Interactions of charged particles with graphene

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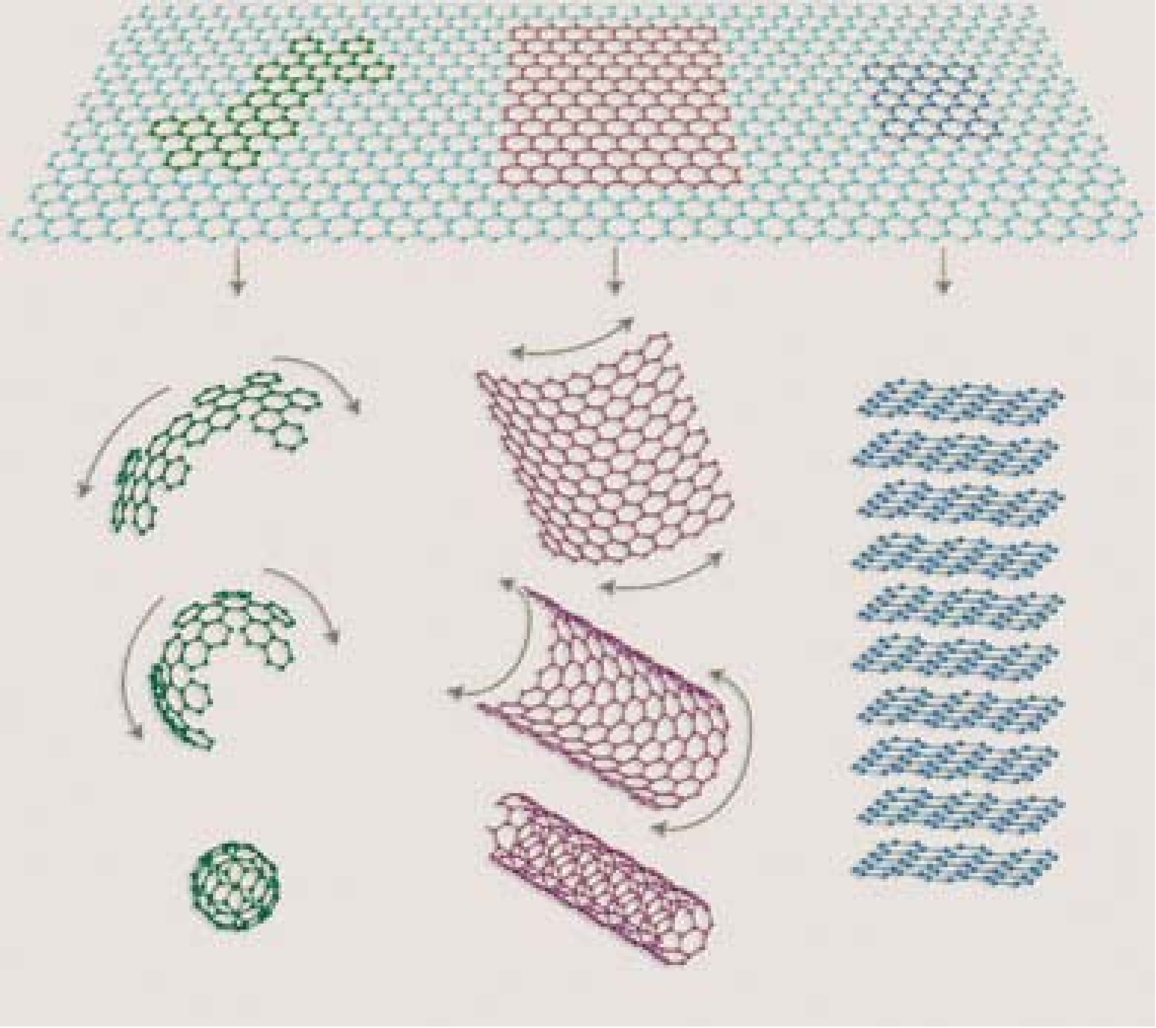
Outline

- ❑ Introduction
- ❑ Ion interactions with graphene
  - Nonlinear screening
  - Image potential
  - Stopping force
  - Wake effect
- ❑ Plasmon excitations by electron beams
  - HREELS, energy *E*~20 eV, substrate phonons
  - EELS, energy *E*~100 keV, layered electron gas
- ❑ Outlook

Graphene as building block of carbon nanostructures

graphene



Fullerene, C_{60} **nanotube, CNT** **graphite, HOPG**

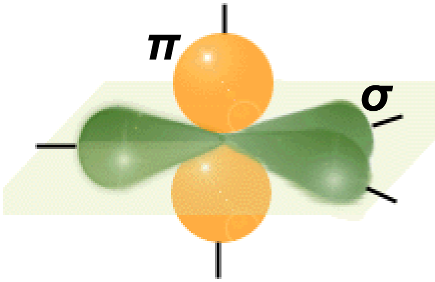
Why study carbon nanostructures ?

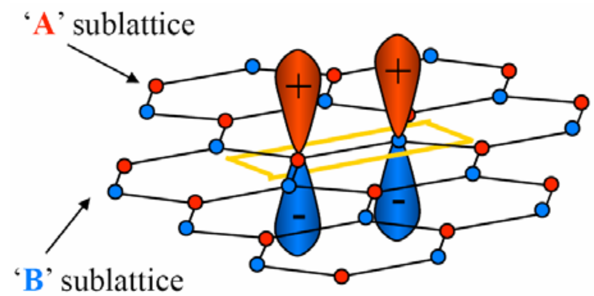
- Physical properties:
  - Electrical, mechanical, thermal
  - Dependent on: molecular structure, dielectric environment, local modification
- Applications:
  - Nanoelectronic devices
  - Biochemical sensors
  - New composite materials
  - Ion storage (H, Li)
  - Nanoelectromechanical systems (NEMS)

Electronic response to external charged particles (regime of electronic stopping for moving charges)

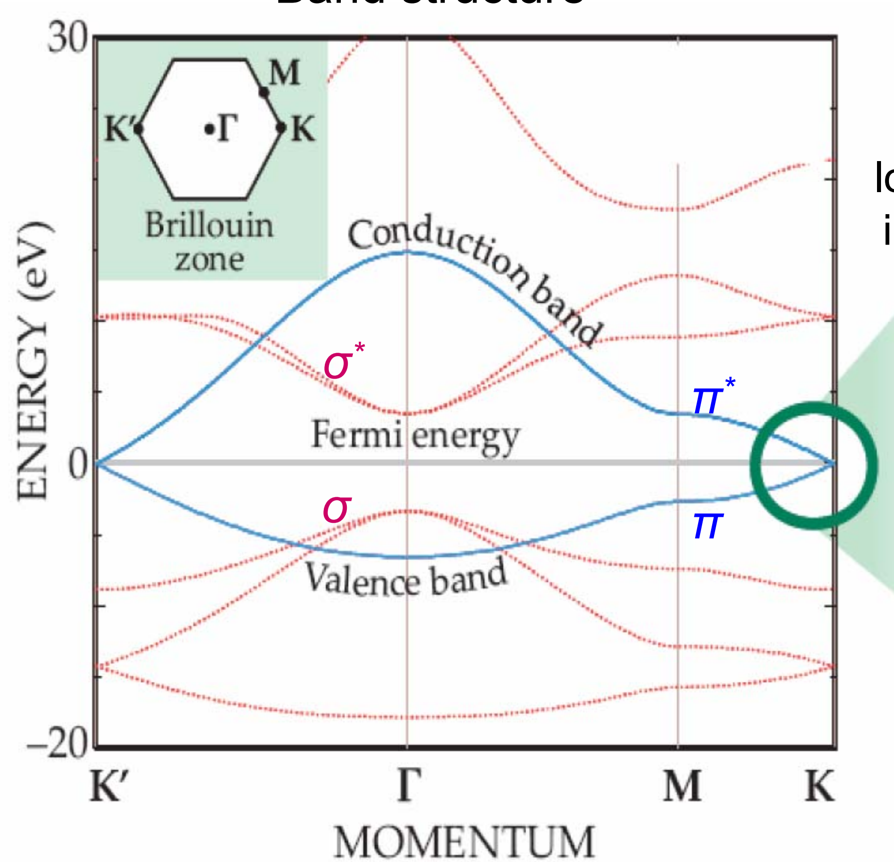
- Electrons
  - EELS in STEM: plasmon excitations of σ and π electrons in CNTs & graphene
  - HREELS: plasmon excitations of π electrons in graphene
  - image potential states: CNTs & graphene
- Ions
  - ion channeling in CNTs
  - grazing scattering of ions on graphene
  - friction forces on slowly moving ions
  - static screening of charged impurities

Electronic structure of graphene

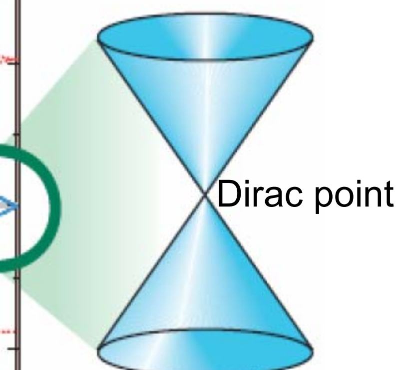




Band structure



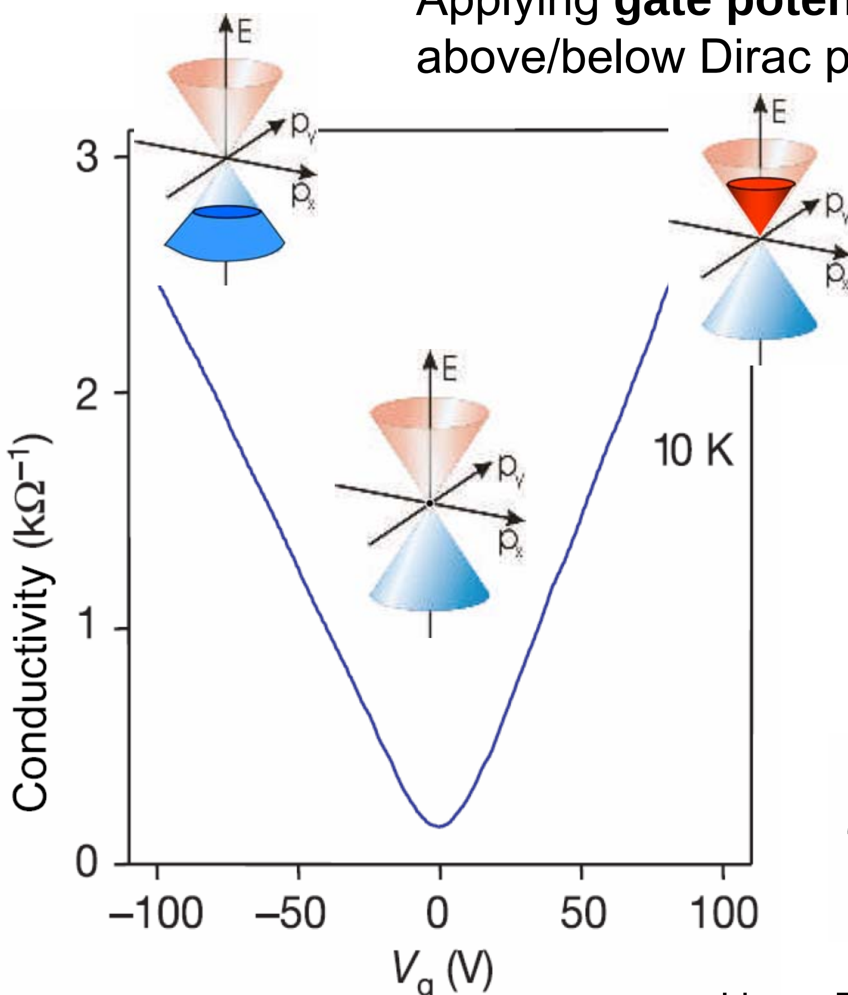
low-energy excitations involve π bands only

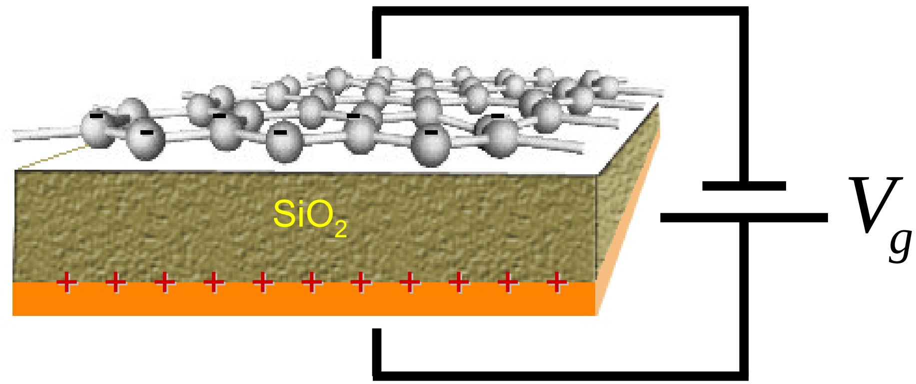


$$\varepsilon_{\mathbf{k}} = \hbar v_F \|\mathbf{k}\|$$

Ambipolar electric field effect in single-layer graphene on top of an oxidized Si wafer

Applying **gate potential** V_g shifts Fermi energy E_F above/below Dirac point (electron or hole doping)





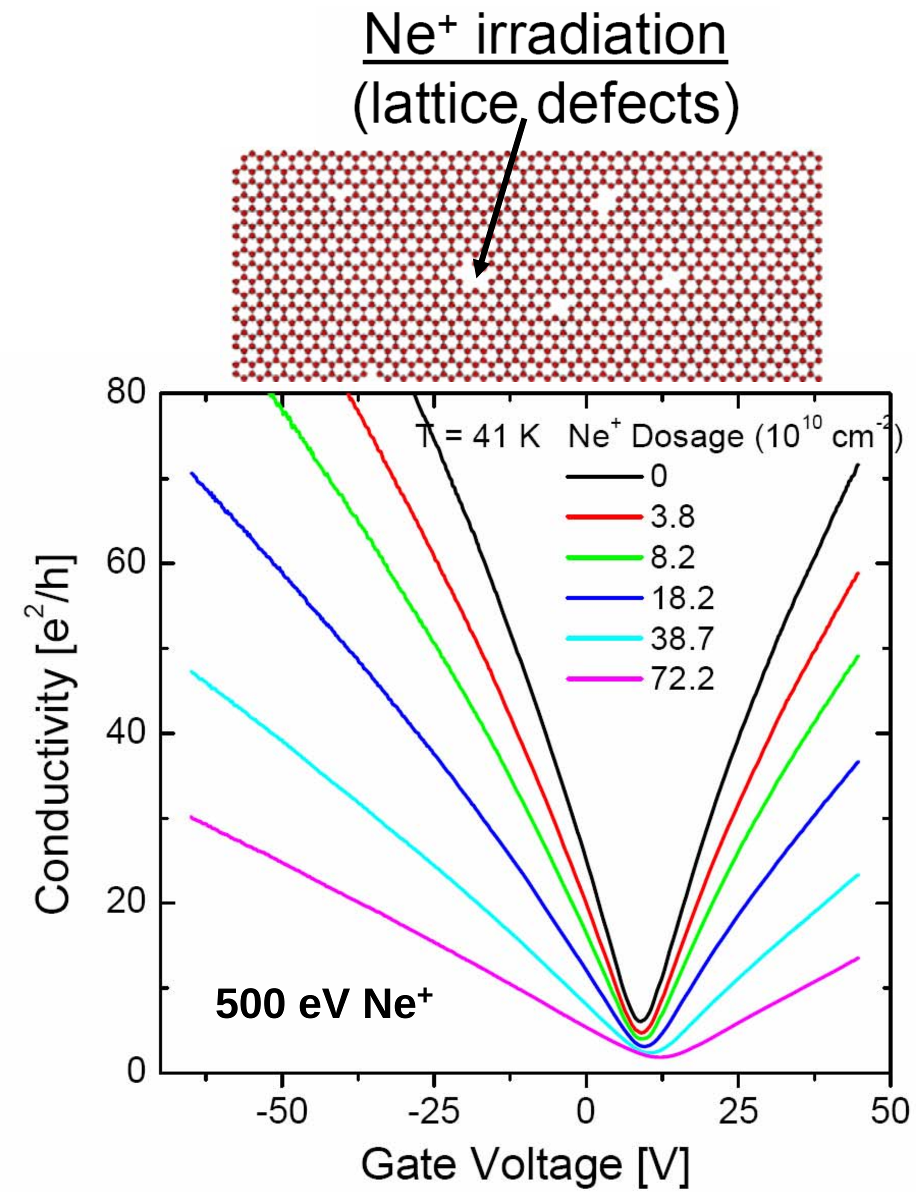
Equilibrium charge carrier density vs chemical potential μ may be >0 and <0

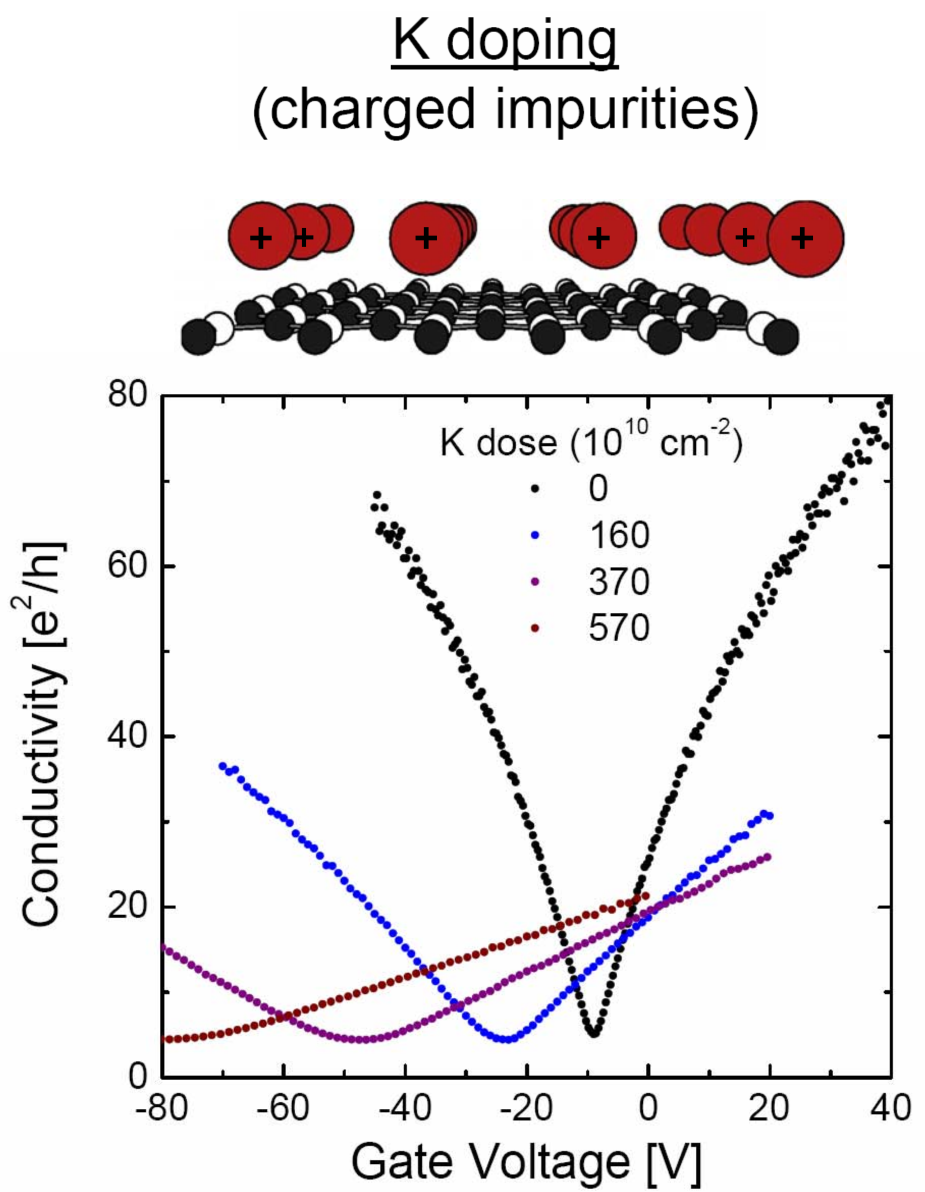
$$n(\mu) = \int_0^{\infty} d\varepsilon\, \rho(\varepsilon) \left[\frac{1}{1+\mathrm{e}^{\beta(\varepsilon-\mu)}} - \frac{1}{1+\mathrm{e}^{\beta(\varepsilon+\mu)}} \right]$$

Linear DOS: $\rho(\varepsilon) \approx \frac{g_s g_v}{2\pi} \frac{|\varepsilon|}{(\hbar v_F)^2}$, Fermi speed: $v_F \approx \frac{c}{300}$

Ion bombardment vs ion adsorption on graphene

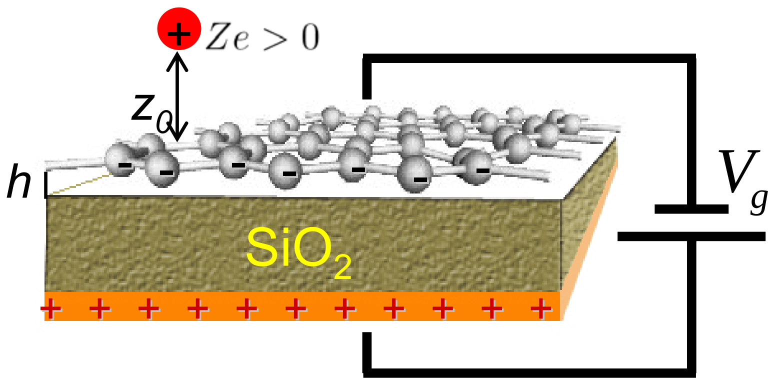
J.H. Chen *et al., Phys. Rev. Lett.* (2009), *Nature Phys.* (2008)





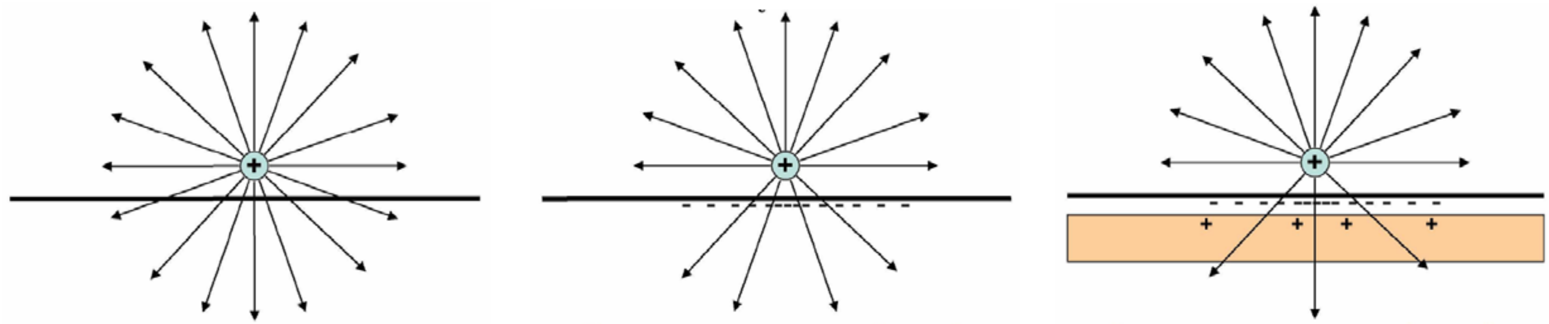
Nonlinear screening of external charge by graphene

M. Ghaznavi *et al.* *Phys. Rev.* B 81 (2010) 085416



Study several effects:

- doping via gate potential
- finite temperature
- exchange interaction effects
- distance z_0 from graphene
- size of gap *h* to the substrate



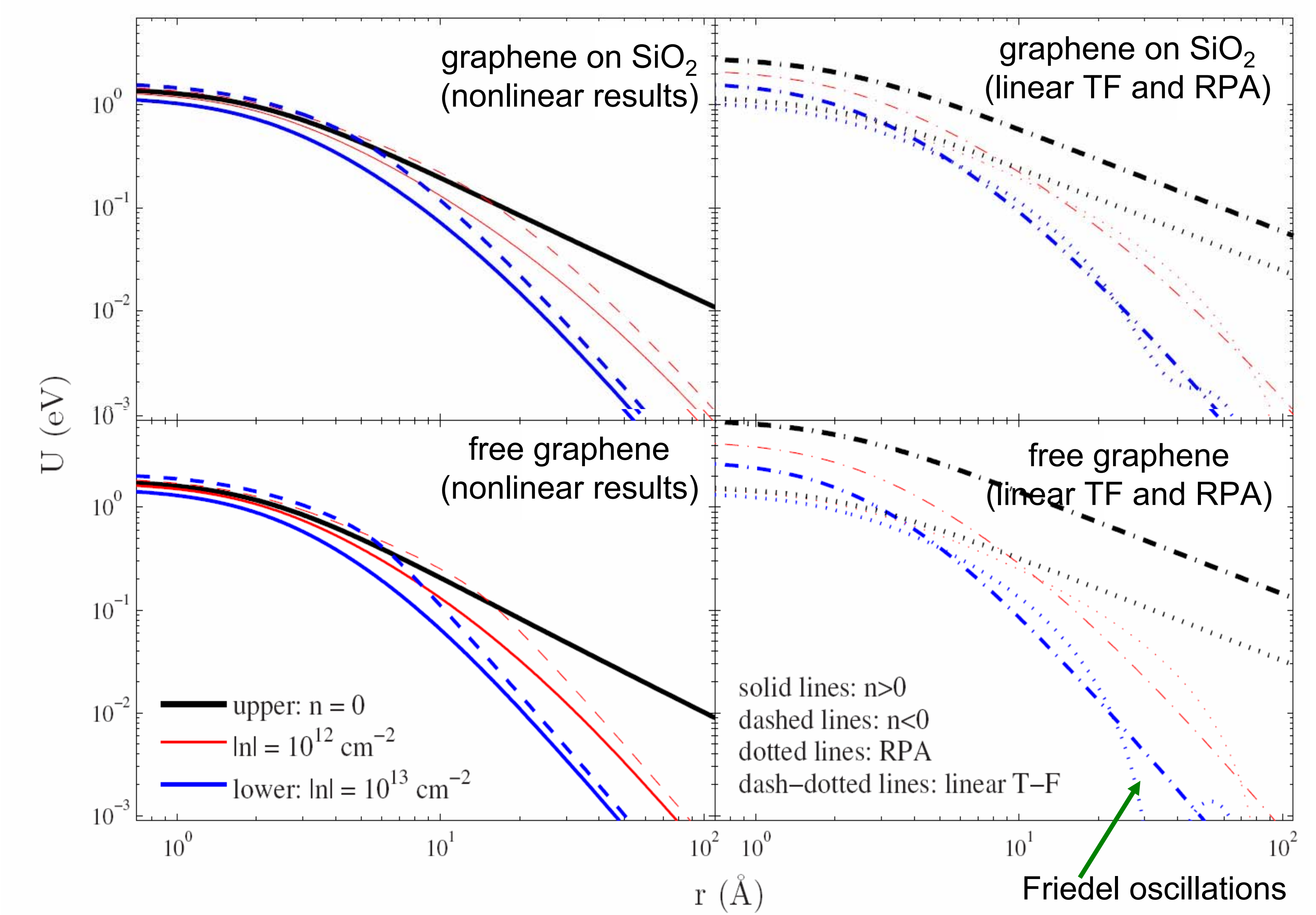
Thomas-Fermi model at finite T

$$U(\mathbf{r}) = U_{\text{ext}}(\mathbf{r}) - e^2 \int d^2\mathbf{r}' \; [n(\mu + U(\mathbf{r}')) - n(\mu)] \left[\frac{1}{\|\mathbf{r} - \mathbf{r}'\|} - \frac{\epsilon_s - 1}{\epsilon_s + 1} \frac{1}{\sqrt{(\mathbf{r} - \mathbf{r}')^2 + 4h^2}} \right]$$

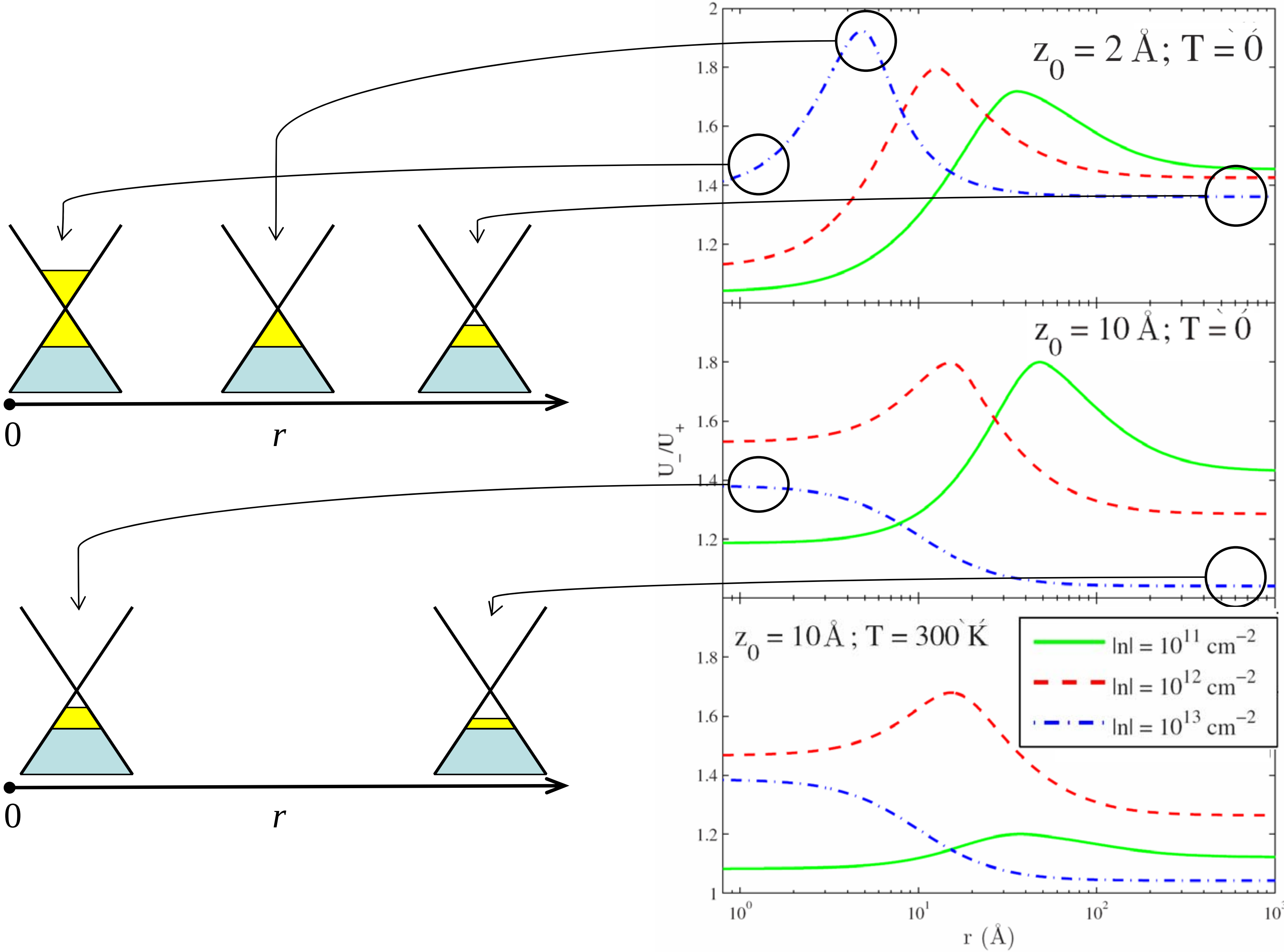
Self-consistent solution for screened potential $U_\pm(r)$ in graphene when $\mu \gtrless 0$

Screened potential in graphene due to external charge

M. Ghaznavi *et al. Phys. Rev.* B 81 (2010) 085416



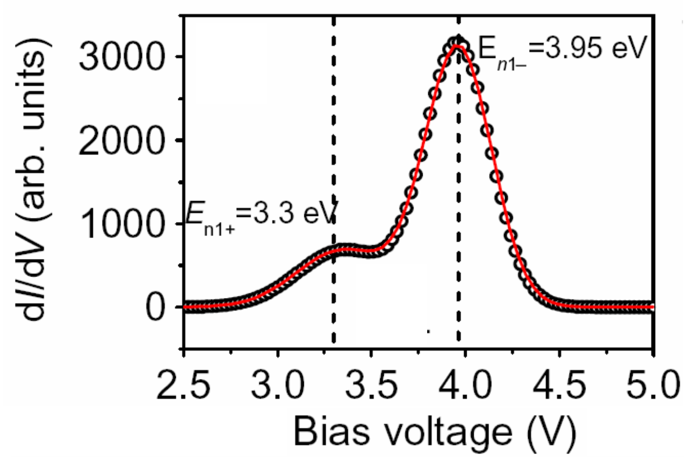
$\boxed{\text{Ratio } U_-(r)/U_+(r) \text{ of screened potentials for } n \lessgtr 0}$

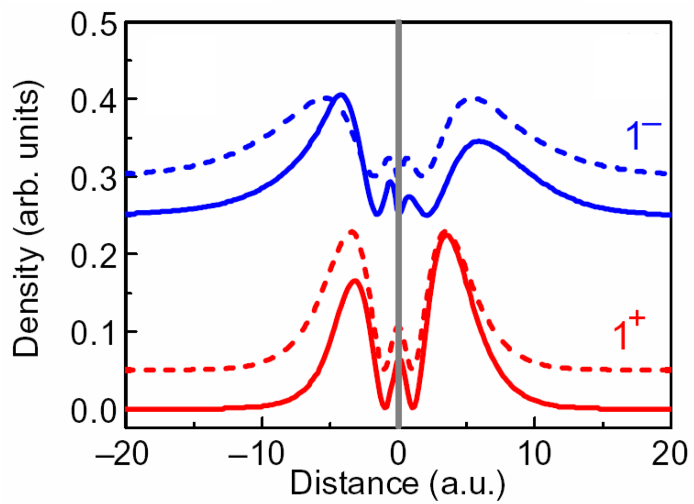


Nonlinear image interaction for external charge

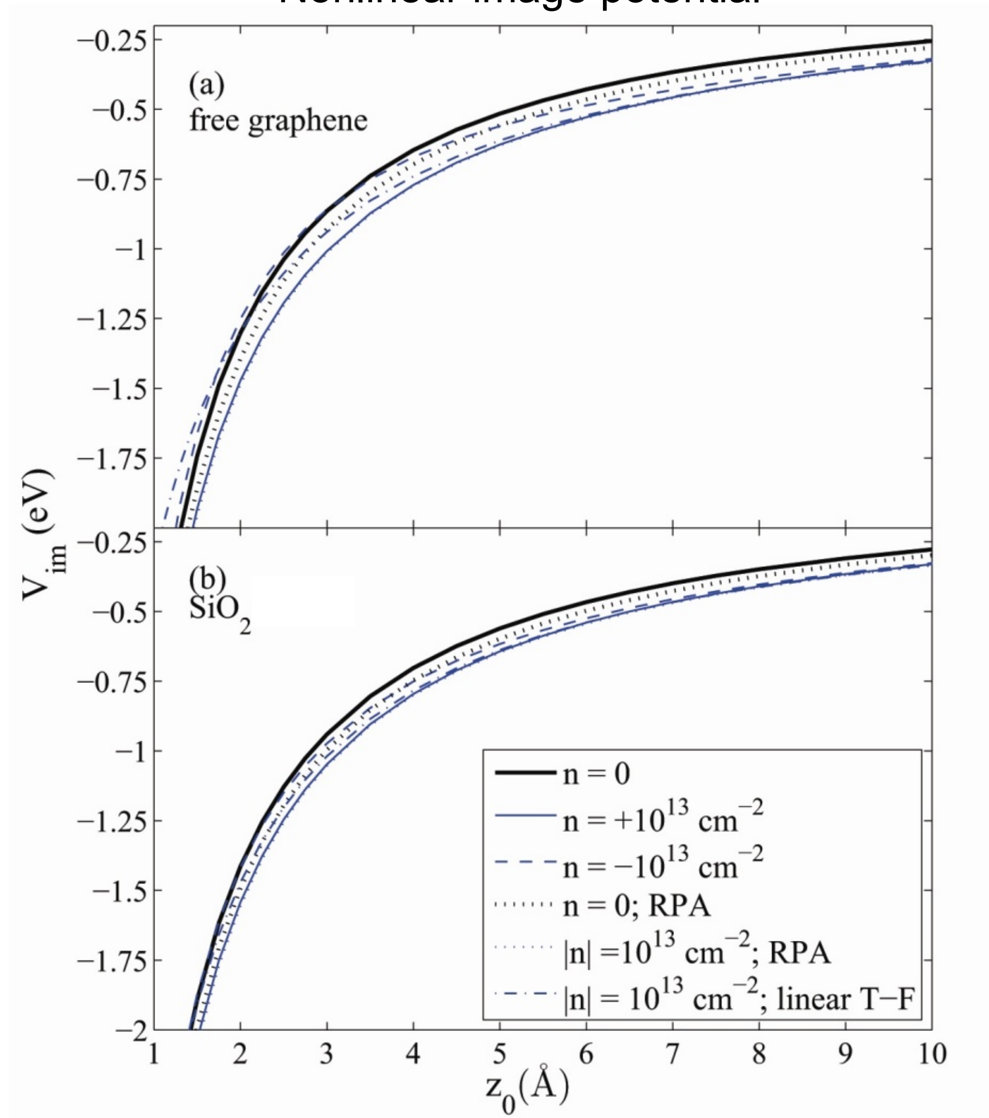
M. Ghaznavi *et al. Phys. Rev.* B 81 (2010) 085416

Observation of image potential states on epitaxial graphene: S. Bose *et al. New J. Phys.* 12 (2010) 023028



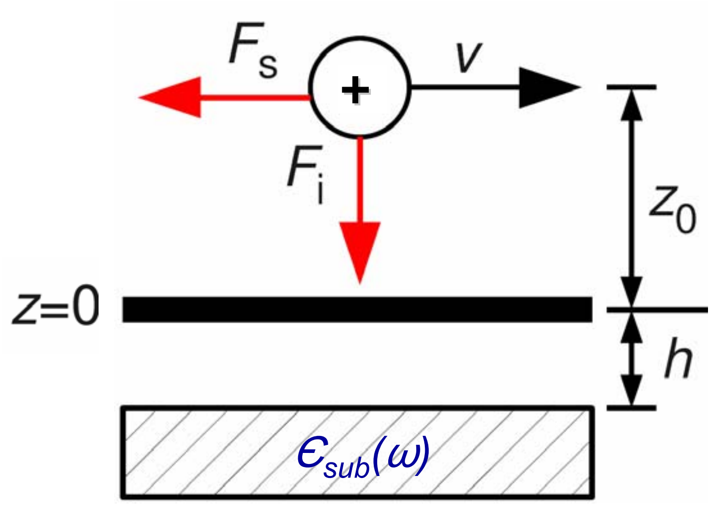


Nonlinear image potential



Interaction of moving external charge with graphene

K.F. Allison *et al.*,*Phys. Rev.* 80 (2009), *Nanotechnology* 21 (2010) 134017



Study several effects:

- doping via gate potential
- damping via Mermin approach
- local field effects
- phonons in polar substrate
- size of gap to the substrate

Dielectric function of graphene + substrate: Random Phase Approximation

$$\epsilon(q,\omega) = \left[1 - \frac{\epsilon_{\rm sub}(\omega)-1}{\epsilon_{\rm sub}(\omega)+1}\,{\rm e}^{-2qh}\right]^{-1} + \frac{2\pi e^2}{q}\Pi_{\rm gra}(q,\omega;\gamma)$$

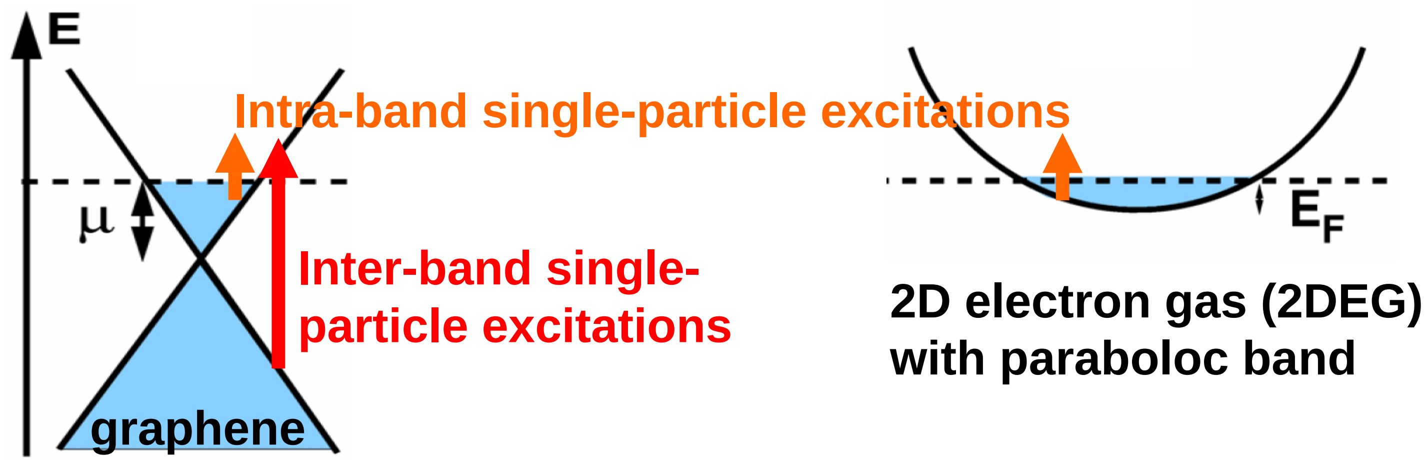
Stopping force

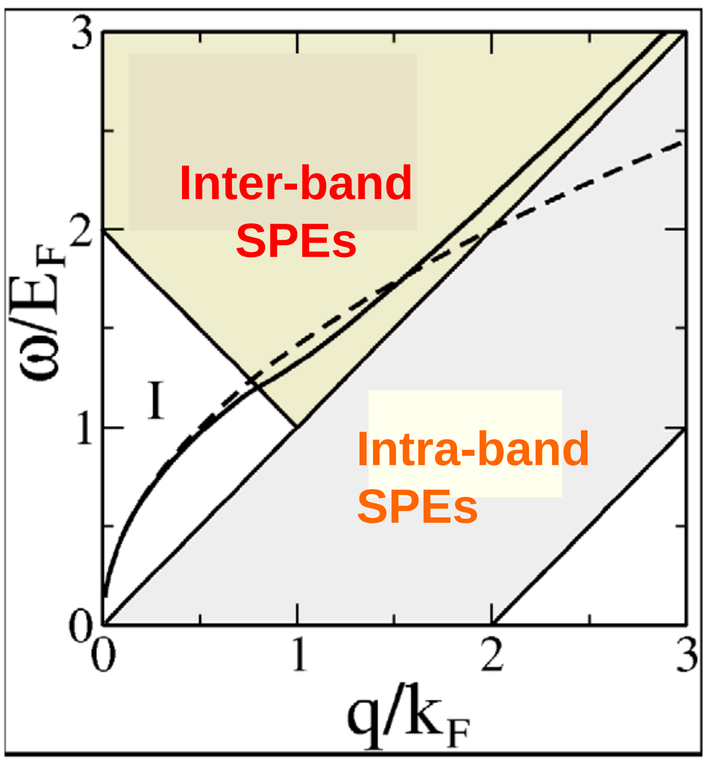
$$F_s = \frac{2}{\pi}\frac{Z^2e^2}{v}\int_0^\infty dq e^{-2qz_0}\int_0^{qv} d\omega \frac{\omega}{\sqrt{q^2v^2-\omega^2}}\Im\left[\frac{1}{\epsilon(q,\omega)}\right]$$

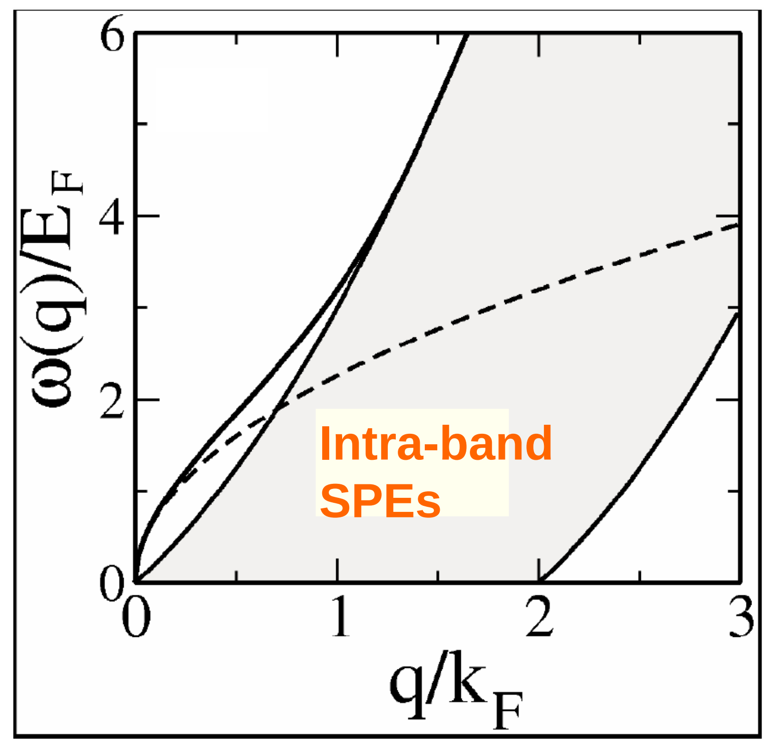
Image force

$$F_i = \frac{2}{\pi}Z^2e^2\int_0^\infty dq q e^{-2qz_0}\int_0^{qv} \frac{d\omega}{\sqrt{q^2v^2-\omega^2}}\Re\left[\frac{1}{\epsilon(q,\omega)}-1\right]$$

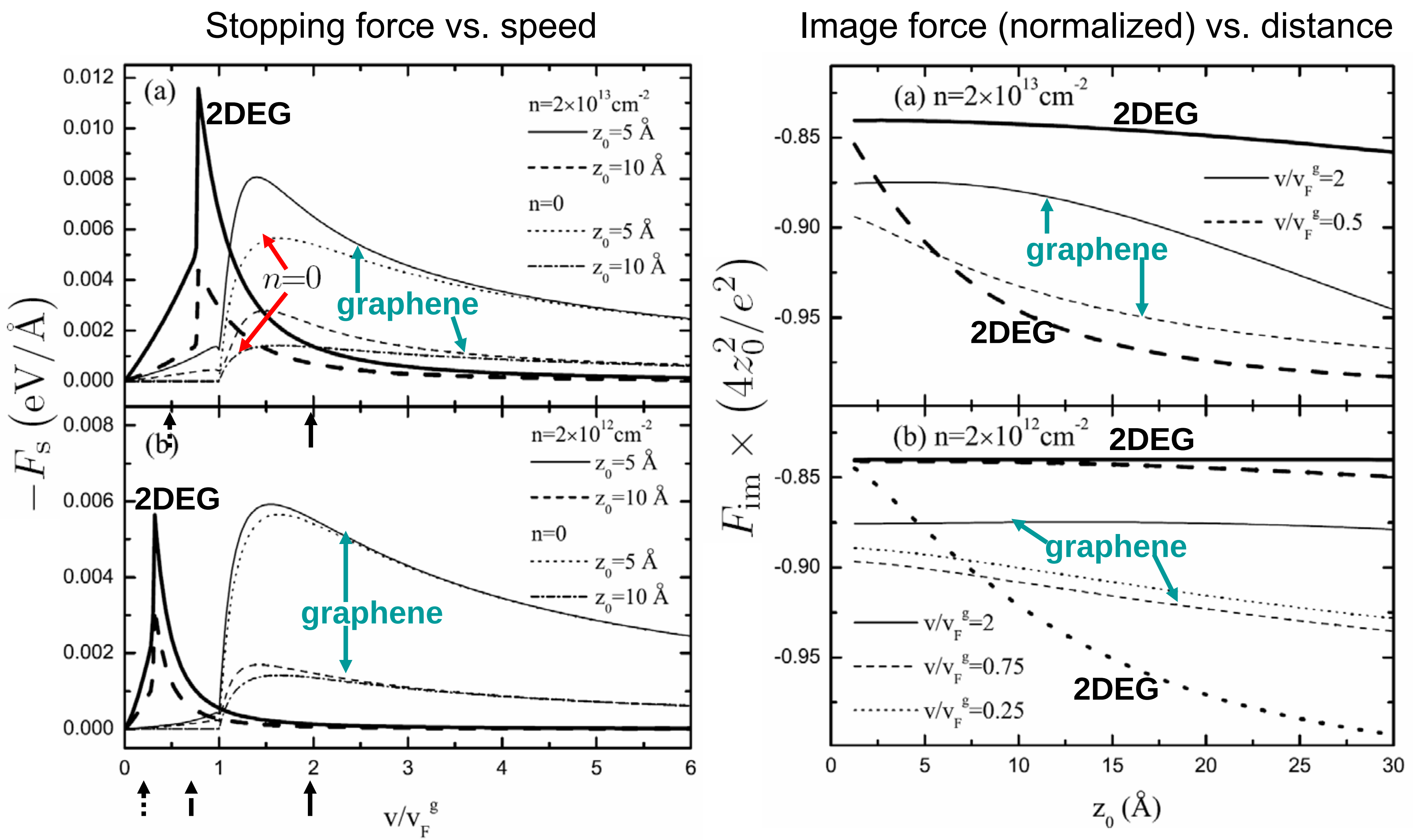
Loss Function $-\Im\{1/\epsilon(q,\omega)\}$ for graphene and 2DEG



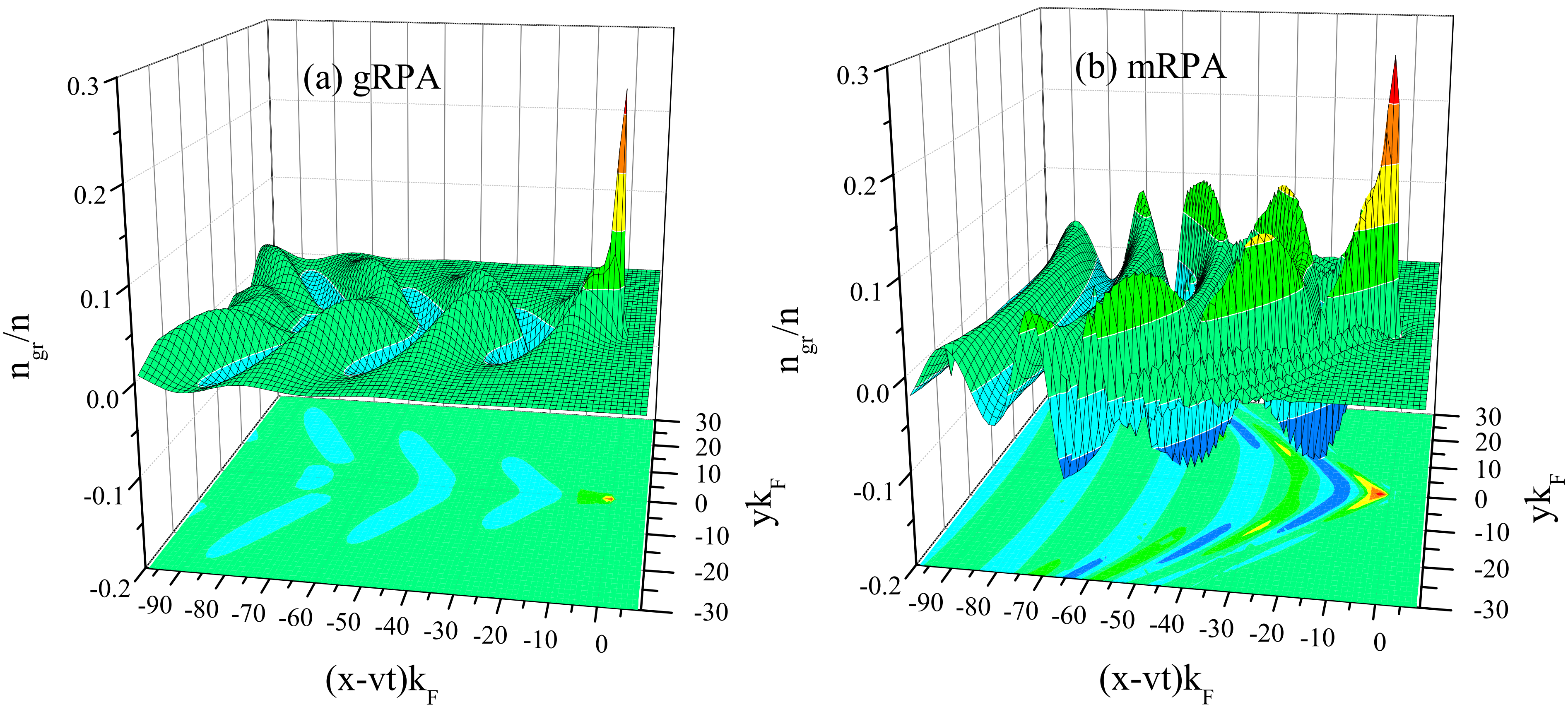




Stopping and image forces on a point charge moving over epitaxial graphene on SiC and 2DEG (Ag on Si)



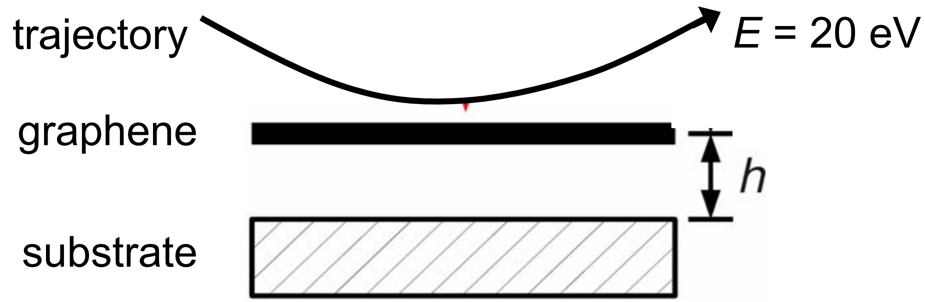
Wake effect due to charge moving at speed *v* = $4v_F$ and distance z_0 = 20 Å from free graphene & 2DEG with doping equilibrium density *n* = 10^{13} cm^{-2}



Fermi wavenumber in graphene: $k_F = \sqrt{\pi n}$

HREEL spectra for graphene on a substrate

K.F. Allison *et al.*, *Nanotechnology* 21 (2010) 134017



Total energy loss:

$$E_{\rm loss} = -\int_{-\infty}^{\infty} dt \int d^2\mathbf{r} \int_{-\infty}^{\infty} dz\, \rho_{\rm ext}(\mathbf{r}, z, t)\, \frac{\partial}{\partial t}\Phi_{\rm ind}(\mathbf{r}, z, t)$$

$$= \int_0^{\infty} d\omega\, \omega \int \frac{d^2\mathbf{q}}{2\pi^2}\, |S(\mathbf{q}, \omega)|^2\, \Im\left[\frac{-1}{\epsilon(q, \omega)}\right]$$

Loss function

Projectile structure factor:

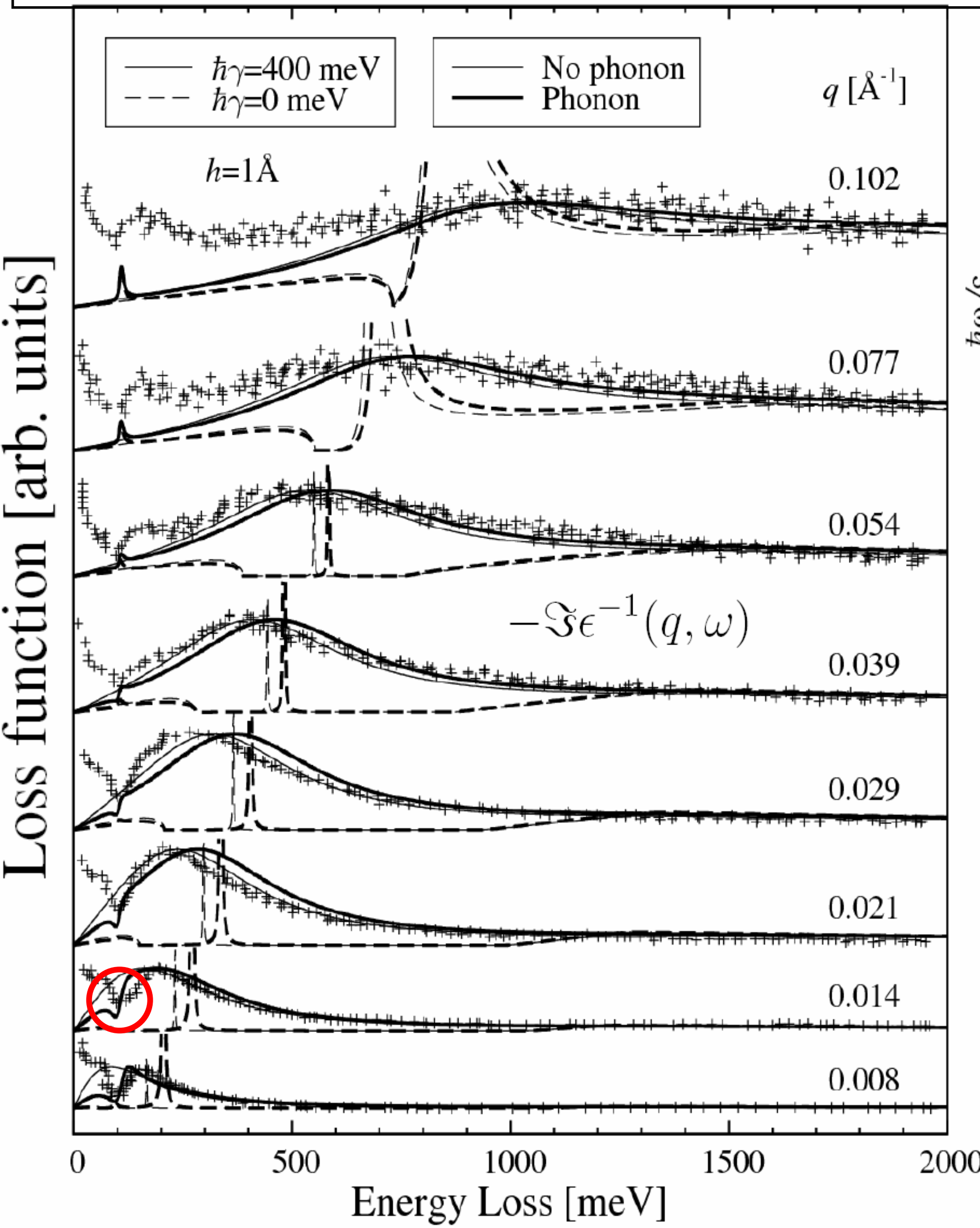
$$S(\mathbf{q}, \omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \int d^2\mathbf{r}\, e^{-i\mathbf{q}\cdot\mathbf{r}} \int_{-\infty}^{\infty} dz\, e^{-q|z|}\, \rho_{\rm ext}(\mathbf{r}, z, t)$$

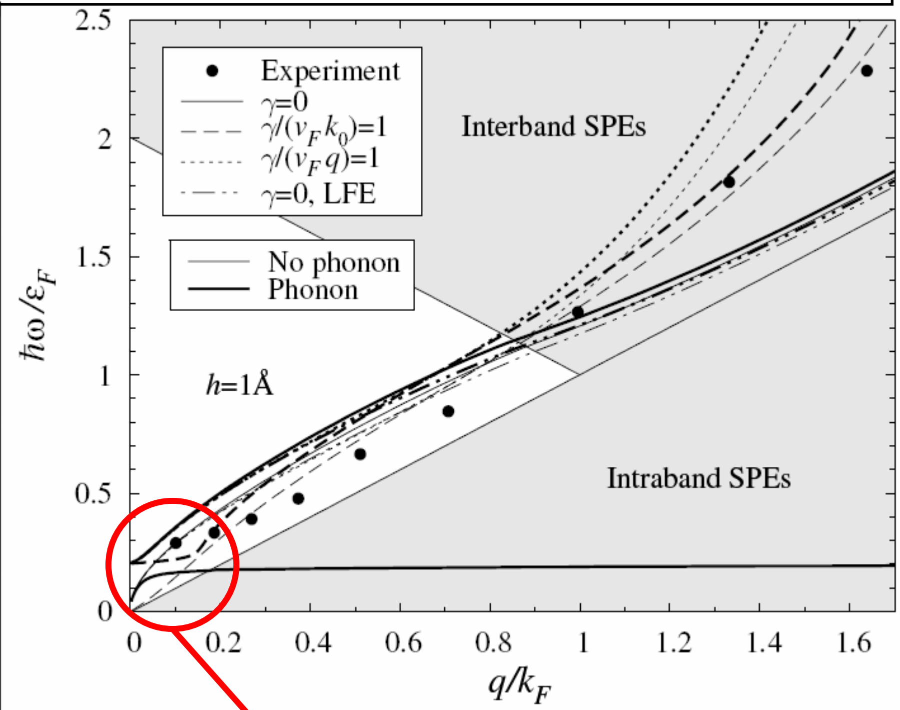
$$\epsilon(q, \omega) = \left[1 - \frac{\epsilon_{\rm surf}(q, \omega) - 1}{\epsilon_{\rm surf}(q, \omega) + 1}\, e^{-2qh}\right]^{-1} + \frac{2\pi e^2}{q}\chi_{\rm gr}(q, \omega)$$

Surface dielectric function:

$$\epsilon_{\rm surf}(q, \omega) = \left[\frac{q}{\pi}\int_{-\infty}^{\infty} \frac{dq_z}{(q^2 + q_z^2)\ \epsilon_{\rm sub}\left(\sqrt{q^2 + q_z^2}, \omega\right)}\right]^{-1}$$

HREELS spectra for graphene on SiC, exper. by Liu *et al.* PRB 78 (2008) 201403





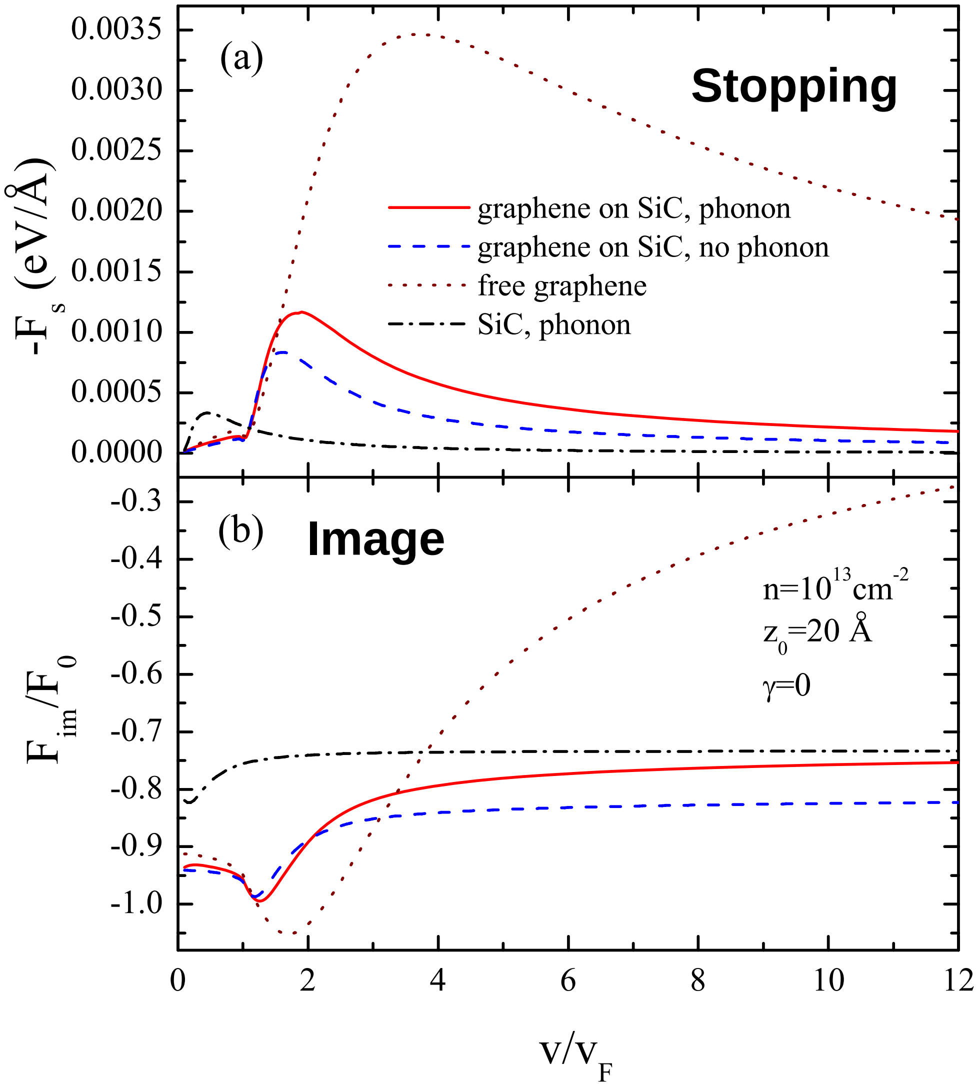
Plasmon coupling with substrate phonon

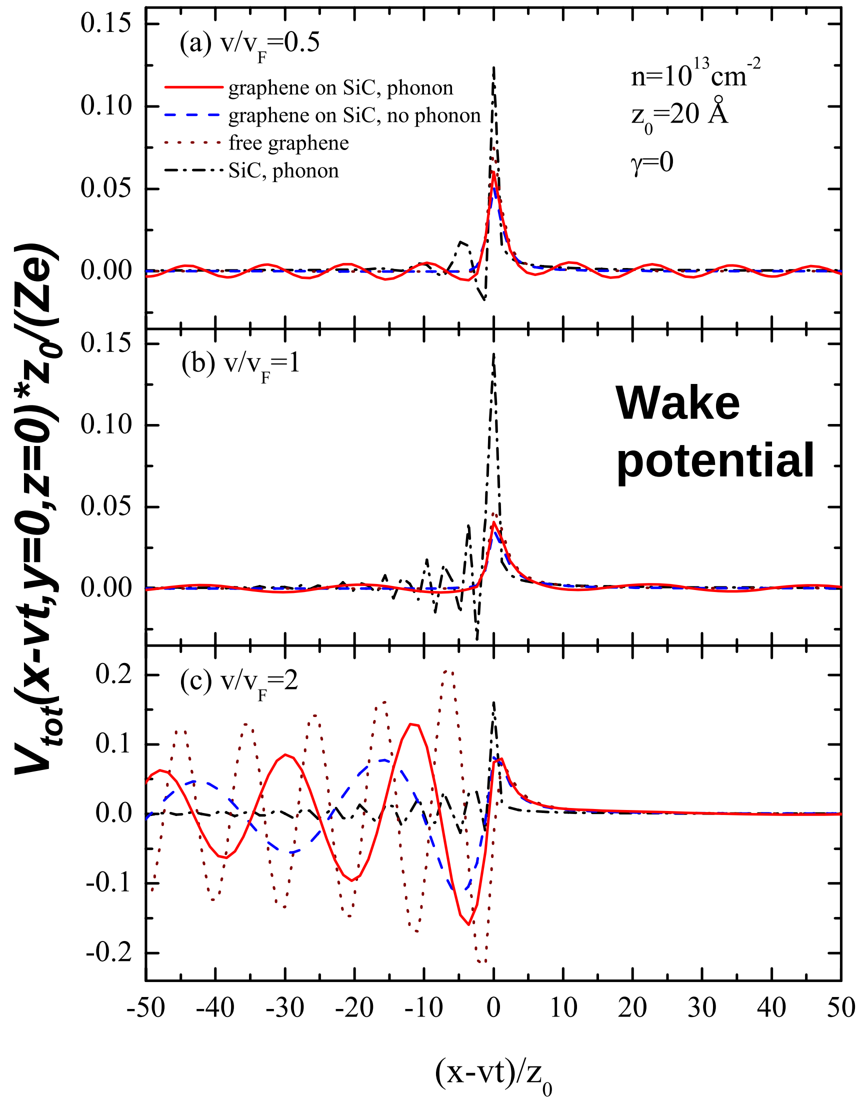
$$\epsilon(q,\omega) \approx 1 + \epsilon_{\text{sub}}(\omega) - 4\frac{e^2}{\hbar}\frac{v_F q}{\omega^2}\sqrt{\pi n} = 0$$

$$\epsilon_{\text{sub}}(\omega) = \epsilon_\infty + (\epsilon_0 - \epsilon_\infty)\,\frac{\omega_{\text{TO}}^2}{\omega_{\text{TO}}^2 - \omega^2}$$

Stopping and image forces on projectile moving over graphene on SiC: effects of **substrate phonons**

CEPAS proceedings





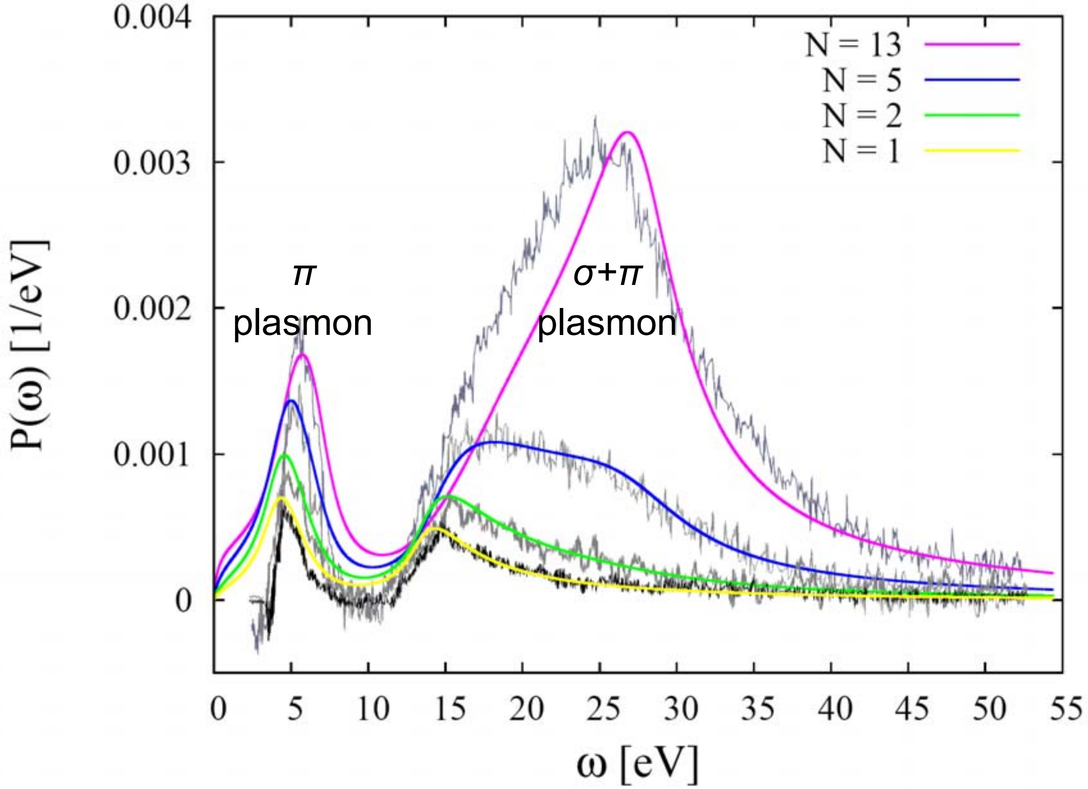
EEL spectra for free-standing *N*-layer graphene

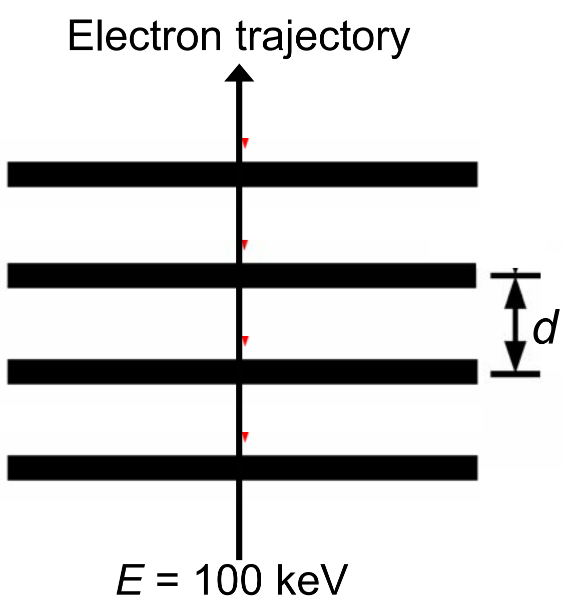
experiment by: T. Eberlein *et al.*, *Phys. Rev.* B 77 (2008) 233406

$$P(\omega) = \frac{4e^2}{\pi v^2} \int_0^{q_{\max}} \frac{dq\, q^2}{\left[q^2 + \left(\frac{\omega}{v}\right)^2\right]^2} \Im \left\{ V(q)\chi(q,\omega) \sum_{n=1}^{N} \sum_{n'=1}^{N} e^{-i(n-1)d\omega/v} \left(\mathcal{M}^{-1}\right)_{nn'} e^{i(n'-1)d\omega/v} \right\}$$

$$\mathcal{M}_{nn'}(q,\omega) = \delta_{nn'} + (1-\delta_{nn'})\, V(q)\chi(q,\omega) e^{-qd|n'-n|}$$

$$\chi(q,\omega) = \chi_0(q,\omega)/\left[1 + V(q)\chi_0(q,\omega)\right], \quad V(q) = 2\pi e^2/q$$

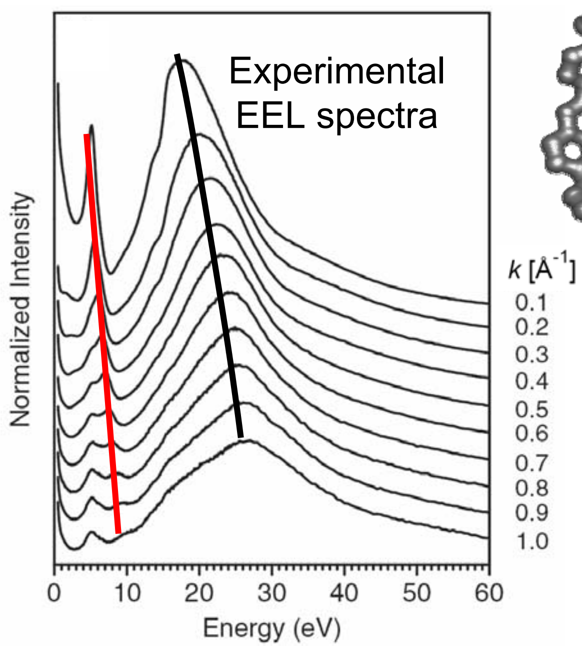


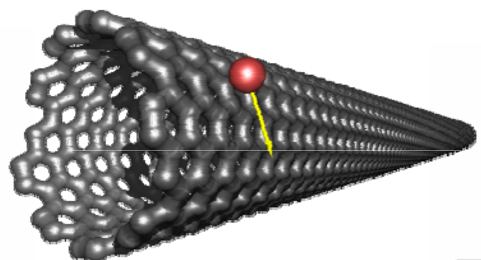


Two-fluid, 2D hydrodynamic model for σ and π electrons: used in single-wall carbon nanotube

experiment: Kramberger *et al., Phys. Rev. Lett.* 100 (2008) 196803
theory: Mowbray *et al., Phys. Rev.* B 82 (2010) 035405

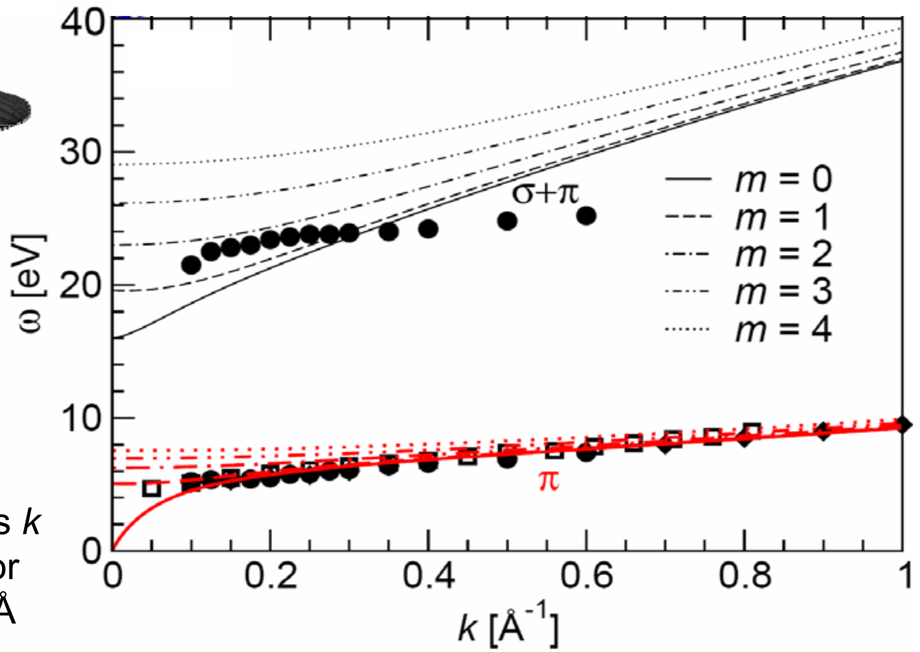
$$\chi_0(q,\omega) = \frac{n_\pi^0 q^2/m_\pi^*}{s_\pi^2 q^2 + \omega_{\pi r}^2 - \omega(\omega + i\gamma_\pi)} + \frac{n_\sigma^0 q^2/m_\sigma^*}{s_\sigma^2 q^2 + \omega_{\sigma r}^2 - \omega(\omega + i\gamma_\sigma)}$$





$$q^2 \to \frac{m^2}{a^2} + k^2$$

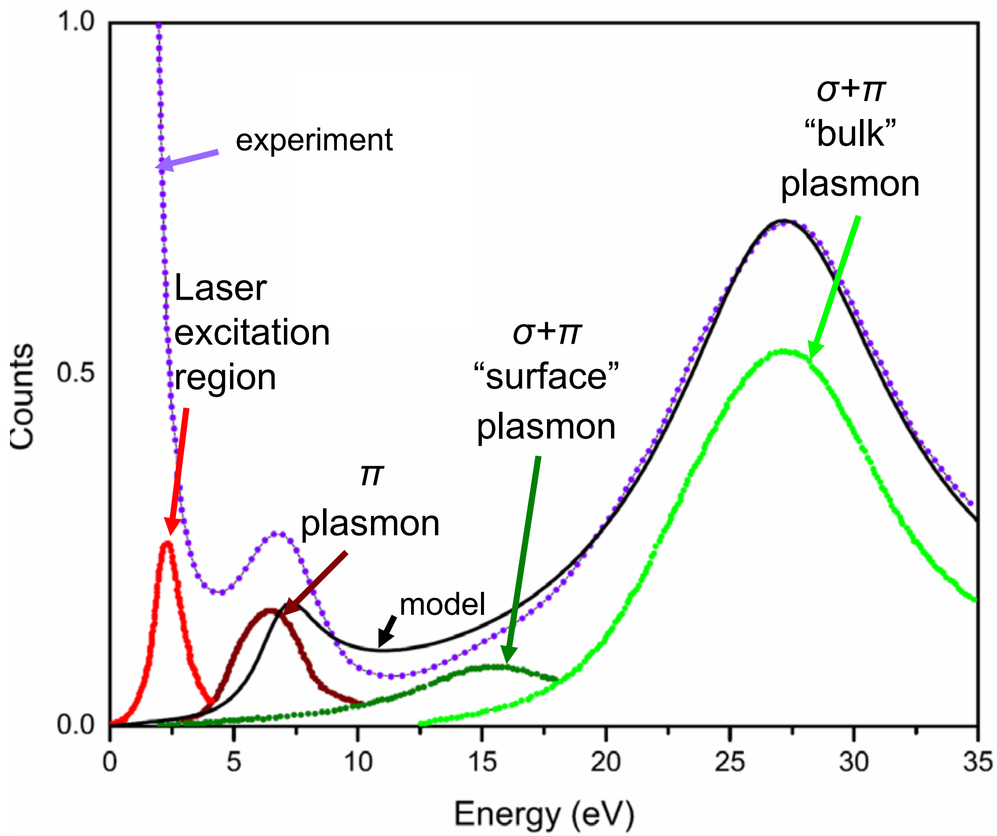
Plasmon dispersions vs k with angular mode m for nanotube radius a = 7 Å



EELS for free-standing **graphite** (*N >> 1)*

F. Carbone *et al., Chem. Phys. Lett.* 468 (2009) 107

$$P(\omega) \approx N \frac{4e^2}{\pi v^2} \int_0^{q_{\max}} \frac{dq\, q^2}{\left[q^2 + \left(\frac{\omega}{v}\right)^2\right]^2} \Im \left[\frac{V(q)\chi_0(q,\omega)}{1 + \frac{\sinh(qd)}{\cosh(qd) - \cos(\omega d/v)} V(q)\chi_0(q,\omega)} \right]$$



Outlook

- Electronic energy loss and image interaction important for charged particle interactions with C-nanostructures: energy deposition & transport
- Plasmon excitations in HREELS and EELS: full theory needed for dielectric response of both σ and π electrons
- Ion charge states during scattering need to be considered
- Effects of dielectric environment, particularly substrate phonons
- Concept of friction and screening or mobile ions in aqueous solution

Thank you for your attention