Surface Plasmon Slowing Down and Cherenkov-Type THz Emission in Graphene Based Structure

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Introduction

Owing to that, **nanosized** elementary circuits components, **nanoscale** integration, and **nanostructured** electromagnetic materials are on demand.

Which are current technological trends in applied electromagnetics?

- Miniaturization of electric circuits & components …
- Energy consumption dropping …
- Opening up the mm-Wave & THz frequency ranges

Owing to that, **nanosized** elementary circuits components, **nanoscale** integration, and **nanostructured** electromagnetic materials are on demand.

Extra-small EM radiation sources are among them.
The Cherenkov radiation is governed by the synchronization condition \( w - ku = 0 \), where \( k \) is the wave vector and \( u \) is the electron velocity.

- an electron gun,
- a focusing structure,
- a slowing-down system,
- an electron collector

S. Maksimenko, INP BSU
Can one provide the generation conditions on nanoscale?

- Ballisticity of electron motion on the proper distance
- Slowing down of EM wave to satisfy the synchronism condition with electron beam
- Large enough current density in a nanoobject
Graphene & Carbon Nanotube

\[ R_c = ma_1 + na_2 \]

SWCNT \((m,n)\)

- **Length:** 1-10 mkm
- **Diameter:** 1-3 nm
- **Conductivity type:** metallic or semiconductor

\((m,0)\) - zigzag, 
\((m,m)\) - armchair
### Graphene & CNTs: Physical properties

<table>
<thead>
<tr>
<th>Property</th>
<th>Si</th>
<th>Cu</th>
<th>SWCNT</th>
<th>MWCNT</th>
<th>Graphene or GNR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max current density (A/cm²)</td>
<td>-</td>
<td>10⁷</td>
<td>&gt; 1x10⁹</td>
<td>&gt; 1x10⁹</td>
<td>&gt; 1x10⁸ Novoselov, et al., Science, 2001</td>
</tr>
<tr>
<td>Melting point (K)</td>
<td>1687</td>
<td>1356</td>
<td>3800 (graphite)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tensile strength (GPa)</td>
<td>7</td>
<td>0.22</td>
<td>22.2 ± 2.2</td>
<td>11-63</td>
<td></td>
</tr>
<tr>
<td>Mobility (cm²/V-s)</td>
<td>1400</td>
<td></td>
<td>&gt; 10000</td>
<td>&gt; 10000</td>
<td></td>
</tr>
<tr>
<td>Thermal conductivity (×10³ W/m-K)</td>
<td>0.15</td>
<td>0.385</td>
<td>1.75-5.8</td>
<td>3.0</td>
<td>3.0-5.0 Balandin, et al., Nano Let., 2008</td>
</tr>
<tr>
<td>Mean free path (nm) @ room temp.</td>
<td>30</td>
<td>40</td>
<td>&gt; 1,000</td>
<td>25,000</td>
<td>~1,000 Bolotin, et al., Phys. Rev. Let., 2008</td>
</tr>
</tbody>
</table>
Dynamical conductivity of CNT

Radial dependence of the conductivity below and in the optical transitions band

$R_h = \frac{\sqrt{3}}{2\pi} d \sqrt{m^2 + mn + n^2}$ where $d = 1.42\text{Å}$ is the interatomic distance in graphene.
Solution of the conductivity problem accounting for the spatial confinement effects couples classical electrodynamics and physics of nanostructures.
Surface Wave in CNTs

The problem statement:
consider the propagation of surface waves along an isolated, infinitely long CNT in vacuum. The CNT conductivity is assumed to be axial. The investigated eigenwaves satisfy the Maxwell equations, EBCs and the radiation condition (absence of external field sources at the infinity).

The statement is analogous to the problem of macroscopic spiral slow-down systems for microwave range [L. Weinstein, Electromagnetic waves, 1988].

Dispersion equation of surface waves
\[
\frac{\kappa^2}{k^2} K_q(\kappa R) I_q(\kappa R) = \frac{ic}{4\pi R \sigma_{zz}} \left( 1 - \frac{\kappa^2 + k^2}{(\omega + i/\tau)^2 c^2 l_0} \right).
\]
Complex-valued slow-wave coefficient $b$ for a polar-symmetric surface wave

\[
\text{Re} \beta = \frac{v_{ph}}{c} = \text{Re} \frac{k}{h}
\]

Axial component of the time-averaged Poynting vector
Carbon Nanotube as EM device (primarily in THz range):

- Electromagnetic slow-wave line: $v_{ph}/c \approx 0.02$
- Dispersionless surface wave nanowaveguide and high-quality interconnects (PRB 1999)
- Thermal antenna (PRL 2008)
- Monomolecular traveling wave tube (PRB 2009)
- Strong influencing the spontaneous decay rate (PRL 2002)

Antenna resonances for 1 mkm CNT are in the THz range because the plasmon slowing
Theory predicts the slowing-down of surface wave in a metallic SWCNT as much as 50-100 times;

*Is it true?*

The answer can be found from observing antenna resonances red shift.
Experimental observations of THz peak in CNT-based composites

One can suppose that THz finite-length (antenna) resonances explain THz conductivity peak in CNT composites.


FIG. 3. (Color online) Temperature dependence of the optical conductivity of the two samples.

T. Kampfrath, phys. stat. sol. (b) 244, No. 11, 3950–3954 (2007)
THz peak: experiment

Direct experimental demonstration of the correlation between the THz peak frequency and the SWCNT length. That is, the direct experimental evidence of the slowing down in CNTs and the FIR-THz antenna.
**Method of the calibrated CNTs fabrication**

Distribution of the CNT bundles before (a) and after (b) treatment

**Soft cutting of single-wall carbon nanotubes by low temperature ultrasonication in a mixture of sulfuric and nitric acids**

M V Shuba¹, A G Poddubskaya¹, P P Kuzhir¹, S A Maksimenko¹,
V K Ksenovich², G Niaura³, D Seliuta³, I Kasalynas³ and G Valulis³

**Short-length carbon nanotubes as building blocks for high dielectric constant materials in the terahertz range**

M V Shuba¹,², A G Poddubskaya², P P Kuzhir¹,³, S A Maksimenko¹,³,
E Flahaut⁴, V Fiero⁵, A Cejlard⁶ and G Valulis¹,⁶

*Shuba et al., PRB 2013*
The possibility of strong slowing down of EM wave


Ballistic electron transport (up to tens of µm)


Very large current density (up to $10^{10}$ A/cm$^2$)

- M. Radosavljević, J. Lefebvre, and A. T. Johnson, Phys. Rev. B 64, 241307, 2001;
Čerenkov lasing in a CNT

Dispersion equation

$$k - k_m = -\frac{\omega_L^2}{8\pi k_m c^2} \sum_l B_{nl} \left\{ \frac{1}{-\hbar \omega + \epsilon_n(p_n) - \epsilon_l(p_n - \hbar k)} + \frac{1}{\hbar \omega + \epsilon_n(p_n) - \epsilon_l(p_n + \hbar k)} \right\}$$

$\omega_L^2$ is the Langmuir frequency squared, proportional to the current density.

$$\epsilon_l(p_n \pm \hbar k) \approx \epsilon_l(p_n) \pm \hbar k \frac{\partial \epsilon_l}{\partial p_n} + \frac{\hbar^2 k^2}{2} \frac{\partial^2 \epsilon_l}{\partial p_n^2}$$

$v_l = \frac{\partial \epsilon_l(p_n)}{\partial p_n}$ \quad \rightarrow \quad group \ velocity \ of \ nanotube \ electron$

$$\pm \hbar \omega + \epsilon_n(p_n) - \epsilon_l(p_n \pm k) = \pm \hbar (\omega - kv_l \pm \Omega_{nl}) + \frac{\hbar^2}{2} \frac{\partial^2 \epsilon_l}{\partial p_n^2} k^2$$

Typical dispersion equation for second order Cherenkov instability:

$$k - k_m \sim -\frac{\omega_L^2}{(\omega - kv_l)^2}$$
Threshold Current and Instability Increment

Radiation generation is already possible at the current stage of the nanotechnology development

\[ j = 10^{10} \text{ A/cm}^2 \]

\[ L = 10 - 30 \text{ mm} \]

Gain per unit length is extremely large comparing with macrodevices

It allows proposing CNTs as candidates for the development of nano-sized Chernekov-type emitters for THz range
Advantage of using MWCNTs or multi-layered graphene

Estimated wave retardation in a SWCNT reaches 50-100 times
• G. Slepyan et al, PRB 1999

In double-walled CNT:
For long wavelength, when l >> d (d is the distance between CNTs),
frequencies can be approximately written as:

\[ \omega_+ \sim \omega_1(R_1) + \omega_2(R_2) \quad \text{and} \quad \omega_- \sim \left| \omega_1(R_1) - \omega_2(R_2) \right| \]

Phase velocity corresponding to frequency \( \omega_- \), \( v_{ph-} = \omega_- / k \), can be significantly less than the phase velocities in a single walled nanotube

However \( \omega_1(R_1) \neq \omega_2(R_2) \), therefore decreasing of phase velocity in two-walled CNT is limited.

Does bilayer graphene show no such drawback?.....
Decreasing the phase velocity in two-walled CNT is limited.

Does bilayer graphene show no such drawback?.....

It turns out that the phase velocity in bilayer graphene exceeds phase velocity in graphene monolayer. Electron tunneling between graphene layers is guilty.

Tunneling should be suppressed.

It can be realized in spatially separated double-layer or multilayer graphene.

- K. Batrakov, V. Saroka, S. Maksimenko, Chr. Thomsen, J. of Nanophotonics, 6, 061719 (2012)
Schematic representation of graphene sandwich fabrication, consisting of a number of repeating steps, and final graphene/PMMA multilayer structure containing here four graphene sheets. The lateral dimensions of the samples are 7.2 mm * 3.4 mm for MW measurements and cycle sample with diameter 1 cm for THz measurements.
Graphene-like films being 100-1000 times thinner than skin depth provide reasonably high EM attenuation in microwave frequency range, caused by absorption mechanism.

EM absorption is as high as 50% for PyC film of 75 nm thickness and a few layers graphene, 1.5-2 nm thick.
Fabrication of multi-layered PMMA/Graphene structures

Graphene/PMMA (~700 nm) sandwich

30 GHz

dielectric substrate

APPLIED PHYSICS LETTERS 108, 123101 (2016)

Enhanced microwave-to-terahertz absorption in graphene

K. Batrakov,¹,a) P. Kuzhir,¹ S. Maksimenko,¹ N. Volynets,¹ S. Voronovich,¹ A. Paddubskaya,¹ G. Valusis,² T. Kaplas,³ Yu. Svirko,³ and Ph. Lambin⁴
The problem statement

Consider an electron beam propagating along the x axis parallel to a graphene sheet or multilayer graphene sandwich comprising graphene sheets separated by layers of a medium with a dielectric functions $\varepsilon$.

Let us examine the propagation of surface waves along the sandwich in free space, assuming the distances between the graphene layers to be large on the atomic scale and, therefore, neglecting electron interlayer tunneling in the sandwich.

The eigenwaves under study satisfy the Maxwell equations, the boundary conditions at the graphene surfaces in each layer, and the condition that there are no exterior current sources at infinity.
The advantage achieved by graphene doubling is the appearance of an acoustic mode among the plasmon oscillations inherent in the system. This mode’s frequency is proportional to the difference in frequencies of the plasmonic oscillations in the layers.

As a result, the phase velocity of this wave appears to be much less than that achievable in monolayers. Owing to such a large slowing down, one can meet Cherenkov synchronism even for graphene $\pi$ electrons whose velocity is $\approx 300$ less than the speed of light.
Solution of boundary problem with boundary conditions on graphene layers gives dispersion equation for eigenwaves

\[ H(z_i + 0) - H(z_i - 0) = \frac{4\pi}{c} [j_t(z_i) \times n] \]

magnetic field discontinuity on opposite sides of graphene sheet

Dispersio equation for surface TM mode in two-layer graphene system

\[ 2 + \frac{4\pi k_z c}{\omega} \pm \frac{4\pi k_z c}{\omega} \exp\left\{- \sqrt{q^2 - \omega^2/c^2 l}\right\} = 0 \]

Here \( q \) is wave number, \( k_z = \sqrt{\omega^2/c^2 - q^2} \), \( l \) is distance between graphene layers

Graphene conductactivity:

\[ \sigma_{\text{intra}}(\omega, \mu, T, \gamma_{\text{trans}}) = \frac{e^2 g_s g_v}{16 \hbar} \frac{8T}{\pi \hbar} \ln \left[ 2 \cosh \left( \frac{\mu}{2T} \right) \right]\frac{i}{\omega + i \gamma_{\text{trans}}} \]

\[ \sigma_{\text{inter}}(\omega, \mu, T, \gamma_{\text{opt}}) = -\frac{ie^2 g_s g_v}{16 \pi \hbar} \tilde{\Omega} \int_0^\infty \frac{\sinh(x |\mu|/T)}{\cosh(\mu/T) + \cosh(x \mu/T)} \frac{dx}{x^2 - \tilde{\Omega}^2/4} \]

\( m \) is chemical potential, \( \gamma_{\text{trans}}, \gamma_{\text{opt}} \) are broadening parameters
Chemical potential can be tuned by:

1) electrostatic doping (smooth frequency tuning);  
2) number of graphene layers, because in acoustic mode potential is proportional to this number (discrete tuning).

**Frequency tuning**

by varying the chemical potential

Cherenkov resonant frequency $\nu$ vs chemical potential at the electron beam energy 4 KeV (1), 10 KeV (2) and 60 KeV (3).

by varying the electron beam energy

The Cherenkov frequency dependence on the electron beam energy. Chemical potential 0.1 eV (1) and 0.2 eV (2).
Generation equation

Continuity conditions on the beam boundaries and discontinuity conditions for magnetic field on graphene layers give dispersion equation for system “sandwich + electron beam” (generation equation)

\[ I_b = -\frac{(2 + \sigma_0)^2 - (\sigma_0)^2 \exp\{-2\sqrt{q^2 - \omega^2/c^2l}\}}{\sigma_0 \left[ 2 + \sigma_0 + \exp\{-2\sqrt{q^2 - \omega^2/c^2l}\}(2 - \sigma_0) \right]} \]

\[ \sigma_0 = (4\pi/\omega)k_z\sigma \]

In this equation

\[ I_b = \exp(2ik_zh) \frac{(k_{bz}^2 - k_z^2) \{\exp(ik_{bz}\delta) - \exp(-ik_{bz}\delta)\}}{(k_{bz} - k_z)^2 \exp(ik_{bz}\delta) - (k_{bz} + k_z)^2 \exp(-ik_{bz}\delta)} \]

- \( h \) is the distance between sandwich and electron beam,
- \( d \) is the beam thickness,
- \( w_l \) is the electron beam Langmuir frequency,

\[ k_{bz} = k_z \sqrt{1 - \frac{\omega_l^2}{\gamma^3(\omega - qu)^2}} \]
Quasi-Cherenkov radiation of an electron beam passing over the graphene/polymer sandwich structure.

**Graphene layered systems as a terahertz source with tuned frequency**

K. Batrakov* and S. Maksimenko

**Slowing down of the acoustic mode vs interlayer distance in a double graphene at different chemical potentials**

**Frequency dependence of the instability increment for 4 (1), 8 (2), and 9 (3) layers**
Conclusions

- We theoretically predicted and proved experimentally strong slowing down of surface EM waves in CNTs and graphene

- We theoretically demonstrate feasibility of extra small THz range sources based on CNTs

- We theoretically predicted and experimentally proved strong absorption of GHz- & THz waves in graphene

- We propose more realistic THz-range source on the base of multilayered graphene/polymer structures
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AF20-15-61804-1 CRDF Global Agreement
Thank you for your attention!
Nanoelectromagnetics

A research discipline studying the behaviour of high-frequency electromagnetic radiation on nanometer scale is currently emerging as a synthesis of macroscopic electrodynamics and microscopic theory of electronic properties of different nanostructures.

**FUNDAMENTAL CHALLENGE in NANOSCALE ELECTROMAGNETICS** is unusual constitutive properties of structural materials due to spatial confinement of the charge carriers motion or INTERPLAY of SCHROEDINGER and MAXWELL EQUATIONS.

Maksimenko & Slepyan, Nanoelectromagnetics of low-dimensional structures
Commercial THz time-domain spectrometer T-Spec by EKSPLA.
A 1050+/-40 nm wave length pumping laser having 50-150 fs pulse duration and more than 40 mW output power at approximately 80 MHz pulse repetition rate is used to excite a photoconductor antenna and produced THz radiation up to 2 THz.
The spectrometer, THz emitter, and detector consist of a microstrip antenna integrated with a photoconductor and silicon lens. The sample in the form of a plane parallel plate is placed between emitter and detector normally to the initial EM wave. The THz detector output is proportional to the instant electrical field strength of the THz pulse.
Axial surface conductivity of isolated single-wall carbon nanotube

\[
\sigma_{zz}(\omega) = -\frac{ie^2\omega}{\pi^2\hbar R} \left\{ \frac{1}{\omega(\omega+i/\tau)} \sum_{s=1}^{m} \int_{1stBZ} \frac{\partial E_c}{\partial p_z} \frac{\partial F_c}{\partial p_z} dp_z - 2\sum_{s=1}^{m} \int |R_{cv}|^2 E_c \frac{F_c - F_v}{\hbar^2 \omega(\omega+i/\tau)} dp_z \right\},
\]

Drude regime for metallic SWCNT

Interband transitions

\[ \tau_1 = 2 \times 10^{-11} \text{s} \]
Our result has been confirmed in *Nano Letters* **13**, 5991 (2013):

**Plasmonic Nature of the Terahertz Conductivity Peak in Single-Wall Carbon Nanotubes**

Qi Zhang, † Erik H. Hároz, † Zehua Jin, † Lei Ren, † Xuan Wang, † Rolf S. Arvidson, ‡ Andreas Lüttge, ‡,§ and Junichiro Kono*, †, ‡

samples. Our experimental results show that the broad THz peak originates from a plasmon resonance in both the metallic and the doped semiconducting carbon nanotubes rather than the interband excitation of the curvature-induced gap in nonarmchair metallic nanotubes. The intraband free electron