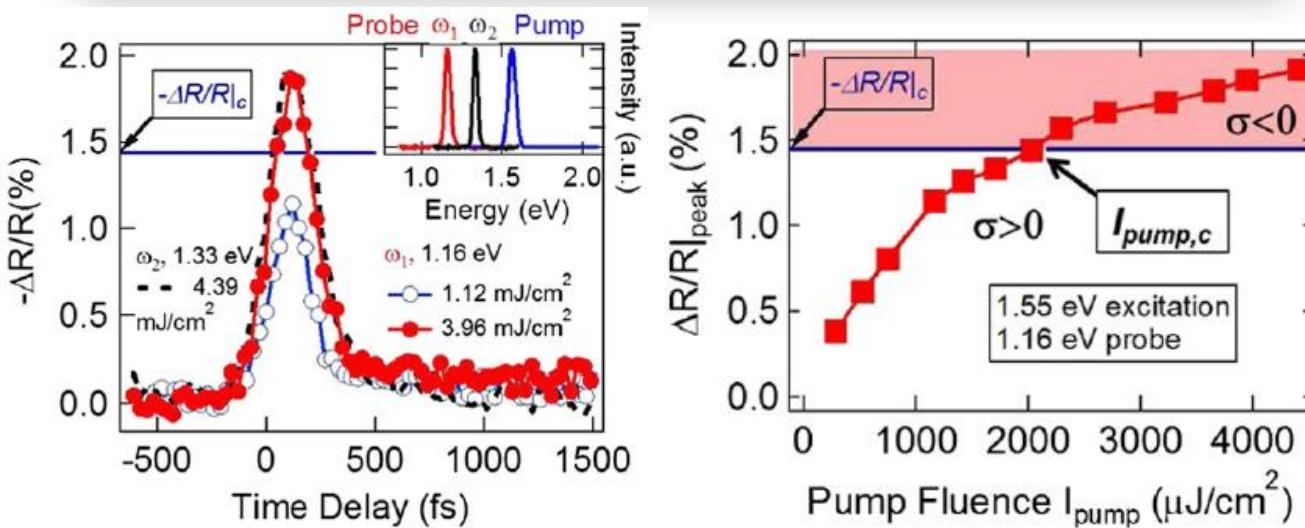
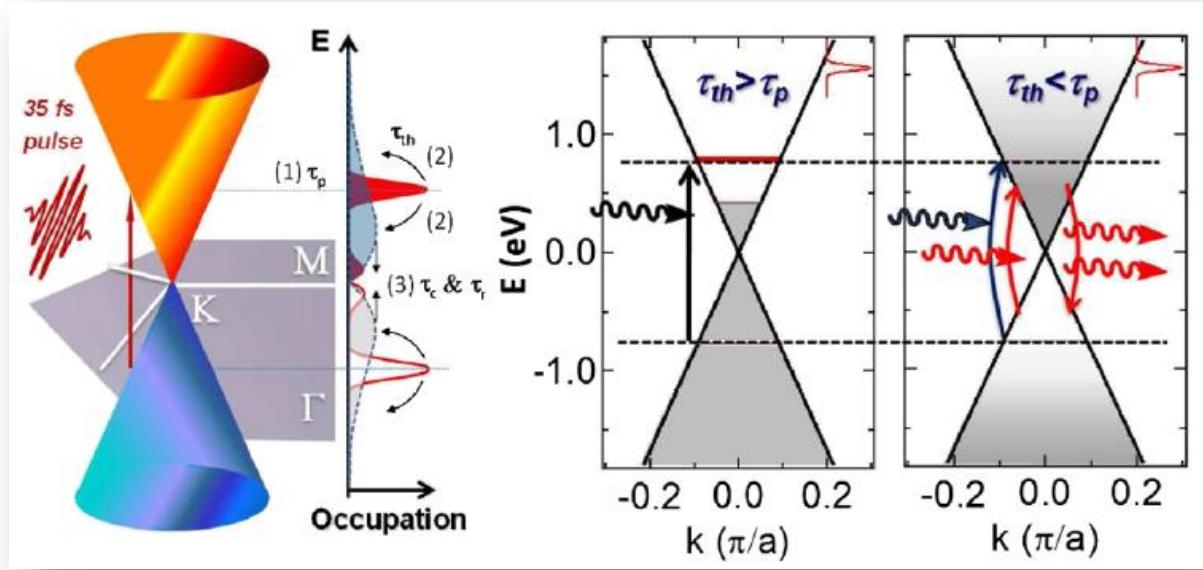
Narrower Emission Spectra at a Longer Probe Delay, Reflecting Equilibration

S. Boubanga Tombet et al., PRB 85, 035443 (2012).



Observation of IR Stimulated Emission in fs Regime before Quasi-Equilibration

T. Li et al., *PRL 108*, 167401 (2012).

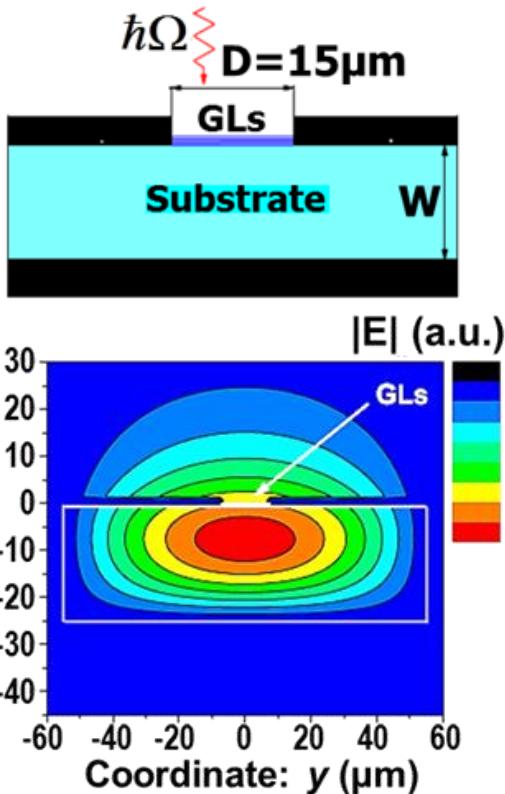


Gain Profile of MGL Laser Calculated for Slot-Line Waveguides

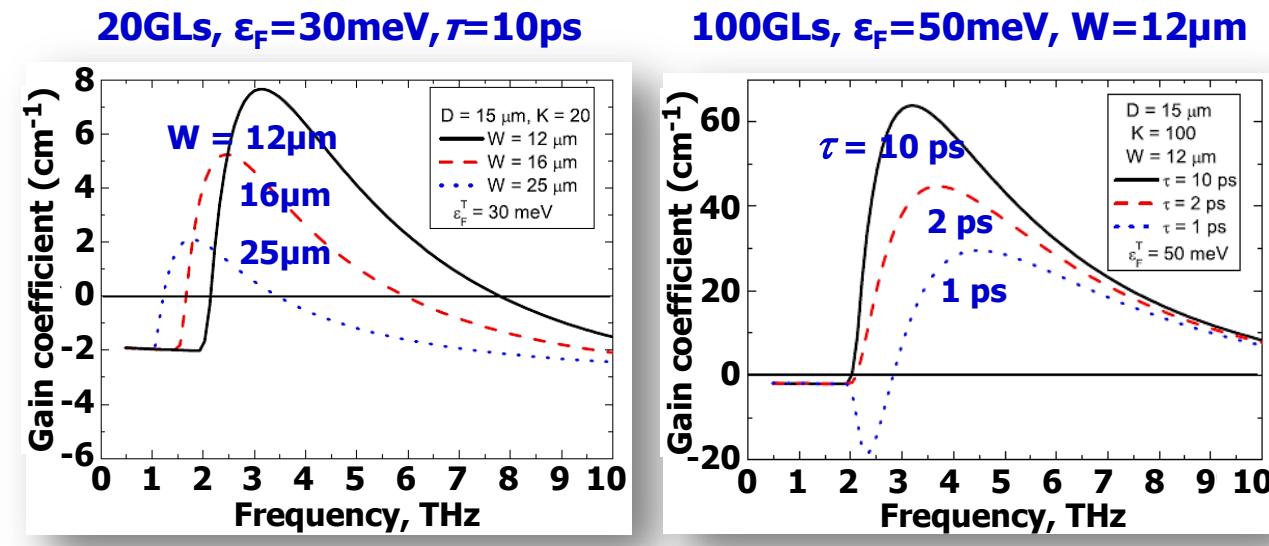
JOURNAL OF APPLIED PHYSICS 107, 054505 (2010)

Terahertz lasers based on optically pumped multiple graphene structures with slot-line and dielectric waveguides

V. Ryzhii,^{1,2,a)} A. A. Dubinov,^{1,3} T. Otsuji,^{2,4} V. Mitin,⁵ and M. S. Shur⁶



$$g_\omega = \frac{4\pi \operatorname{Re} \sigma_\omega}{c\sqrt{\eta_S}} \Gamma_\omega - \alpha_\omega, \quad \Gamma_\omega = \frac{\int_{-D/2}^{D/2} |E_\omega(y, 0)|^2 dy}{\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} |E_\omega(y, z)|^2 dy dz}$$

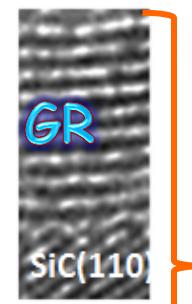
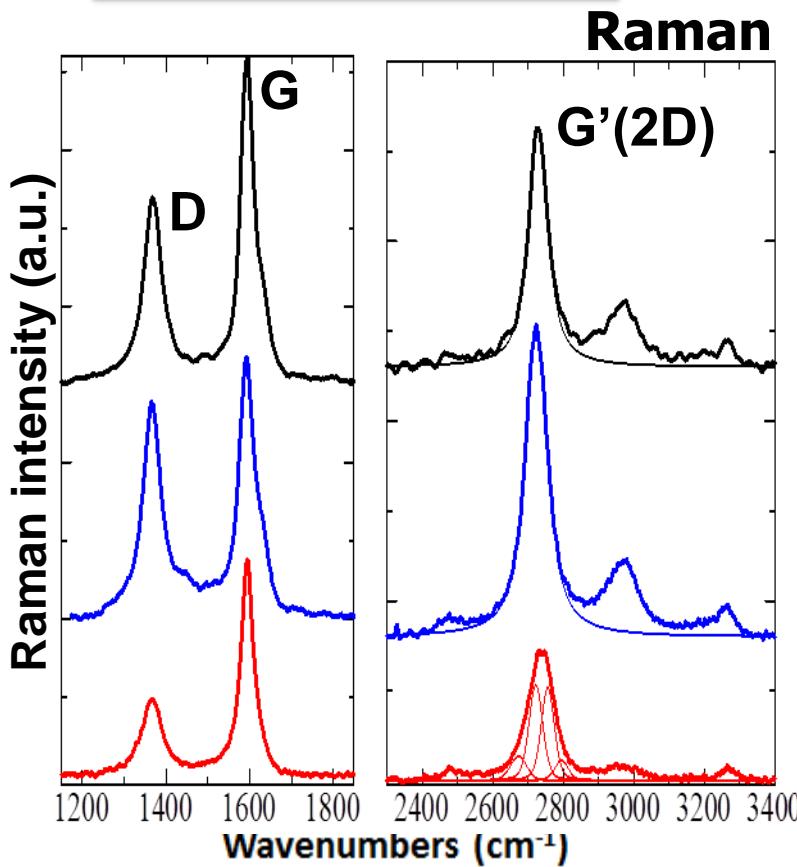


MLG: Epitaxial Graphene on SiC/Si can Control the GR-Layer Stacking

GOS: Graphene on Si

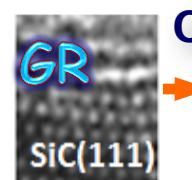
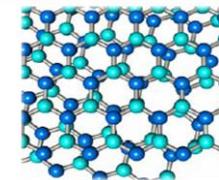


H. Fukidome et al., *J. Mat. Chem.* **21**, 17242 (2011).
M. Suemitsu and H. Fukidome, *J. Phys. D* **43**, 374012 (2010).
M. Suemitsu et al., *e-J. Surface Sci. Nano.* **7** 311 (2009).



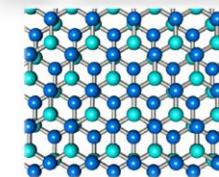
On SiC(100)/Si(100)
SiC(110)/Si(110)

☞ Turbostratic
Non-Bernal



On SiC(111)/SiC(111)

☞ AB stacking



■ Multilayer of monolayer GR

■ High power applications

■ THz photonic applications

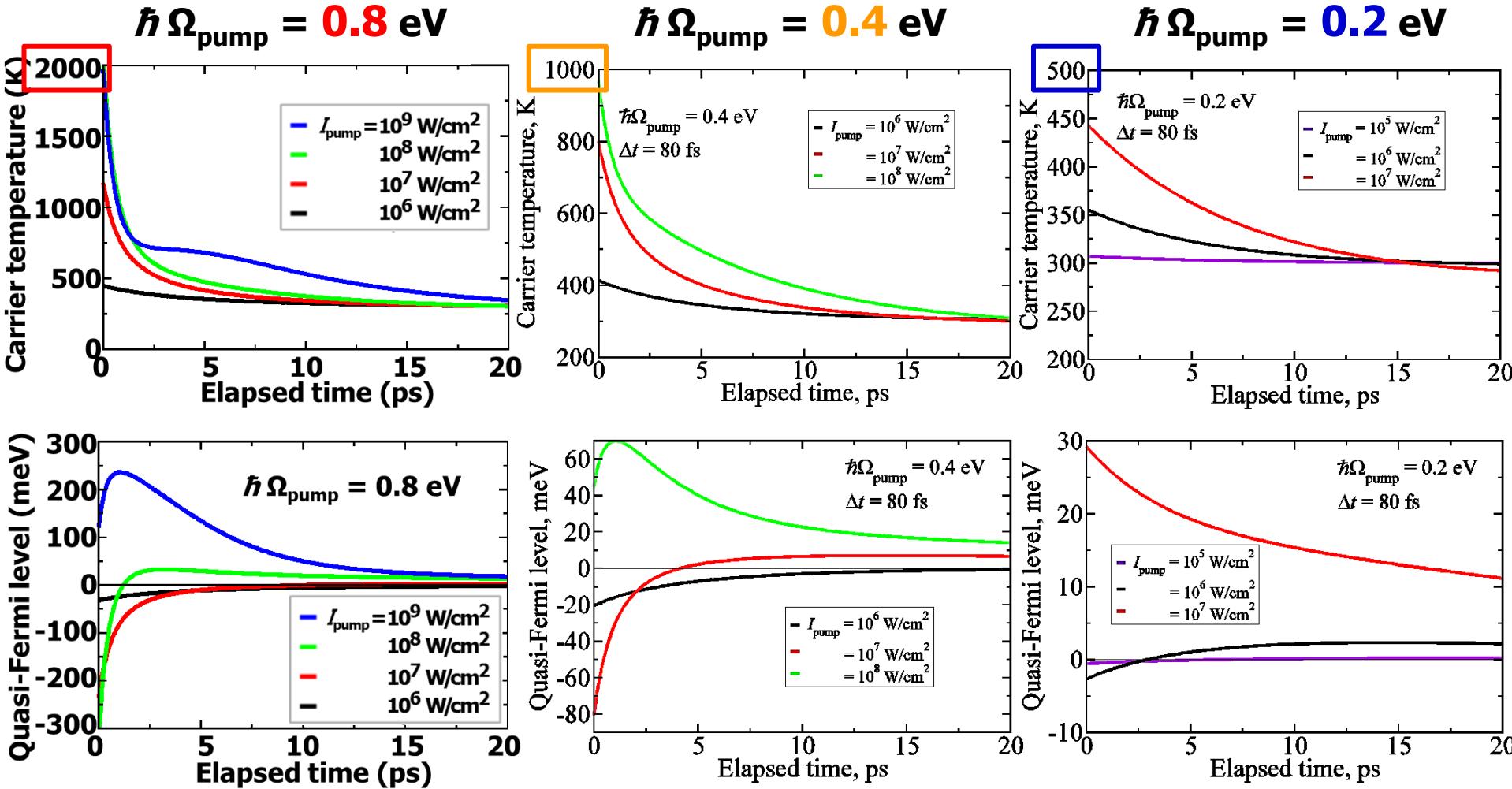
■ High on/off ratio

■ Transistor & logic ICs

Pumping Photon Energy vs. T_c & ϵ_F

- Impulsive Pumping

T. Otsuji et al., IEEE T. THz. Sci. Tech. 3, 63 (2013).



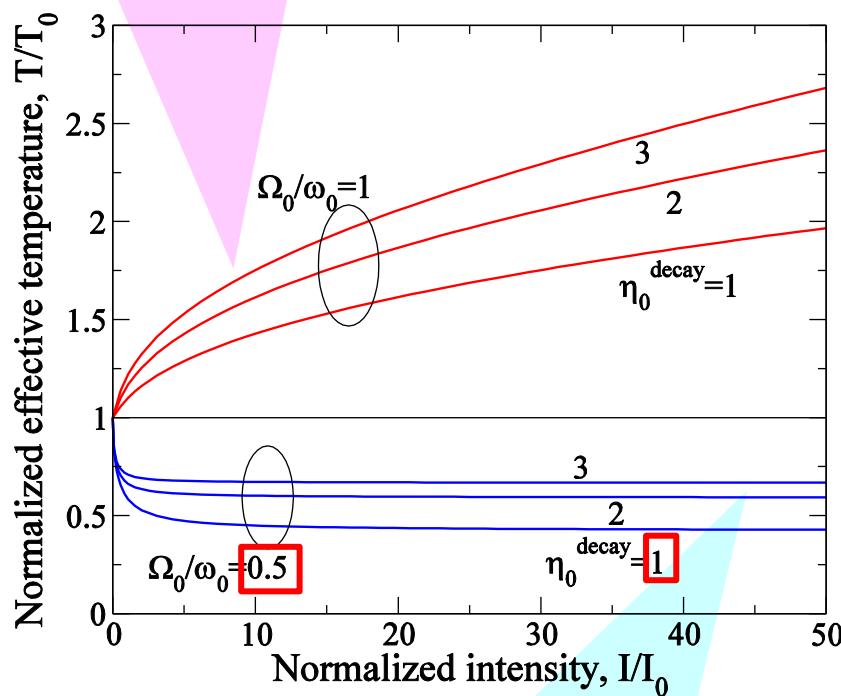
Effects of Optical-Phonon Decay τ_o^{decay} & Effective Pumping Photon Energy Ω_0

V. Ryzhii, M. Ryzhii, V. Mitin, A. Satou, and T. Otsuji, *JJAP* 50, 094001 (2011).

Thermal conductivity⁻¹ \propto

$$\eta_0^{\text{decay}} = \tau_o^{\text{decay}} / \tau_o^{\text{inter}}$$

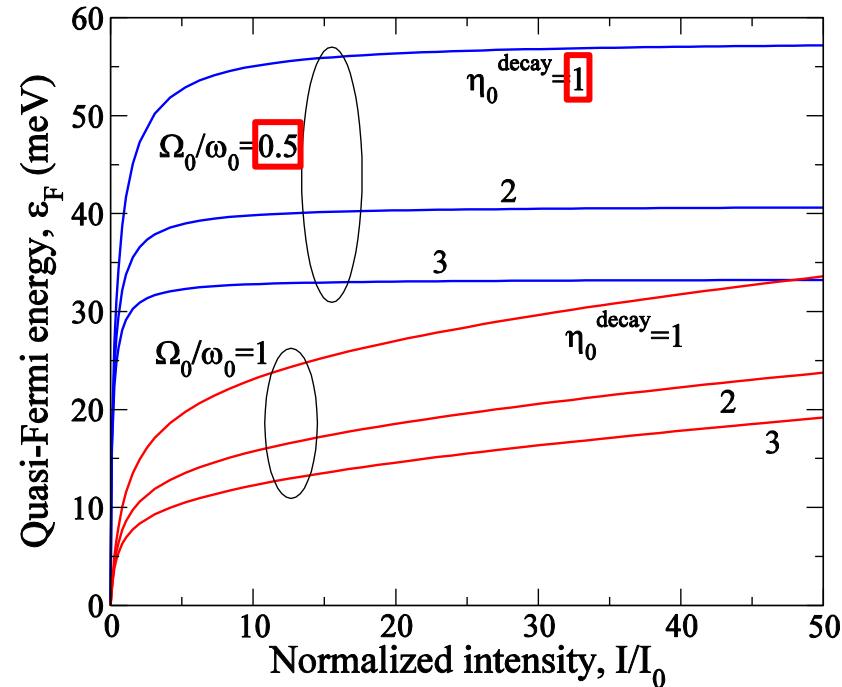
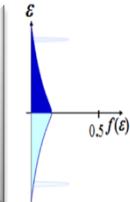
Heating of electron-hole plasma



Cooling of electron-hole plasma

Effective pumping photon energy

$$\Omega_0 = \Omega - \frac{2K\omega_0}{1 + K\tau_{op}/\tau_{cc}} \quad (K = 1, 2, \dots)$$



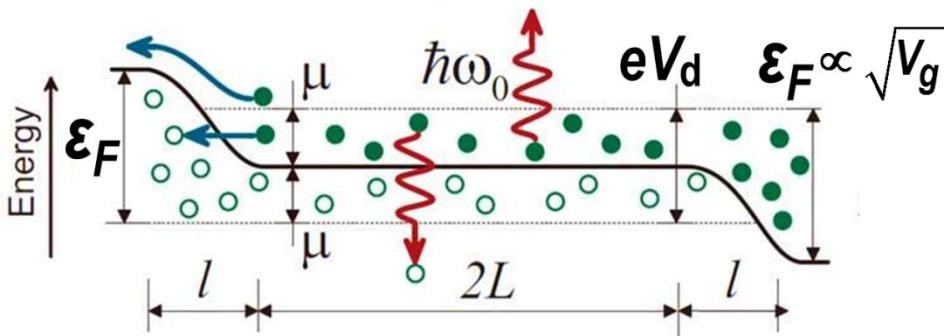
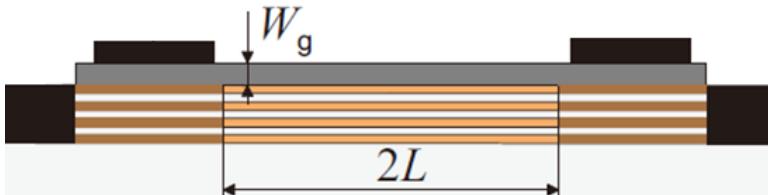
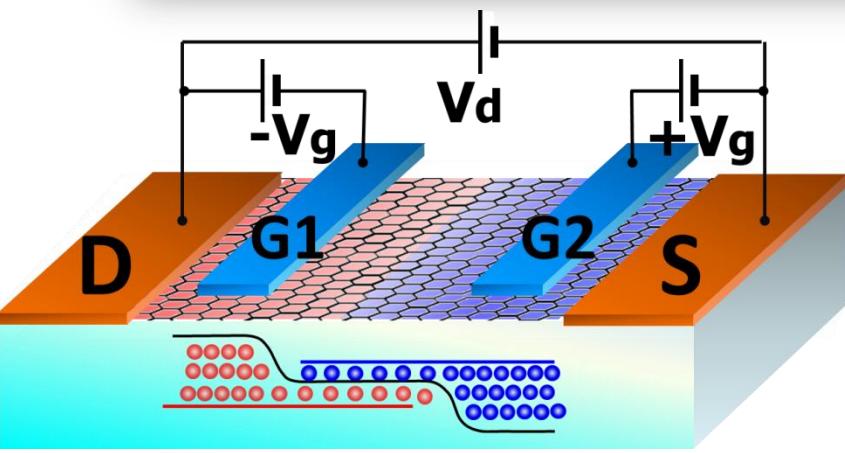
ϵ_F can be elevated by:

- cooling carriers
- lowering pumping photon energy

Toward the creation of terahertz graphene injection laser

V. Ryzhii,^{1,a)} M. Ryzhii,¹ V. Mitin,² and T. Otsuji³

M. Ryzhii and V. Ryzhii, **JJAP 46**, L151 (2007).



Parameters

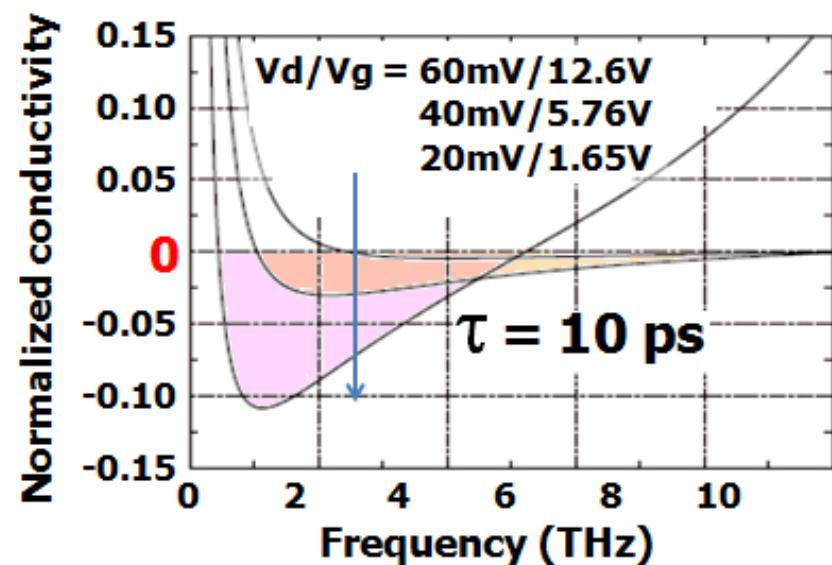
- $+V_G / -V_G \Rightarrow \eta, I_{th}$
- $V_D \Rightarrow \lambda$ at G_{max}
- $L \Rightarrow \eta > 0$

η : injection efficiency = $(I_{inject} - I_{tt})/I_{inject}$

I_{inject} : injection current

I_{tt} : tunneling & thermionic leakage current

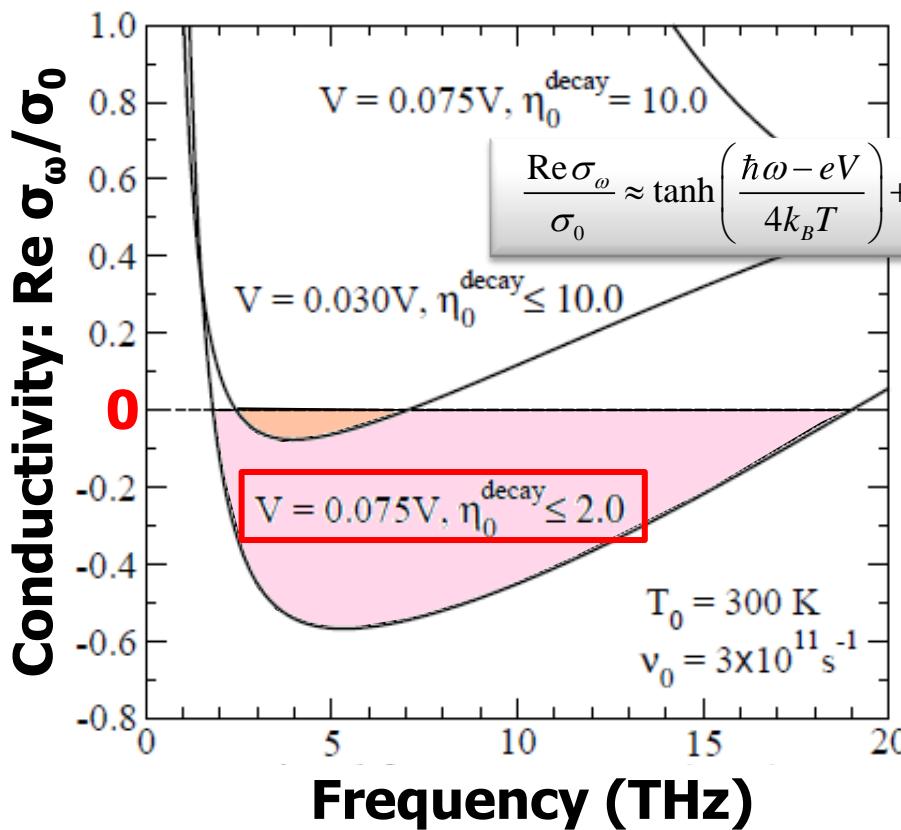
I_{th} : threshold current



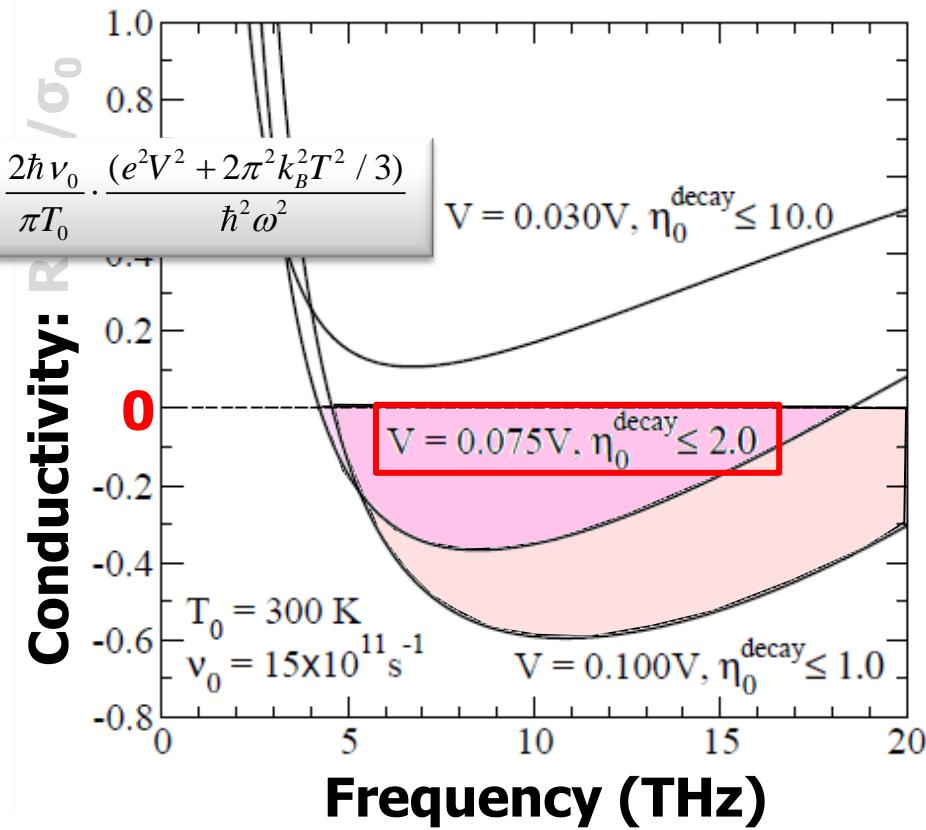
Negative Dynamic Conductivity at Different τ and η_0^{decay} at RT

V. Ryzhii, M. Ryzhii, V. Mitin, and T. Otsuji, **JAP 110**, 094503 (2011).

Excellent quality of GR
with $v_0 = 3 \times 10^{11} \text{ s}^{-1}$ ($\tau = 3.3 \text{ ps}$)



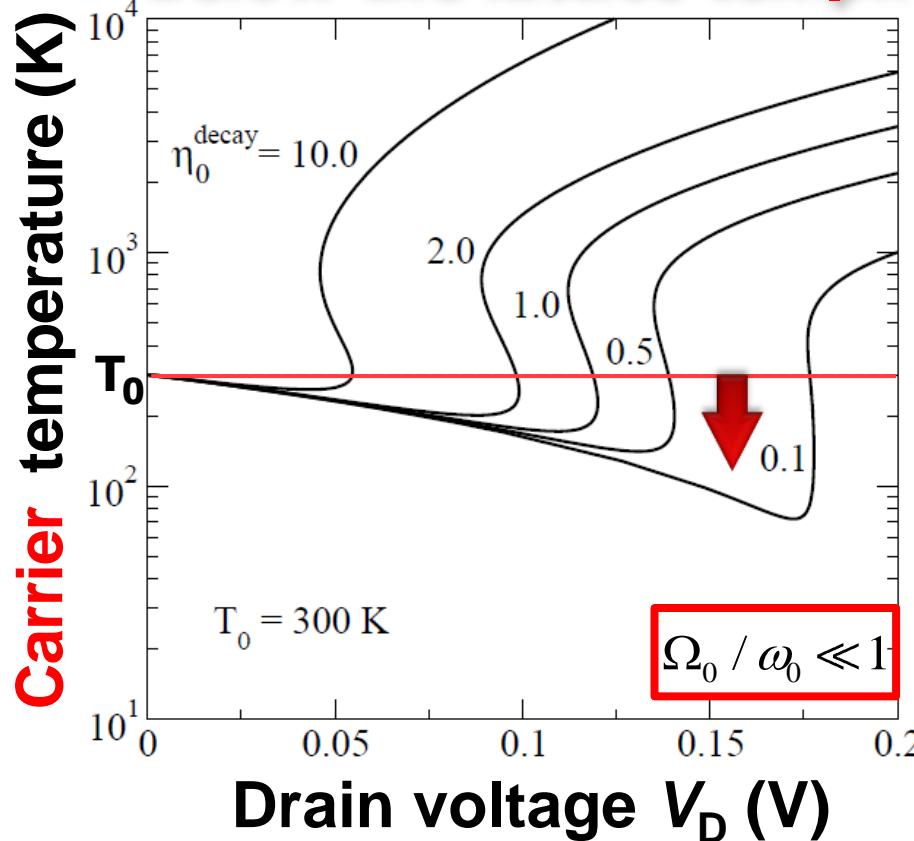
Less quality of GR
with $v_0 = 15 \times 10^{11} \text{ s}^{-1}$ ($\tau = 0.6 \text{ ps}$)



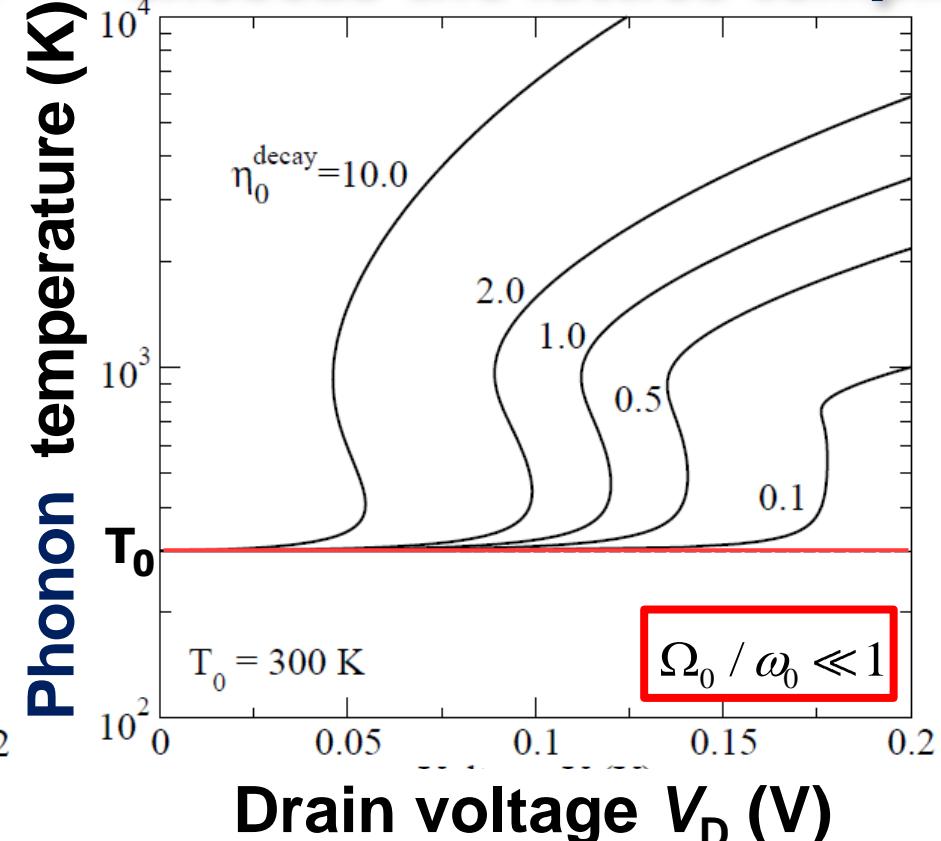
Advantage of Current Injection: Carrier Over-Cooling Effect even at RT

V. Ryzhii, M. Ryzhii, V. Mitin, and T. Otsuji, **JAP 110**, 094503 (2011).

**Carrier temp. can be
below the lattice temp.!**



**Phonon temp. always
exceeds the lattice temp.!**



2D Plasmon Dispersions in Graphene

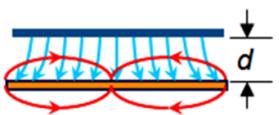
Normal semiconductors

ungated
2D



$$\omega = \sqrt{\frac{e^2 n}{2\epsilon m}} k$$

gated
2D



$$\omega = \sqrt{\frac{e^2 n d}{\epsilon m}} \cdot k \propto k d^{1/2} V_g^{1/2}$$

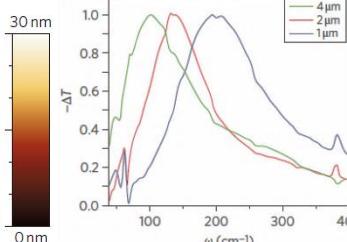
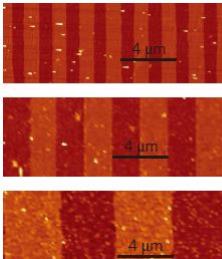
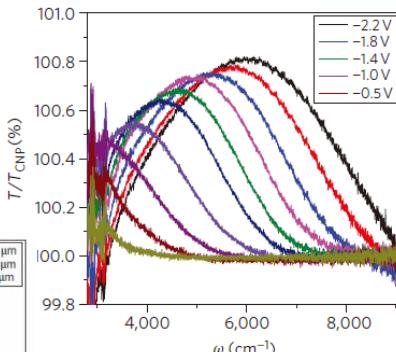
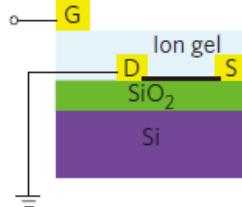
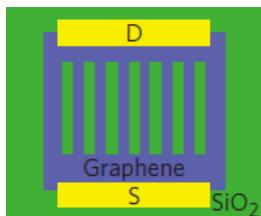
Gated graphene ribbon array

$$\tilde{\omega}_p = \omega_p - i\gamma = \sqrt{\frac{4\pi e^2 n}{\wp m^*} \frac{q \cos^2 \theta}{1 + \coth(qd)} - \frac{1}{4\tau^2} - i \frac{1}{2\tau}}$$

e : elementary charge, \wp : the permittivity

d : the period of the GRA

τ : the momentum relaxation time of electrons

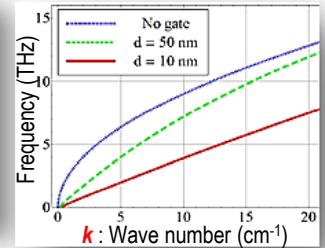


V. Ryzhii, A. Satou, T. Otsuji, **JAP** **101**, 024509 (2007).

V. Popov, T.Y. Bagaeva, T. Otsuji, V. Ryzhii, **PRB** **81**, 073404 (2010).

V. Ryzhii, M. Ryzhii, V. Mitin, A. Satou, T. Otsuji, **JJAP** **50**, 094001 (2011).

Gated graphene

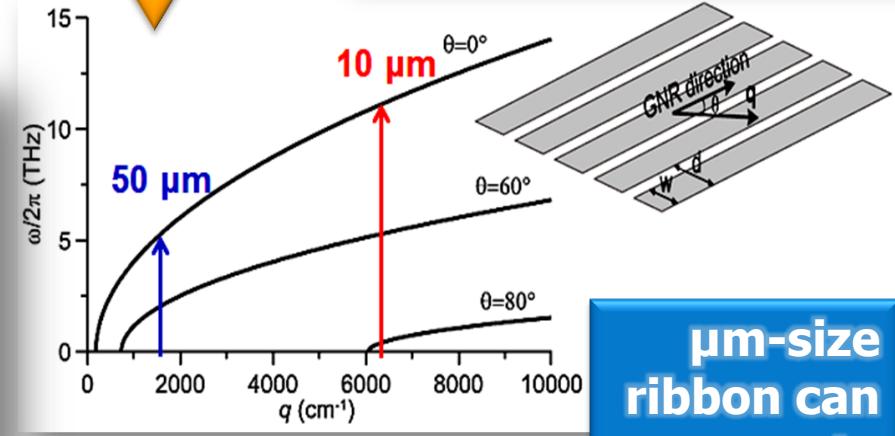
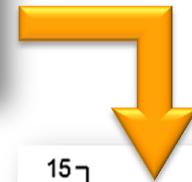


($s > v_F$, $|k|^{-1} \gg d$)

$$\omega = ks \approx k \sqrt{\frac{4 \ln 2 e^2 dk_B T}{\epsilon \hbar^2}} \propto k d^{1/2} T^{1/2}$$

$$\epsilon_F = \hbar v_F \sqrt{\epsilon V_g / 2ed} \begin{cases} \ll k_B T \\ \gg k_B T \end{cases}$$

$$\omega = ks \approx k v_F \sqrt{\frac{\alpha}{2}} \propto k v_F d^{1/4} V_g^{1/4}$$



μm-size ribbon can resonate in THz

L. Ju et al., **Nature Nanotech.** **6**, 630 (2011).

Dispersions and the Damping of Graphene Plasmons

A. D. Svintsov, V. Vyurkov, S. Yurchenko, T. Otsuji, and V. Ryzhii, **JAP 111**, 083715(2012).

Semi-classical Boltzmann's equations for Dirac Fermion

Electron: $\frac{\partial f_e}{\partial t} + v_F \frac{\mathbf{p}}{p} \frac{\partial f_e}{\partial \mathbf{r}} + e \frac{\partial \varphi}{\partial \mathbf{r}} \frac{\partial f_e}{\partial \mathbf{p}} = St\{f_e, f_e\} + St\{f_e, f_h\} + St_i\{f_e\}$

Hole: $\frac{\partial f_h}{\partial t} + v_F \frac{\mathbf{p}}{p} \frac{\partial f_h}{\partial \mathbf{r}} - e \frac{\partial \varphi}{\partial \mathbf{r}} \frac{\partial f_h}{\partial \mathbf{p}} = St\{f_h, f_h\} + St\{f_h, f_e\} + St_i\{f_h\}$

Continuity equations:

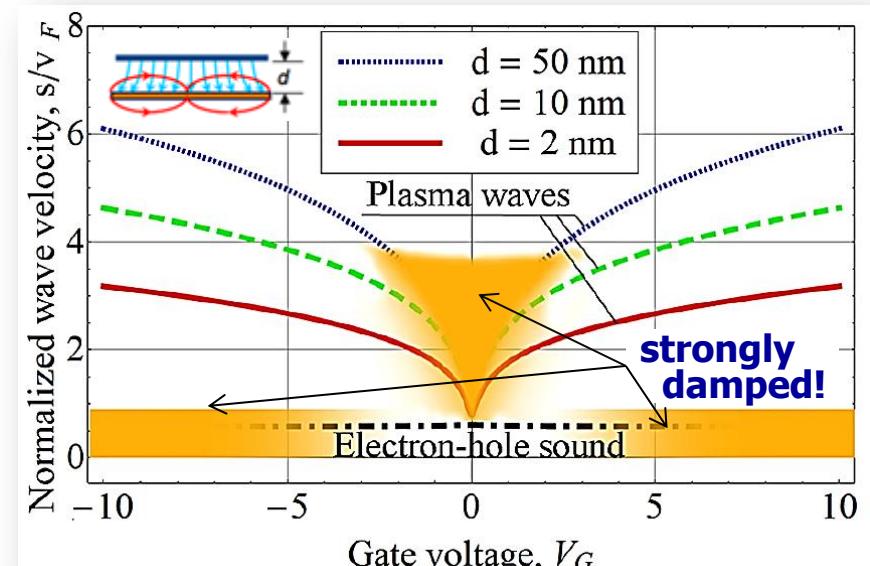
Collision integrals:
inter-carrier, disorder

$$\frac{\partial \Sigma_e}{\partial t} + \frac{\partial \Sigma_e \mathbf{V}_e}{\partial \mathbf{r}} = 0, \quad \frac{\partial \Sigma_h}{\partial t} + \frac{\partial \Sigma_h \mathbf{V}_h}{\partial \mathbf{r}} = 0.$$

Hydrodynamic Euler equations:

Electron: $\frac{3}{2} \frac{\partial}{\partial t} \frac{\langle p_e \rangle \mathbf{V}_e}{v_F} + \frac{\partial}{\partial \mathbf{r}} \frac{v_F \langle p_e \rangle}{2} - e \Sigma_e \frac{\partial \varphi}{\partial \mathbf{r}} = -\beta_e \mathbf{V}_e - \beta_{eh} (\mathbf{V}_e - \mathbf{V}_h),$

Hole: $\frac{3}{2} \frac{\partial}{\partial t} \frac{\langle p_h \rangle \mathbf{V}_e}{v_F} + \frac{\partial}{\partial \mathbf{r}} \frac{v_F \langle p_h \rangle}{2} + e \Sigma_h \frac{\partial \varphi}{\partial \mathbf{r}} = -\beta_h \mathbf{V}_h - \beta_{eh} (\mathbf{V}_h - \mathbf{V}_e).$



Bipolar modes for intrinsic and photoexcited GR

e-h plasma waves: $\omega_+ = -i \left(\frac{\nu}{2} + \frac{\nu_{eh}}{3} \right) + \sqrt{k^2 v^2 (1+2r)^2 - \left(\frac{\nu}{2} + \frac{\nu_{eh}}{3} \right)^2}$

e-h sound waves: $\omega_- = -i \frac{\nu}{2} + \sqrt{k^2 v^2 - \left(\frac{\nu}{2} \right)^2}$

strongly damped!

Unipolar modes for doped / gate-biased GR

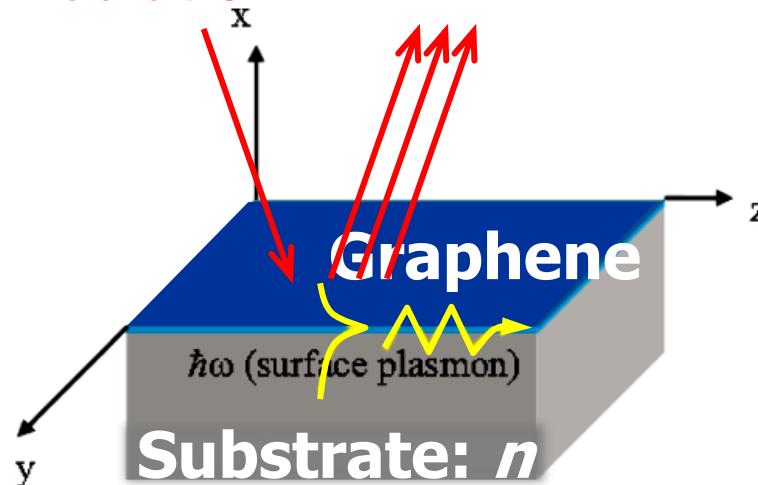
Majority carriers: $\omega_+ = -i \frac{\nu_e}{2} + \sqrt{k^2 v_e^2 (1+r_e)^2 - \left(\frac{\nu_e}{2} \right)^2},$

Minority carriers: $\omega_- = -i \left(\frac{\nu_h}{2} + \frac{\nu_{eh}}{3} \right) + \sqrt{k^2 v_h^2 (1+2r_h)^2 - \left(\frac{\nu_h}{2} + \frac{\nu_{eh}}{3} \right)^2}.$

Amplification of THz-SPPs along Population-Inverted-Graphene Waveguide

A. Dubinov, Y. Aleshkin, V. Mitin, T. Otsuji, V. Ryzhii, *JPCM* **23**, 145302 (2011).
F. Rana, *IEEE T. NanoTechnol.* **7**, 91 (2008).

THz photon
radiation

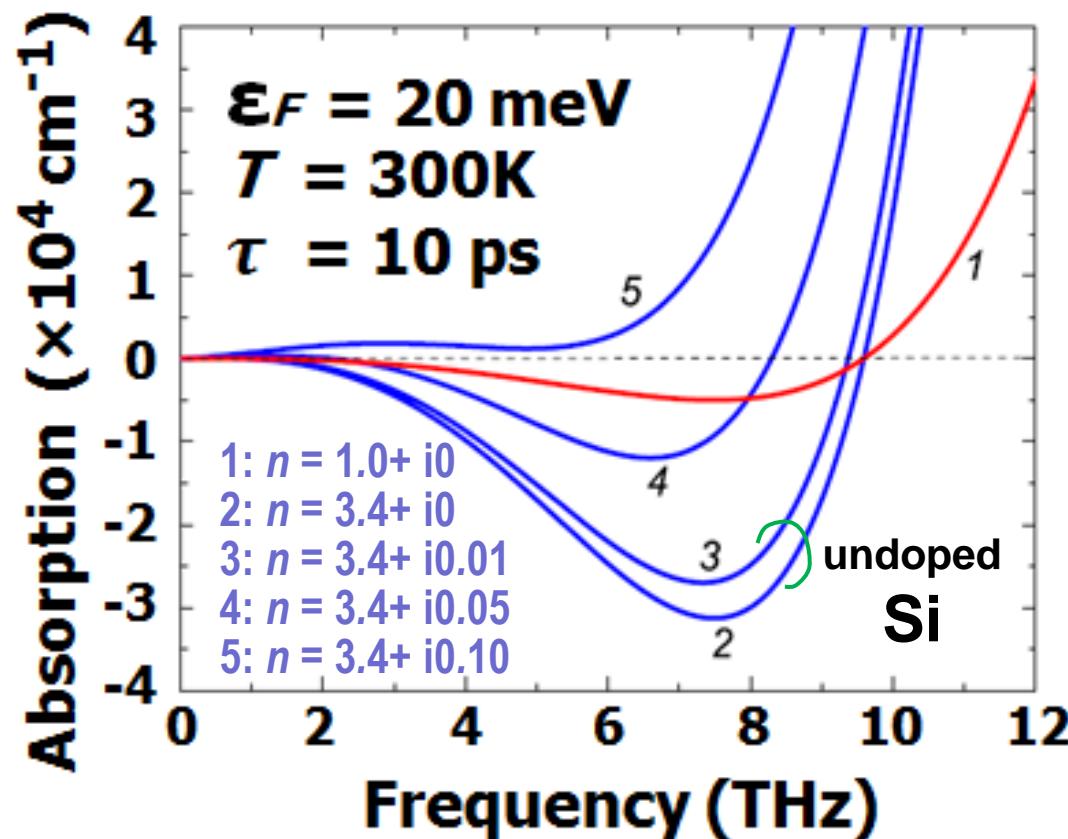


Complex propagation constant
of the SPPs:

$$\rho = \sqrt{1 - \frac{c^2}{4\pi^2\sigma_\omega^2}}$$

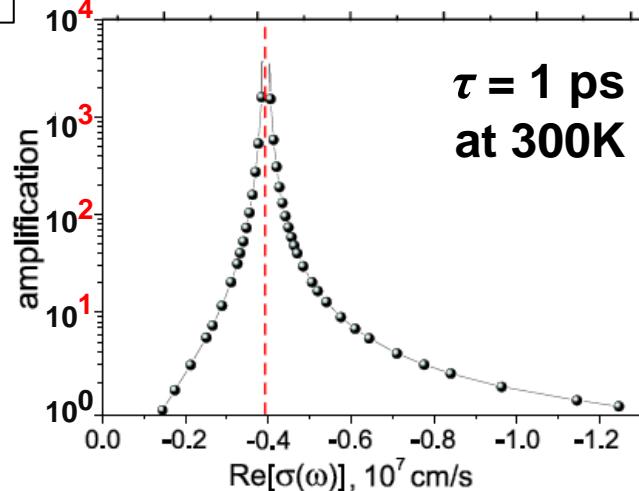
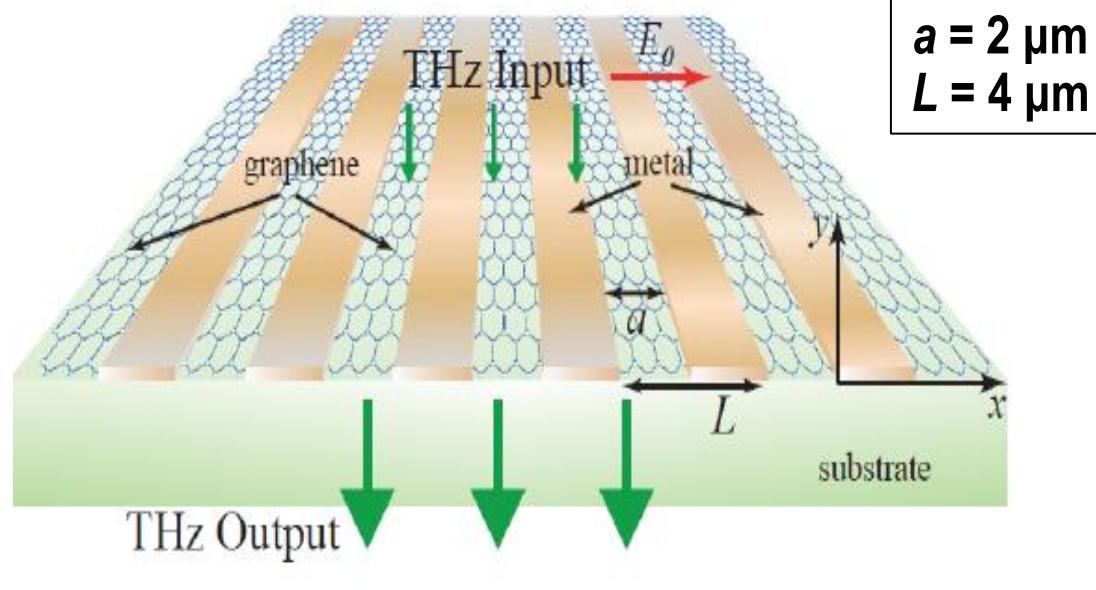
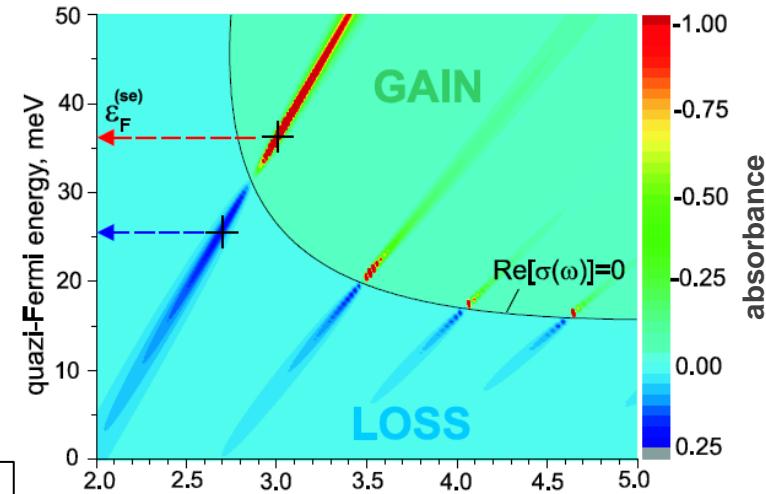
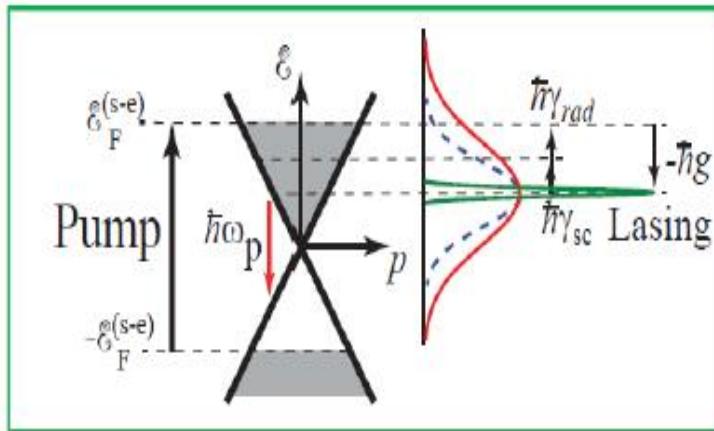
Absorption coefficient of SPPs:

$$\alpha = \text{Im}(q_z) = 2\text{Im}(\rho\omega/c)$$

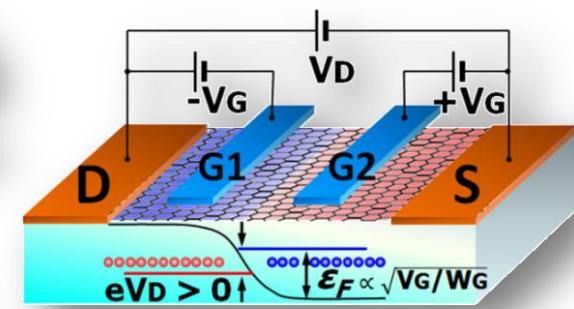
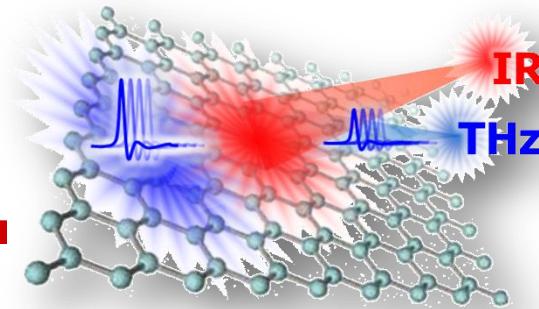


Plasmonic Terahertz Lasing from Nano-Patterned Graphene-Metal Array

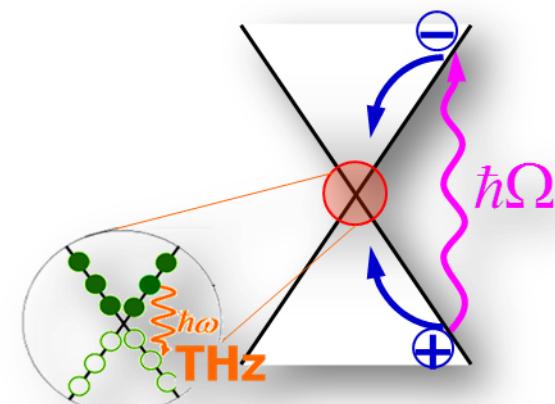
V.V. Popov, O.V. Polischuk, A.R. Davoyan, V. Ryzhii, T. Otsuji, M.S. Shur, *PRB* **86**, 195437 (2012).



Summary



1. Optically/Electrically pumped GR-THz/IR lasers.
2. Carrier heating can be suppressed by reducing the pumping photon energy.
3. Current injection is the best-suited, providing a carrier over-cooling effect.
4. Dual-gate GR-FET is a possible GR-injection THz/IR laser structure.
5. Active plasmonic structures can greatly boost the THz/IR gain.
6. Advantages of graphene THz/IR injection lasers:
 - no need extra care for carrier depopulation.
 - low end of THz range at room temperatures.
 - integrated with simple epitaxy techniques.



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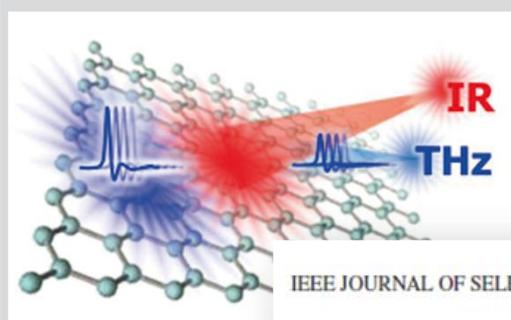
Volume 45 Number 30 1 August 2012

Fast Track Communication

Double graphene-layer plasma resonances terahertz detector
V Ryzhii, T Otsuji, M Ryzhii and M S Shur

Topical review

Graphene-based devices in terahertz science and technology
T Otsuji, S A Boubanga Tombet, A Satou, H Fukidome, M Suemitsu,
E Sano, V Popov, M Ryzhii and V Ryzhii



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Terahertz-Wave Generation Using Graphene: Toward New Types of Terahertz Lasers

Taiichi Otsuji, Member, IEEE, Stephane Boubanga Tombet, Akira Satou, Member, IEEE,
Maxim Ryzhii, Senior Member, IEEE, and Victor Ryzhii, Fellow, IEEE

(Invited Paper)

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Dr. Hiroyuki
Handa



Hiromi
Karasawa



Takayuki
Watanabe



Tetsuya
Fukushima

**Thank you very much
for your attention!**