Quantum plasmonics with atomically-thin materials

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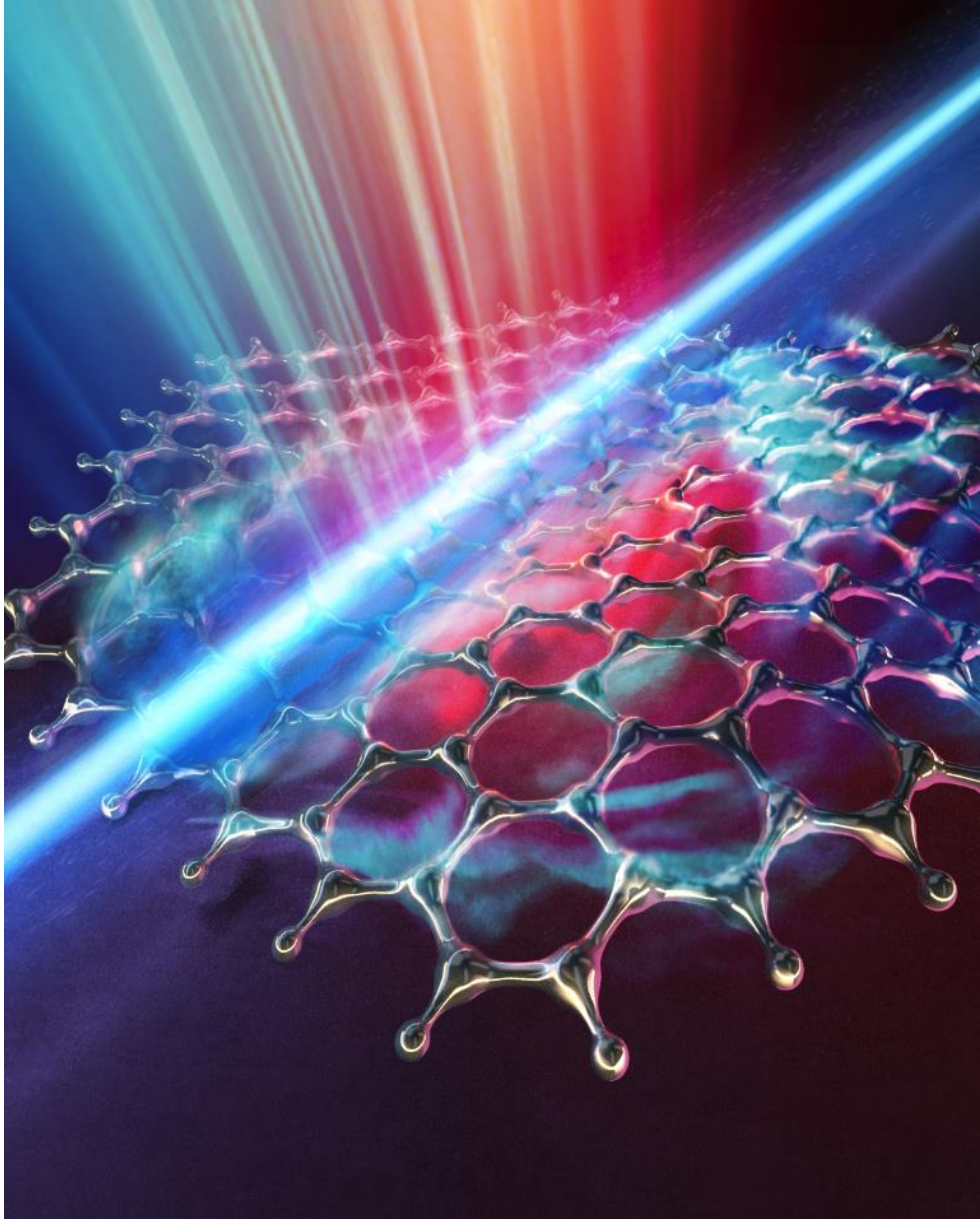
2nd Danish Quantum Community Conference

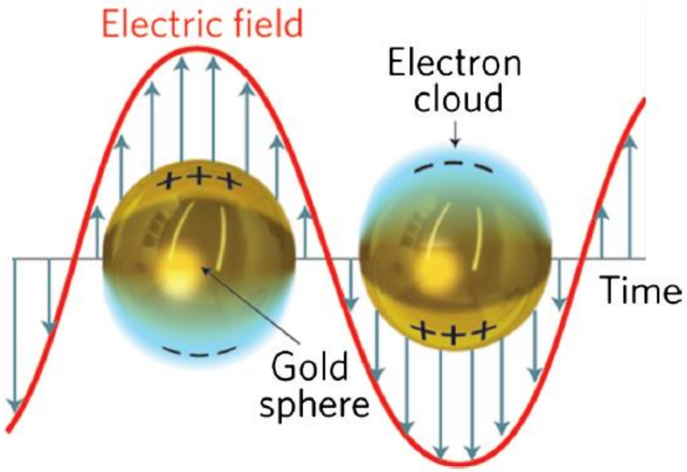
October 7, 2020

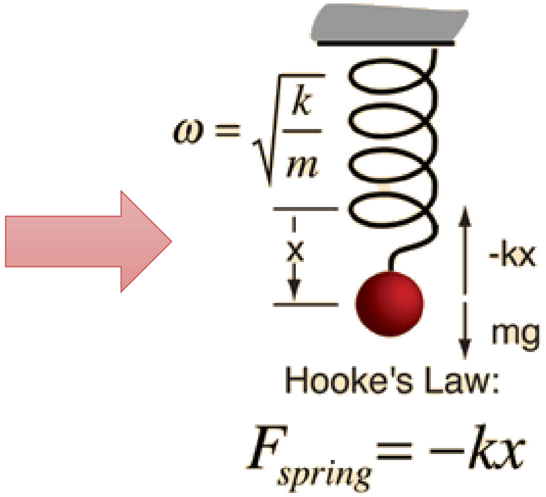


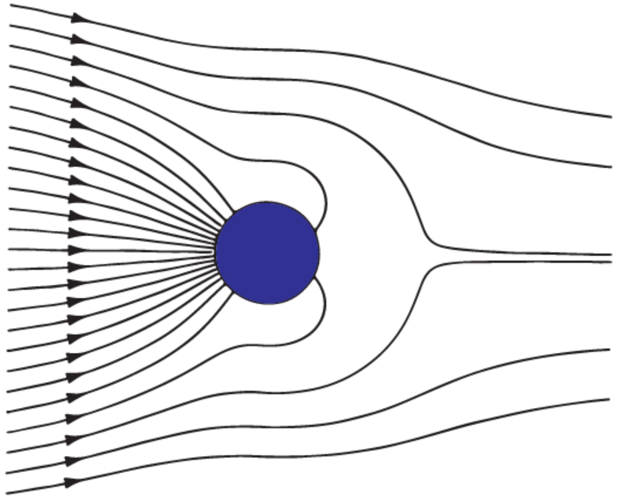
Plasmonics

SDU

Plasmons—collective oscillations of conduction electrons in metals

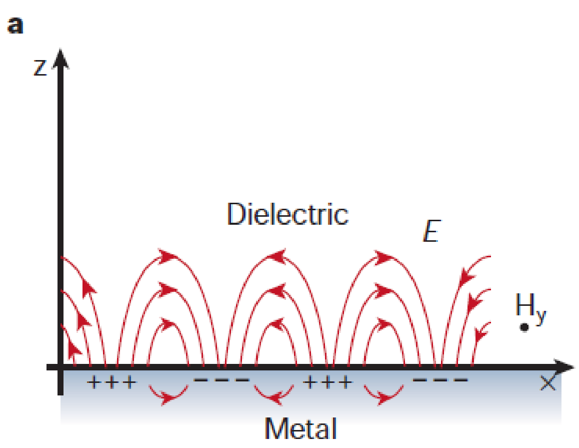


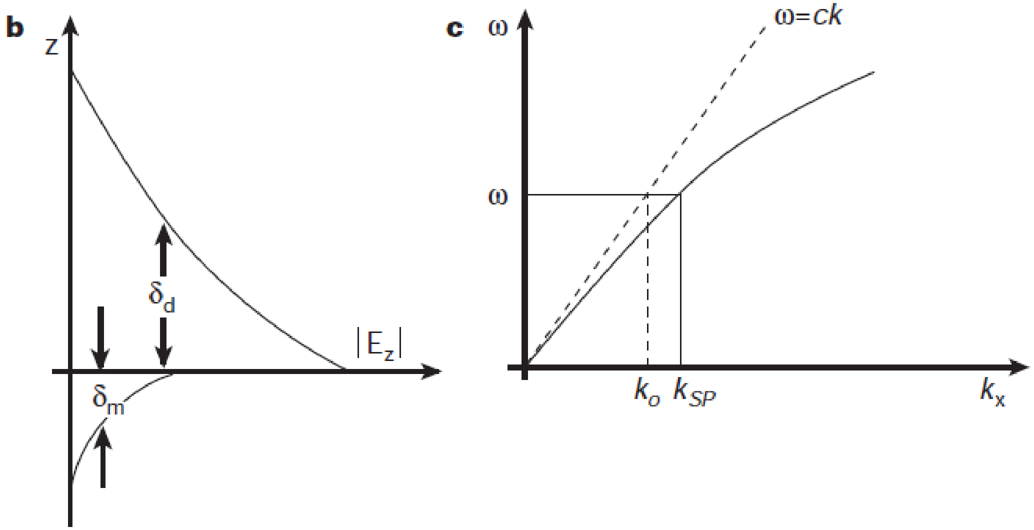


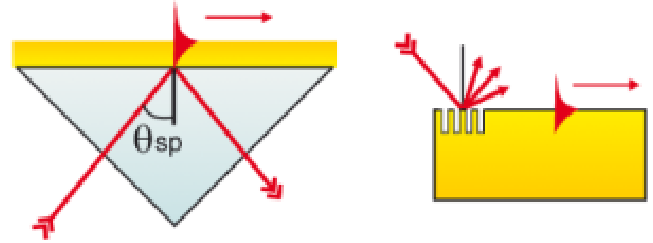


Liang et al., Plasmonics (2014)

Brongersma et al., Nat. Nanotechnol. (2015)





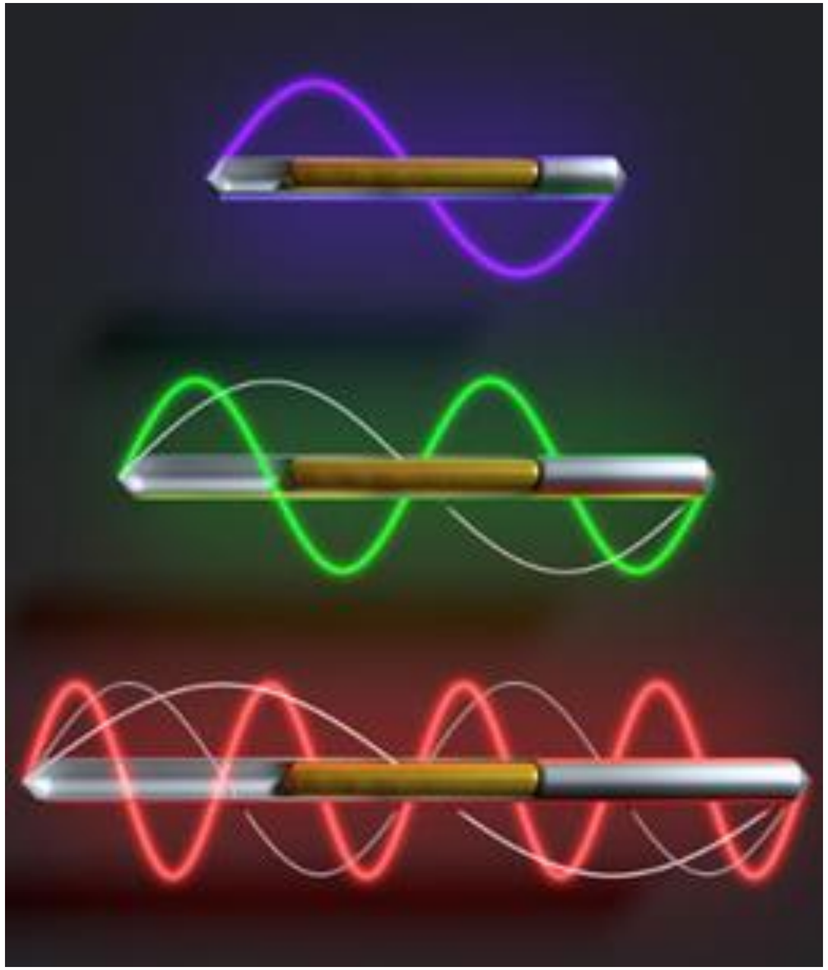


Barnes et al., Nature (2003)

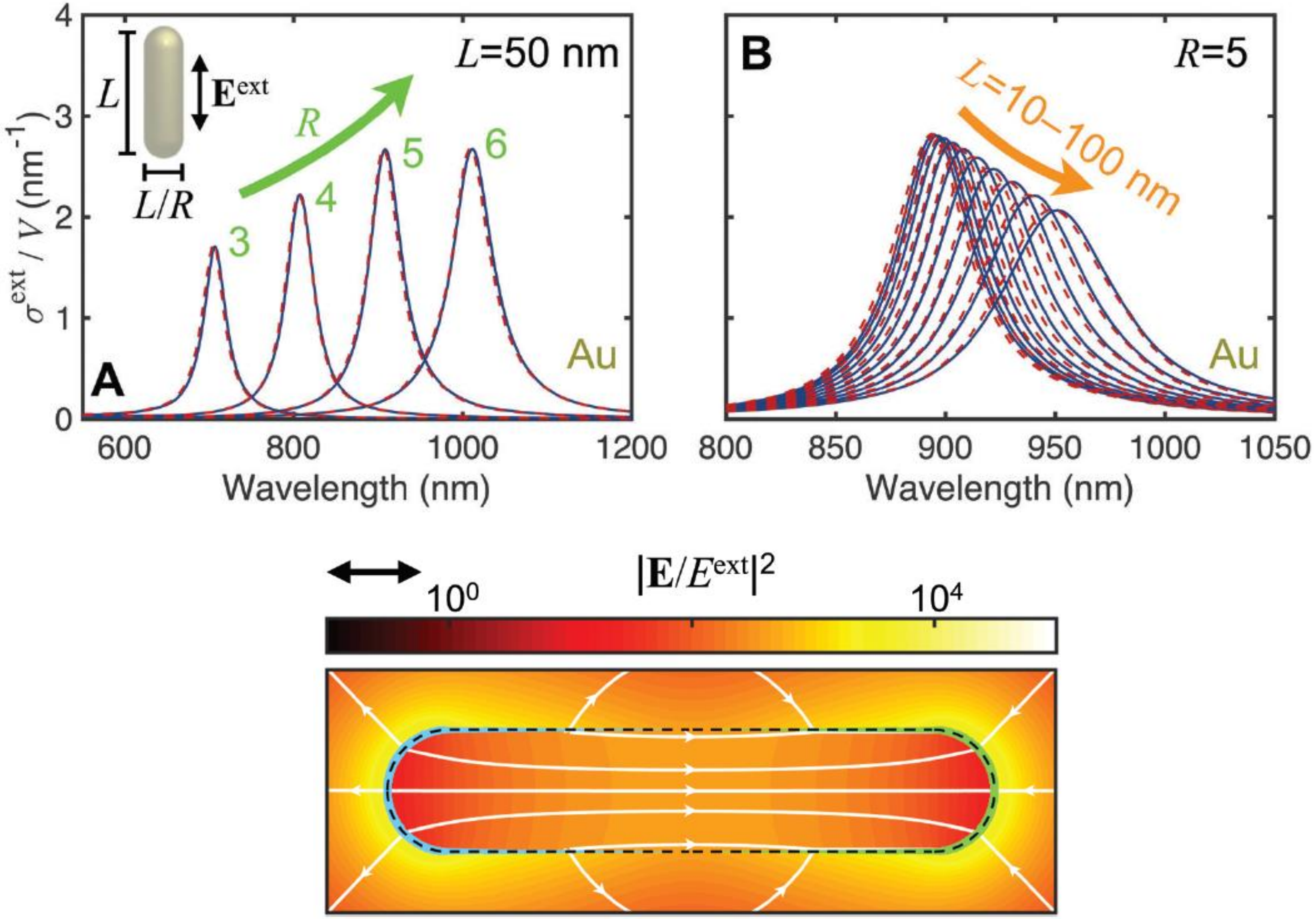
Plasmonics

SDU

Plasmons—collective oscillations of conduction electrons in metals

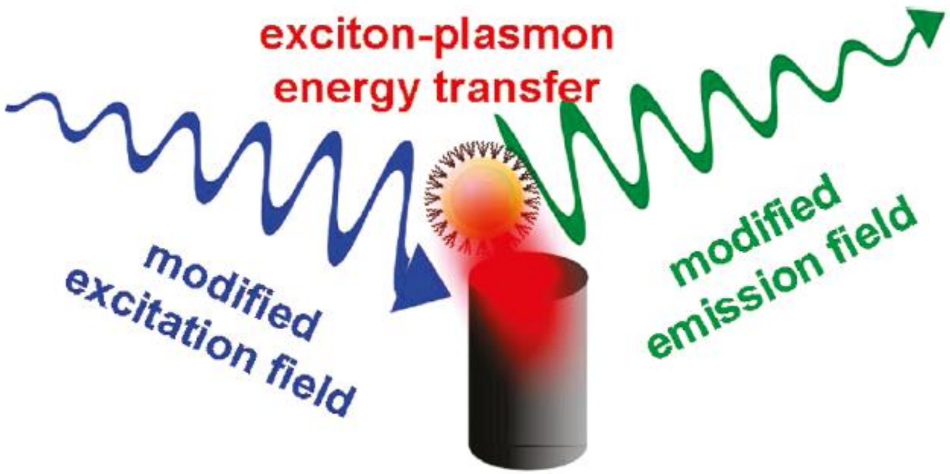


Mayer et al., Nano Lett. (2015)

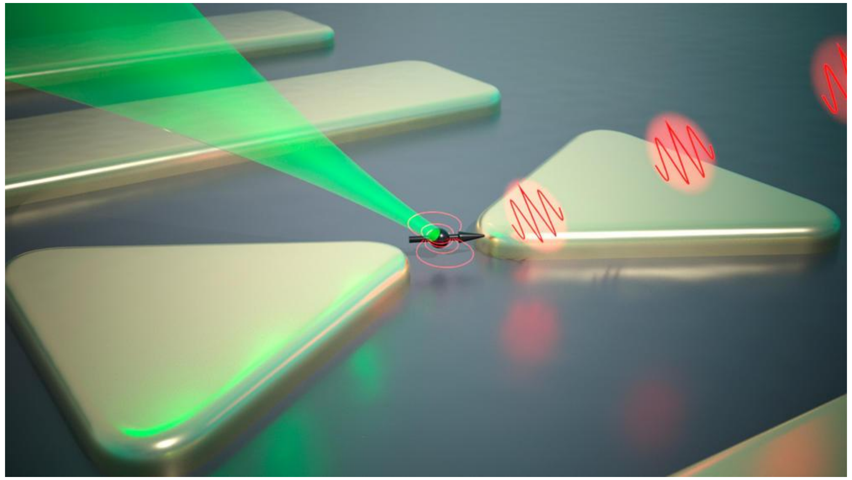


R. Yu et al., Chem. Soc. Rev. (2017)

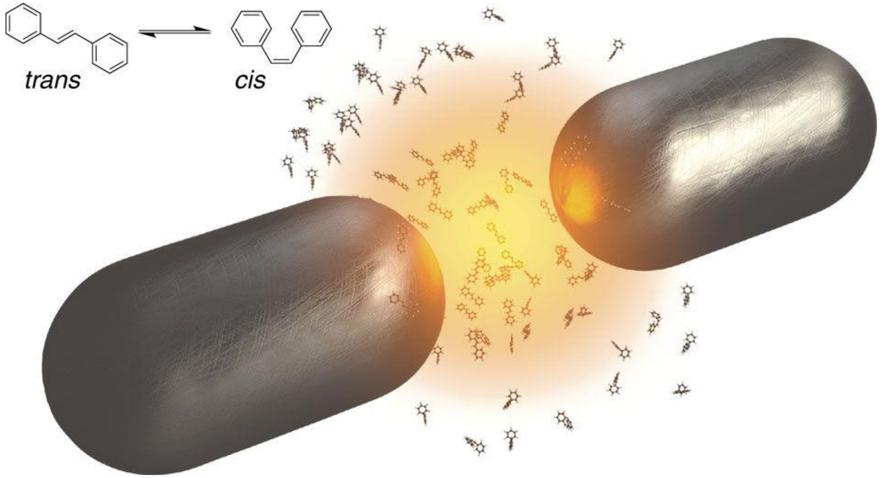
Atom-plasmon interactions



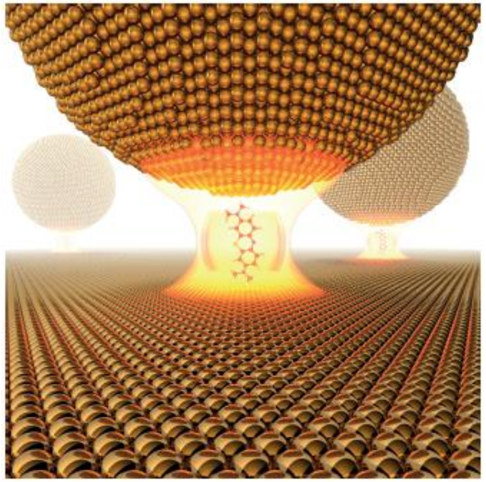
M. Achermann., Phys. Chem. Lett. (2010)



F. Koenderink, ACS Photon. (2017)



J. Galego et al., Nat. Commun. (2016)



R. Chikkaraddy et al., Nature (2016)

Plasmonics

- *Metal losses strongly affect performance*
- *Difficult to "actively tune" plasmons*

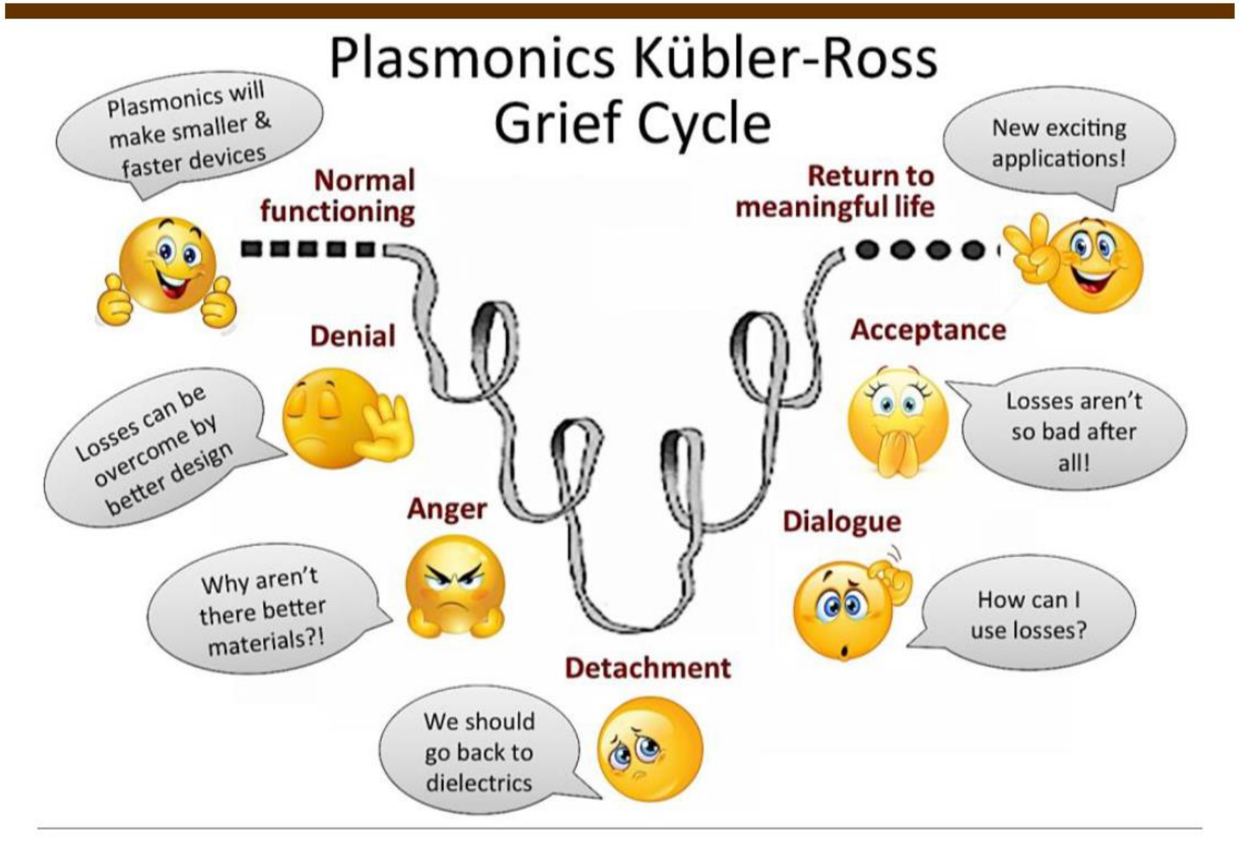
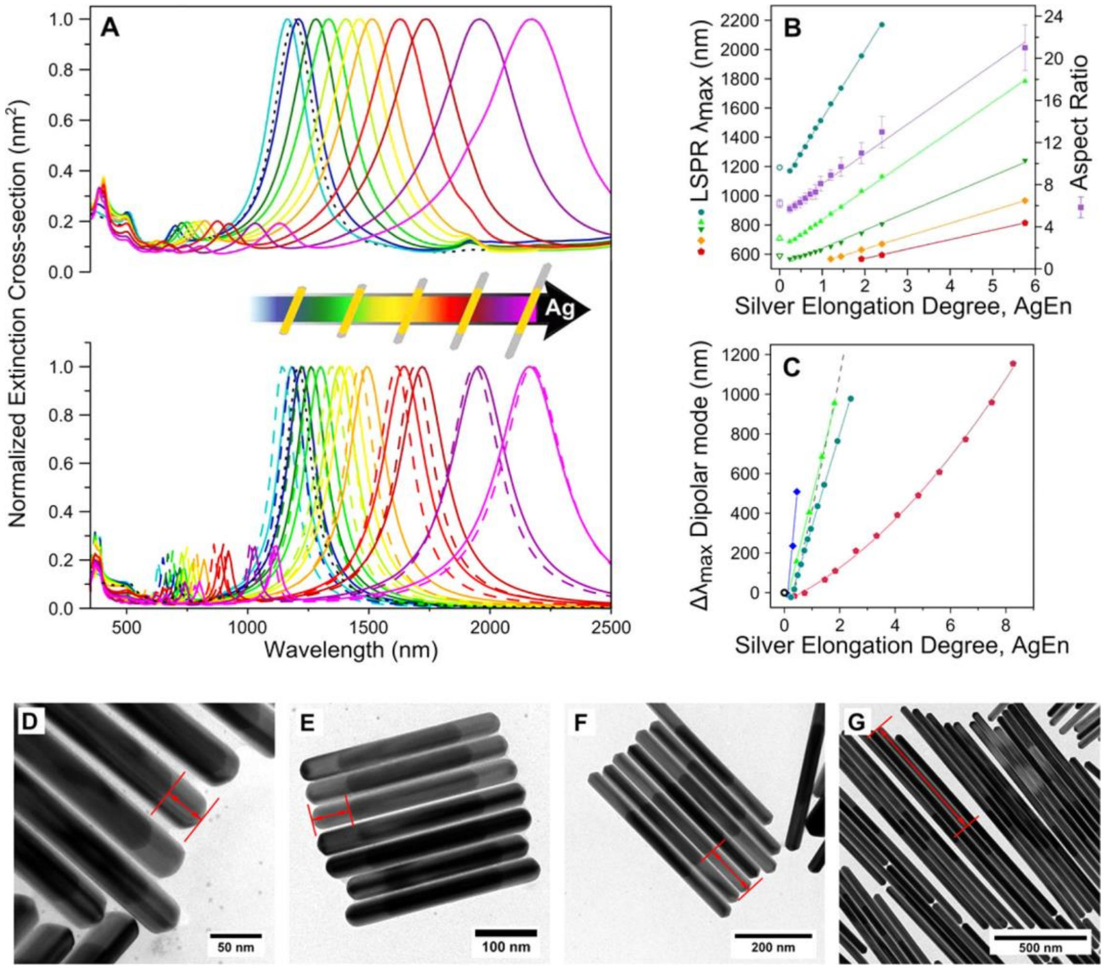


Fig. 1. Plasmonics Kubler-Ross grief cycle: evolution of the attitude within the research community towards the role of dissipative and radiative losses in the success of the plasmonics research enterprise.

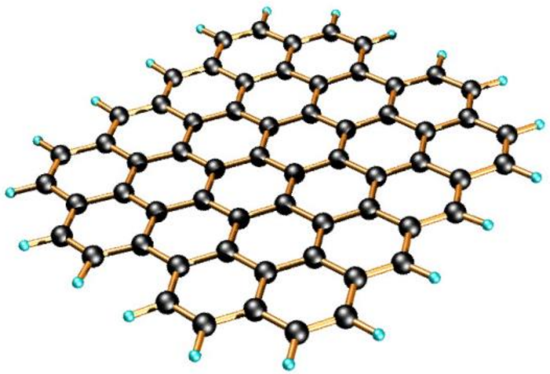
Boriskina, Adv. Opt. Photon. (2017); J. Khurgin Nat. Nanotechnol. (2015)

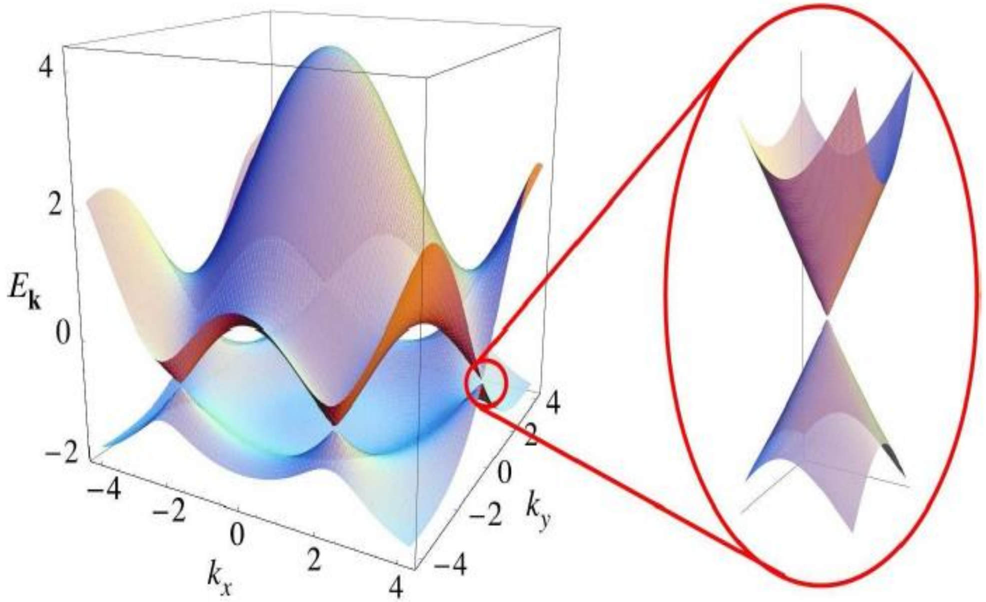


Mayer et al., Nano Lett. (2015)

Graphene

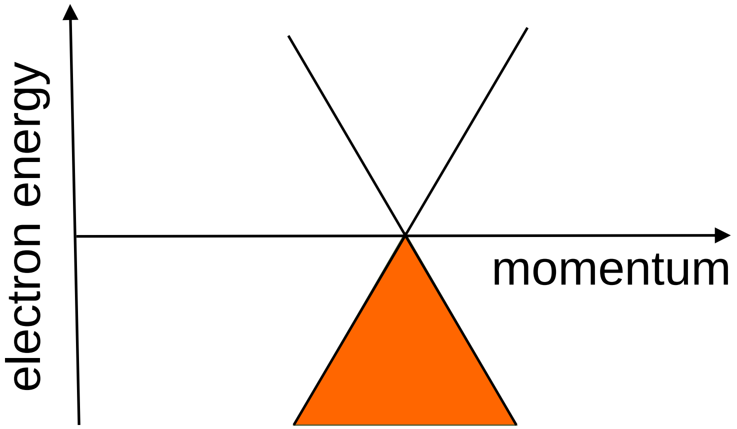
- Atomically-thin carbon layer
- Linear e$^-$ dispersion relation
- High charge-carrier mobility





A. H. Castro Neto et al., *Rev. Mod. Phys.* 81, 109 (2009).

Graphene layer



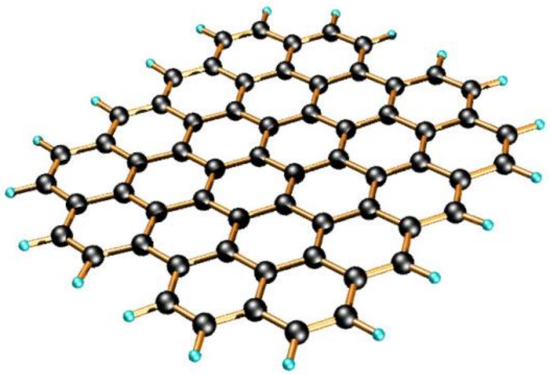
$$E_\mathbf{k} = \hbar v_F |\mathbf{k}|$$

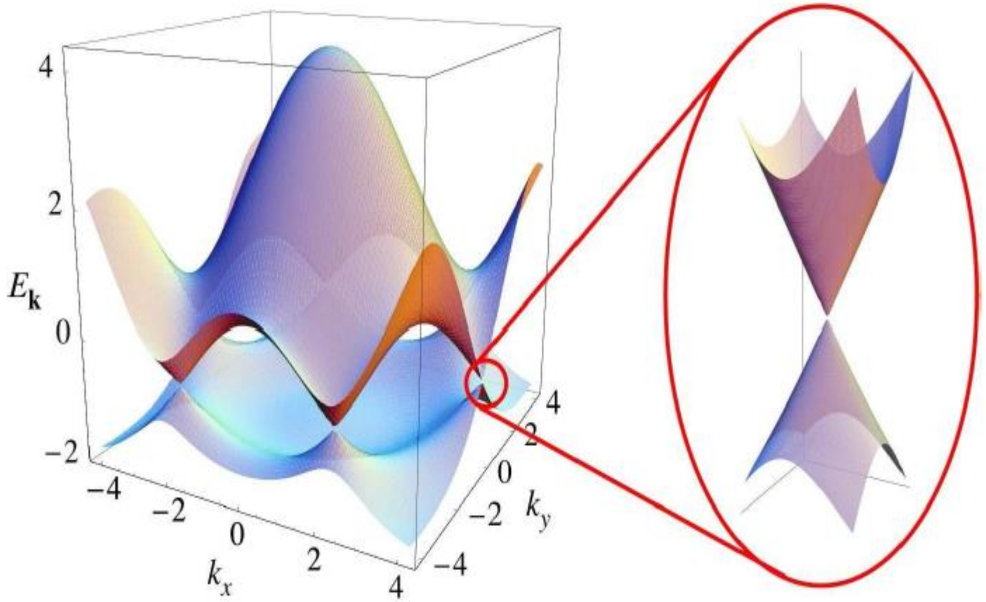
$$v_F \approx c/300$$

$$\sigma \approx \frac{e^2}{4\hbar}$$

Graphene

- Atomically-thin carbon layer
- Linear e⁻ dispersion relation
- High charge-carrier mobility





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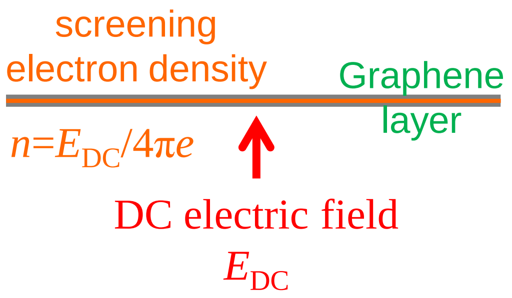
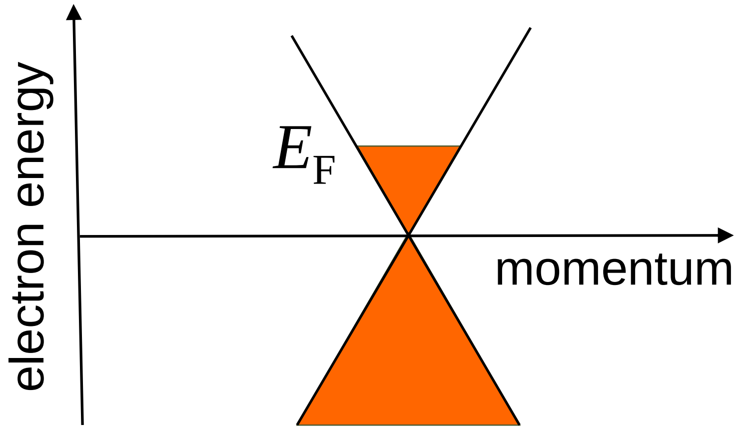
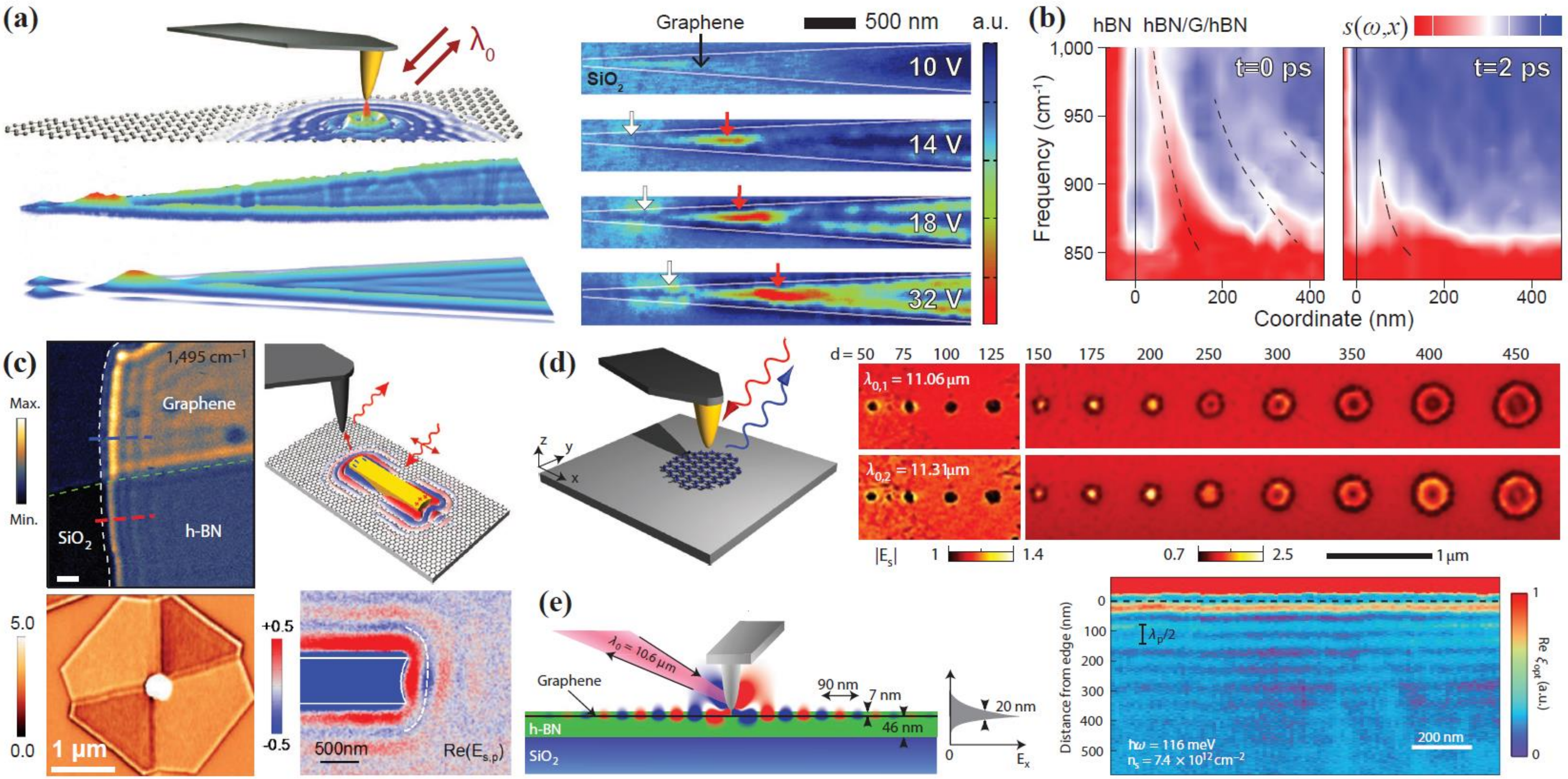




$$\sigma \approx \frac{e^2 E_F}{\hbar^2 \pi}\frac{i}{\omega + i\tau^{-1}}$$

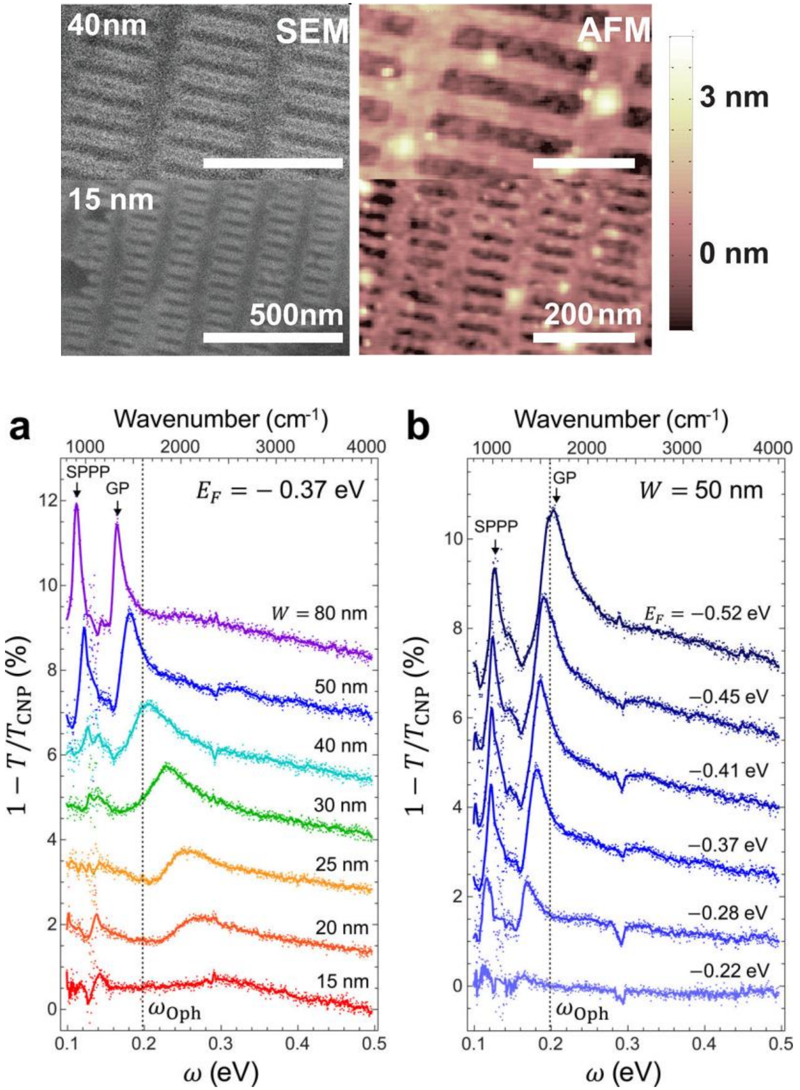
$$\tau = \mu E_F / e v_F^2$$

Graphene plasmonics

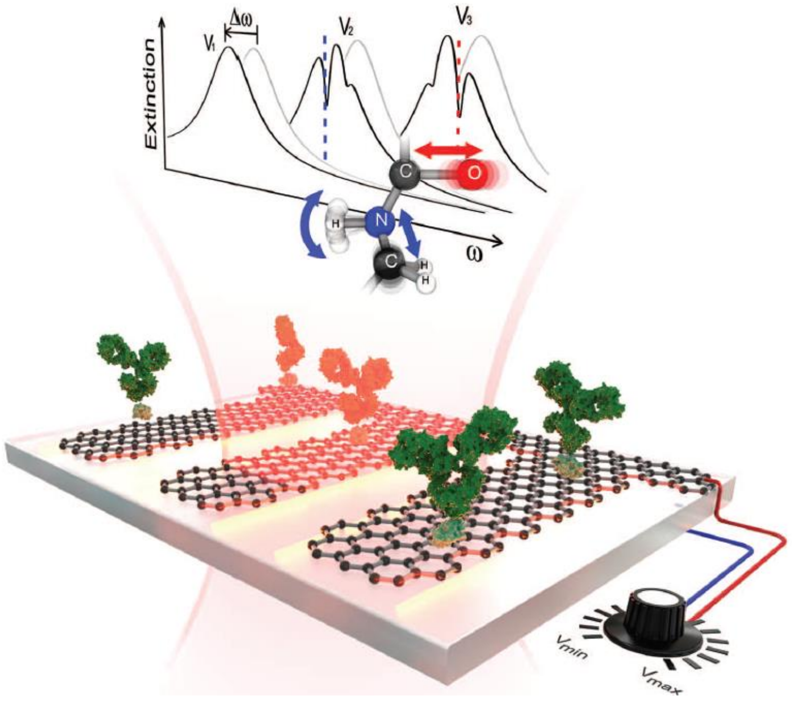


Basov, Koppens, Hillenbrand, et al.

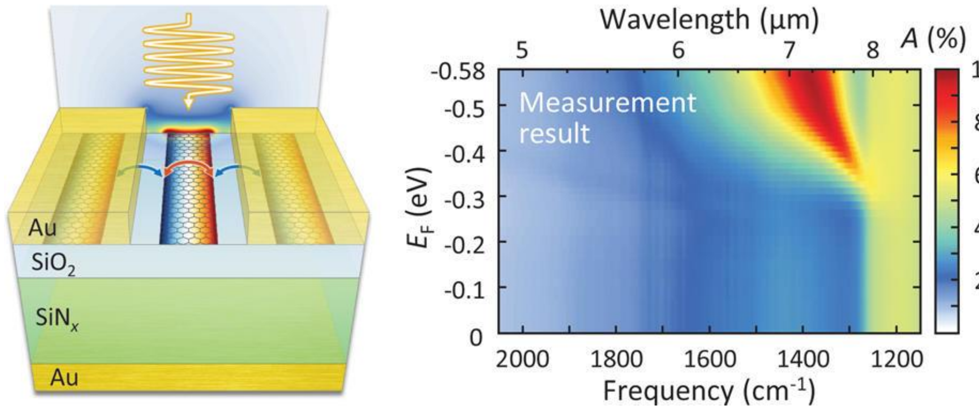
Localized graphene plasmons



V. Brar *et al*., Nano Lett. (2012)

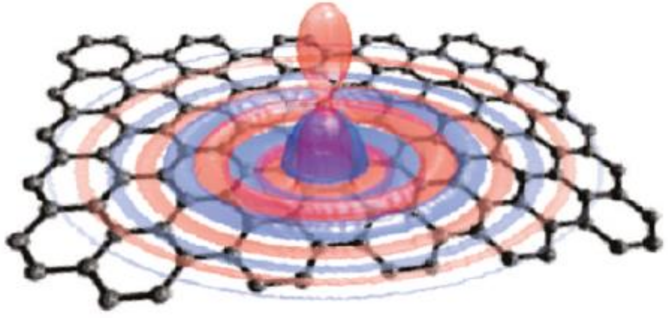


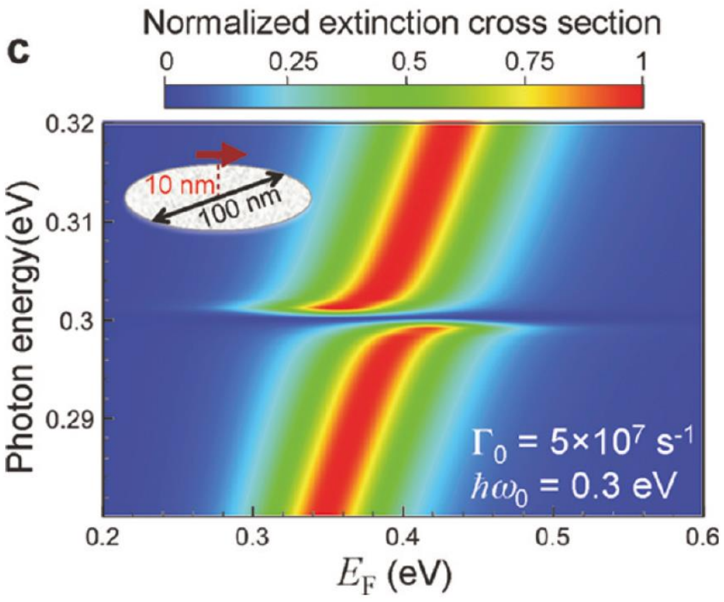
D. Rodrigo *et al*., Science (2015)

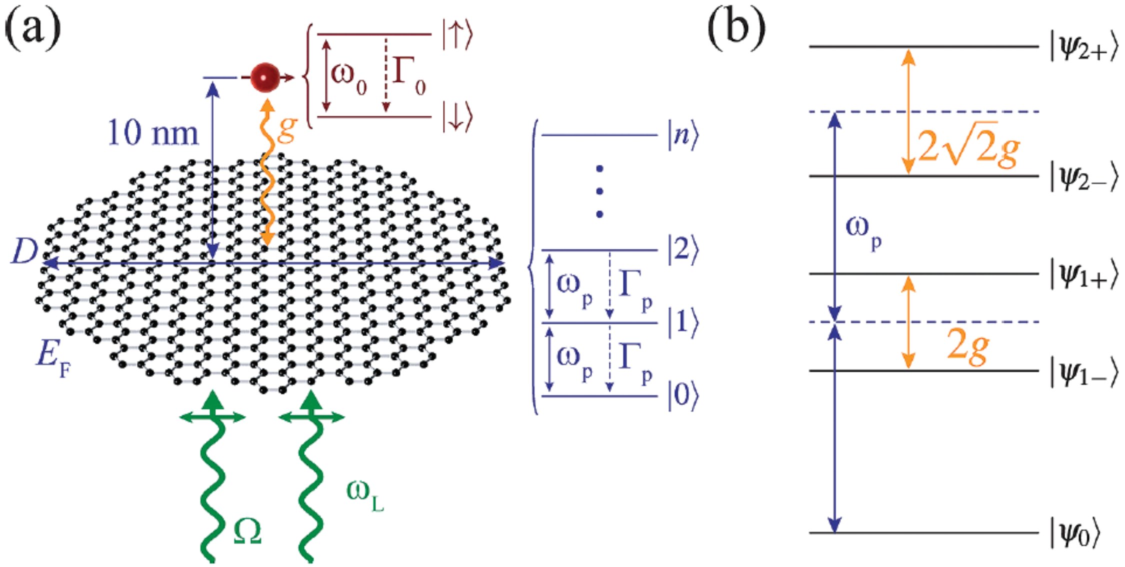


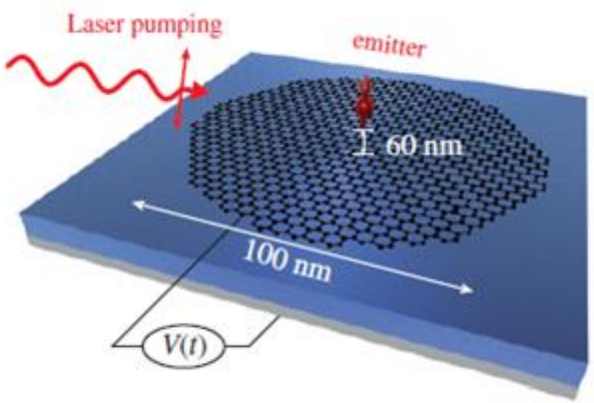
S. Kim *et al*., Nano Lett. (2018)

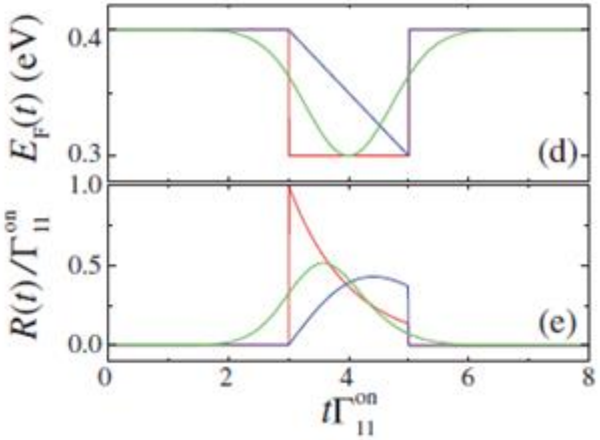
Quantum optics with graphene plasmons









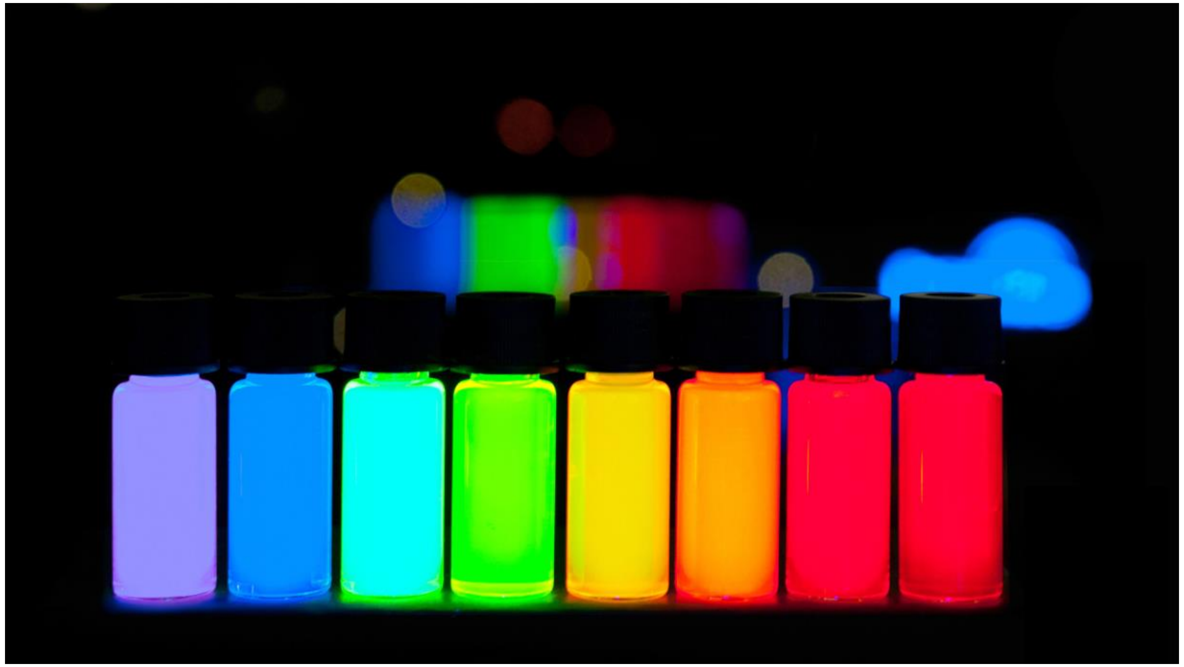


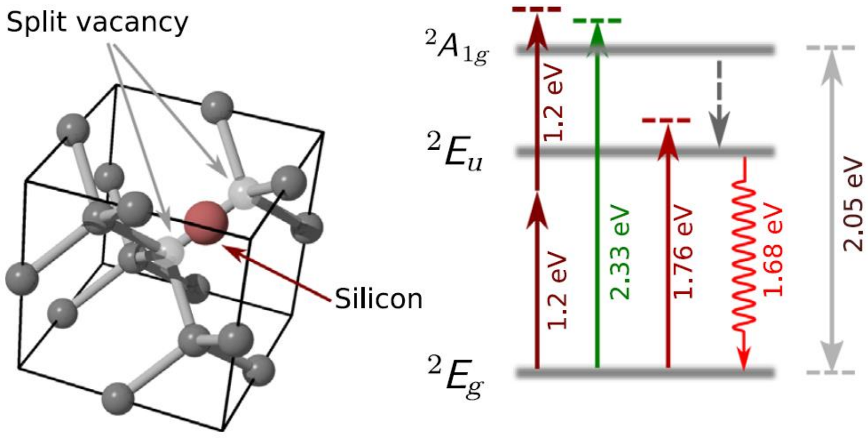
Koppens et al., Nano Lett. (2012)

Manjavacas et al., ACS Nano and New J. Phys. (2012)

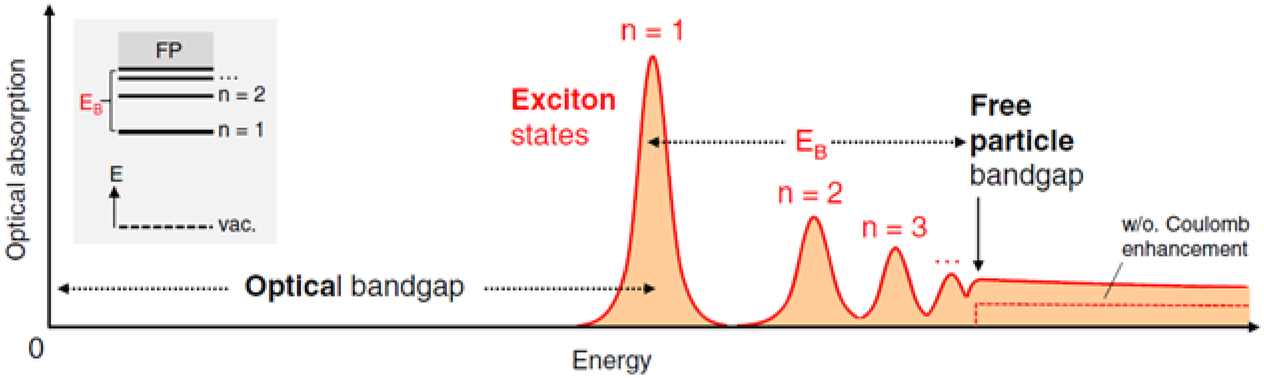
Quantum light emitters

SDU

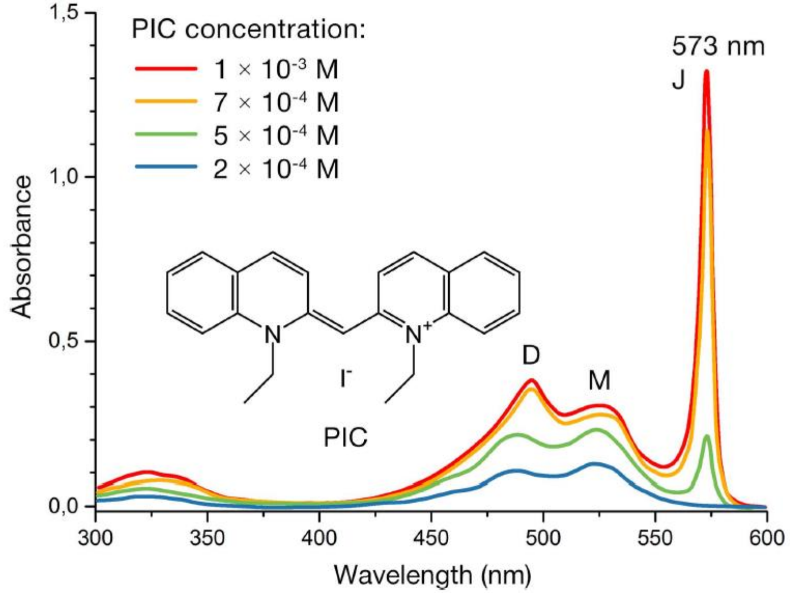




Higbie et al., Phys. Rev. Appl. (2017)

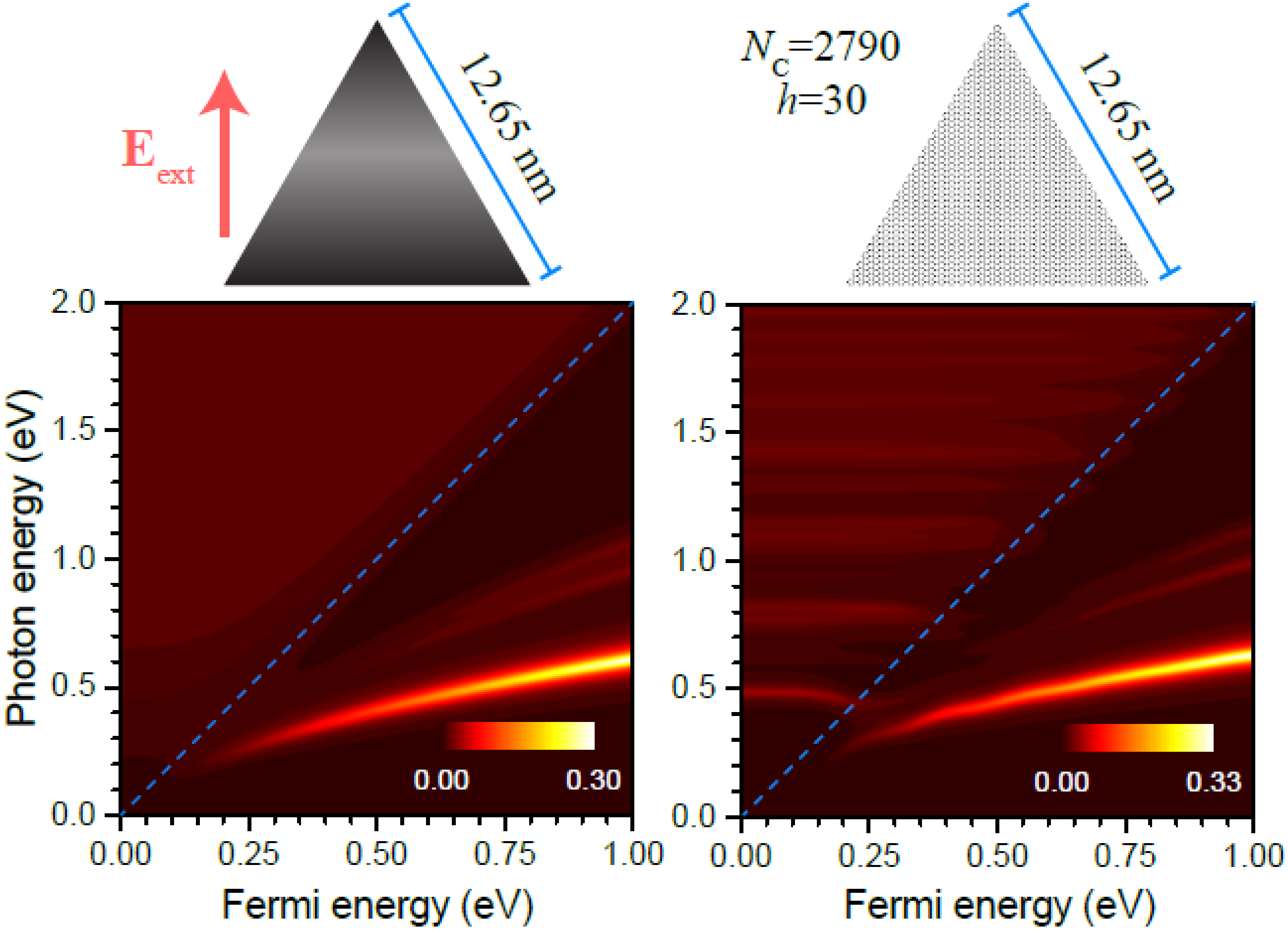


G. Wang et al., Rev. Mod. Phys. (2018)



Bricks et al., Methods Appl. Fluroesc. (2018)

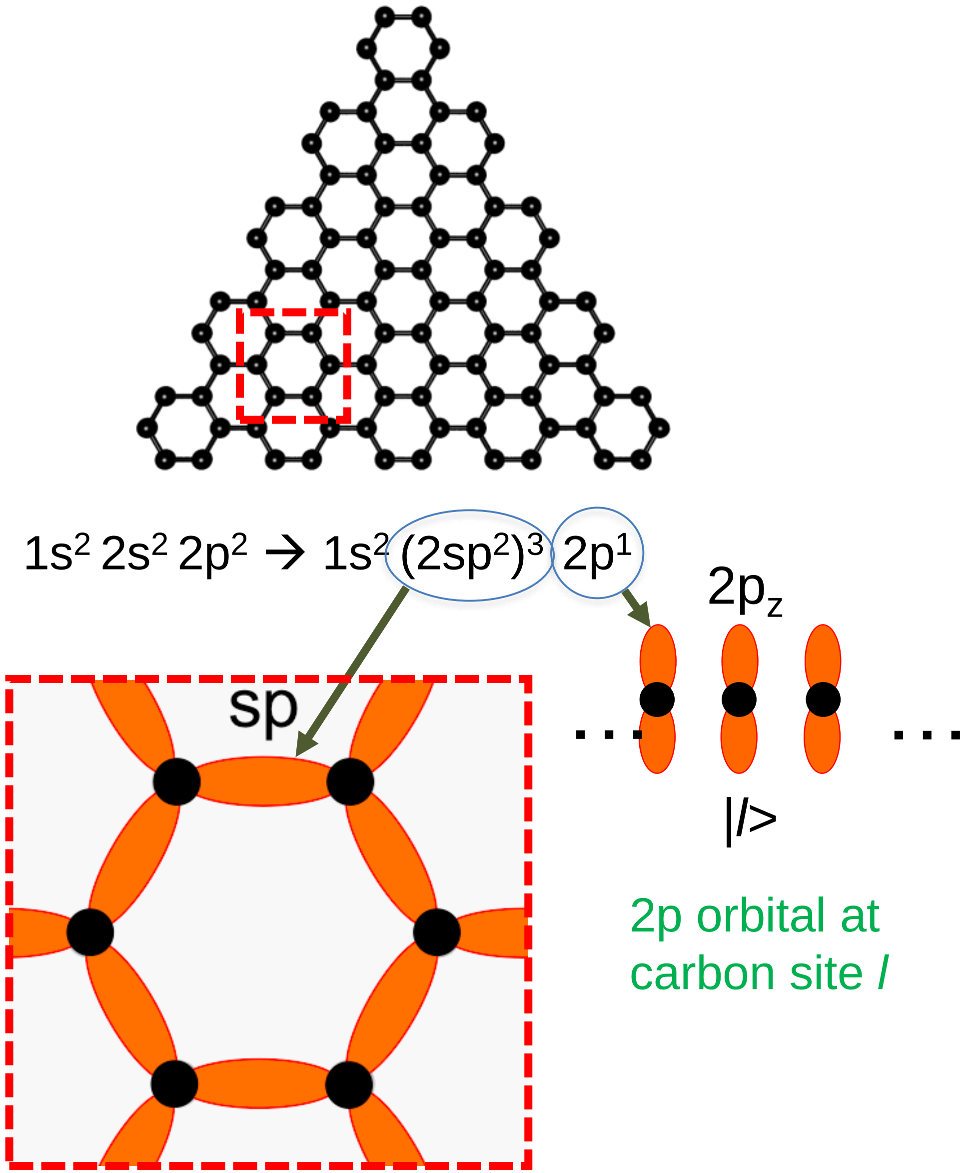
Towards near-IR and visible



$$\omega_{\mathrm{p}} \propto \sqrt{\frac{E_{\mathrm{F}}}{D}}$$

J. D. Cox and F. J. García de Abajo, Acc. Chem. Res. **52**, 2536 (2019)

Atomistic description



Single-electron density matrix EOM:

$$\frac{\partial \rho}{\partial t} = -\frac{\mathrm{i}}{\hbar}[H, \rho] - \frac{1}{\tau}(\rho - \rho^0)$$

$$H = H_{\mathrm{TB}} - e\phi$$

$$\langle l | H_{TB} | l' \rangle = -h\delta_{\langle l, l' \rangle}$$

Tight-binding Hamiltonian

$$\phi_l^{\mathrm{ext}} = -\mathbf{R}_l \cdot \mathbf{E}(t)$$

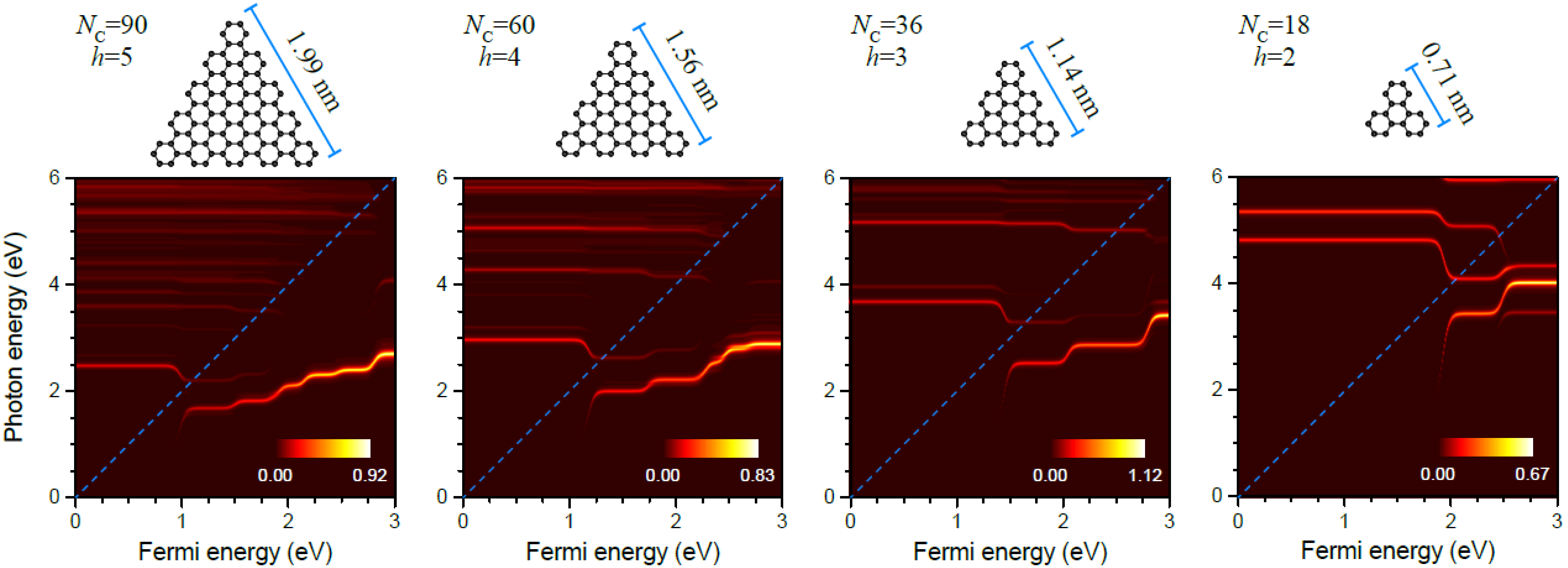
External potential

$$\phi_l^{\mathrm{ind}} = -2e \sum_{l'} v_{ll'} \left(\rho_{l'l'} - \rho^0_{l'l'} \right)$$

Coulombic electron-electron interaction

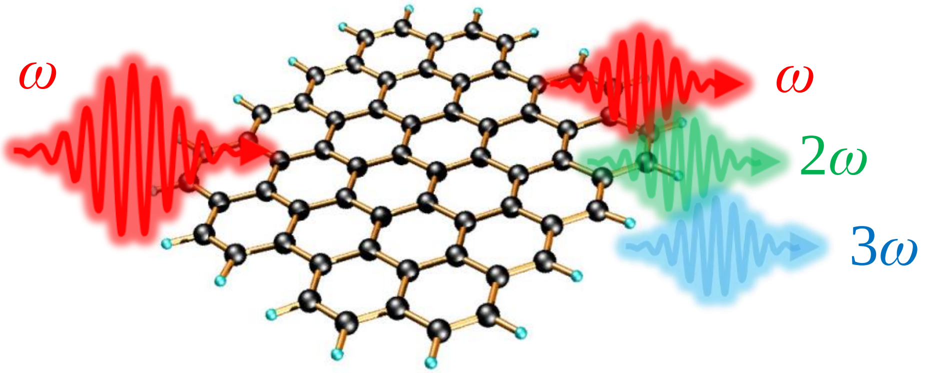
ρ_{ll} → e^- population at carbon site l

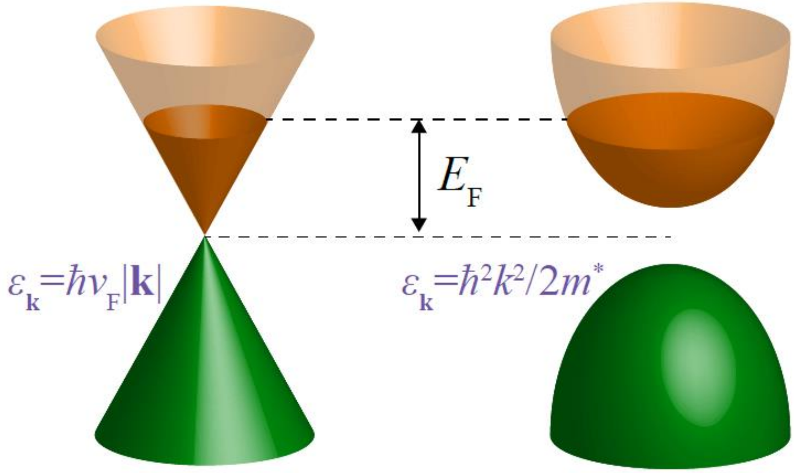
Quantum finite size effects

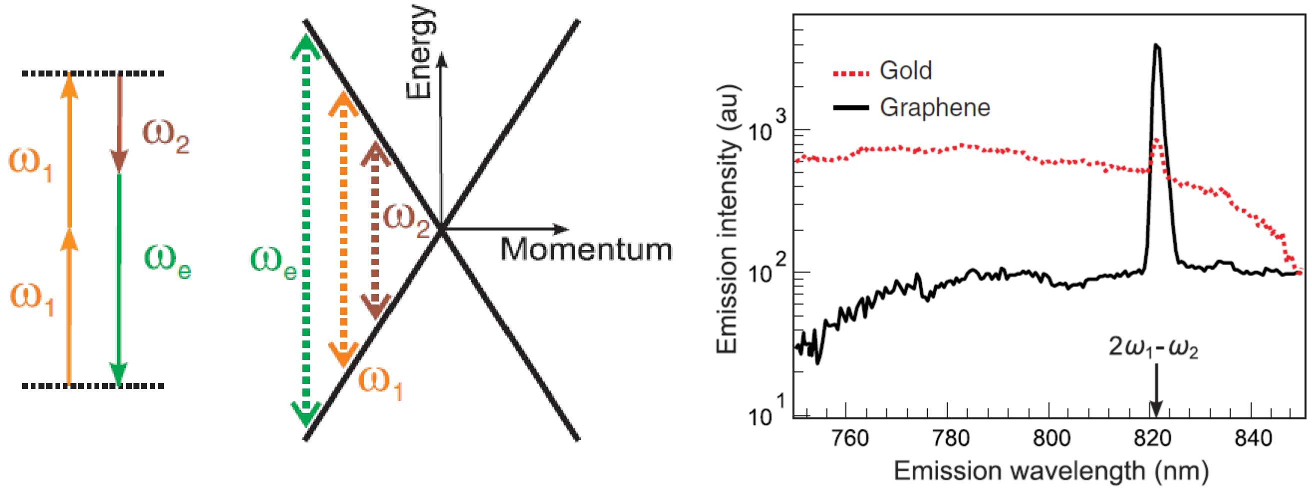


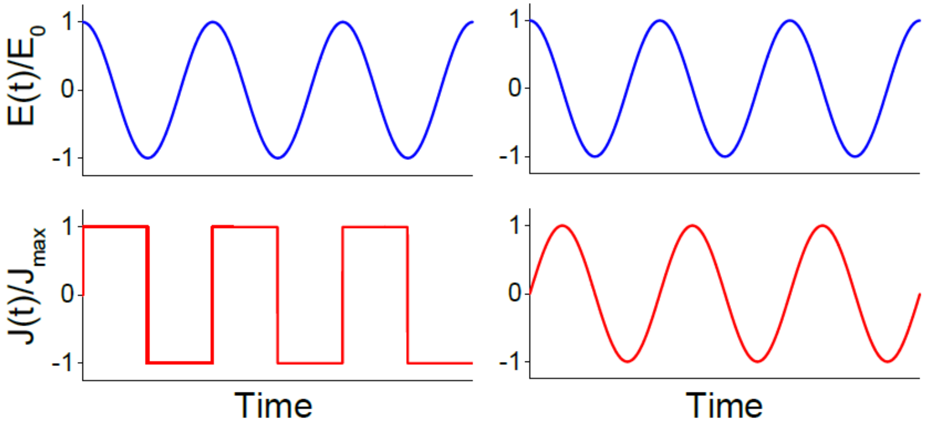
J. D. Cox and F. J. García de Abajo, Acc. Chem. Res. **52**, 2536 (2019)

Graphene nonlinear response







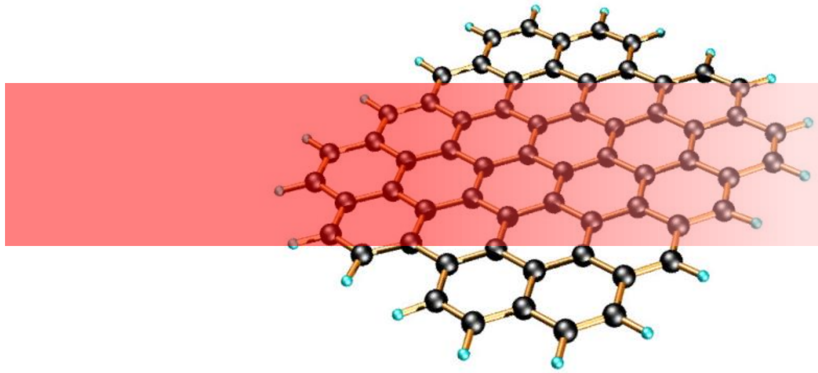


$$f(x) = \frac{4}{\pi} \sum_{n=1,3,5,\ldots}^{\infty} \frac{1}{n} \sin\left(\frac{n\pi x}{L}\right)$$

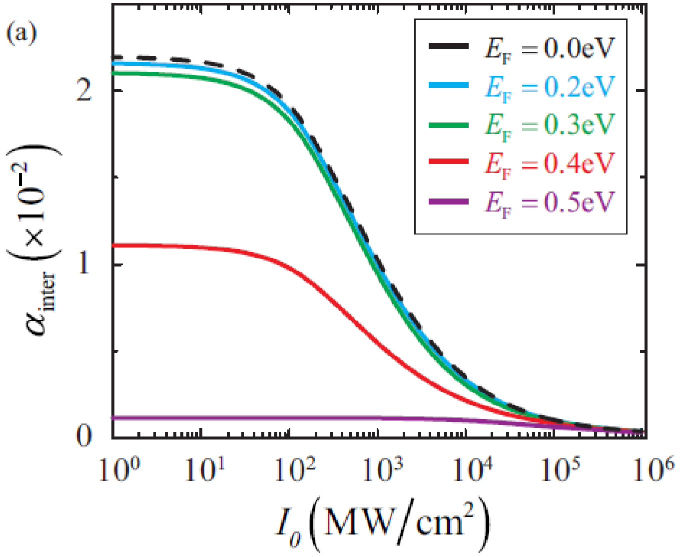
E. Hendry et al., Phys. Rev. Lett. (2010)

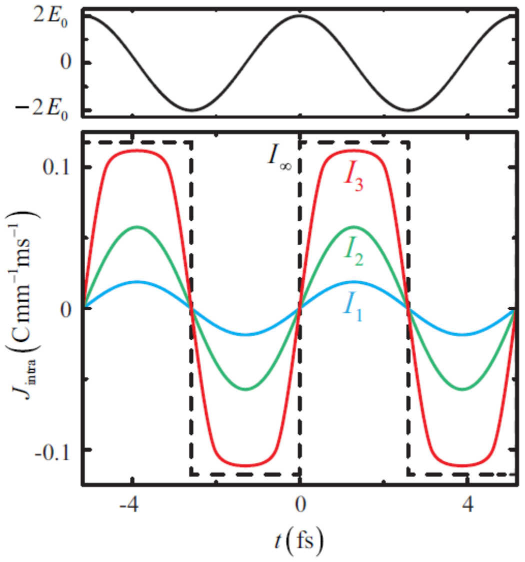
See e.g., S. A. Mikhailov, Europhys. Lett. (2007)

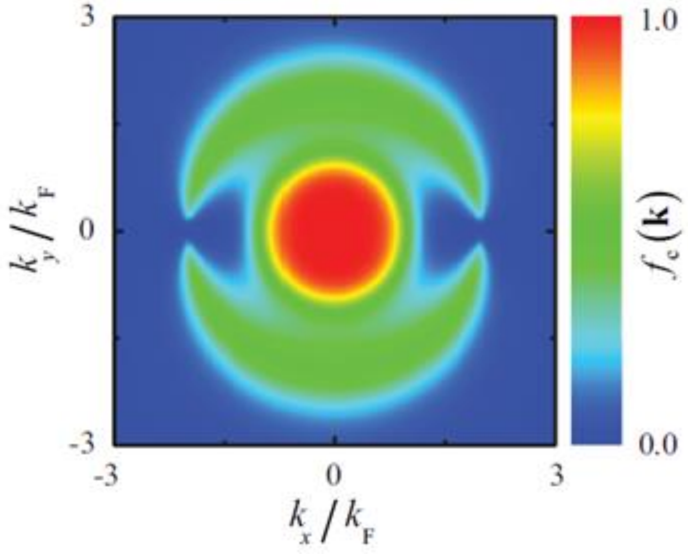
Graphene nonlinear response

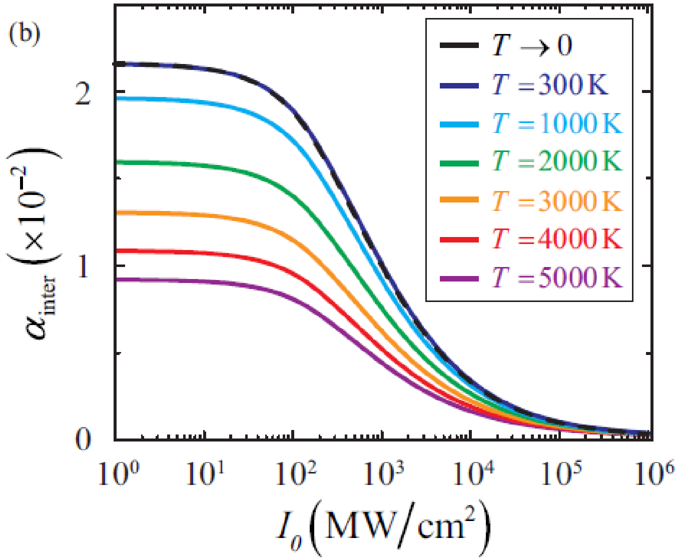


$$I_t = I_0 e^{-\alpha z}$$



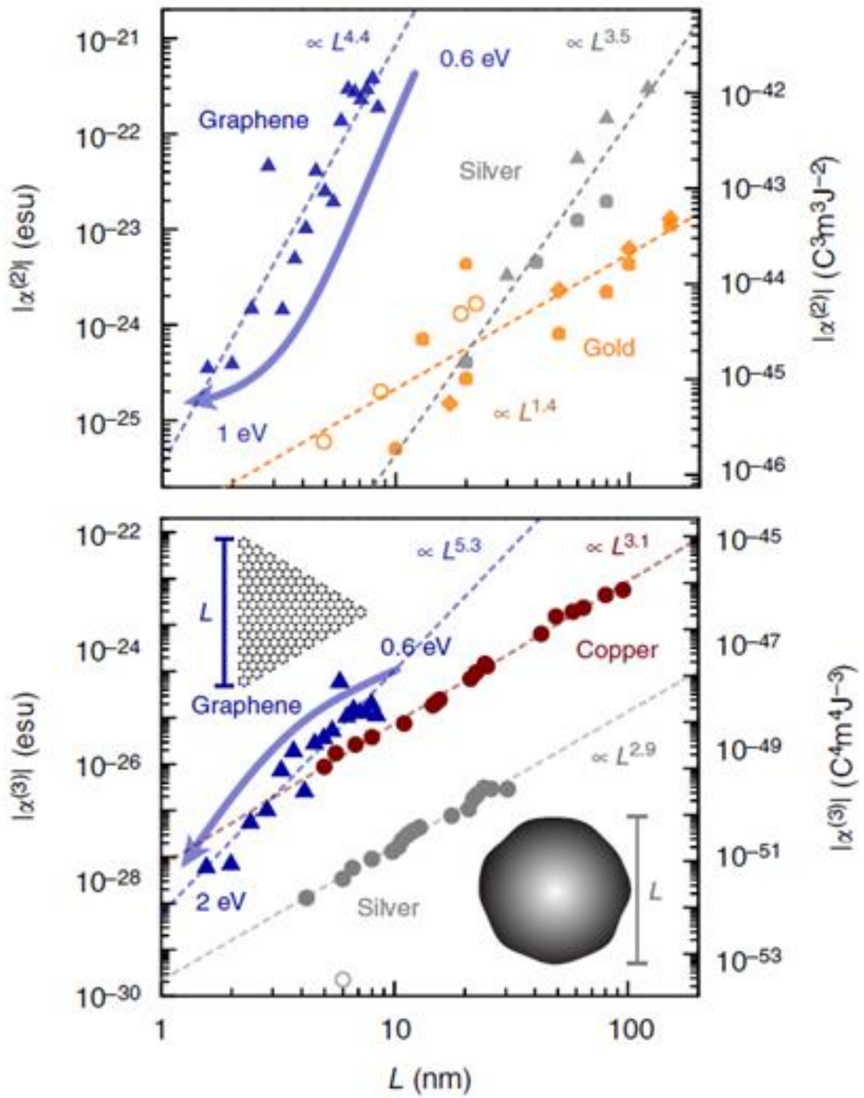


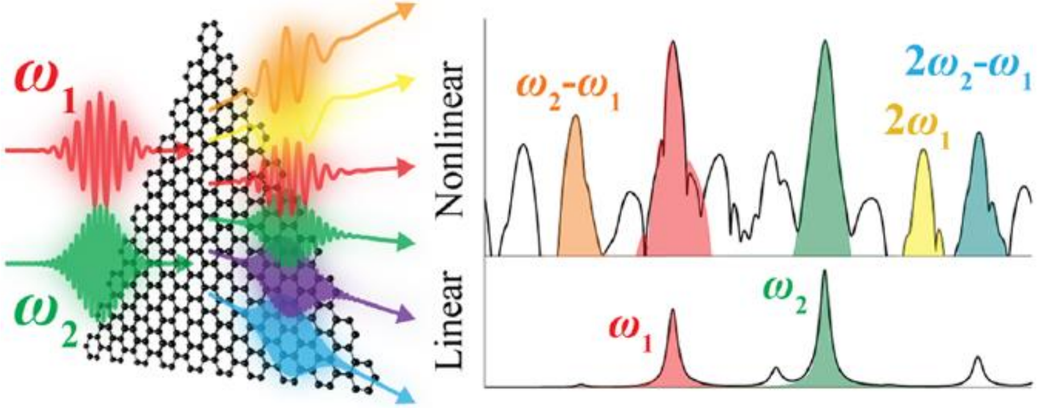


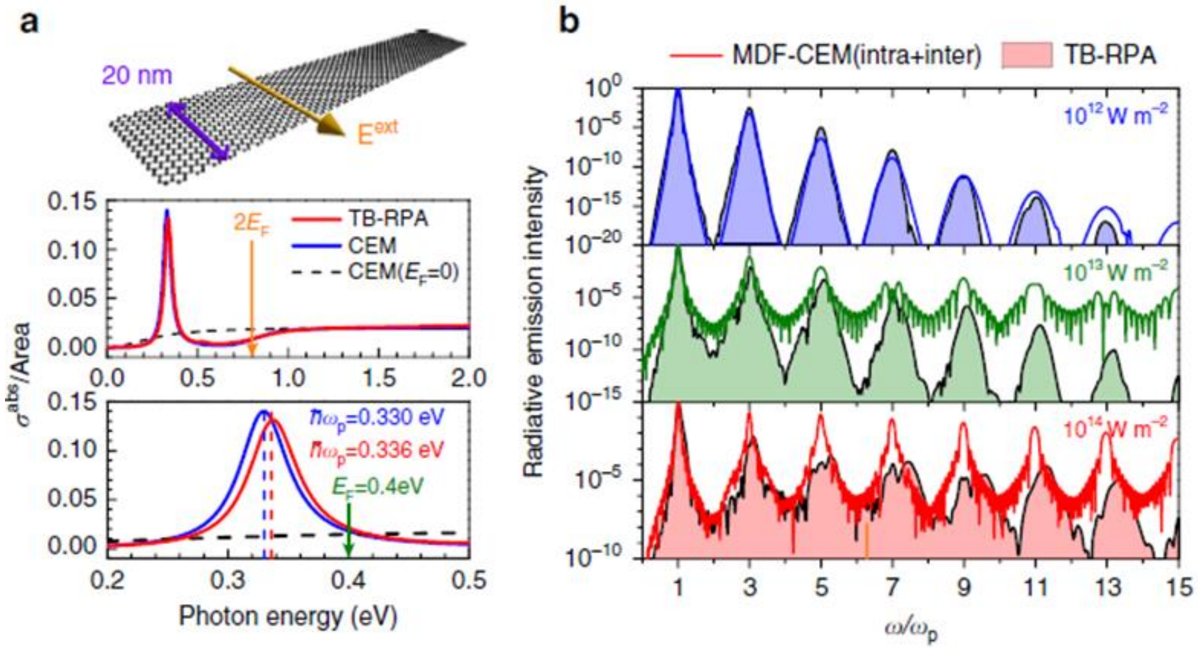


A. Marini, J. D. Cox, and F. J. García de Abajo, Phys. Rev. B **95**, 125408 (2017).

Nonlinear graphene plasmonics







Review article: J. D. Cox and F. J. García de Abajo, Acc. Chem. Res. **52**, 2536 (2019)

Nonlinear graphene plasmonics

SDU

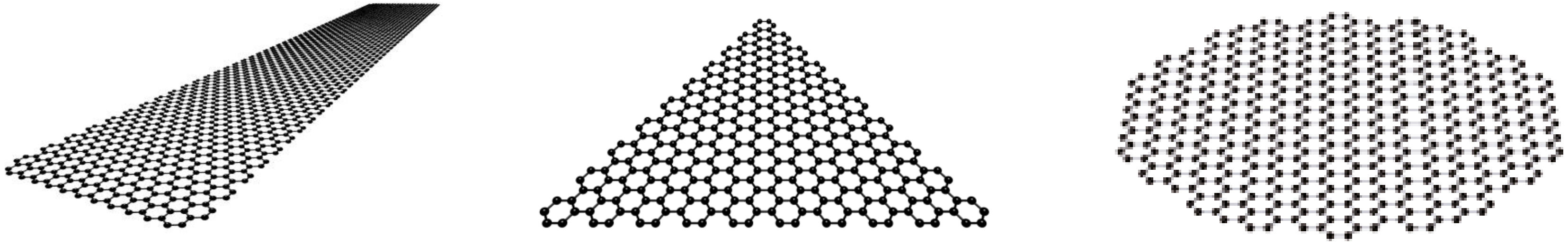
$$\alpha^{11}_{\omega,ab} = \frac{iD^2}{\omega}\sigma^{11}_{\omega} \sum_{j\in \text{res.}} \frac{\zeta_{j,a}\zeta_{j,b}}{1-\eta(\omega)/\eta_1}, \qquad \text{(linear)}$$

$$\alpha^{22}_{\omega,abc} = \frac{-iD}{2\omega} \frac{\sigma^{22,A}_{\omega}\tilde{\xi}^{22,A}_{abc} + \sigma^{22,B}_{\omega}\tilde{\xi}^{22,B}_{abc} + \sigma^{22,C}_{\omega}\tilde{\xi}^{22,C}_{abc}}{\left(1-\eta(\omega)/\eta_1\right)^2}, \qquad \text{(SHG)}$$

$$\alpha^{31}_{\omega,abcd} = \frac{iD^2}{\omega}\sigma^{31}_{\omega} \frac{\tilde{\xi}^{31}_{adcb} + 2\tilde{\xi}^{31}_{abcd}}{\left(1-\eta(\omega)/\eta_1\right)^2 \left|1-\eta(\omega)/\eta_1\right|^2}, \qquad \text{(Kerr)}$$

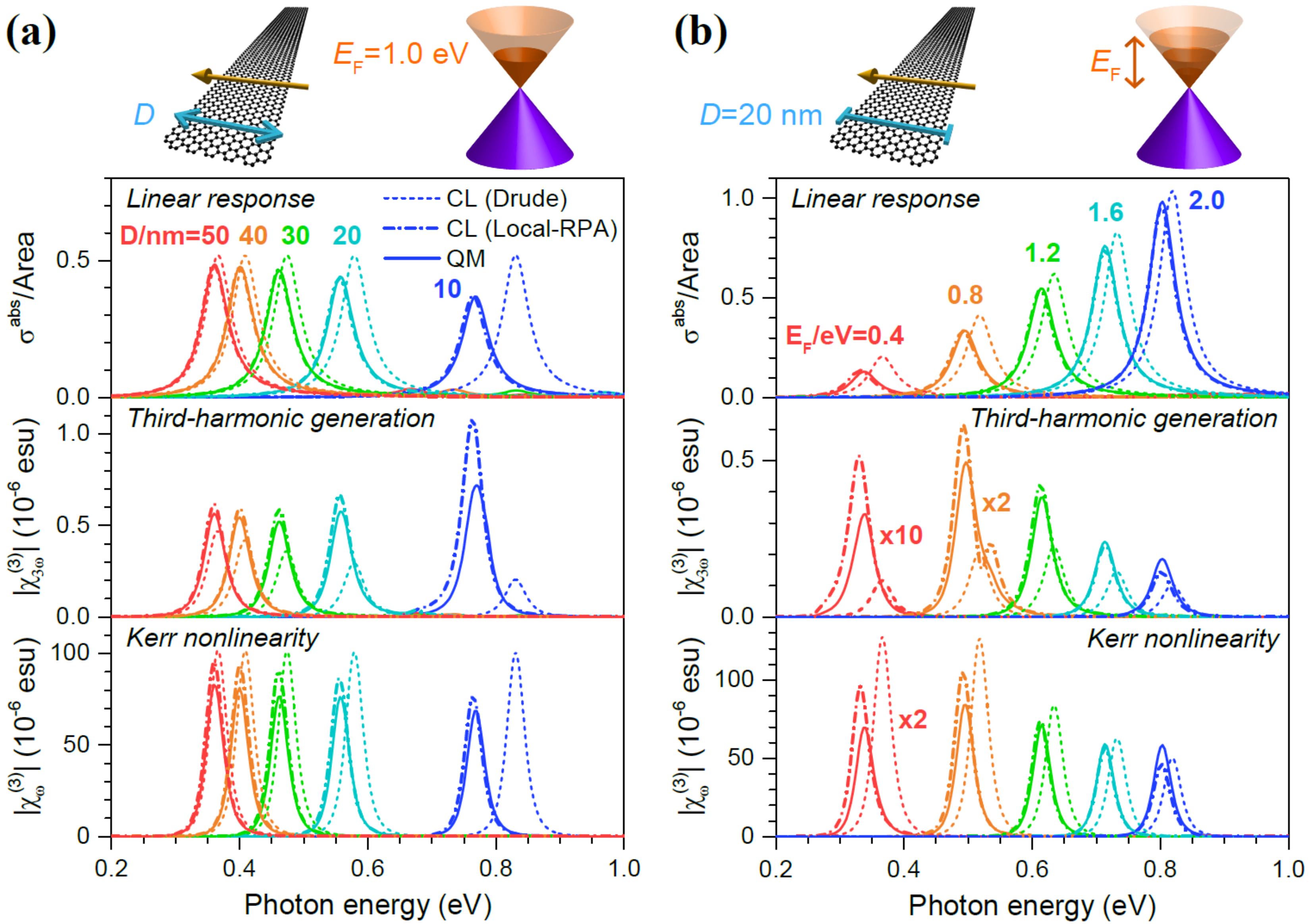
$$\alpha^{33}_{\omega,abcd} = \frac{iD^2}{3\omega}\sigma^{33}_{\omega} \frac{\tilde{\xi}^{33}_{abcd}}{\left(1-\eta(\omega)/\eta_1\right)^3}, \qquad \text{(THG)}$$

	η_1	ζ_{1x}	$\tilde{\xi}^{22,A}_{xxx}$	$\tilde{\xi}^{22,B}_{xxx}$	$\tilde{\xi}^{22,C}_{xxx}$	$\tilde{\xi}^{31}_{xxxx}$	$\tilde{\xi}^{33}_{xxxx}$
Ribbon	-0.06873	0.9428	0	0	0	1.031	-0.9415
Triangle	-0.08780	0.5400	0.3080	-0.3610	-0.7227	0.2622	0.2472
Disk	-0.07310	0.8461	0	0	0	0.7376	0.7082



J. D. Cox, R. Yu, and F. J. García de Abajo, Phys. Rev. B 96, 045442 (2017)

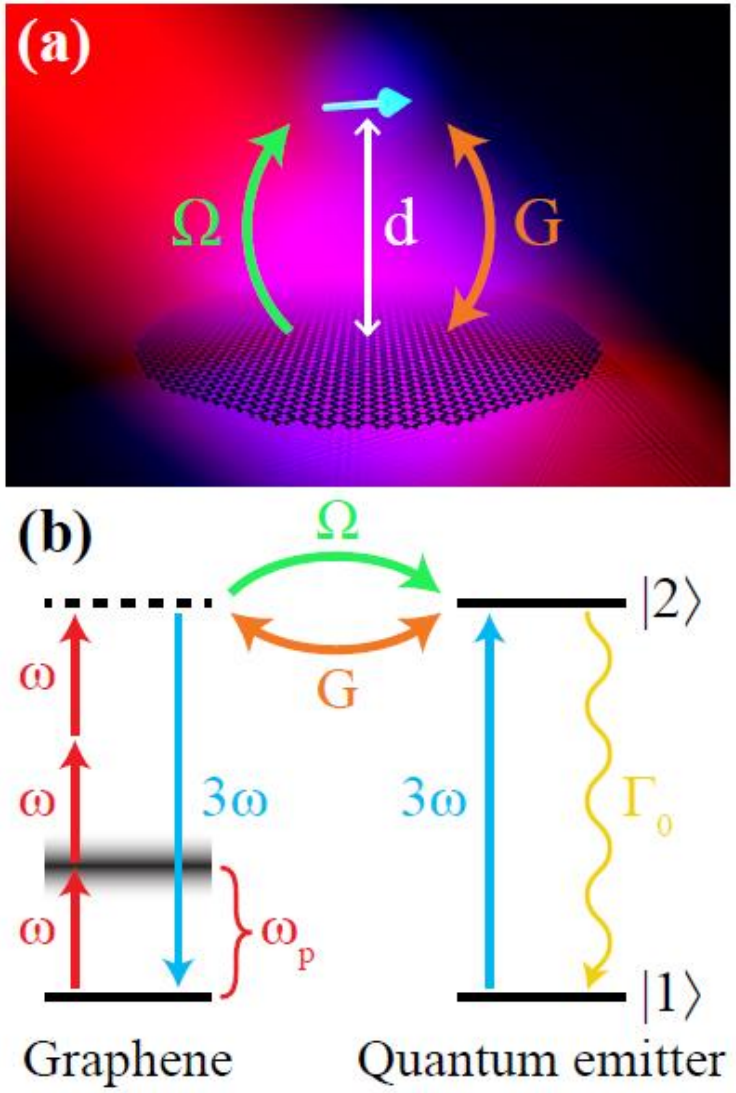
Nonlinear graphene plasmonics



J. D. Cox, R. Yu, and F. J. García de Abajo, Phys. Rev. B 96, 045442 (2017)

Nonlinear near-field coupling

SDU



$$\mathbf{E}^{ns}_{\rm ind}(\mathbf{r},\omega) = \frac{\rm i}{s\omega\epsilon^{\rm a}_{s\omega}D}\sum_m \frac{b^{ns}_m(\omega)}{1-\eta^{(1)}_{s\omega}/\eta_m}\mathbf{e}_m(\mathbf{r}) + \frac{\rm i}{s\omega\epsilon^{\rm a}_{s\omega}D}\sum_m \frac{\mathbf{d}_{s\omega}\cdot\mathbf{e}_m(\mathbf{r})}{1-\eta^{(1)}_{s\omega}/\eta_m}\mathbf{e}_m(\mathbf{r})$$

$$\mathcal{H} = \hbar\sum_{i=1}^{2}\varepsilon_i\sigma_{ii} - \vec{\mu}_{12}\cdot\mathbf{E}(\mathbf{r},t)\left(\sigma_{12}+\sigma_{21}\right)$$

$$\dot{\rho} = -\frac{\rm i}{\hbar}[\mathcal{H},\rho] + \mathcal{L}[\rho]$$

$$\frac{\partial\rho_{11}}{\partial t} = \Gamma_0\rho_{22} + {\rm i}\left(\Omega^* + G^*\tilde{\rho}_{12}\right)\tilde{\rho}_{21} - {\rm i}\left(\Omega + G\tilde{\rho}_{21}\right)\tilde{\rho}_{12}$$

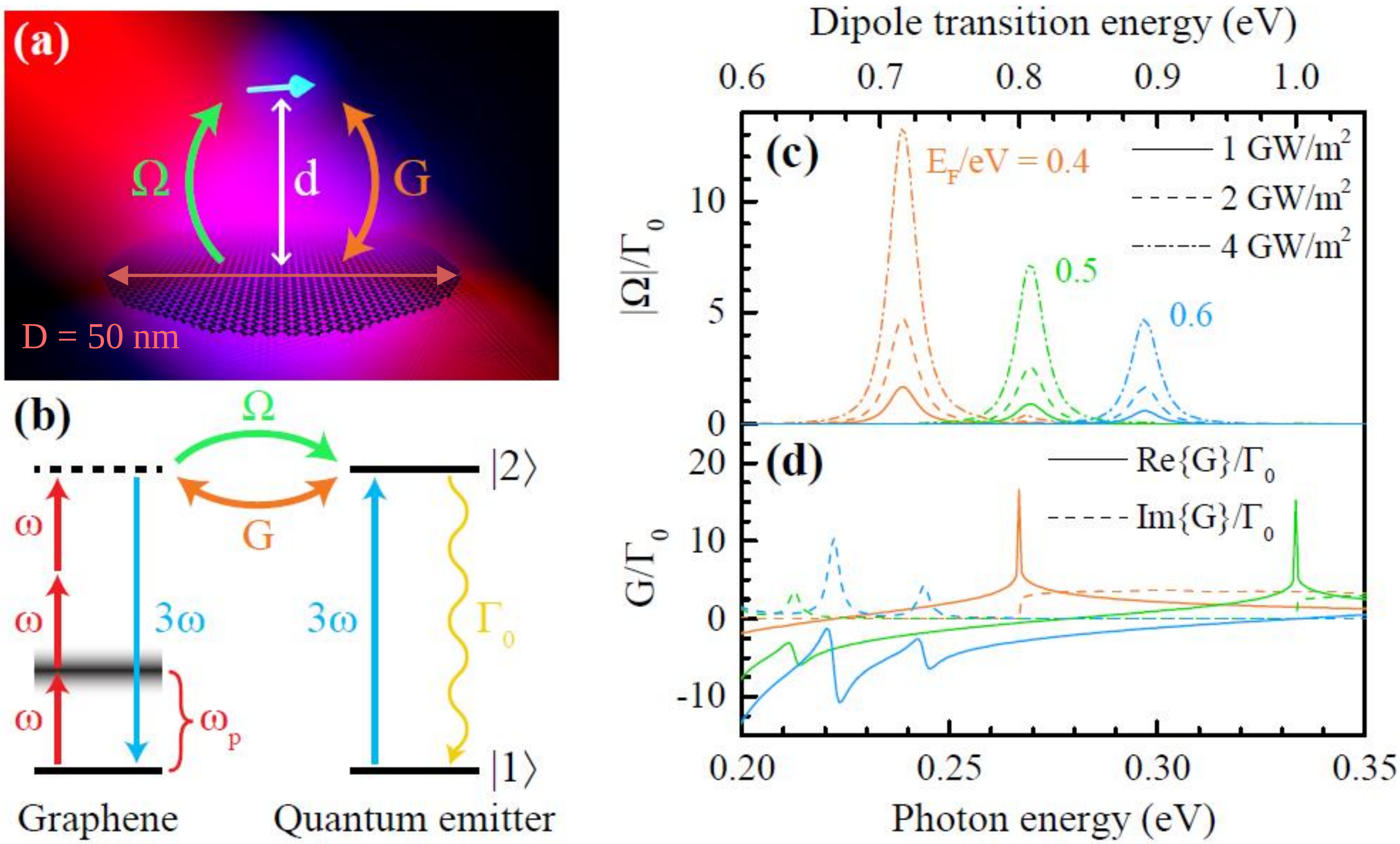
$$\frac{\partial\tilde{\rho}_{21}}{\partial t} = \left({\rm i}\Delta - \frac{\Gamma_0}{2}\right)\tilde{\rho}_{21} - {\rm i}\left(\Omega + G\tilde{\rho}_{21}\right)\left(\rho_{22}-\rho_{11}\right)$$

$$\Omega = \frac{\rm i}{s\hbar\omega\epsilon^{\rm a}_{s\omega}D}\sum_m b^{ns}_m(\omega)\frac{\vec{\mu}_{12}\cdot\mathbf{e}_m}{1-\eta^{(1)}_{s\omega}/\eta_m}$$

$$G = \frac{{\rm i}\sigma^{(1)}_{s\omega}}{s\hbar\omega(\epsilon^{\rm a}_{s\omega})^2D^4}\sum_m\frac{(\vec{\mu}_{12}\cdot\mathbf{e}_m)^2}{1-\eta^{(1)}_{s\omega}/\eta_m}$$

J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

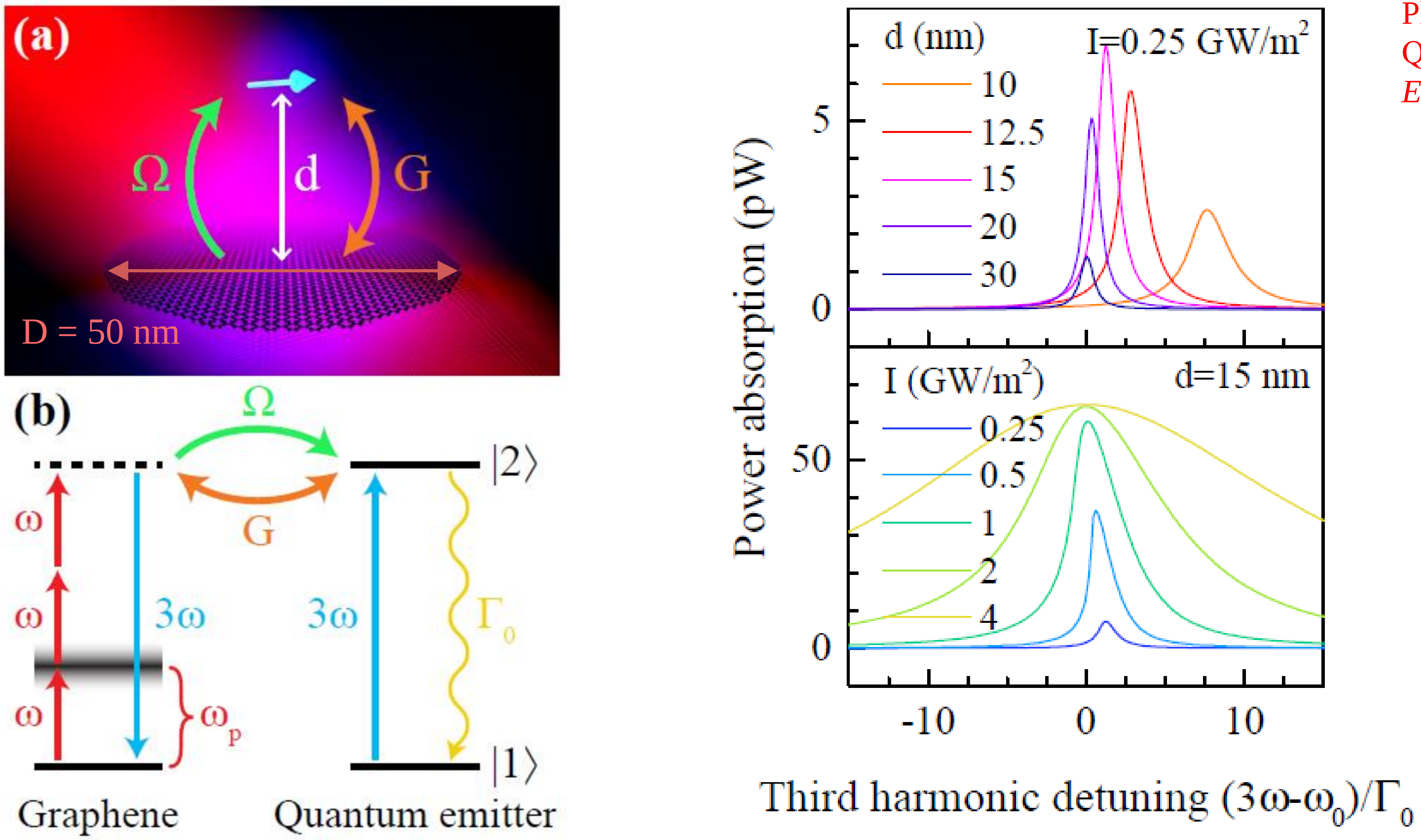
Nonlinear near-field coupling



Plasmon $\tau_p \approx 66$ fs;
QE decay $1/\Gamma_0 = 1$ ns;
Spacing $d = 20$ nm

J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

Power absorption

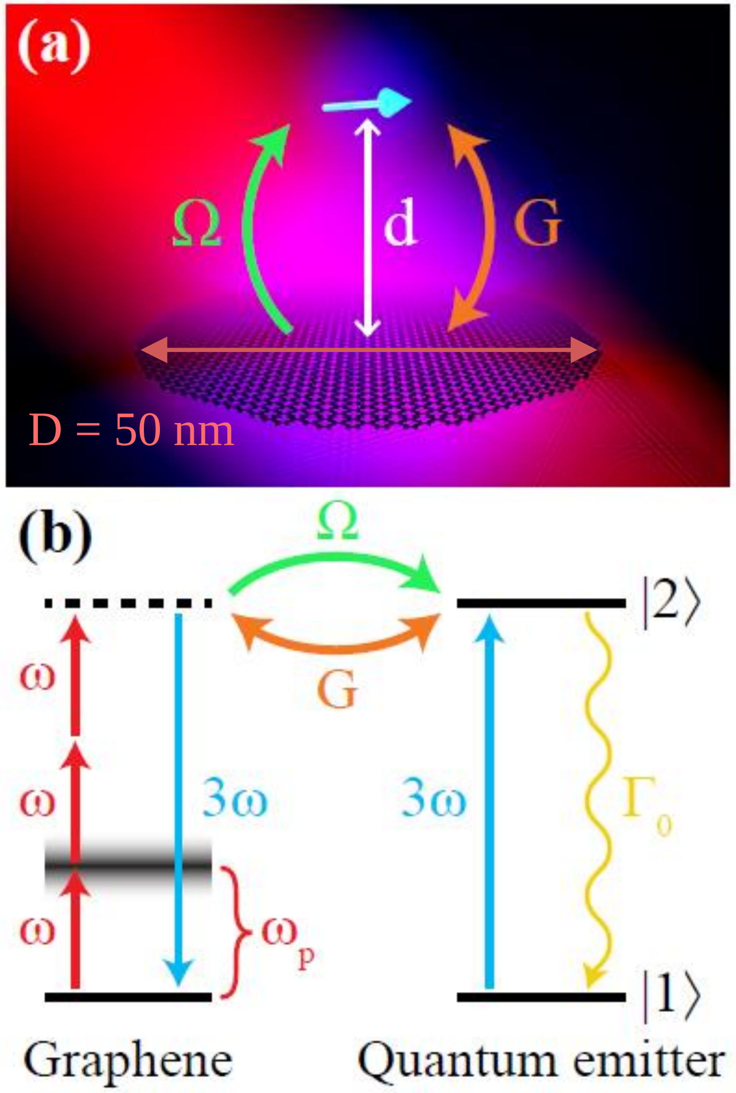


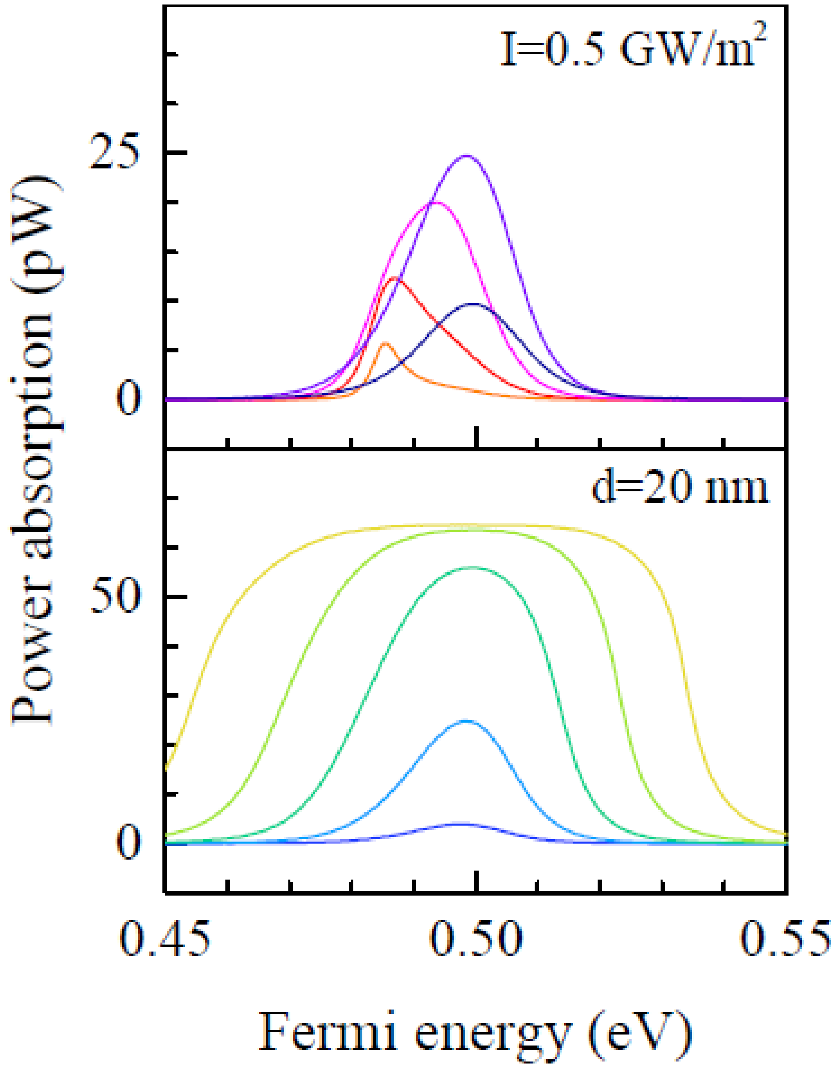
Plasmon $\tau_p \approx 66$ fs;
QE decay $1/\Gamma_0 = 1$ ns;
$E_F = 0.5$ eV

J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

Power absorption

SDU



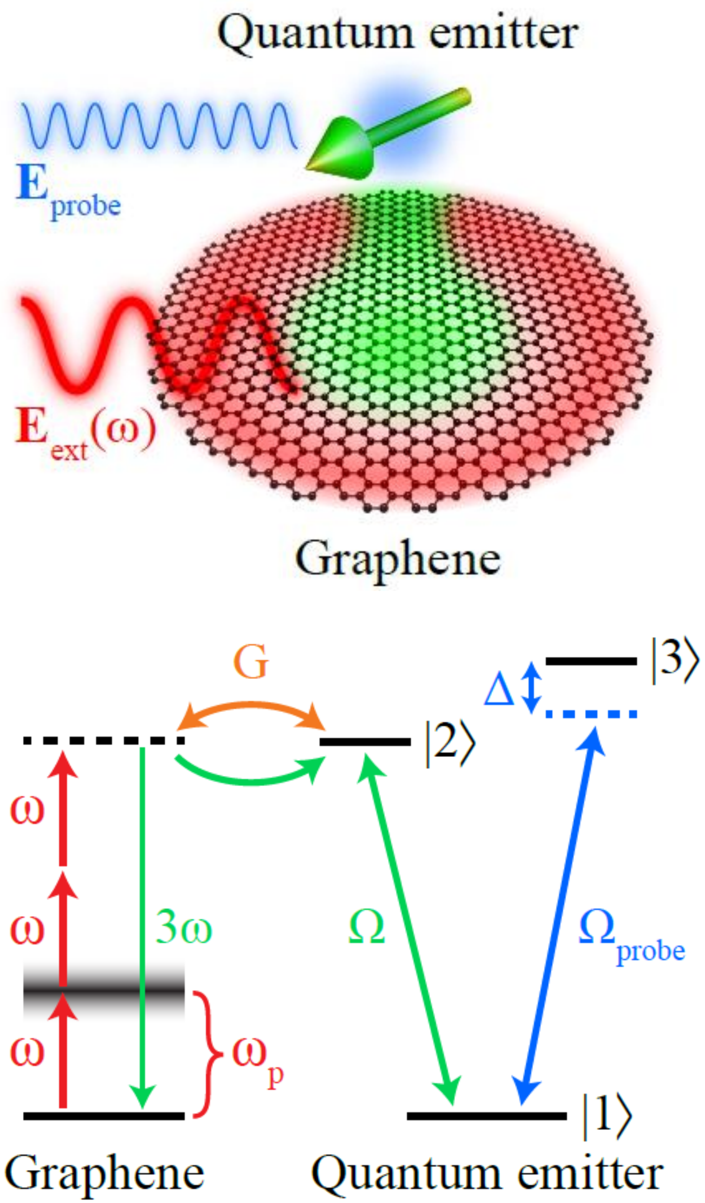


Plasmon $\tau_p \approx 66$ fs;
QE decay $1/\Gamma_0 = 1$ ns;
On resonance

J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

EIT

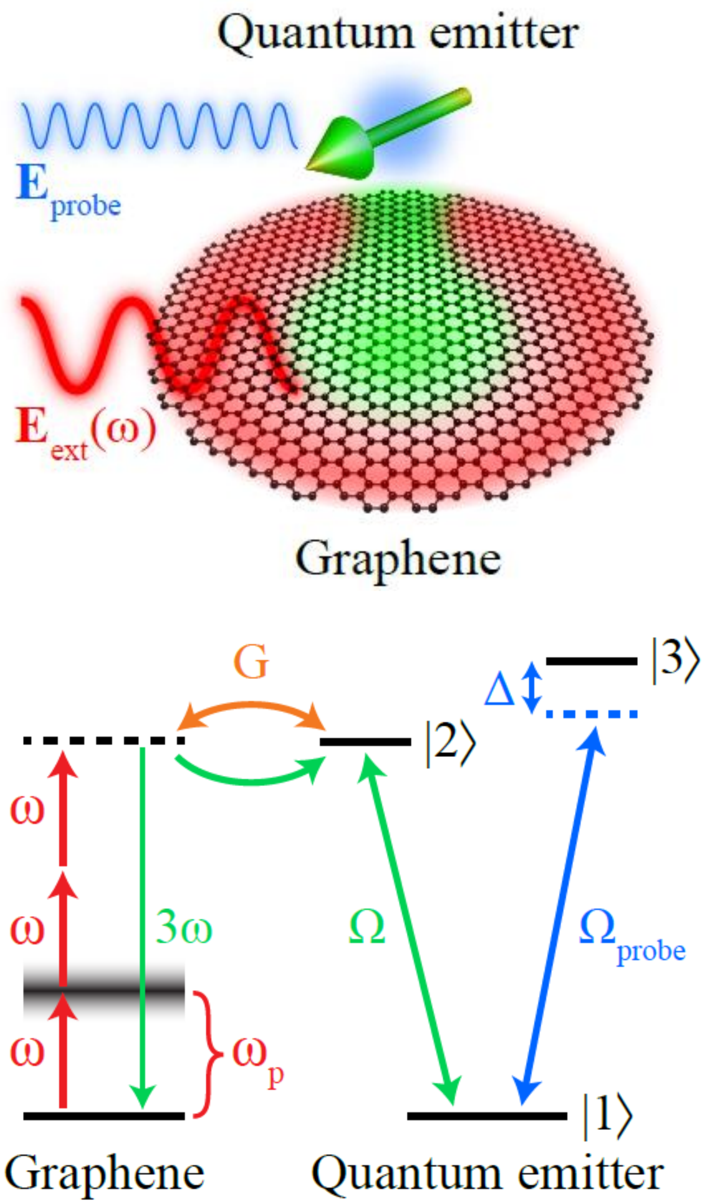
SDU

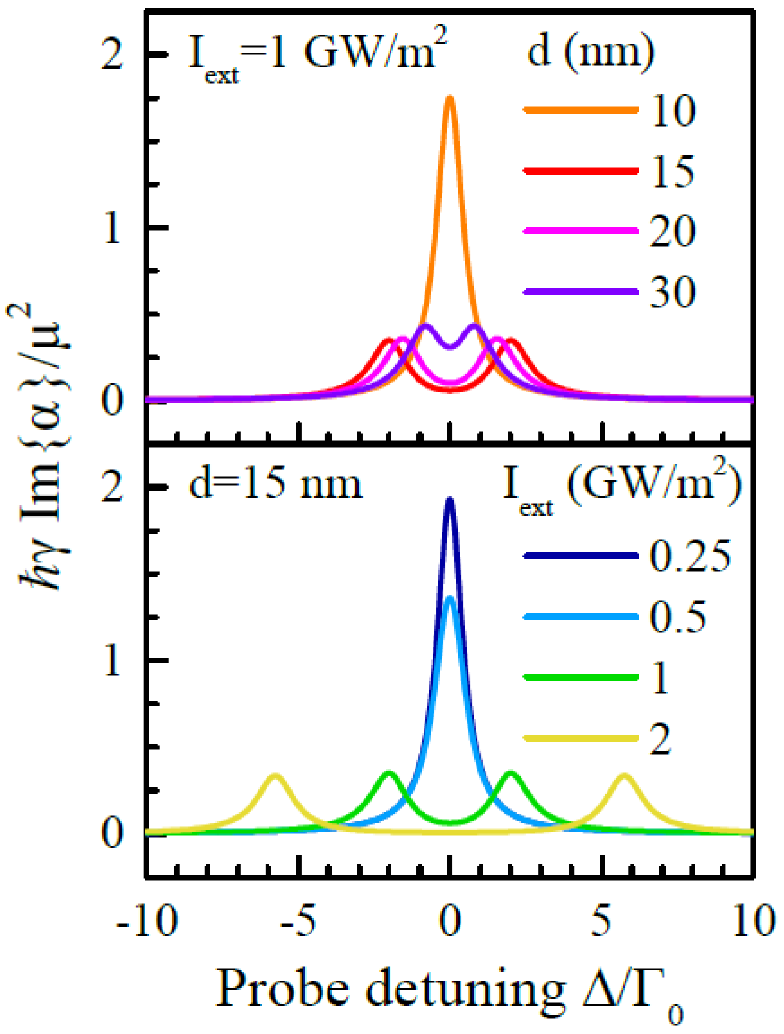


J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

EIT

SDU





J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

EIT

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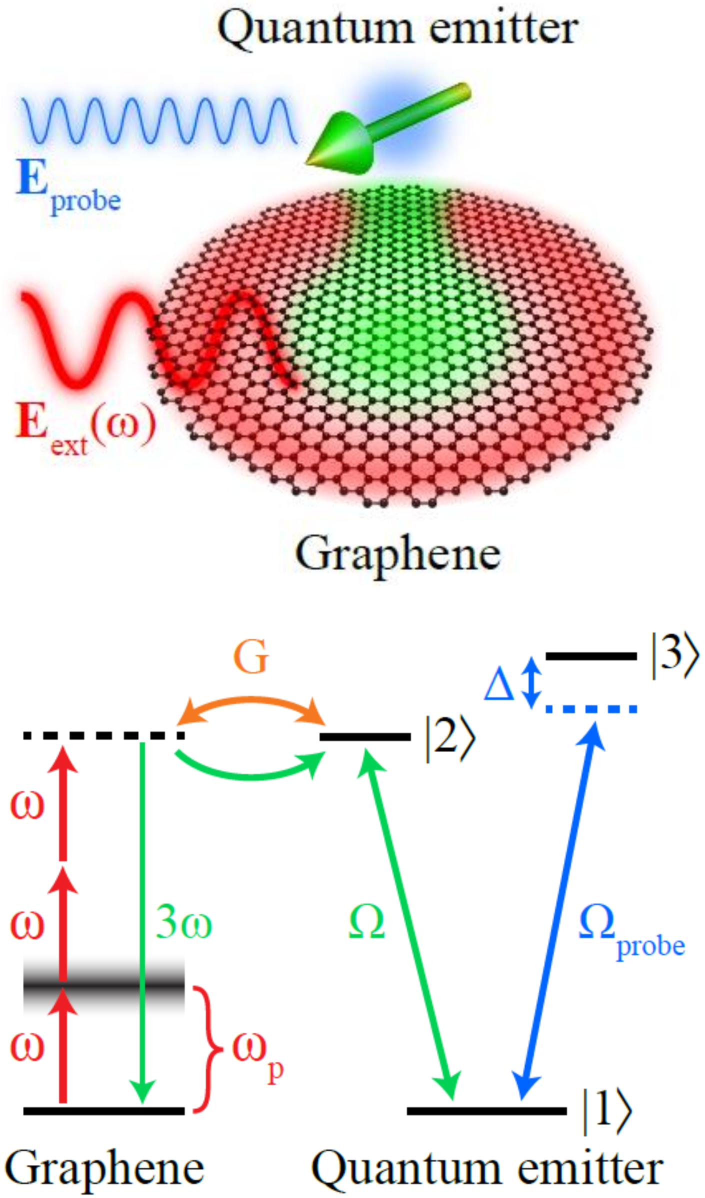
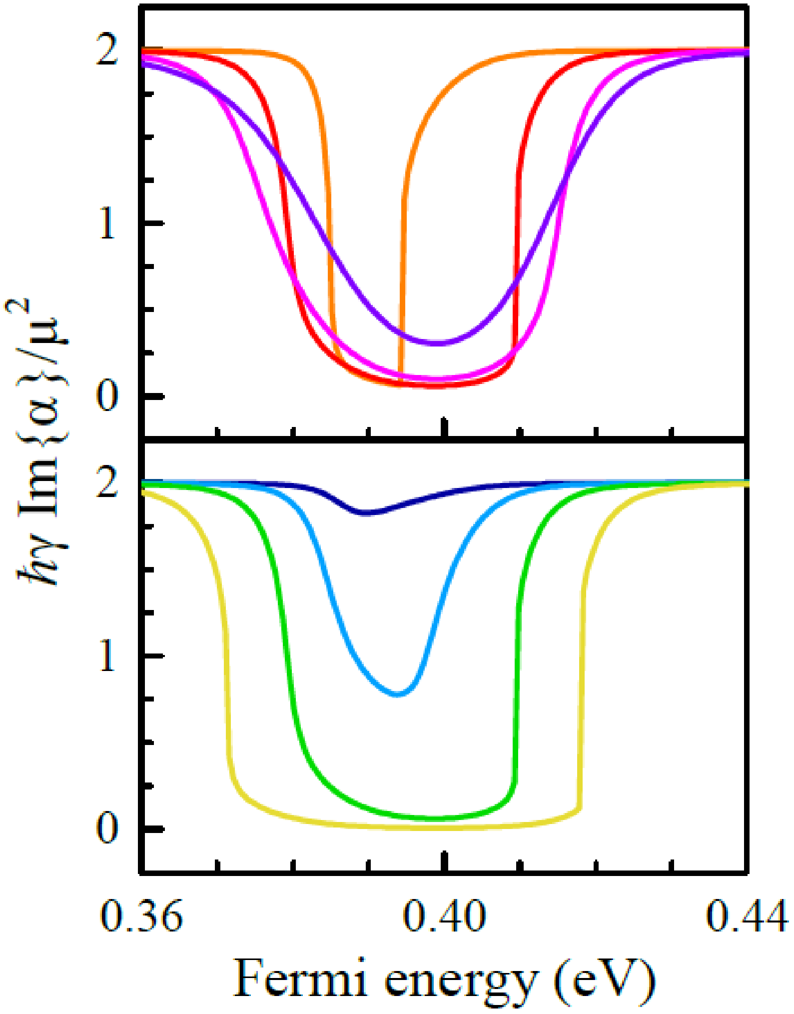
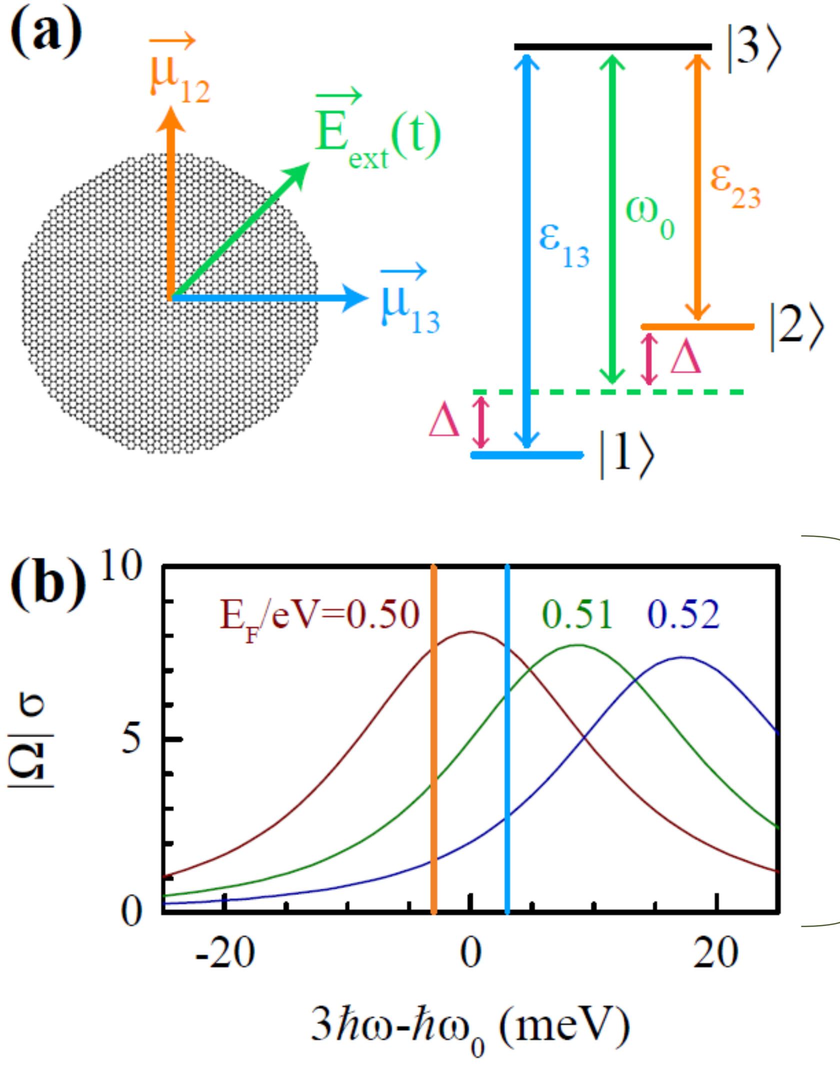




J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

Temporal control

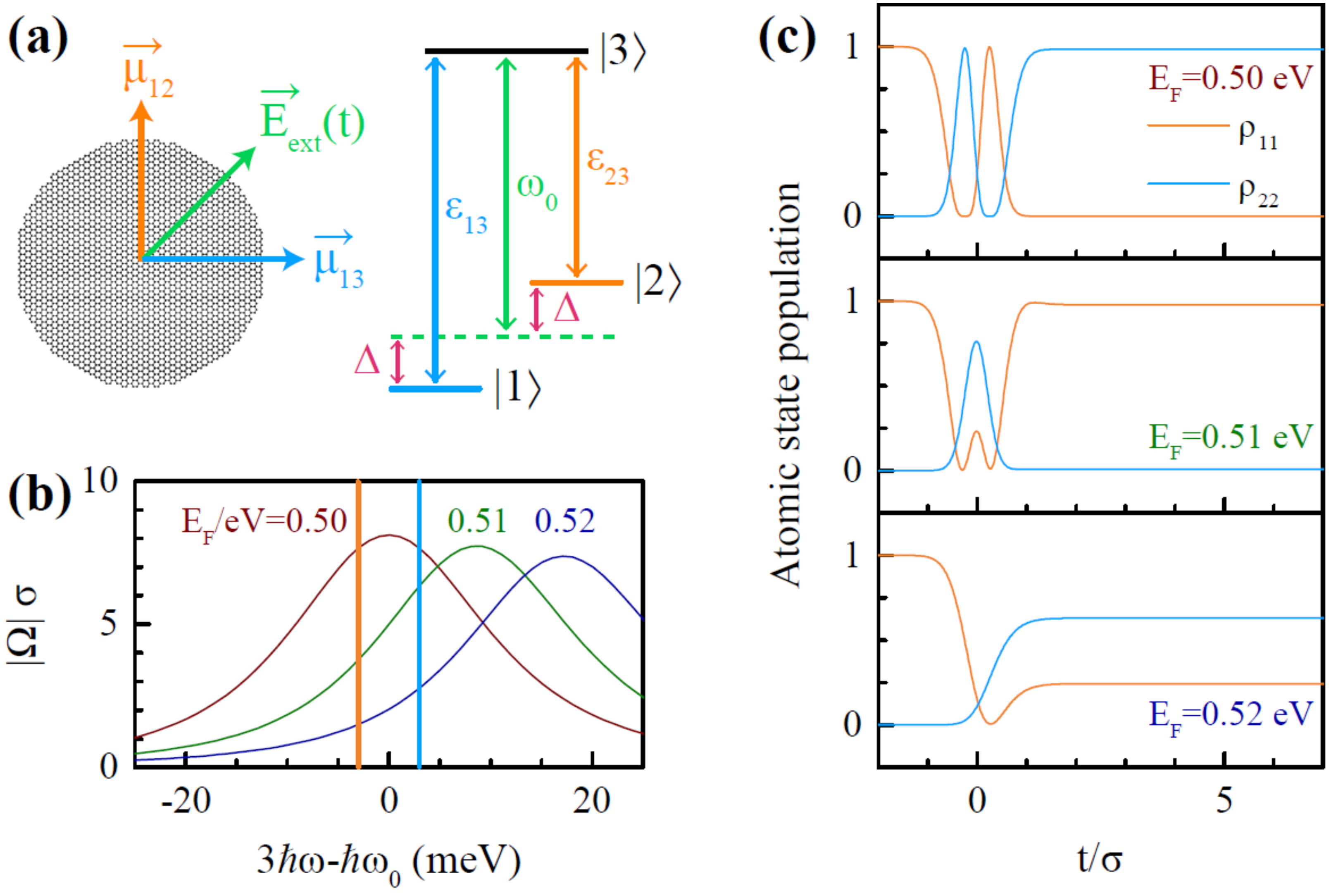
SDU



Peak intensity 1 TW/m^2
FWHM duration 100 fs

J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

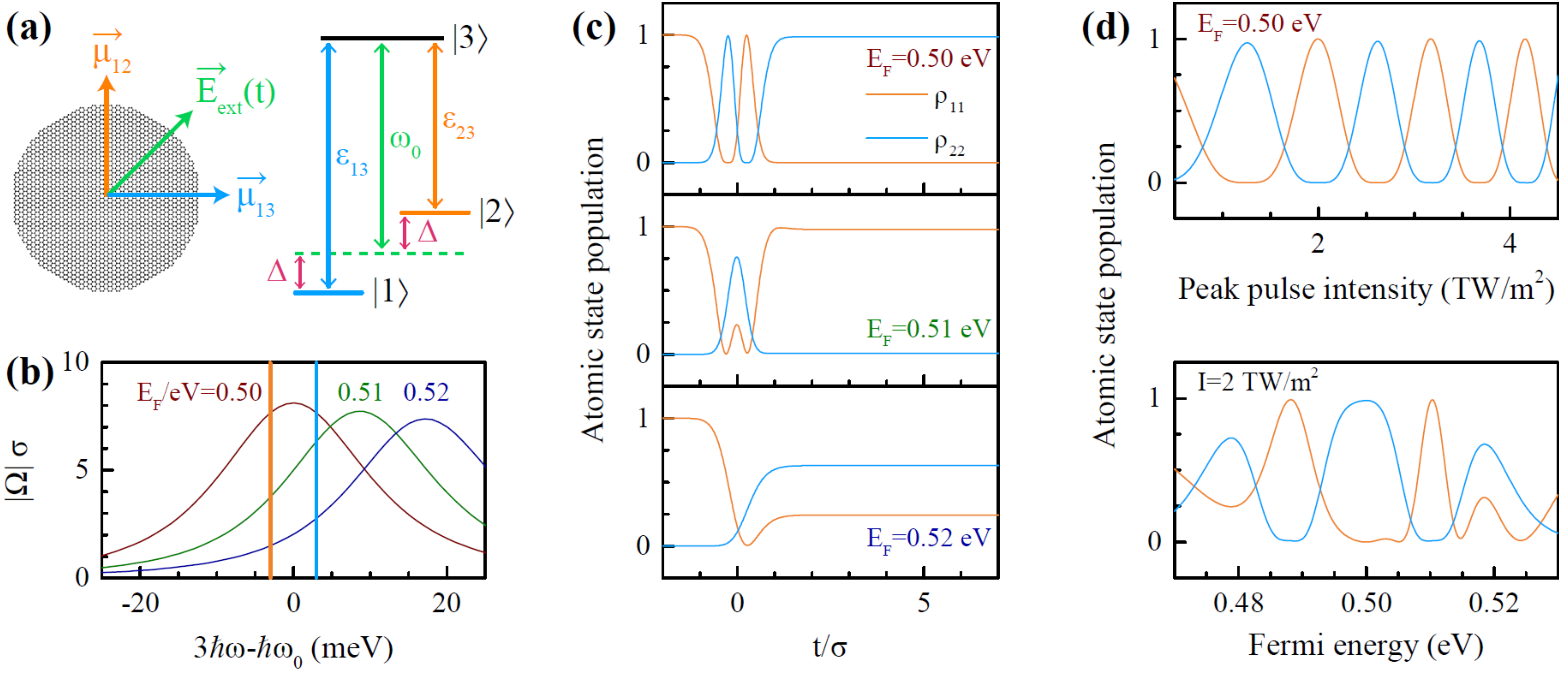
Temporal control



J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

Temporal control

SDU



J. D. Cox and F. J. García de Abajo, PRL 121, 257403 (2018)

Single-photon-level nonlinear optics

SDU

PRL **111**, 247401 (2013) PHYSICAL REVIEW LETTERS week ending 13 DECEMBER 2013

Single-Photon Nonlinear Optics with Graphene Plasmons

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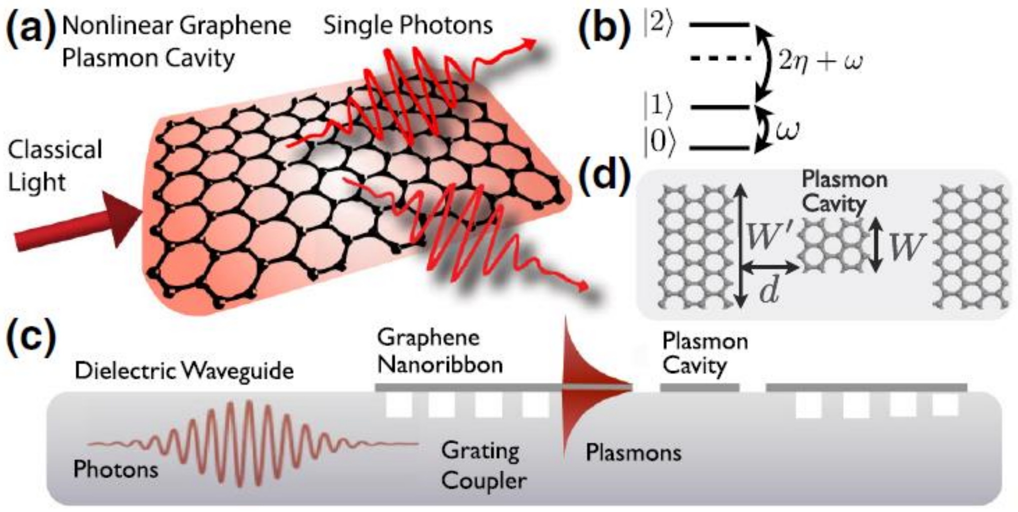
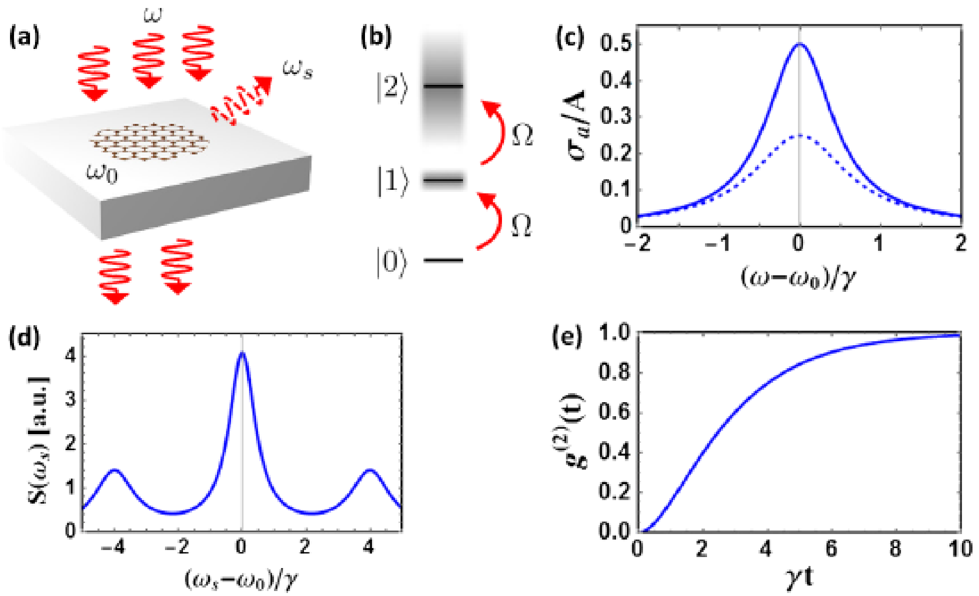
[1]*Department of Physics, Harvard University, Cambridge, Massachusetts 02138, USA*
[2]*ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain*
[3]*ICREA-Institući Catalana de Recerca i Estudis Avanats, 08010 Barcelona, Spain*
(Received 13 September 2013; published 11 December 2013)

We show that it is possible to realize significant nonlinear optical interactions at the few photon level in graphene nanostructures. Our approach takes advantage of the electric field enhancement associated with the strong confinement of graphene plasmons and the large intrinsic nonlinearity of graphene. Such a system could provide a powerful platform for quantum nonlinear optical control of light. As an example, we consider an integrated optical device that exploits this large nonlinearity to realize a single photon switch.





PRL **114**, 236801 (2015) PHYSICAL REVIEW LETTERS week ending 12 JUNE 2015

Multiplasmon Absorption in Graphene

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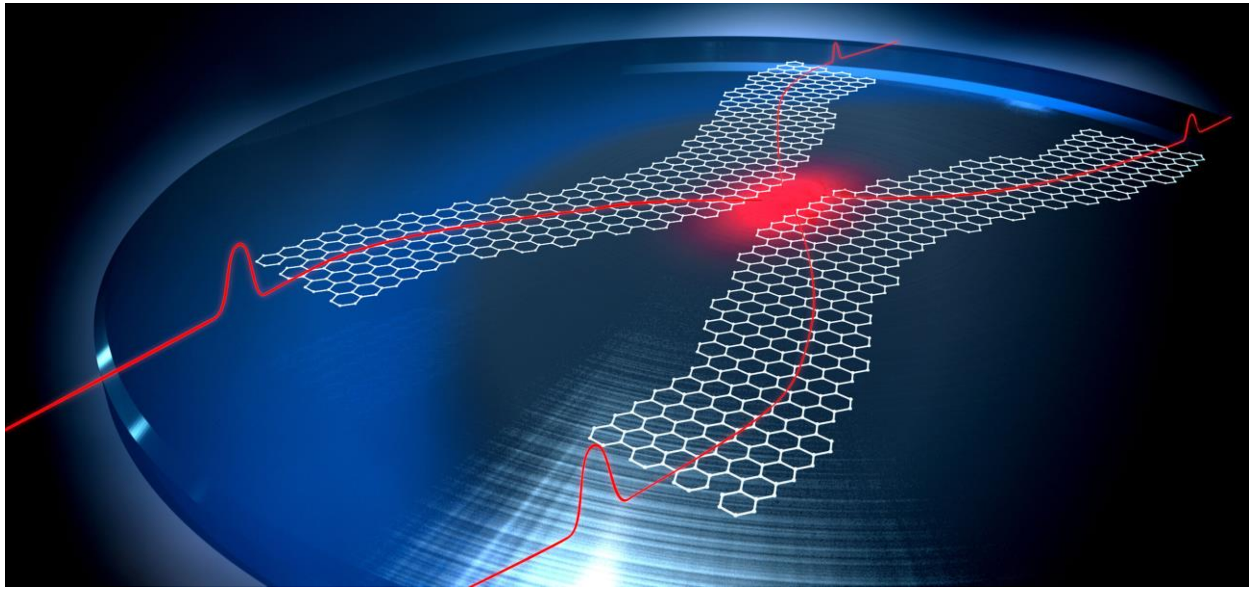
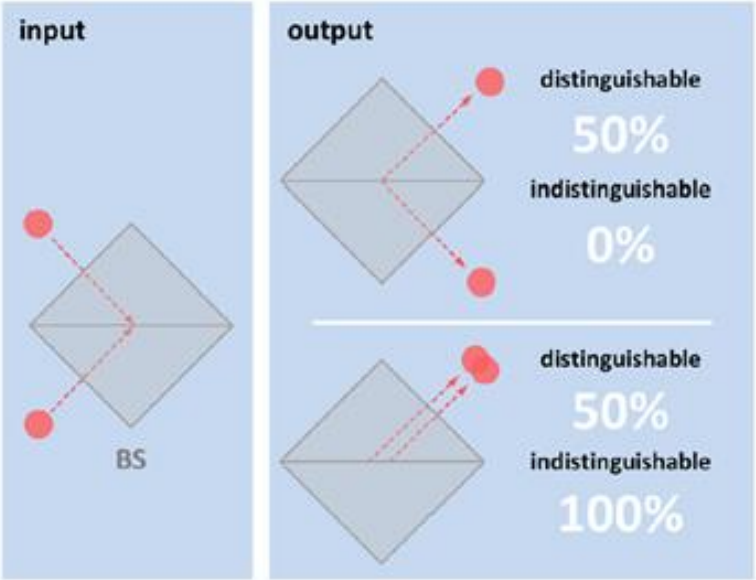
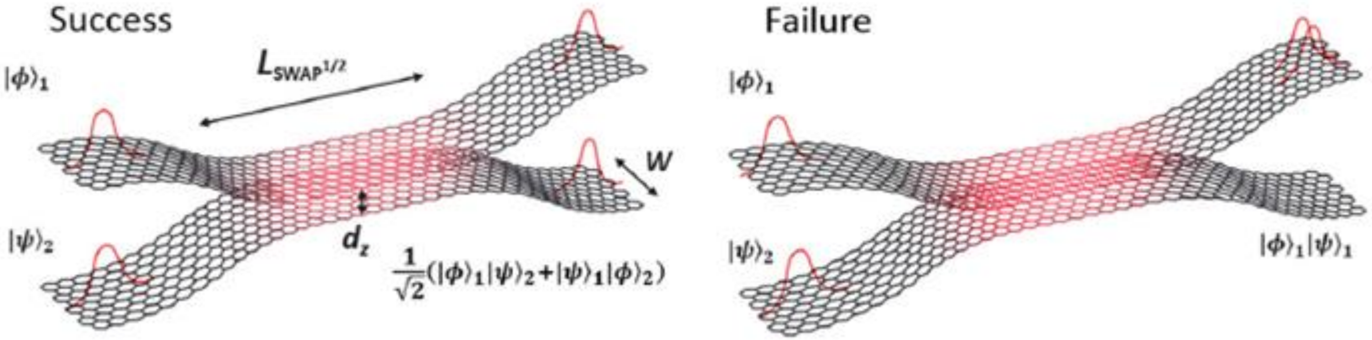
[1]*ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels (Barcelona), Spain*
[2]*Department of Physics, University of Zagreb, 10000 Zagreb, Croatia*
(Received 21 January 2015; published 10 June 2015)

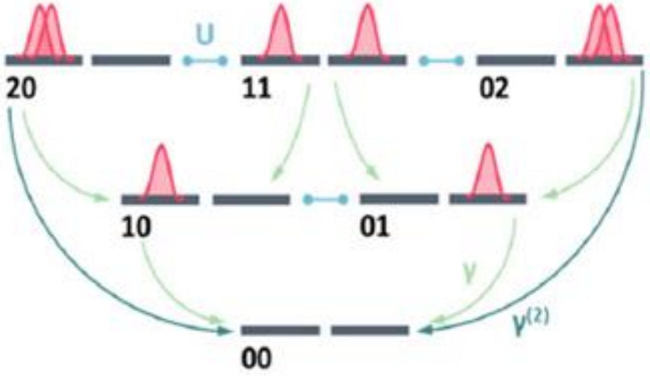
We show that graphene possesses a strong nonlinear optical response in the form of multiplasmon absorption, with exciting implications in classical and quantum nonlinear optics. Specifically, we predict that graphene nanoribbons can be used as saturable absorbers with low saturation intensity in the far-infrared and terahertz spectrum. Moreover, we predict that two-plasmon absorption and extreme localization of plasmon fields in graphene nanodisks can lead to a plasmon blockade effect, in which a single quantized plasmon strongly suppresses the possibility of exciting a second plasmon.

Quantum logic



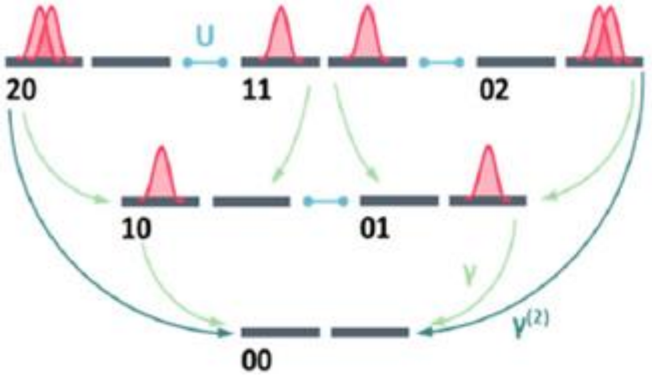


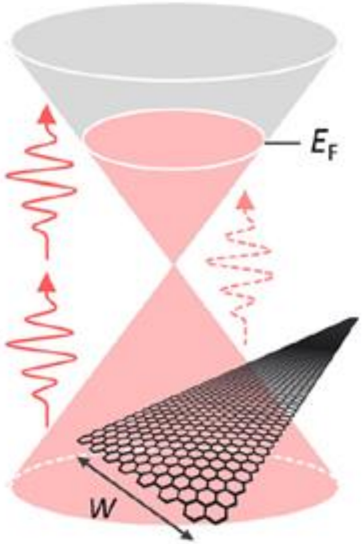


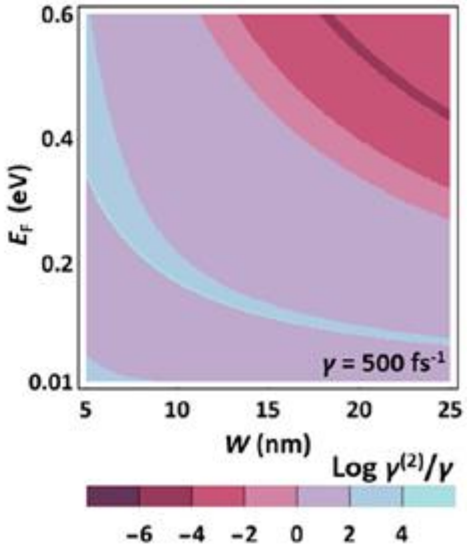


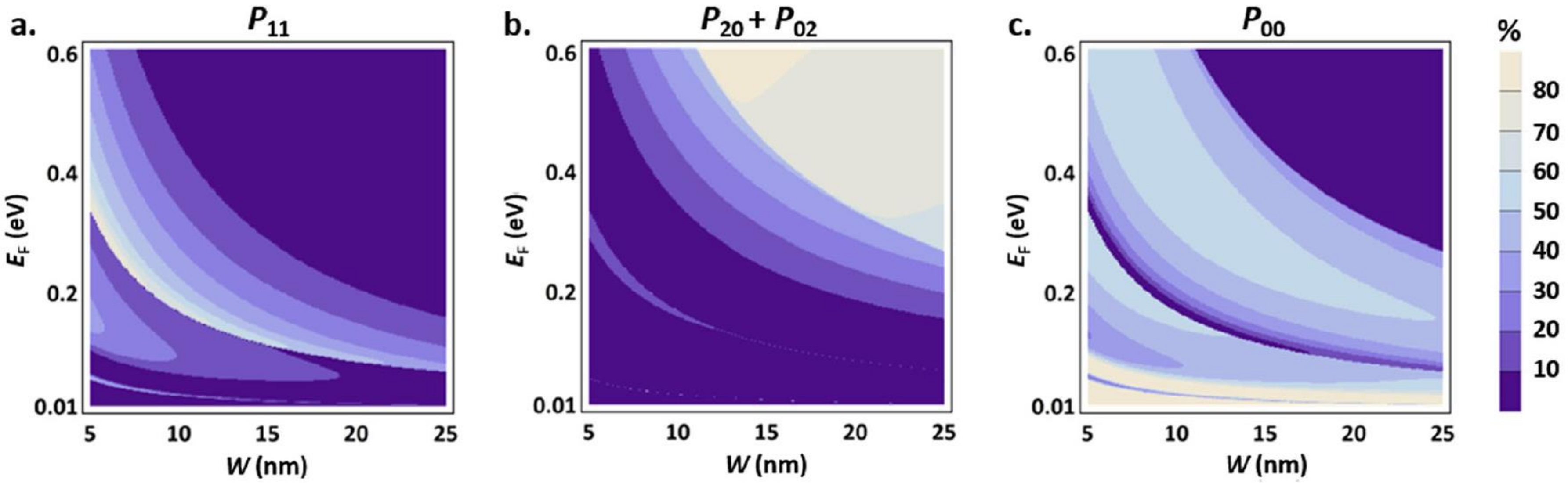
I. Alonso Calafell, J.D. Cox et al., npj Quantum Inf. 37 (2019)

Quantum logic





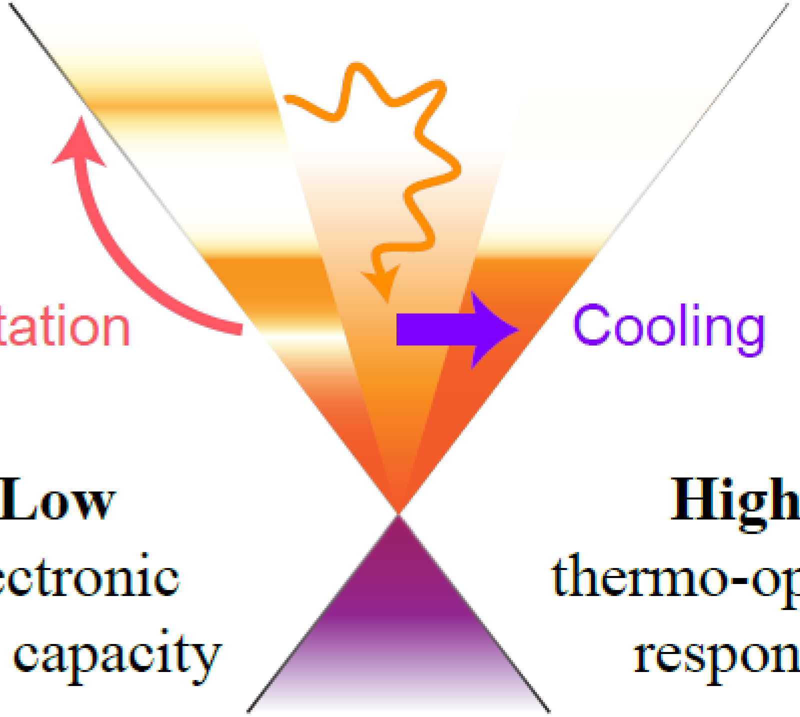


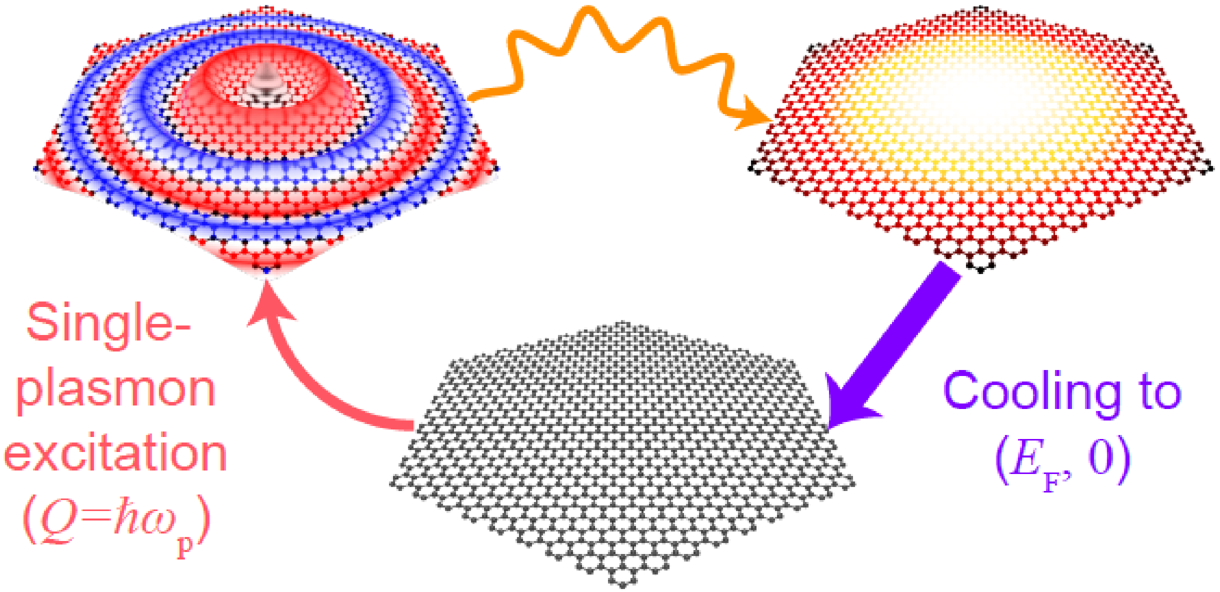


I. Alonso Calafell, J.D. Cox et al., npj Quantum Inf. 37 (2019)

Thermo-optical switching

SDU

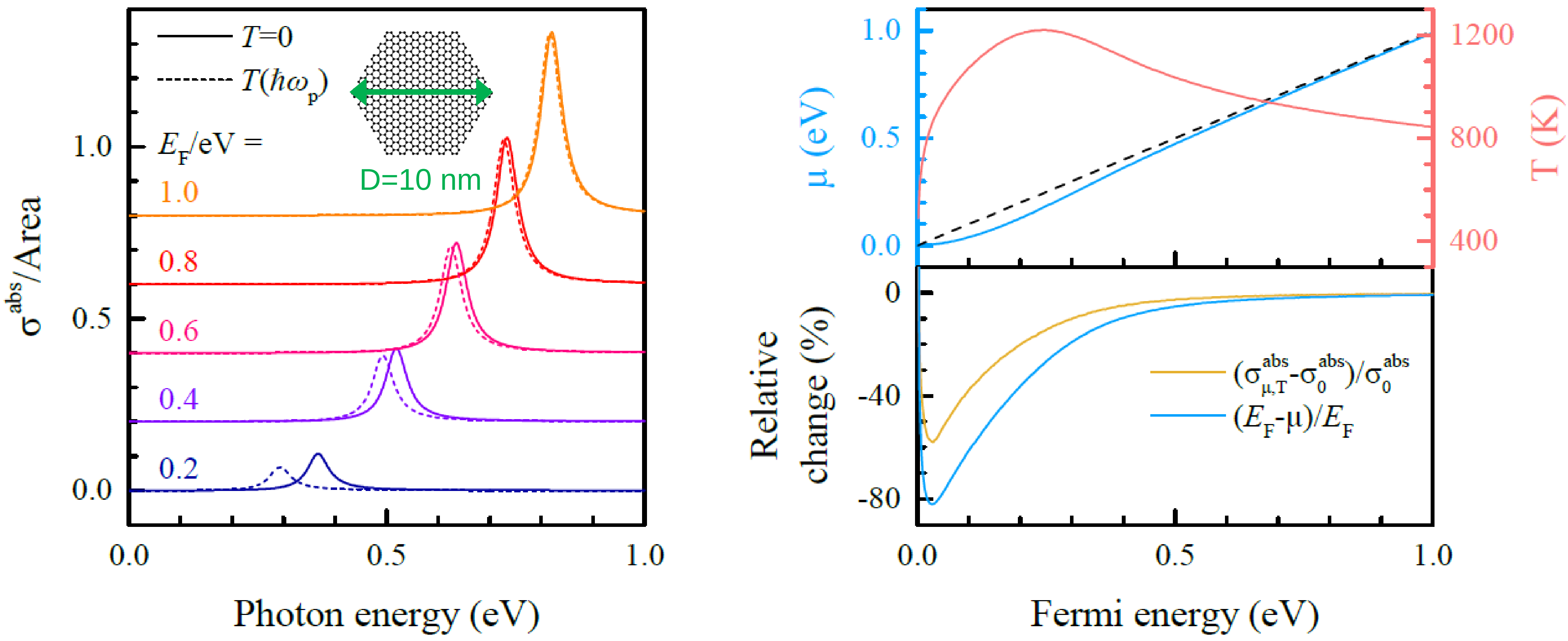




J. D. Cox and F. J. García de Abajo, Nano Lett. **19**, 3743 (2019)

Thermo-optical switching

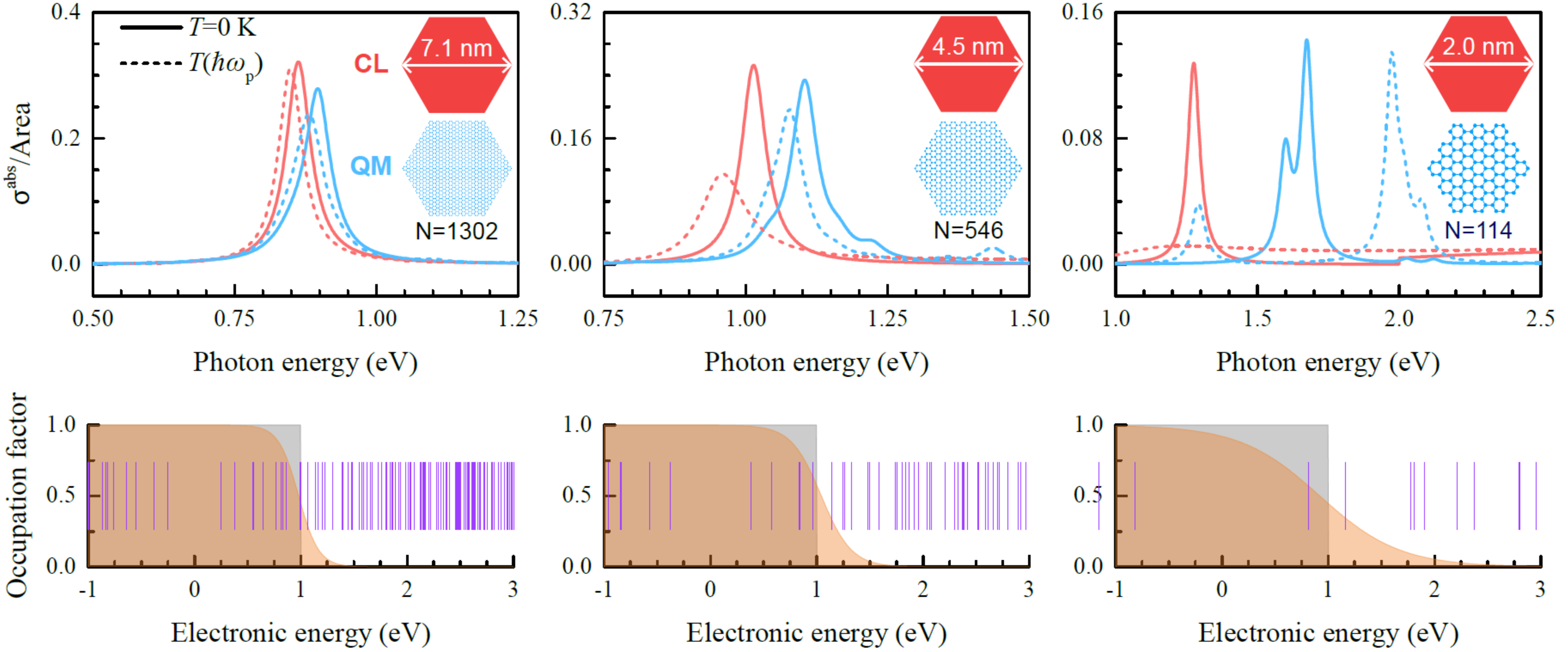
SDU



J. D. Cox and F. J. García de Abajo, Nano Lett. **19**, 3743 (2019)

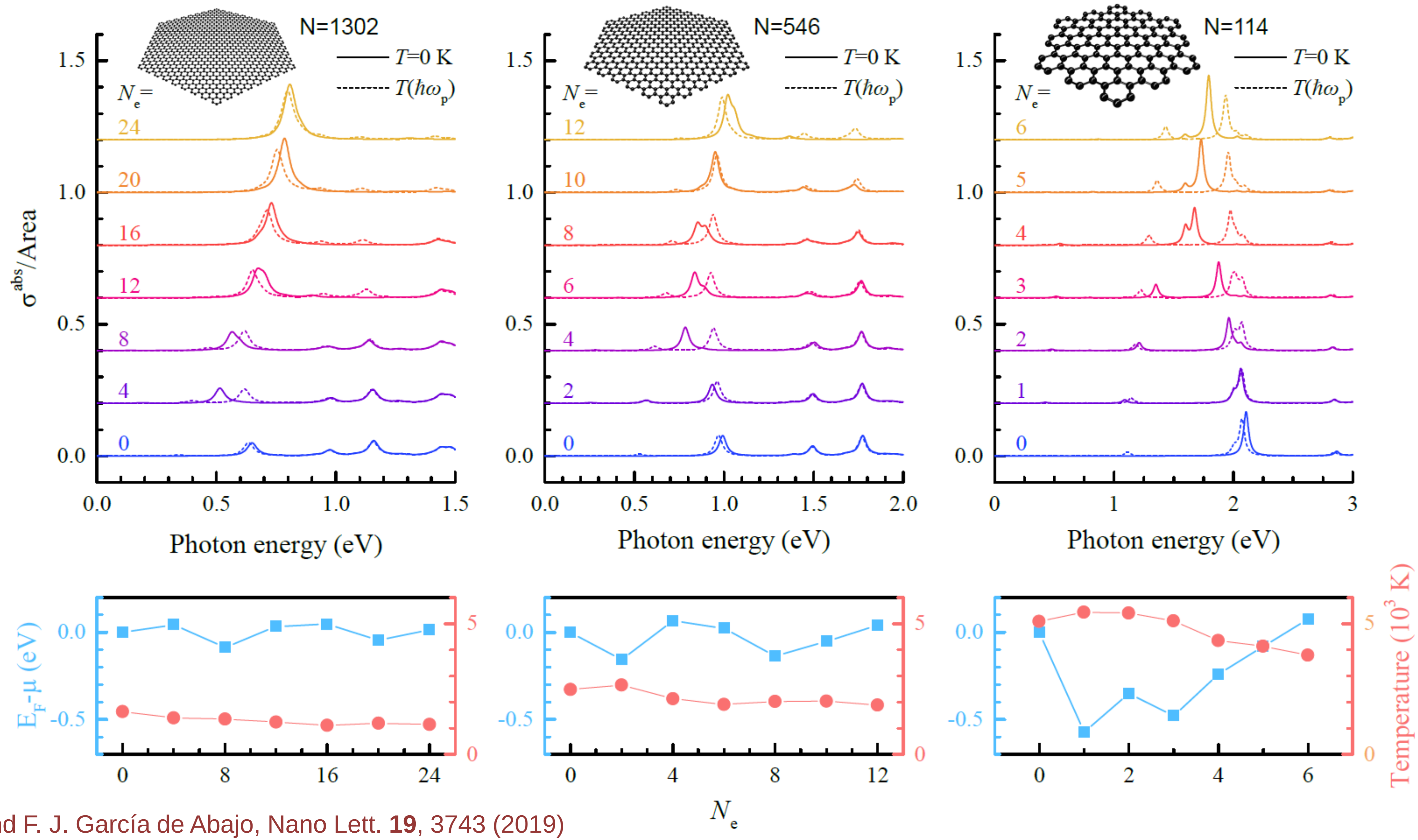
Thermo-optical switching

SDU



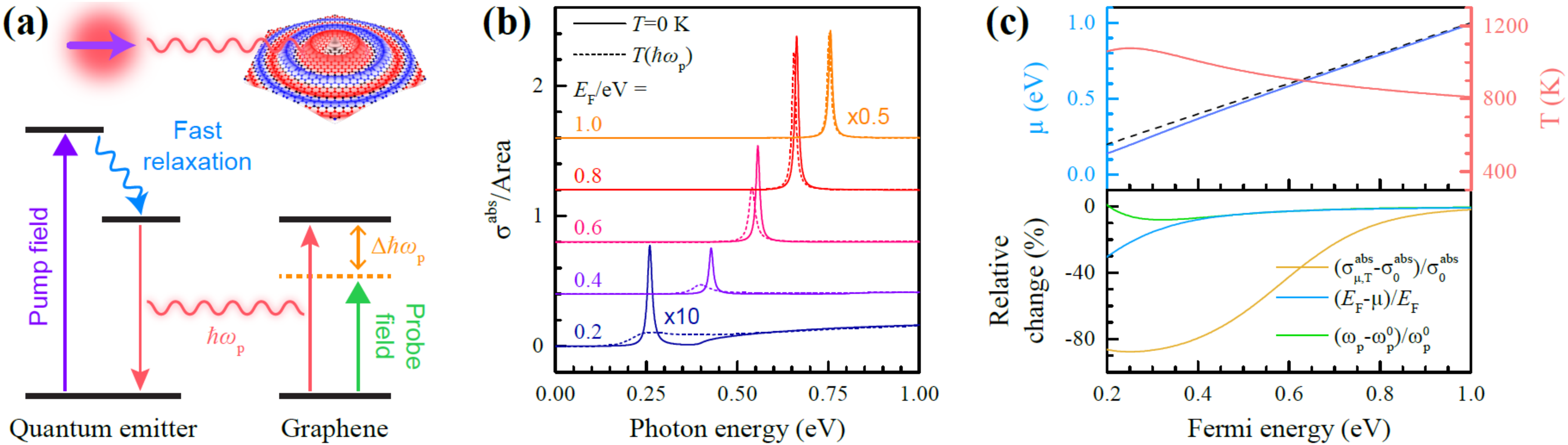
J. D. Cox and F. J. García de Abajo, Nano Lett. 19, 3743 (2019)

Thermo-optical switching



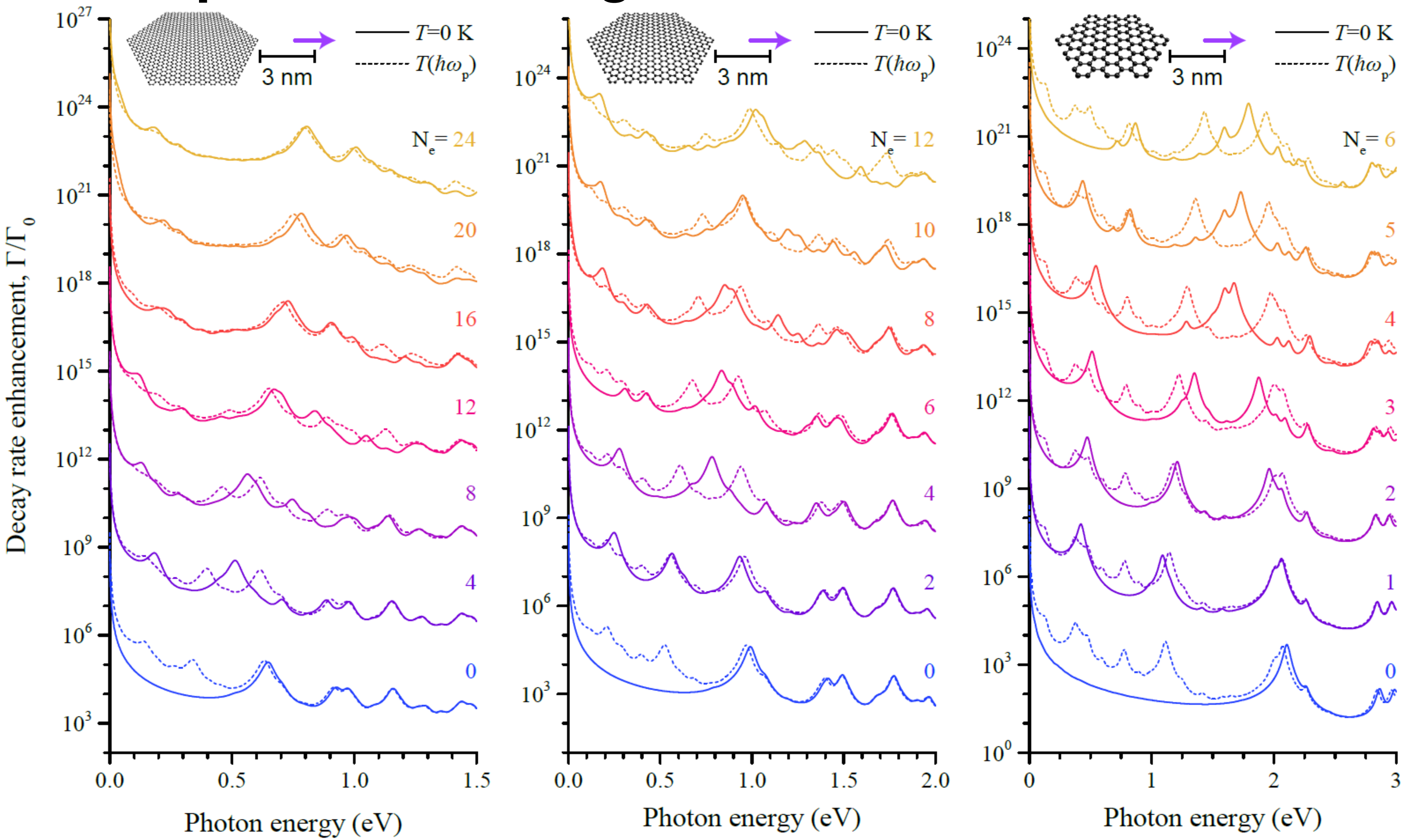
J. D. Cox and F. J. García de Abajo, Nano Lett. **19**, 3743 (2019)

Thermo-optical switching



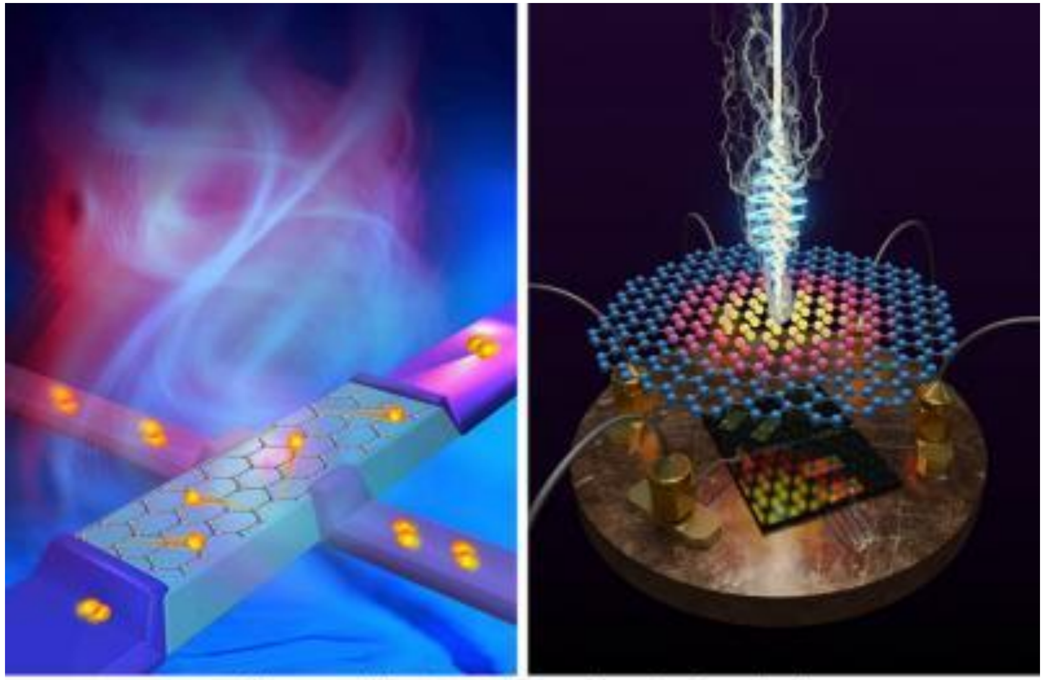
J. D. Cox and F. J. García de Abajo, Nano Lett. **19**, 3743 (2019)

Thermo-optical switching

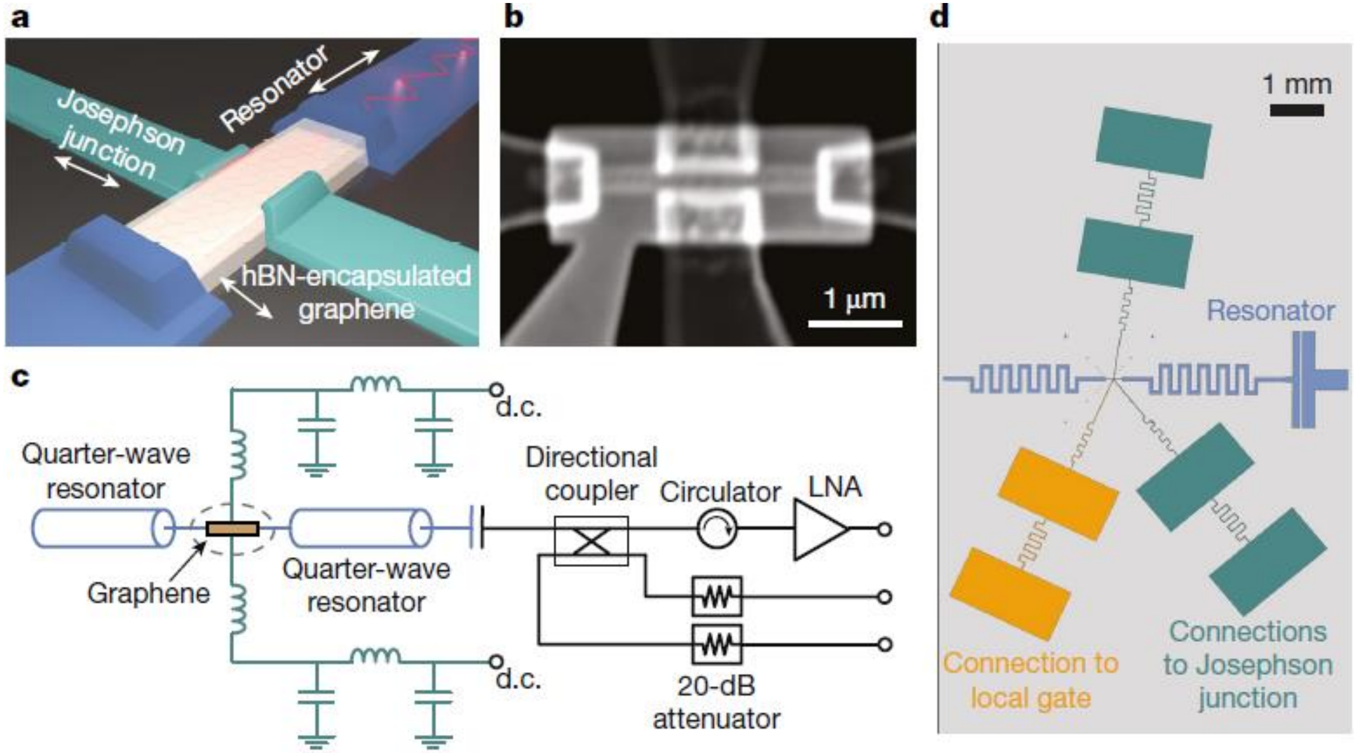


J. D. Cox and F. J. García de Abajo, Nano Lett. **19**, 3743 (2019)

Thermo-optical switching



Microwave bolometer based on graphene Josephson junction
(Credit: Graham Rowlands from Raytheon BBN Technologies, Sampson Wilcox from MIT)



Lee et al., Nature (2020)

Nonlinear interactions with electrons

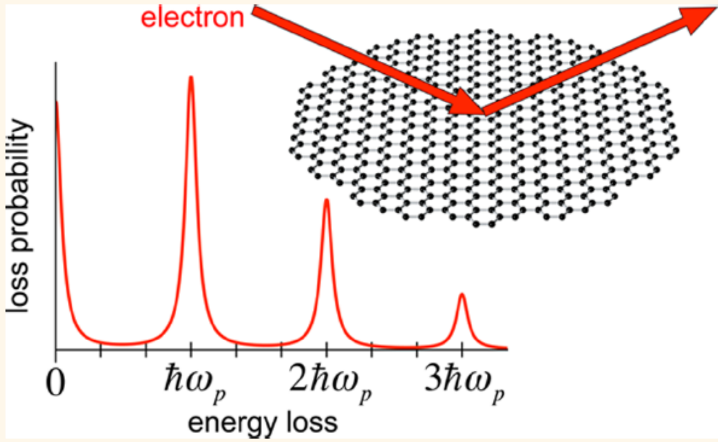
SDU

Multiple Excitation of Confined Graphene Plasmons by Single Free Electrons

F. Javier García de Abajo*

ICFO-Institut de Ciencies Fotoniques, Mediterranean Technology Park, 08860 Castelldefels, Barcelona, Spain, and ICREA-Institució Catalana de Recerca i Estudis Avançats, 08010 Ciutat Vella, Barcelona, Spain

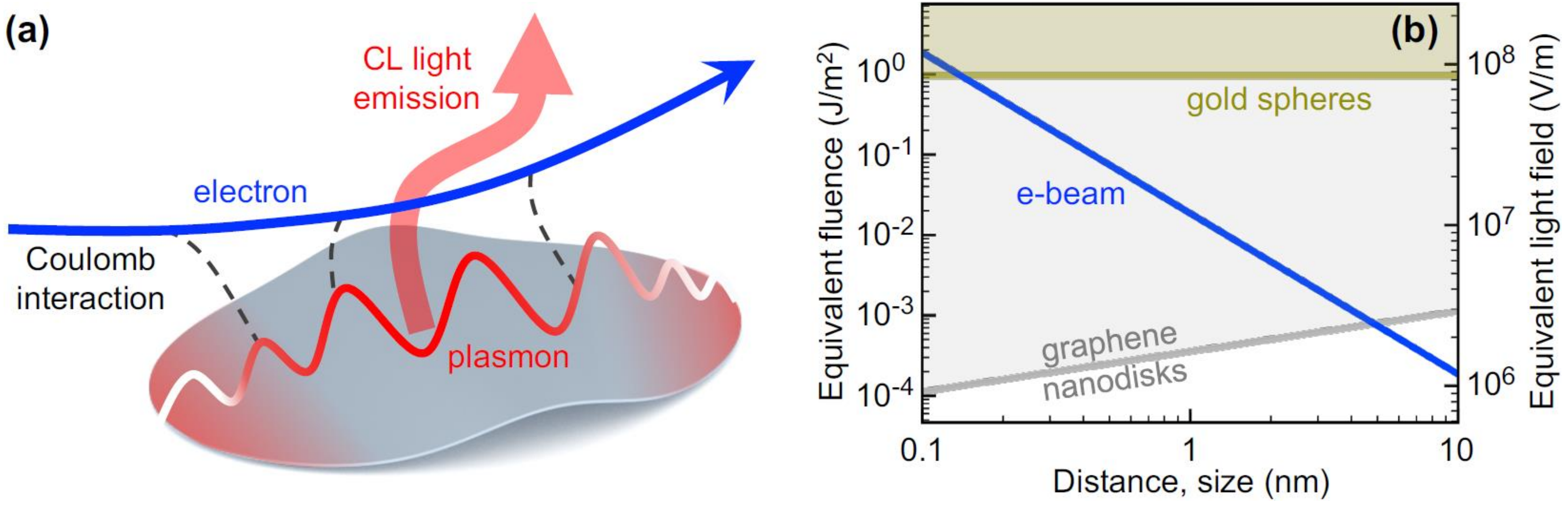
ABSTRACT We show that free electrons can efficiently excite plasmons in doped graphene with probabilities in the order of one per electron. More precisely, we predict multiple excitations of a single confined plasmon mode in graphene nanostructures. These unprecedentedly large electron-plasmon couplings are explained using a simple scaling law and further investigated through a general quantum description of the electron—plasmon interaction. From a fundamental viewpoint, multiple plasmon excitations by a single electron provide a unique platform for exploring the bosonic quantum nature of these collective modes. Not only does our study open a viable path toward multiple excitation of a single plasmon mode by a single electron, but it also reveals electron probes as ideal tools for producing, detecting, and manipulating plasmons in graphene nanostructures.



KEYWORDS: graphene · plasmons · multiple plasmon excitation · electron energy loss · quantum plasmonics · nanophotonics

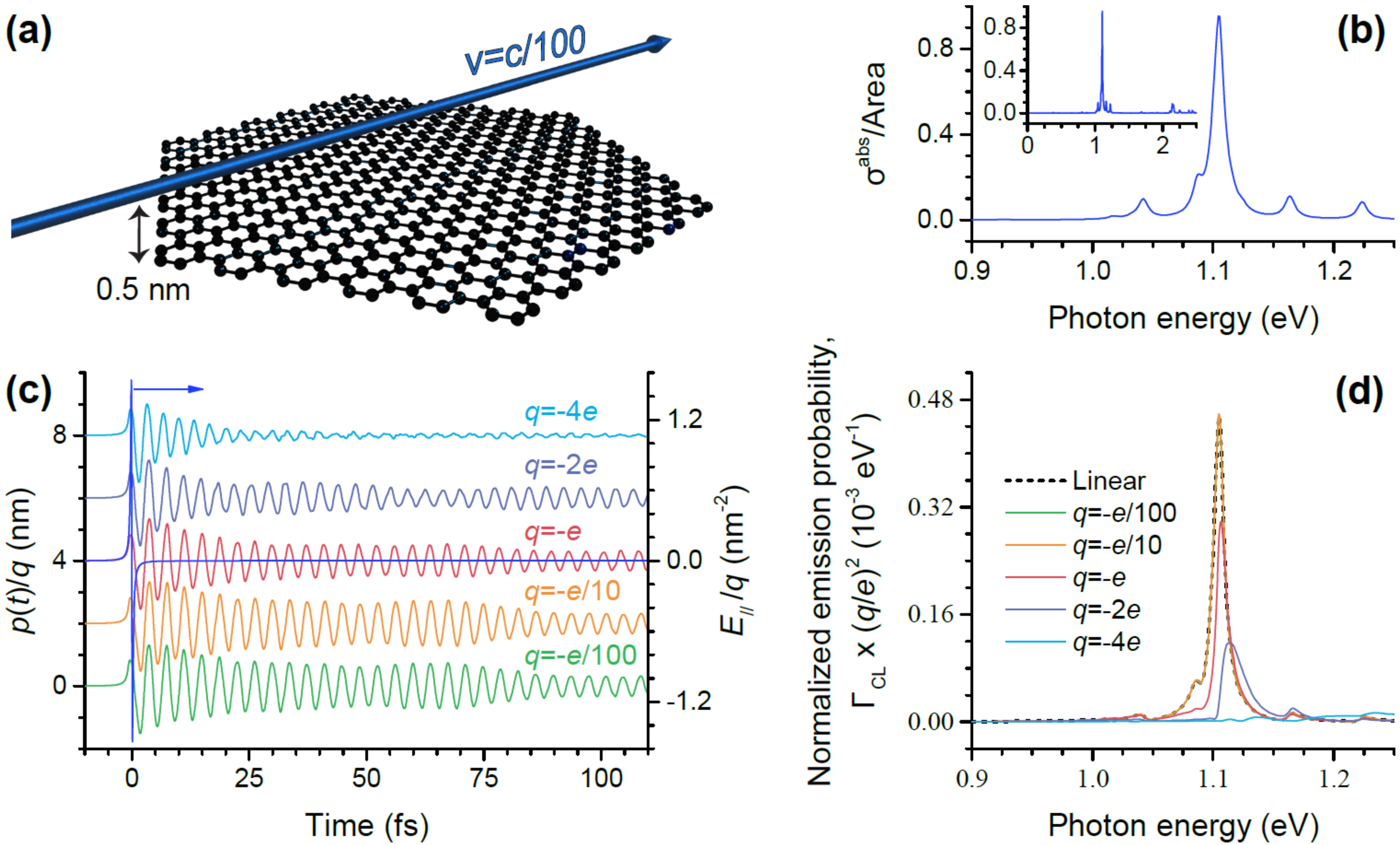
F. J. García de Abajo, ACS Nano **7**, 11409 (2013)

Estimating electron-induced nonlinearity



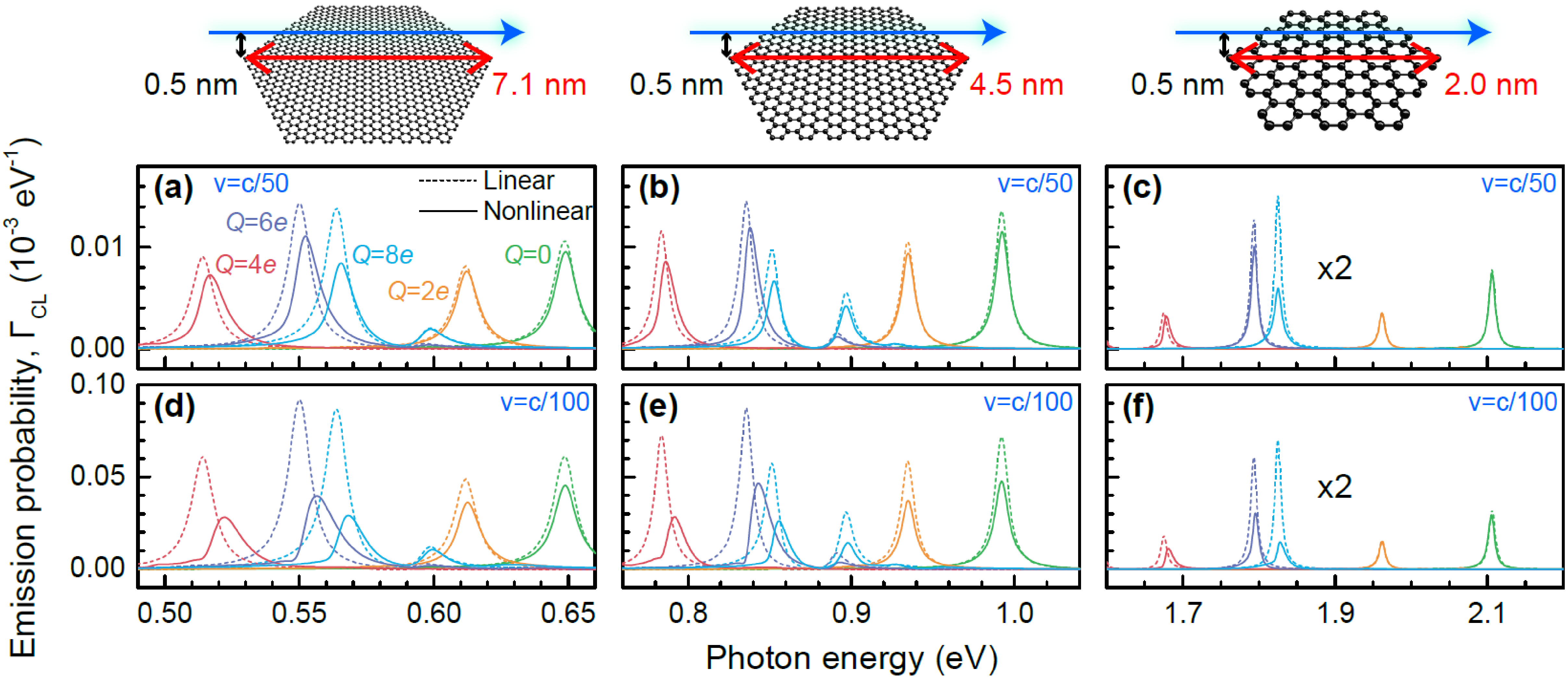
J. D. Cox and F. J. García de Abajo, Nano Lett. **20**, 4792 (2020)

Electron-induced nonlinear phenomena



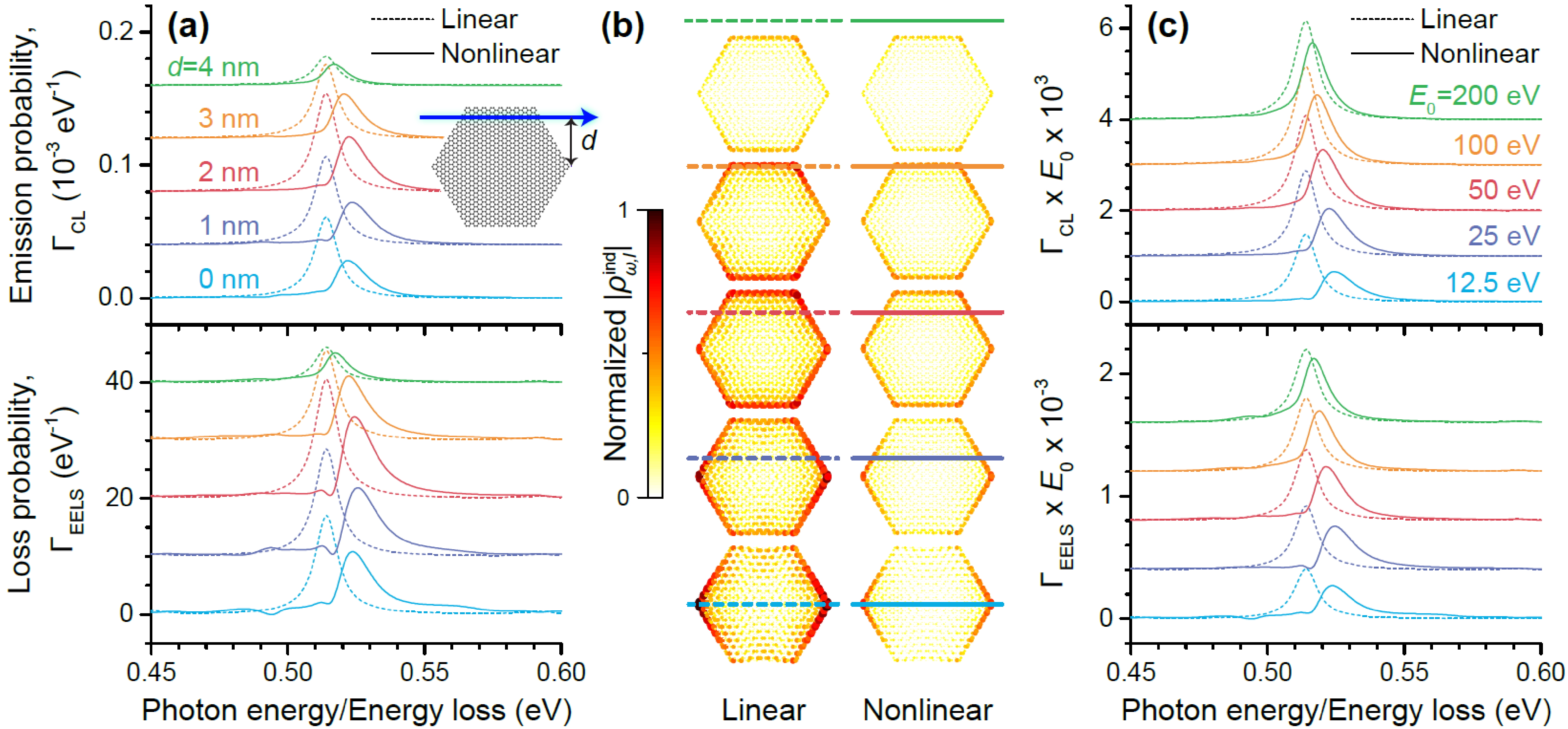
J. D. Cox and F. J. García de Abajo, Nano Lett. **20**, 4792 (2020)

Electron-induced nonlinear phenomena



J. D. Cox and F. J. García de Abajo, Nano Lett. **20**, 4792 (2020)

Electron-induced nonlinear phenomena



J. D. Cox and F. J. García de Abajo, Nano Lett. **20**, 4792 (2020)

Summary

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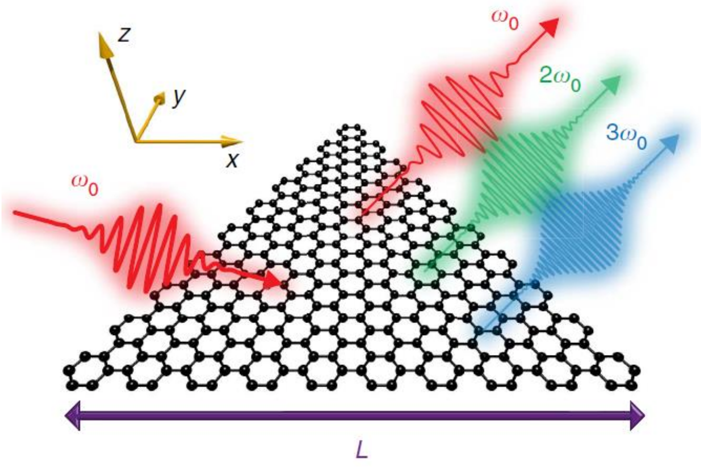
Appealing prospects for nonlinear graphene plasmonics:

- Electrical control over quantum optical states
- Single-photon-level optical nonlinearity



Further details: http://physics.sdu.dk/people/cox

J. D. Cox and F. J. García de Abajo, Acc. Chem. Res. **52**, 2536 (2019)
J. D. Cox and F. J. García de Abajo, Nano Lett. **20**, 4792 (2020)

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Fundació Privada CELLEX