Nonlinear electrodynamic phenomena in graphene

Sergey Mikhailov

University of Augsburg, Germany

International Conference “Graphene Nanophotonics”
Benasque, March 6, 2013

Graphene as a nonlinear material

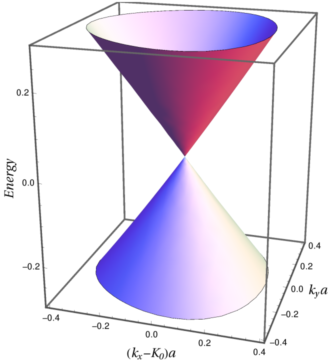
Important graphene properties

- Linear energy dispersion

$$E_{\pm}(\mathbf{p}) = \pm v_F |\mathbf{p}|$$

- Two bands (electrons and holes)
- Large Fermi velocity

$$v_F \simeq 10^8 \, cm/s$$



Outline

1. Frequency multiplication and mixing
2. Nonlinear broadening of “linear” resonances
3. Plasmon enhanced harmonics generation
4. Graphene based tunable terahertz emitter
5. Summary and Conclusions

Outline

1. Frequency multiplication and mixing
2. Nonlinear broadening of “linear” resonances
3. Plasmon enhanced harmonics generation
4. Graphene based tunable terahertz emitter
5. Summary and Conclusions

Frequency multiplication in graphene

Linear energy dispersion ⇒ nonlinear electromagnetic response:

$$\dot{p}_x = -eE\cos\omega t, \quad p_x(t) \sim -(eE/\omega)\sin\omega t$$

$$v_x = v_F \frac{p_x}{p} \sim v_F \mathrm{sgn}(\sin\omega t)$$

$$\sim v_F \frac{4}{\pi}\left\{\sin\omega t + \frac{1}{3}\sin 3\omega t + \frac{1}{5}\sin 5\omega t + \dots\right\}$$

- Higher harmonics generation $\omega \Rightarrow m\omega$
- Nonlinearity in graphene should be seen at much lower electric fields than in many other materials

Frequency multiplication in graphene

Linear energy dispersion $\Rightarrow$ nonlinear electromagnetic response:

$$\dot{p}_x = -eE\cos\omega t, \quad p_x(t) \sim -(eE/\omega)\sin\omega t$$

$$v_x = v_F\frac{p_x}{p} \sim v_F \operatorname{sgn}(\sin\omega t)$$
$$\sim v_F\frac{4}{\pi}\left\{\sin\omega t + \frac{1}{3}\sin 3\omega t + \frac{1}{5}\sin 5\omega t + \dots\right\}$$

- Higher harmonics generation $\omega \Rightarrow m\omega$
- Nonlinearity in graphene should be seen at much lower electric fields than in many other materials

Frequency multiplication in graphene

Typical nonlinear electric field?

$$v_x = v_F \frac{p_x}{\sqrt{p_x^2 + p_y^2}}, \qquad -p_F \lesssim p_y \lesssim p_F, \qquad p_F = \hbar\sqrt{\pi n_s}$$

$$\Rightarrow \frac{v_x}{v_F} = \frac{p_x(t)}{|p_y|}\left(1 - \frac{p_x^2(t)}{2|p_y^2|}\right) \sim \frac{p_x(t)}{p_F}\left(1 - \frac{p_x^2(t)}{2|p_F^2|}\right)$$

Dimensionless electric field parameter in graphene

$$\mathcal{E}_{gr} \simeq \frac{eE}{p_F|\omega + i\gamma|}$$

if $\omega \gtrsim \gamma$, $f \simeq 1$ THz and $n_s \simeq 10^{11}$ cm^{-2}, then $\mathcal{E}_{gr} \simeq 1$ if

$$E \simeq 2 \times 10^3 \text{ V/cm}$$

Conventional plasma vs. Graphene

- Graphene:

$$\mathcal{E}_{gr} \simeq \frac{eE}{p_F|\omega + i\gamma|} \qquad E_{typical} \simeq 2 \times 10^3 \text{ V/cm}$$

- Conventional 3D plasma:

$$\mathcal{E}_{par} \simeq \frac{eE}{mc|\omega + i\gamma|} \qquad E_{typical} \simeq 10^8 \text{ V/cm}$$

- Five orders of magnitude difference!
- 2nd and 3rd order effects $\propto \mathcal{E}^2$ and $\mathcal{E}^3 \Rightarrow$

Ten – fifteen orders of magnitude difference!

Frequency multiplication

External field $E(t) = E_0 \cos \omega t \Rightarrow$

- Low frequencies $\hbar\omega \ll 2|\mu|$, quasiclassical theory

$$j_{3\omega}(t) = \frac{1}{32} \frac{n_s e^2 v_F^2}{\omega|\mu|} E_0 \left(\frac{eE_0 v_F}{\omega \times |\mu|} \right)^2 \sin 3\omega t$$

- High frequencies $\hbar\omega \gg 2|\mu|$, quantum theory

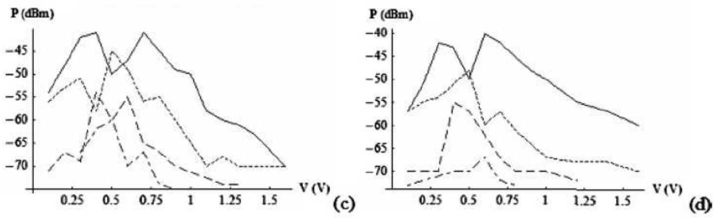
$$j_{3\omega}(t) \simeq \frac{e^2}{4\hbar} E_0 \left(\frac{eE_0 v_F}{\omega \times \hbar\omega} \right)^2 \cos(3\omega t)$$

- Harmonics amplitudes get smaller at higher frequencies
- But: interband transitions $\Rightarrow$ resonances at

$$\hbar\omega = 2|\mu|, \quad \hbar\omega = |\mu|, \quad \hbar\omega = 2|\mu|/3$$

Frequency multiplication: Microwave experiment

Dragoman et al, APL’10



Output power vs dc bias for the second to fifth harmonics of an excitation frequency (c) 3 GHz and (d) 5 GHz (coplanar waveguide over graphene monolayer). Up to 7th harmonics have been observed (frequency up to 40 GHz)

Frequency mixing

External electric field: $E_1 \cos\omega_1 t + E_2 \cos\omega_2 t$

3rd order response at $3\omega_1,\ 3\omega_2,\ 2\omega_1 \pm \omega_2,\ 2\omega_2 \pm \omega_1$

- Low frequencies $\hbar\omega_{1,2} \ll 2|\mu|$, quasiclassical theory

$$j_{(2\omega_1 \pm \omega_2)}(t) = -\frac{3}{32}\frac{n_s e^2 v_F^2}{|\mu|\omega_2} E_2 \left(\frac{e v_F E_1}{\omega_1 |\mu|}\right)^2 \sin[(\omega_2 \pm 2\omega_1)t]$$

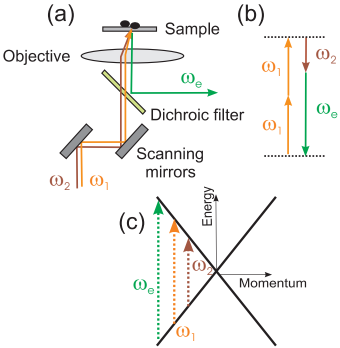
- High frequencies $\hbar\omega_{1,2} \gg |\mu|$, quantum theory

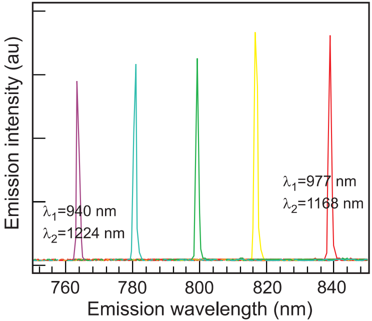
$$j_{(2\omega_1 - \omega_2)}(t) = -\frac{3}{8}\frac{e^2}{4\hbar} E_2 \left(\frac{e v_F E_1}{\omega_1 \hbar\omega_2}\right)^2 F(\omega_1, \omega_2) \cos[(2\omega_1 - \omega_2)t]$$

- Intensity dependence $I_{(2\omega_1 - \omega_2)} \propto I_{\omega_1}^2 I_{\omega_2}$
- Polarization dependence $I_{||}/I_{\perp} = 9$

Experiment: Optical frequency mixing

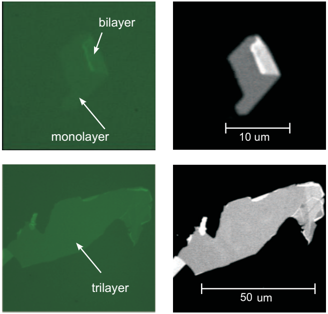
Hendry et al, PRL’10





Experiment: Optical frequency mixing

(a) Reflection (b) Four-wave mixing



Nonlinear susceptibility $\chi^{(3)}_{graphene}$:

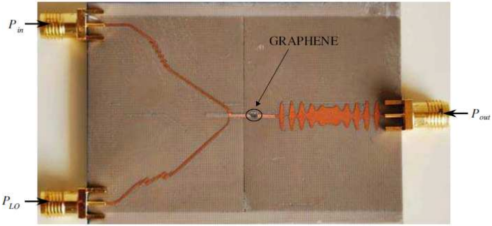
$$\chi^{(3)}_{gr} \simeq 10^{-7}\ \mathrm{esu}$$

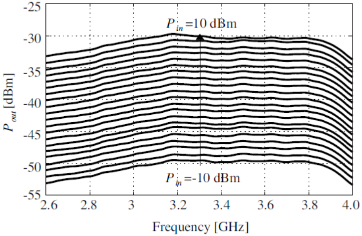
- eight orders larger than in insulators
- $\sim$ 10 times larger than in gold
- about four orders larger than in InSb

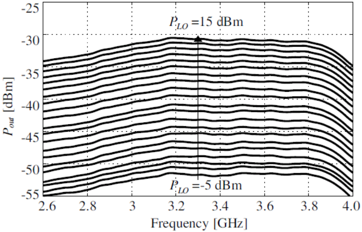
Experiment: Microwave frequency mixing

Hotopan et al, Prog. Electromagn. Res.'11

Local oscillator
$f_{LO} = 36$ GHz
$f_{RF} \simeq 39.3$ GHz







Outline

1 Frequency multiplication and mixing

2 Nonlinear broadening of “linear” resonances

3 Plasmon enhanced harmonics generation

4 Graphene based tunable terahertz emitter

5 Summary and Conclusions

Nonlinear broadening of “linear” resonances

System of particles in a weak external field F:

$$\frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial t}+\mathbf{v}_{\mathbf{p}}\frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial \mathbf{r}}+\mathbf{F}(\mathbf{r},t)\frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial \mathbf{p}}=0$$

- Conventional (perturbative) way to solve the problem:

$$f_{\mathbf{p}}(\mathbf{r},t)=f_{\mathbf{p}}^{(0)}+f_{\mathbf{p}}^{(1)}(\mathbf{r},t),\quad f^{(1)}\propto F(\mathbf{r},t)$$

$$\Rightarrow \text{if } F\propto e^{i\omega t} \text{ then } f^{(1)}\propto e^{i\omega t}$$

- Nonperturbative way gives different result!

Nonlinear broadening of “linear” resonances

System of particles in a weak external field F:

$$\frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial t} + \mathbf{v}_{\mathbf{p}} \frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial \mathbf{r}} + \mathbf{F}(\mathbf{r},t) \frac{\partial f_{\mathbf{p}}(\mathbf{r},t)}{\partial \mathbf{p}} = 0$$

- Conventional (perturbative) way to solve the problem:

$$f_{\mathbf{p}}(\mathbf{r},t) = f_{\mathbf{p}}^{(0)} + f_{\mathbf{p}}^{(1)}(\mathbf{r},t), \quad f^{(1)} \propto F(\mathbf{r},t)$$

$$\Rightarrow \text{if } F \propto e^{i\omega t} \text{ then } f^{(1)} \propto e^{i\omega t}$$

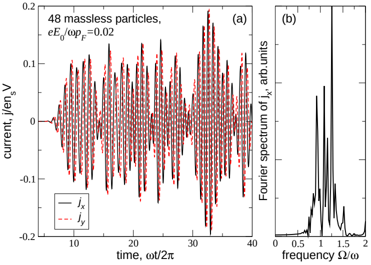
- Nonperturbative way gives different result!

Example 1: Classical cyclotron resonance

Nonperturbative way is equivalent to the solution of the system of equations

$$\dot{\mathbf{r}} = \mathbf{v} = v_F \frac{\mathbf{p}}{p}, \quad \dot{\mathbf{p}} = -e\mathbf{E}(t) - \frac{e}{c}\mathbf{v} \times \mathbf{B}$$

$\Rightarrow$ Nonlinear system, results depend on initial conditions



Example 2: Plasma oscillations

Overview: plasma waves in low-dimensional electron systems in semiconductors

Theory

- F. Stern (PRL’67 first paper on the theory of 2D plasmons)
- group of J. Quinn (PRL, PRB, 1972-..., magnetoplasmons and more, plasmons in superlattices)
- A. Fetter (Annals of Physics (NY), 1973-74, excellent textbook-type articles; PRB 1985-86, edge magnetoplasmons)
- A. V. Chaplik (Sov. Phys. JETP, about 1970-1985, plasmons in MOSFETs)
- Volkov, Mikhailov (JETP Lett, Sov. Phys. JETP, 1985-88, edge magnetoplasmons)
- Mikhailov (JETP Lett, J Phys Condens Matt, Phys Rev B, 1991-present, edge magnetoplasmons, plasmons in wires (stripes), quantum dots, antidots, rings)
- group of E. Zaremba (PRB, plasmons in dots, rings, etc)
- Books and book chapters:
  - Volkov, Mikhailov, in: "Landau Level Spectroscopy", ed. by G.Landwehr and E.I.Rashba North-Holland, Amsterdam, ch. 15 (1991).
  - Mikhailov, in Edge excitations in low-dimensional charged systems, ed. by Kirichek, Horizons in World Physics, Vol. 236, Nova Science Publisher (2000)

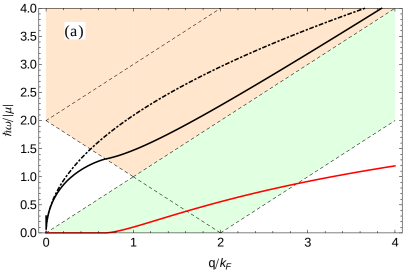
Example 2: Plasma oscillations

Experiment

- I. Grodnensky et al (JETP Lett., PRL, PRB, 1986-...; edge magnetoplasmons, plasmons in wires, dots, etc)
- group of D. Heitmann (PRB, PRL, a lot of works on bulk and edge plasmons and magnetoplasmons in wires, dots, antidots, etc; FIR transmission technique)
- group of J. Kotthaus (PRB, PRL, a lot of works on plasmons in rings, elliptic quantum dots (Claus Dahl, also theory), etc; FIR and microwave transmission technique)
- group of K. von Klitzing (PRB, PRL, also many works, microwaves, FIR, also photoresistance response)
- I. Kukushkin (PRL, PRB, JETP Lett. etc, many works on plasmons in microwave frequency range; retardation effects, proposal of edge-magnetoplasmon based frequency sensitive detector of microwave radiation - patent - company terasense.com - produces microwave cameras operating at room T at GHz-THz frequencies)

Example 2: Plasma oscillations

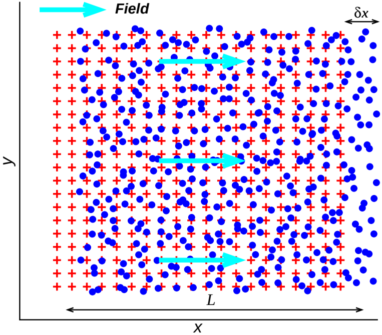
Conventional (perturbative) way (RPA, Wunsch'06; Hwang'07):



At low q and ω:

$$\omega_p^2(q) = \frac{2\pi n_s e^2}{m^\star} q, \quad m^\star = \frac{p_F}{v_F}, \quad \text{no damping}$$

Example 2: Plasma oscillations



$$m\frac{d^2(\delta x)}{dt^2} = -eE_x \sim -e\frac{en_s\delta x}{L\kappa} \quad \Rightarrow \quad \boxed{\omega_{p2}^2(q) = \frac{2\pi n_s e^2}{m\kappa}q}$$

Example 2: Plasma oscillations

Plasma waves as oscillations of particles in a parabolic potential $U(x) = Kx^2/2$, $K \propto$ background density

- Parabolic dispersion:

$$\dot{\mathbf{r}} = \frac{\mathbf{p}}{m}, \quad \dot{\mathbf{p}} = -Kx\mathbf{e}_x$$

$$\Rightarrow \quad \ddot{\mathbf{r}} = -\frac{K}{m}\mathbf{r}$$

- Linear dispersion:

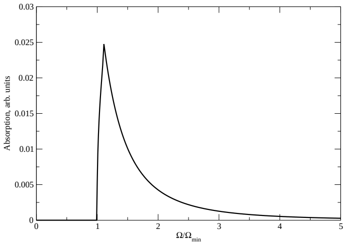
$$\dot{\mathbf{r}} = v_F\frac{\mathbf{p}}{p}, \quad \dot{\mathbf{p}} = -Kx\mathbf{e}_x$$

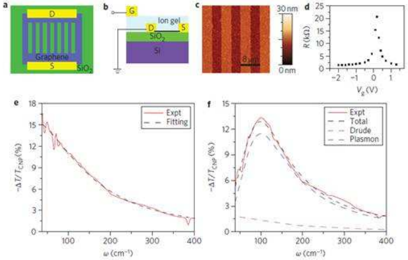
$\Rightarrow$ Oscillation frequency depends on initial conditions

Example 2: Plasma oscillations

⇒ Nonperturbative method gives a finite linewidth at

$$T = 0,\ c = \infty \text{ and } \gamma = 0$$





Experiment: Plasmon line asymmetric, plasmon frequency $\simeq$ 3 THz, linewidth $\simeq$ 4 THz

Example 2: Plasma oscillations

The difference is:

$$\frac{\partial f}{\partial t} + \frac{\mathbf{p}}{m}\frac{\partial f}{\partial \mathbf{r}} + \mathbf{F}\frac{\partial f}{\partial \mathbf{p}} = 0$$

$$\frac{\partial f}{\partial t} + v_F\frac{\mathbf{p}}{p}\frac{\partial f}{\partial \mathbf{r}} + \mathbf{F}\frac{\partial f}{\partial \mathbf{p}} = 0$$

- Nonanalitical term $v_F\mathbf{p}/p$ in the kinetic equation for graphene!
- The response problem should be solved non-perturbatively even at $F \to 0$

Example 2: Plasma oscillations

The difference is:

$$\frac{\partial f}{\partial t} + \frac{\mathbf{p}}{m}\frac{\partial f}{\partial \mathbf{r}} + \mathbf{F}\frac{\partial f}{\partial \mathbf{p}} = 0$$

$$\frac{\partial f}{\partial t} + v_F\frac{\mathbf{p}}{p}\frac{\partial f}{\partial \mathbf{r}} + \mathbf{F}\frac{\partial f}{\partial \mathbf{p}} = 0$$

- Nonanalitical term $v_F\mathbf{p}/p$ in the kinetic equation for graphene!
- The response problem should be solved non-perturbatively even at $F \to 0$

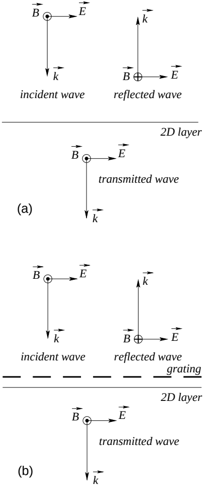
Outline

1. Frequency multiplication and mixing
2. Nonlinear broadening of “linear” resonances
3. Plasmon enhanced harmonics generation
4. Graphene based tunable terahertz emitter
5. Summary and Conclusions

Nonlinearity and plasma resonance

- Normal incidence of radiation, uniform electric field, $\mathbf{q} = \mathbf{0} \Rightarrow$ only 3rd (5th,...) order effects can be observed
- 2D layer with grating or array of stripes, $\mathbf{q} \neq \mathbf{0}$

  $\Rightarrow$ 2nd order effects can be observed too
- In addition, plasma resonances can be excited



1st and 2nd order polarizability

External field $\phi(\mathbf{r},t) = \phi_{\mathbf{q}\omega} e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + c.c.$

$$\Rightarrow \quad \rho(\mathbf{r},t) = \rho_{\mathbf{q}\omega} e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + \rho_{2\mathbf{q},2\omega} e^{2i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + c.c.$$

$$\rho_{\mathbf{q}\omega} = \alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} \phi_{\mathbf{q}\omega}, \quad \rho_{2\mathbf{q},2\omega} = \alpha^{(2)}_{2\mathbf{q},2\omega;\mathbf{q}\omega,\mathbf{q}\omega} \phi_{\mathbf{q}\omega} \phi_{\mathbf{q}\omega}$$

Polarizability: Semiconductors vs Graphene

- Conventional 2D electron system (parabolic spectrum):

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} \approx \frac{n_s e^2 q^2}{m\omega^2} \qquad \alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} \approx -\frac{3n_s e^3 q^4}{2m^2\omega^4}$$

- Graphene (linear spectrum):

$$\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega} = \frac{e^2 g_s g_v q^2 T}{2\pi\hbar^2\omega^2} \ln\left(2\cosh\frac{\mu}{2T}\right)$$

$$\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega} = -\frac{3e^3 g_s g_v q^4 v_F^2}{32\pi\hbar^2\omega^4} \tanh\frac{\mu}{2T}$$

Self-consistent screening and nonlinear response

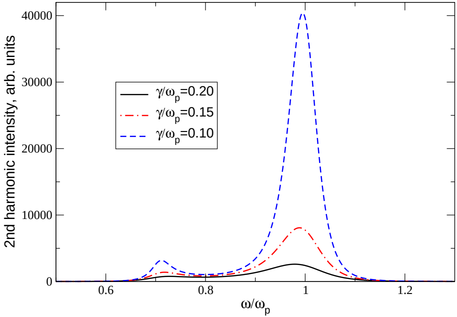
- First order

$$\phi^{tot}_{\mathbf{q}\omega} = \frac{\phi^{ext}_{\mathbf{q}\omega}}{\epsilon(\mathbf{q},\omega)}, \quad \epsilon(\mathbf{q},\omega) = 1 - \frac{2\pi}{q}\alpha^{(1)}_{\mathbf{q}\omega;\mathbf{q}\omega}$$

- Second order

$$\phi^{tot}_{2\mathbf{q}2\omega} = \frac{\pi}{q}\frac{\alpha^{(2)}_{2\mathbf{q}2\omega;\mathbf{q}\omega,\mathbf{q}\omega}}{\epsilon(2\mathbf{q},2\omega)\left[\epsilon(\mathbf{q},\omega)\right]^2}\phi^{ext}_{\mathbf{q}\omega}\phi^{ext}_{\mathbf{q}\omega}$$

Plasmon enhancement of 2nd harmonic



huge resonance at $\omega \simeq \omega_p(q)$, weak at $\omega \simeq \omega_p(q)/\sqrt{2}$

$$\frac{\alpha^{(2)}_{graphene}}{\alpha^{(2)}_{semicond}} = \frac{(v_F^2)_{graphene}}{2(v_F^2)_{semicond}} \approx 10-30 \qquad \frac{I^{graphene}_{tot}}{I^{GaAs}_{tot}} \simeq \left(\frac{\alpha^{(2)}_{graphene}}{\alpha^{(2)}_{GaAs}}\right)^2 \simeq 100-900.$$

Outline

1 Frequency multiplication and mixing

2 Nonlinear broadening of “linear” resonances

3 Plasmon enhanced harmonics generation

4 Graphene based tunable terahertz emitter

5 Summary and Conclusions

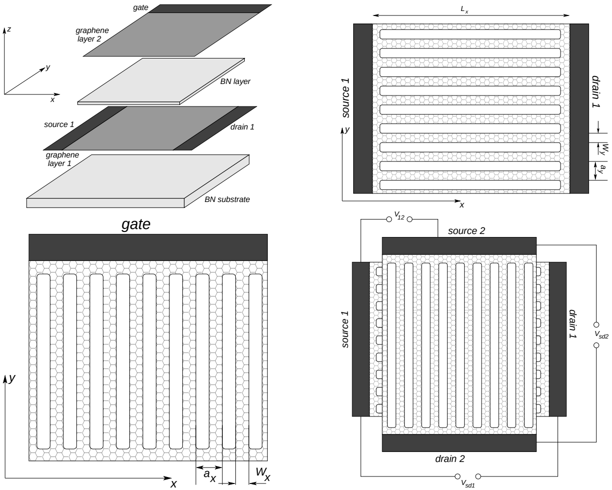
Graphene based terahertz emitter

- Fermi velocity in graphene $v_F = 10^8$ cm/s
- if to force electrons to move with the drift velocity $v_{dr} \simeq v_F = 10^8$ cm/s in a periodic potential with the period $a \simeq 1 - 0.1$ μm they should emit electromagnetic waves with the frequency

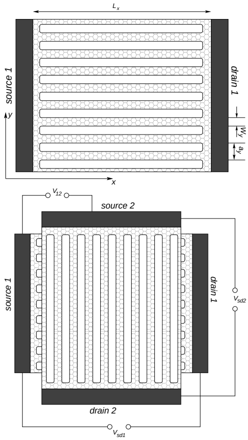
$$f = \frac{v_{dr}}{a} \simeq 1 - 10 \text{ THz}$$

- $\Rightarrow$ solid-state microscopic tunable free electron laser!

Graphene free electron laser



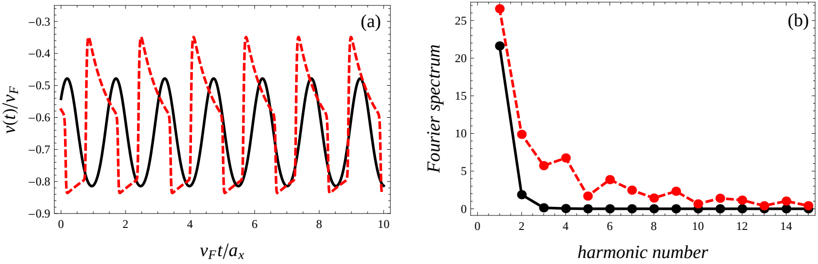
Graphene free electron laser: Estimates



- Estimated frequency: $1 - 30$ THz (including harmonics)
- Room temperature operation
- Radiation power $\simeq 0.5$ W/cm^2
- Efficiency $\simeq 1\%$

Graphene free electron laser: Estimates

- Distance graphene – grating can be only a few monolayers
  - electrons move in a step-like potential (non-sinusoidal)
  - higher harmonics



$f_1 \simeq 0.6 - 3$ THz for $a_x \simeq 1 - 0.2\ \mu$m

Graphene based THz emitter

- voltage-tunable
- broad frequency range at THz
- operating around room temperature
- high power
- high efficiency
- almost transparent
- bendable (power concentration)

Outline

1. Frequency multiplication and mixing
2. Nonlinear broadening of “linear” resonances
3. Plasmon enhanced harmonics generation
4. Graphene based tunable terahertz emitter
5. Summary and Conclusions

Summary and Conclusions

- Higher harmonics generation
- Frequency mixing effects
- Nonlinear broadening of conventional resonances (cyclotron, plasma)
- Plasmon enhanced second harmonic
- Tunable THz emitter

Graphene:

- A lot of interesting nonlinear physics
- A lot of possible electronics and optoelectronic applications

References

- Frequency multiplication
  - SM, EPL **79**, 27002 (2007)
  - SM & K. Ziegler, J Phys Condens Matter **20**, 384204 (2008)
- Frequency mixing
  - Hendry et al, Phas Rev Lett **105**, 097401 (2010)
  - SM, in *Physics and Applications of Graphene - Theory*, Ch. 25 (InTech, Rijeka, Croatia, 2011)
  - SM, Physica E **44**, 924 (2012)
- Nonlinear broadening of resonances
  - SM, Phys Rev B **79**, 241309 (2009)
  - SM & D. Beba, New J Phys **14**, 115024 (2012)
- Plasmon enhanced harmonics generation
  - SM, Phys Rev B **84**, 045432 (2011)
  - SM, Ann Phys (Berlin) **524**, 182 (2012)
- Graphene based terahertz emitter
  - SM, arXiv 1203.3983; Phys Rev B (2013), accepted

Thanks

- **Support:**
  - Deutsche Forschungsgemeinschaft
  - European Community’s 7th Framework Programme, Grant No. 265114

- **THANK YOU FOR YOUR ATTENTION**

References

- Frequency multiplication
  - SM, EPL **79**, 27002 (2007)
  - SM & K. Ziegler, J Phys Condens Matter **20**, 384204 (2008)
- Frequency mixing
  - Hendry et al, Phys Rev Lett **105**, 097401 (2010)
  - SM, in *Physics and Applications of Graphene - Theory*, Ch. 25 (InTech, Rijeka, Croatia, 2011)
  - SM, Physica E **44**, 924 (2012)
- Nonlinear broadening of resonances
  - SM, Phys Rev B **79**, 241309 (2009)
  - SM & D. Beba, New J Phys **14**, 115024 (2012)
- Plasmon enhanced harmonics generation
  - SM, Phys Rev B **84**, 045432 (2011)
  - SM, Ann Phys (Berlin) **524**, 182 (2012)
- Graphene based terahertz emitter
  - SM, arXiv 1203.3983; Phys Rev B (2013), accepted