Anomalous growth of thermoelectric power in gapped graphene

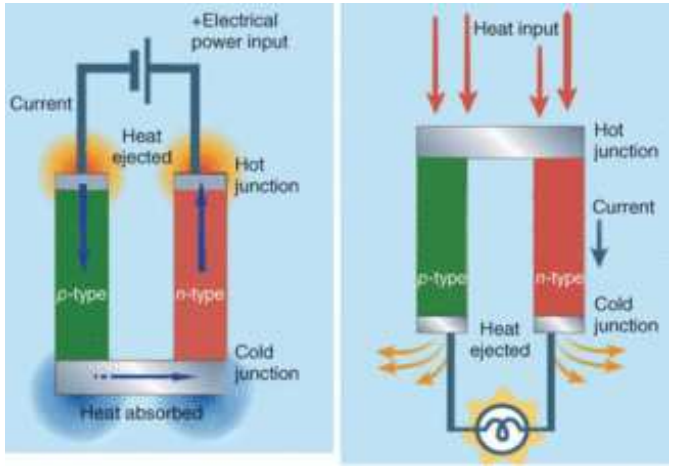
Sergei G. Sharapov

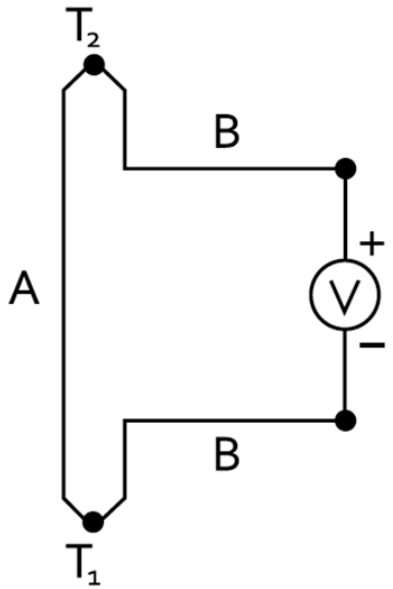
Bogolyubov Institute for Theoretical Physics
of the National Academy of Sciences of Ukraine

Talk is based on: S.G. Sh., A.A. Varlamov, arXiv:1202.1362

28 May - 2 June, 2012

Thermo-electricity – Peltier-Seebeck effect





$$V = (S_B - S_A)(T_2 - T_1)$$
$$S = -\frac{\Delta V}{\Delta T} = \frac{E}{\nabla T}$$

Thomas Johann Seebeck, born in Reval (today Tallinn, Estonia) (1770 – 1831) was a physicist who in 1821 discovered the thermoelectric effect, where a junction of dissimilar metals produces an electric current when exposed to a temperature gradient. This effect is the basis of thermocouples and thermopiles.

Large thermoelectric effect in graphene

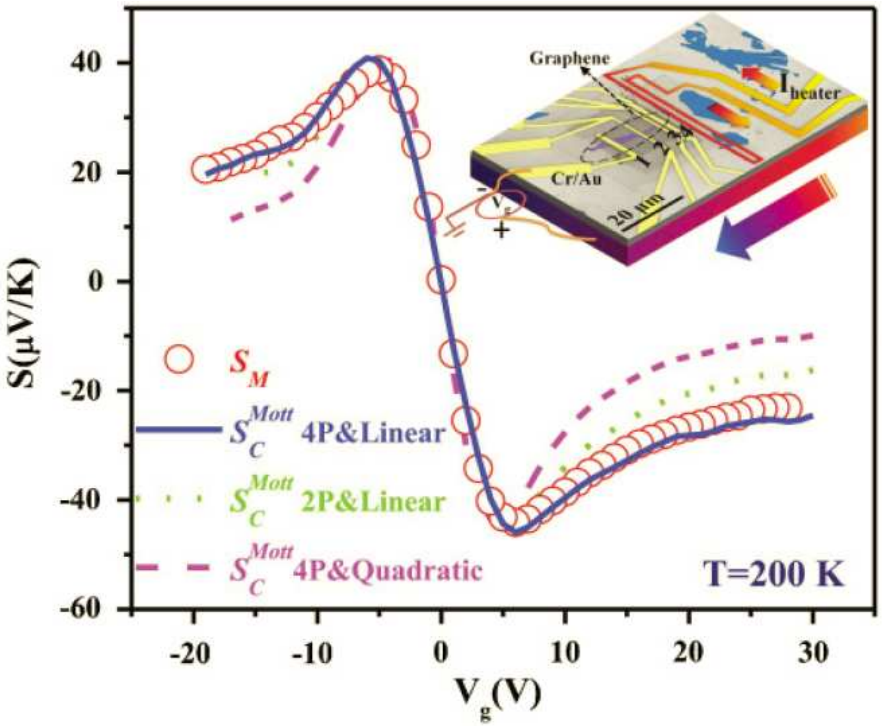


FIG. 1. (Color online) Comparison of experimentally measured Seebeck coefficient S_M (open circles) and three Seebeck curves S_C^{Mott} calculated from measured electrical conductivity using the Mott relation. The solid line is calculated with the 4P resistivity and a linear dispersion relation; the dotted line is with the two-point (2P) resistivity and a linear dispersion relation; and the dashed line is with the 4P resistivity and a quadratic dispersion relation. μ_c of this device is $\sim$1500 cm^2/Vs. The inset shows a false colored scanning electron microscopy image.

Wang, Shi, PRB **83**, 113403 (11).

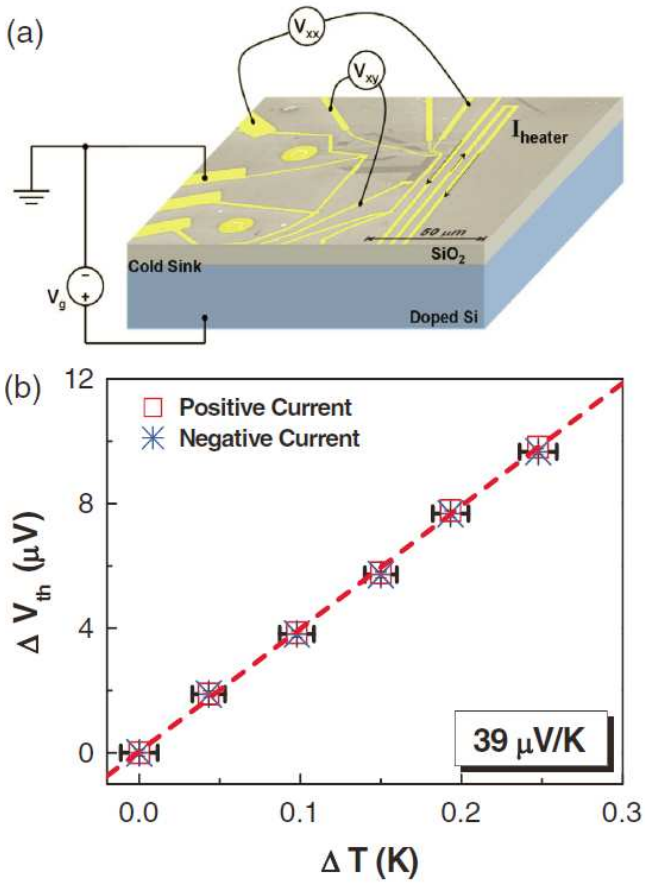


FIG. 1 (color). (a) SEM image and circuit schematic of a graphene device for thermoelectric measurements. (b) ΔT vs thermovoltage change ΔV_{th} for a series of heater power steps at 255 K and zero gate voltage. The linear fit of this curve gives the thermopower of 39 μV/K.

Wei *et al.*, PRL **102**, 166808 (09).

Heat and electric transport equations

Electric field $\mathbf{E}$ and temperature gradient ∇T result in electric and heat currents.

$$\begin{cases} \mathbf{j} &= \sigma \mathbf{E} + \beta \nabla T, \\ \mathbf{q} &= \gamma \mathbf{E} + \zeta \nabla T, \end{cases}$$

It is easier to control $\mathbf{j}$ rather than $\mathbf{E}$, express via $\mathbf{j}$.

$$\begin{cases} \mathbf{E} &= \rho \mathbf{j} + S \nabla T, \\ \mathbf{q} &= \Pi \mathbf{j} - \kappa \nabla T, \end{cases}$$

Onsager relation: $\gamma = -\beta T$

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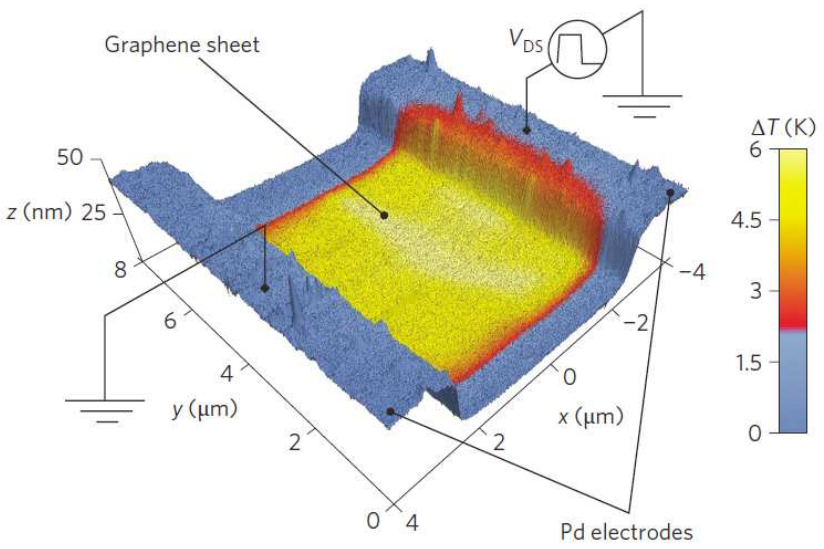
Approximate Mott's formula:

$$\beta = \frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\partial \sigma}{\partial \mu} \quad \Longrightarrow \quad S = -\frac{\pi^2}{3} \frac{k_B^2 T}{e} \frac{\partial \ln \sigma}{\partial \mu}$$

Notice that $k_B/e \approx 86 \mu V/K$ close to observed in graphene which is much larger than in metals.

Peltier effect in graphene nanoelectronics



The temperature of the graphene device during device operation.
K.L. Grosse *et al.*, Nature Nanotechnology **6**, 287 (11).

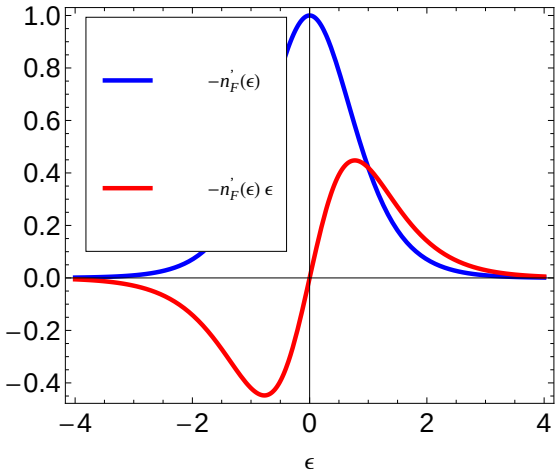
Specific heat per unit time $W = \rho j^2 + (\Pi_a - \Pi_b) j$, where $\rho = 1/\sigma$ is the resistivity.
Thermoelectric effect in graphene accounts for up to one-third of the contact temperature changes and thus it can play significant role in cooling down of such systems.

Odd- and evenness of transport coefficients

Normal metal case

$$\sigma = \frac{e^2}{3}\int_{-\infty}^{\infty} d\epsilon[-n_F'(\epsilon)]v_F^2\nu(\mu+\epsilon)\tau(\mu+\epsilon) \approx \frac{e^2}{3}[v_F^2\nu(\mu)\tau(\mu)]$$

$$\beta = \frac{e}{3T}\int_{-\infty}^{\infty} d\epsilon\,\epsilon\,[-n_F'(\epsilon)]v_F^2\nu(\mu+\epsilon)\tau(\mu+\epsilon)$$

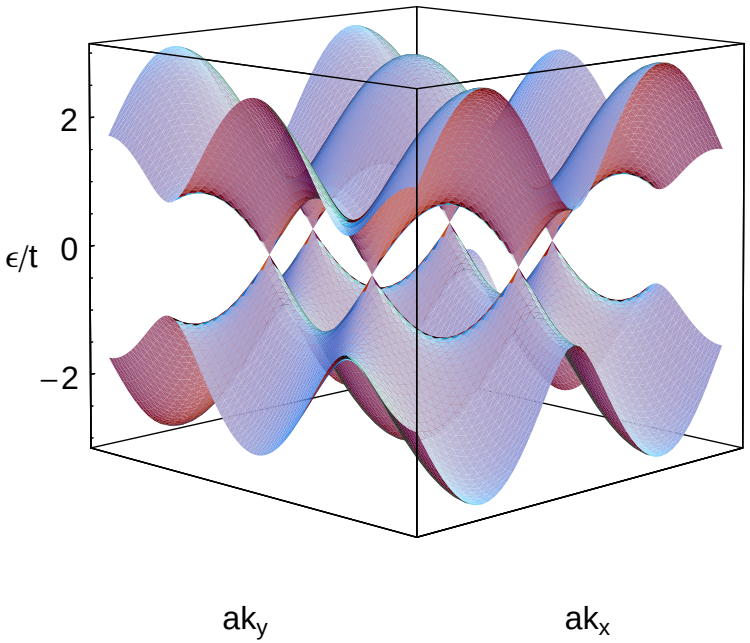


If the product $v_F^2\nu(\mu+\epsilon)\tau(\mu+\epsilon)$ is a smooth function of ϵ, one can expand it:
$v_F^2\nu(\mu+\epsilon)\tau(\mu+\epsilon) \approx$
$v_F^2\nu(\mu)\tau(\mu) + \epsilon\frac{d}{d\mu}[v_F^2\nu(\mu)\tau(\mu)]$
1st term $= 0$ due to oddness, and contributes 2nd.

Arrive at Mott's formula and $S = -\frac{\pi^2}{3}\frac{k_B}{e}\frac{k_B T}{\mu} \sim 10^{-3}\mu V/K$ much smaller than observed in graphene.

Band structure of graphene

$$H = -t \sum_{\mathbf{n},\boldsymbol{\delta}_i,\sigma} \left[a^{\dagger}_{\mathbf{n},\sigma} b_{\mathbf{n}+\boldsymbol{\delta},\sigma} + \text{c.c.} \right]$$



Two bands touch each other and cross the Fermi level in six **K** points located at the corners of the hexagonal 2D Brillouin zone.

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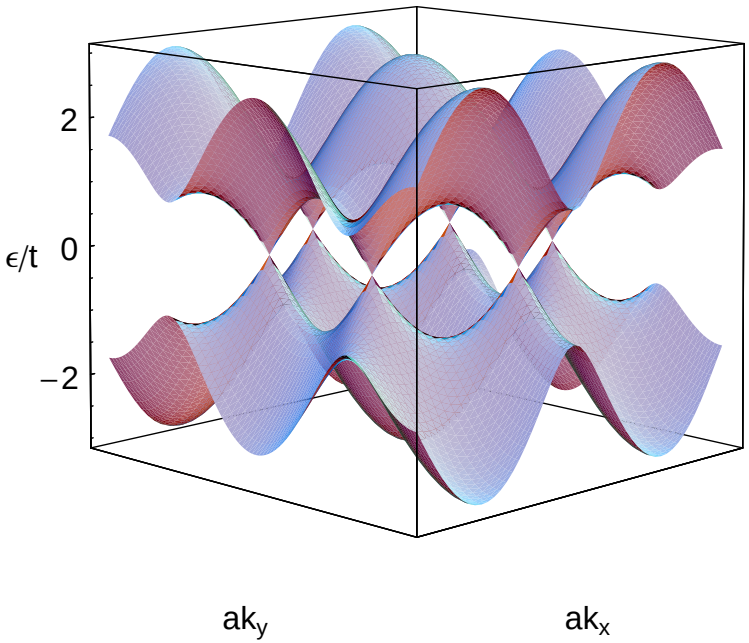

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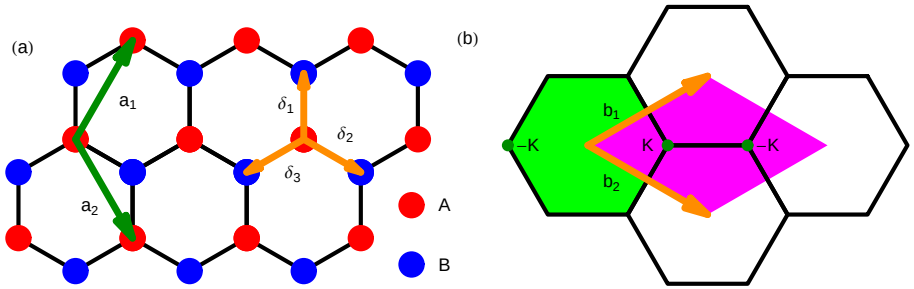

(a) Graphene hexagonal lattice can be described in terms of two triangular sublattices, A and B.
(b) Hexagonal and rhombic extended Brillouin zone (BZ).
Two non-equivalent **K** points in the extended BZ, $\mathbf{K}_- = -\mathbf{K}_+$.
P.R. Wallace, PR **71**, 622 (1947).

Low-energy excitations in graphene

Low-energy excitations at two inequivalent K_+, K_- points have a linear dispersion $E_p = \pm\hbar v_F|\mathbf{p}| - \mu$ with $v_F \approx 10^6\,\mathrm{m/s}$ and μ being the chemical potential.
Each K point described by the spinor: $\psi^T_{K,\sigma} = \left(\psi_{KA\sigma}, \psi_{KB\sigma} \right)$

$$H_{K_+} = \sum_{\sigma=\pm1} \int \frac{d^2p}{(2\pi)^2} \psi^\dagger_{K_+\sigma} \begin{pmatrix} 0 & \hbar v_F(p_x - ip_y) \\ \hbar v_F(p_x + ip_y) & 0 \end{pmatrix} \psi_{K_+\sigma}.$$

where the momentum $\mathbf{p} = (p_x, p_y)$ is already given in a local coordinate system
Semenoff, PRL **53**, 2449 (1984)

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K_+ and K_- points are related by time-reversal symmetry.
What is about spatial inversion $\mathcal{P}$? Graphene's Hamiltonian does not break it, but let's break it by making sublattices inequivalent...

Is there a gap in graphene?

$$H_{K_+} = \sum_{\sigma=\pm 1} \int \frac{d^2p}{(2\pi)^2} \psi^\dagger_{K_+\sigma} \begin{pmatrix} \Delta & \hbar v_F(p_x - ip_y) \\ \hbar v_F(p_x + ip_y) & -\Delta \end{pmatrix} \psi_{K_+\sigma}.$$

The presence of $\Delta \neq 0$ breaks $\mathcal{P} : [x \to -x, y \to -y, A \leftrightarrows B]$ and makes the spectrum $E(\mathbf{p}) = \pm\sqrt{\hbar^2 v_F^2 \mathbf{p}^2 + \Delta^2}$ with the mass Δ.

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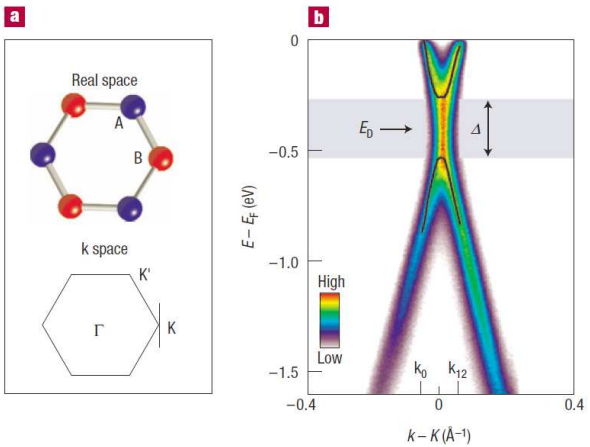
Gap can be induced by interaction with substrate.



S.Y. Zhou *et al.*, Nature Mat. **6**, 770 (07).

D.A. Siegel *et al.* (12), graphene on Cu.

Observation of the gap opening in single-layer epitaxial graphene on a SiC substrate at the K point.
(a) Structure of graphene in the real and momentum space.
(b) ARPES intensity map taken along the black line in the inset of (a).

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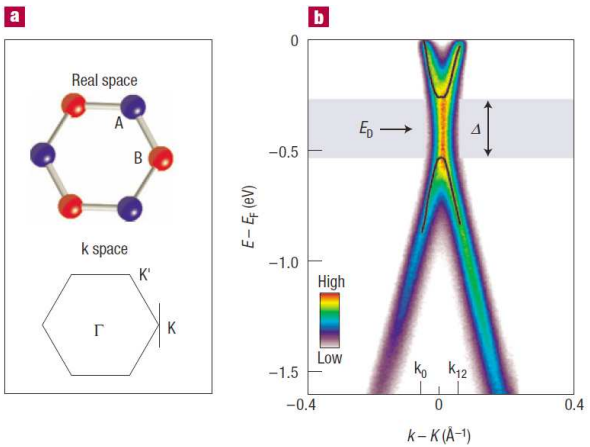

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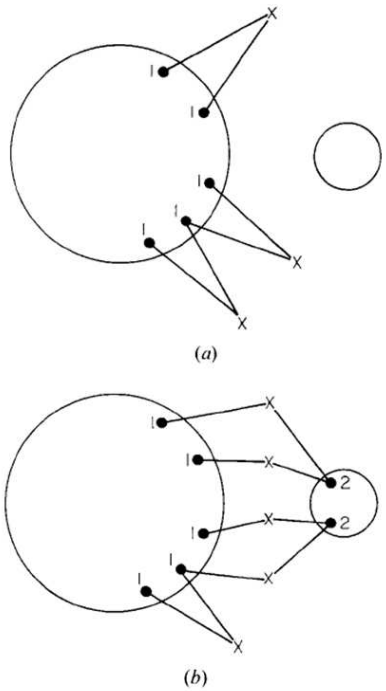
How the gap affects thermopower?

Quasiparticle scattering near ETT



Possible types of electron scattering for a double valley Fermi surface.
A.A. Varlamov, V.S. Egorov, and A.V. Pantsulaya, Adv. in Phys. **38**, 469 (1989).

(a) Scattering processes which do not involve the small valley.
(b) Scattering processes where electron gets to the small void, but then gets back to the continuous part of the Fermi surface.

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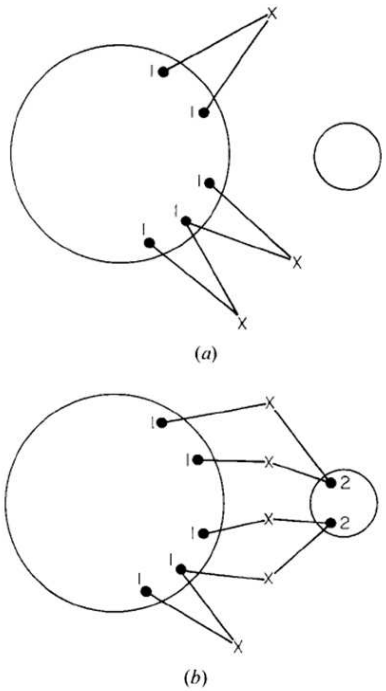

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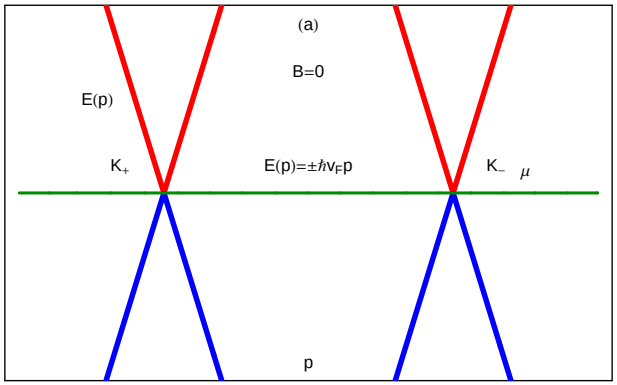
In vicinity of the critical point $\mu = \mu_c$, when the Fermi surface connectivity changes, the quasiparticle relaxation rate $\tau^{-1}(\varepsilon) \equiv \Gamma(\varepsilon)$ also acquires the contribution strongly depending on energy, what generates kinks in conductivity and peaks in thermopower.

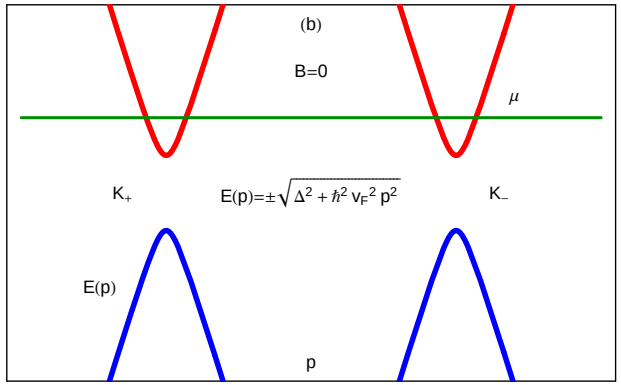
Scattering in gapped graphene

Zero mass, $\Delta = 0$



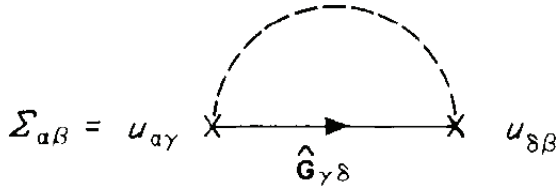
(a) Linear dispersion, $\mu = 0$ as in compensated graphene.

Gapped, $\Delta \neq 0$



(b) A possible modification of the spectrum by the finite gap Δ. μ is shifted from zero by the gate voltage.

Self-consistent equation for self-energy:



Use relatively long-range potential $\widehat{V}(\mathbf{q})$, i.e. ignore scattering between $\mathbf{K}_{\pm}$, but assume $\widehat{V}(\mathbf{q})$ to be momentum independent for the intra-valley scattering.

Control parameter: $|\Delta| < ?? > |\mu|$

Quasiparticle scattering in graphene

The self-energy $\widehat{\Sigma}(\mathbf{p},\varepsilon_n)=\sum_{i=0}^{3}\sigma_i(\mathbf{p},\varepsilon_n)\widehat{\tau}_i$ Since $\sigma_{1,2}=0$, arrive at the system

$$\left\{\begin{array}{c}\sigma_0^R(\varepsilon)\\ \sigma_3^R(\varepsilon)\end{array}\right\}=\frac{4\hbar}{\pi\tau_0|\mu|}\int_0^W\frac{\left\{\begin{array}{c}\varepsilon+\mu-\sigma_0^R(\varepsilon)\\ \Delta+\sigma_3^R(\varepsilon)\end{array}\right\}\xi d\xi}{\left[\varepsilon+\mu-\sigma_0^R(\varepsilon)\right]^2-\xi^2-\left[\Delta+\sigma_3^R(\varepsilon)\right]^2},$$

A new feature, in addition to the usually considered Eq. for σ_0 we also consider Eq. for σ_3 in the gap channel. Then approximately include both channels together:

$$\frac{1}{\tau(\varepsilon)}\equiv\Gamma(\varepsilon)=-\mathrm{Im}\,\sigma_0^R(\varepsilon)-\frac{\Delta}{\varepsilon+\mu}\mathrm{Im}\,\sigma_3^R(\varepsilon)$$
$$=\Gamma_0\left[\frac{|\varepsilon+\mu|}{|\mu|}+\frac{\Delta^2}{|\varepsilon+\mu||\mu|}\right]\theta\left[(\varepsilon+\mu)^2-\Delta^2\right].$$

The relaxation rate acquires the $\theta\left[(\varepsilon+\mu)^2-\Delta^2\right]$ contribution.

Transport coefficients in graphene

Using Kubo formula:

$$\left\{ \begin{array}{c} \sigma \\ \beta \end{array} \right\} = \frac{e^2}{\hbar} \int_{-\infty}^{\infty} \frac{d\varepsilon \mathcal{A}(\varepsilon, \Gamma(\varepsilon), \Delta)}{2T \cosh^2 \frac{\varepsilon}{2T}} \left\{ \begin{array}{c} 1 \\ \varepsilon/(eT) \end{array} \right\},$$

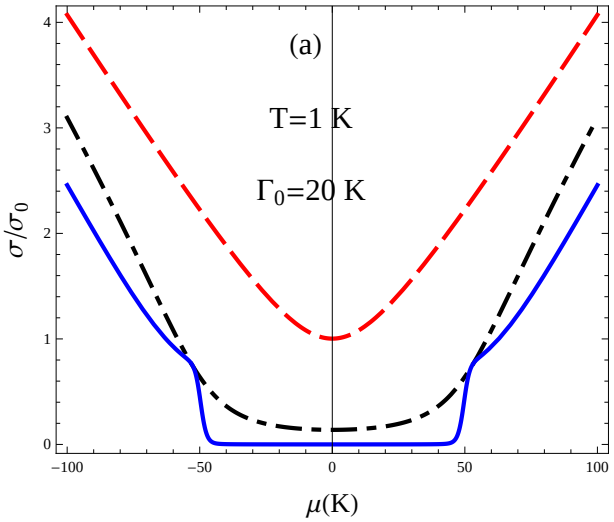
where the function

$$\mathcal{A}(\varepsilon, \Gamma(\varepsilon), \Delta) = \frac{1}{2\pi^2} \left[1 + \frac{(\mu+\varepsilon)^2 - \Delta^2 + \Gamma^2(\varepsilon)}{2|\mu+\varepsilon|\Gamma(\varepsilon)} \times \left(\frac{\pi}{2} - \arctan \frac{\Delta^2 + \Gamma^2(\varepsilon) - (\mu+\varepsilon)^2}{2|\mu+\varepsilon|\Gamma(\varepsilon)} \right) \right].$$

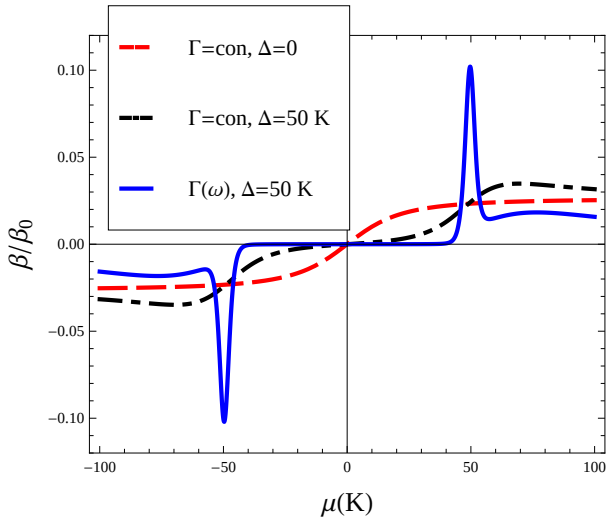
We use regularized scattering rate: $\Gamma^{\mathrm{full}}(\varepsilon) = \Gamma(\varepsilon) + \gamma_0$.

Results

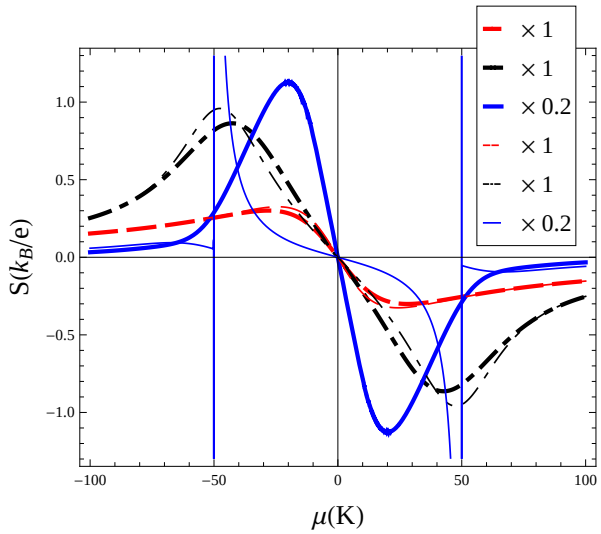
Conductivity $\sigma(\mu)$



Tm.-el. coefficient $\beta(\mu)$



Thermopower $S(\mu)$



In units $\sigma_0 = \frac{2e^2}{\pi^2\hbar}$ $\beta_0 = k_B e/\hbar,\ T = 1\,K$ $S_0 = k_B/e,\ T = 5\,K$

— $\Delta = 0$, $\Gamma(\varepsilon) =$ const - reference case: restore normal metal case, $S = -(\pi^2/3e)\,T/\mu$ in the limit $|\mu| \gg T, \Gamma_0$.

— $\Delta = 50\,K$, $\Gamma(\varepsilon) =$ const: E. Gorbar *et al.*, PRB **66**, 045108 (02).

— $\Delta = 50\,K$, $\Gamma(\varepsilon)$ - present work.

Thin lines – from Mott formula.

Conclusions

- Opening a gap results in appearance of a fingerprint bump of the Seebeck signal when the chemical potential approaches the gap edge.
- Magnitude of the bump can be up 10 times higher than already large value of $S \sim 50 \mu V/K$ at room temperatures observed in graphene.
- Effect is related to a new channel of quasi-particle scattering from impurities with the relaxation time strongly dependent on the energy.
- One can exploit the predicted giant peak of the Seebeck signal as a signature of the gap opening in monolayer graphene.
- Similar phenomenon already observed in bilayer graphene.