Nonlinear electrodynamic phenomena in graphene

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International Conference “Graphene Nanophotonics”
Benasque, March 6, 2013
Graphene as a nonlinear material

Important graphene properties

- Linear energy dispersion
  \[ E_{\pm}(p) = \pm v_F|p| \]
- Two bands (electrons and holes)
- Large Fermi velocity
  \[ v_F \sim 10^8 \text{ cm/s} \]
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
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Linear energy dispersion $\Rightarrow$ nonlinear electromagnetic response:

$$\begin{align*}
\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 \text{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 + \ldots \right\}
\end{align*}$$

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

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\]

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?

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

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

Dimensionless electric field parameter in graphene

\[ \mathcal{E}_{gr} \sim \frac{eE}{p_F|\omega + i\gamma|} \]

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

\[ E \sim 2 \times 10^3 \text{ V/cm} \]
Conventional plasma vs. Graphene

**Graphene:**

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

**Conventional 3D plasma:**

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

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 and mixing
Nonlinear broadening of “linear” resonances
Plasmon enhanced harmonics generation
Graphene

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 \( I_\parallel / I_\perp = 9 \)
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Experiment: Optical frequency mixing

Hendry et al, PRL’10

(a) Sample
Objective
Dichroic filter
Scanning mirrors

(b) \( \omega_1 \) \( \omega_2 \) \( \omega_e \)

(c) Energy vs. Momentum
\( \omega_1 \) \( \omega_2 \) \( \omega_e \)

Emission spectrum:
- \( \lambda_1 = 977 \text{ nm} \)
- \( \lambda_2 = 1168 \text{ nm} \)
- \( \lambda_3 = 940 \text{ nm} \)
- \( \lambda_4 = 1224 \text{ nm} \)

Emission wavelength (nm)
Emission intensity (au)
Experiment: Optical frequency mixing

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

$$\chi^{(3)}_{\text{gr}} \approx 10^{-7} \text{ esu}$$

- eight orders larger than in insulators
- $\sim 10$ times larger than in gold
- about four orders larger than in InSb
**Experiment: Microwave frequency mixing**


Local oscillator

\[ f_{LO} = 36 \text{ GHz} \]

\[ f_{RF} \simeq 39.3 \text{ GHz} \]
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Nonlinear broadening of “linear” resonances

System of particles in a weak external field $F$:

$$\frac{\partial f_p(r, t)}{\partial t} + v_p \frac{\partial f_p(r, t)}{\partial r} + F(r, t) \frac{\partial f_p(r, t)}{\partial p} = 0$$

- Conventional (perturbative) way to solve the problem:

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

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

- Nonperturbative way gives different result!
Nonlinear broadening of “linear” resonances

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- Nonperturbative way gives different result!
Nonperturbative way is equivalent to the solution of the system of equations

\[ \dot{r} = v = v_F \frac{p}{m}, \quad \dot{p} = -eE(t) - \frac{e}{c} v \times B \]

⇒ 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. V. Chaplik (Sov. Phys. JETP, about 1970-1985, plasmons in MOSFETs)
- Group of E. Zaremba (PRB, plasmons in dots, rings, etc)

Books and book chapters:

Example 2: Plasma oscillations

Experiment

- 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):

\[ \omega_p^2(q) = \frac{2\pi n_s e^2}{m^*} q, \quad m^* = \frac{\rho_F}{v_F}, \quad \text{no damping} \]
Example 2: Plasma oscillations

\[ m \frac{d^2(\delta x)}{dt^2} = -eE_x \sim -e \frac{en_s \delta x}{L\kappa} \Rightarrow \]

\[ \omega_{p2}(q) = \frac{2\pi n_s e^2}{m\kappa} q \]
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:

$$\dot{r} = \frac{p}{m}, \quad \dot{p} = -Kxe_x$$

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

- Linear dispersion:

$$\dot{r} = v_F \frac{p}{p}, \quad \dot{p} = -Kxe_x$$

$$\Rightarrow$$ 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:

\[ \frac{\partial f}{\partial t} + \frac{p}{m} \frac{\partial f}{\partial r} + F \frac{\partial f}{\partial p} = 0 \]

\[ \frac{\partial f}{\partial t} + v_F p \frac{\partial f}{\partial r} + F \frac{\partial f}{\partial p} = 0 \]

いますが非線形的成分 \( v_F p/p \) をグラフェンの動的方程式に含まれる。

逆応答問題は、\( F \to 0 \) について非perturbatively 解決しなければならない。
Example 2: Plasma oscillations

The difference is:

\[ \frac{\partial f}{\partial t} + \frac{p}{m} \frac{\partial f}{\partial r} + F \frac{\partial f}{\partial p} = 0 \]

\[ \frac{\partial f}{\partial t} + \nu_F \frac{p}{p} \frac{\partial f}{\partial r} + F \frac{\partial f}{\partial p} = 0 \]

🔹 Nonanlitical term $\nu_F p/p$ in the kinetic equation for graphene!

🔹 The response problem should be solved non-perturbatively even at $F \to 0$
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Nonlinearity and plasma resonance

- Normal incidence of radiation, uniform electric field, $q = 0 \Rightarrow$ only 3rd (5th,...) order effects can be observed
- 2D layer with grating or array of stripes, $q \neq 0$
  $\Rightarrow$ 2nd order effects can be observed too
- In addition, plasma resonances can be excited
1st and 2nd order polarizability

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

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

$\rho_{q\omega} = \alpha^{(1)}_{q\omega; q\omega} \phi_{q\omega}$, $\rho_{2q,2\omega} = \alpha^{(2)}_{2q,2\omega; q\omega,q\omega} \phi_{q\omega} \phi_{q\omega}$
Polarizability: Semiconductors vs Graphene

• Conventional 2D electron system (parabolic spectrum):

\[ \alpha_{q\omega; q\omega}^{(1)} \approx \frac{n_s e^2 q^2}{m \omega^2} \quad \alpha_{2q2\omega; q\omega, q\omega}^{(2)} \approx -\frac{3n_s e^3 q^4}{2m^2 \omega^4} \]

• Graphene (linear spectrum):

\[ \alpha_{q\omega; q\omega}^{(1)} = \frac{e^2 g_s g_v q^2 T}{2\pi \hbar^2 \omega^2} \ln \left( 2 \cosh \frac{\mu}{2T} \right) \]

\[ \alpha_{2q2\omega; q\omega, q\omega}^{(2)} = -\frac{3e^3 g_s g_v q^4 v_F^2}{32\pi \hbar^2 \omega^4} \tanh \frac{\mu}{2T} \]
Self-consistent screening and nonlinear response

- First order

\[ \phi_{q\omega}^{tot} = \frac{\phi_{q\omega}^{ext}}{\epsilon(q, \omega)}, \quad \epsilon(q, \omega) = 1 - \frac{2\pi}{q} \alpha^{(1)}_{q\omega; q\omega} \]

- Second order

\[ \phi_{2q2\omega}^{tot} = \frac{\pi}{q} \frac{\alpha^{(2)}_{2q2\omega; q\omega, q\omega}}{\epsilon(2q, 2\omega) [\epsilon(q, \omega)]^2} \phi_{q\omega}^{ext} \phi_{q\omega}^{ext} \]
Plasmon enhancement of 2nd harmonic

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

$$\frac{\alpha^{(2)}_{\text{graphene}}}{\alpha^{(2)}_{\text{semicond}}} = \frac{(v_F^2)_{\text{graphene}}}{2(v_F^2)_{\text{semicond}}} \approx 10 - 30$$

$$\frac{I_{\text{graphene}}^{\text{tot}}}{I_{\text{GaAs}}^{\text{tot}}} \approx \left( \frac{\alpha^{(2)}_{\text{graphene}}}{\alpha^{(2)}_{\text{GaAs}}} \right)^2 \approx 100 - 900.$$
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Graphene based terahertz emitter

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

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

- $\Rightarrow$ 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 $\sim 0.5$ W/cm$^2$
- Efficiency $\sim 1\%$
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

\[ f_1 \approx 0.6 - 3 \text{ THz for } a_x \approx 1 - 0.2 \mu\text{m} \]
Graphene based THz emitter

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

- Frequency mixing
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- Nonlinear broadening of resonances

- Plasmon enhanced harmonics generation
  - SM, Phys Rev B 84, 045432 (2011)

- 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
Frequency multiplication

Frequency mixing

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- SM, Phys Rev B 84, 045432 (2011)

Graphene based terahertz emitter
- SM, arXiv 1203.3983; Phys Rev B (2013), accepted