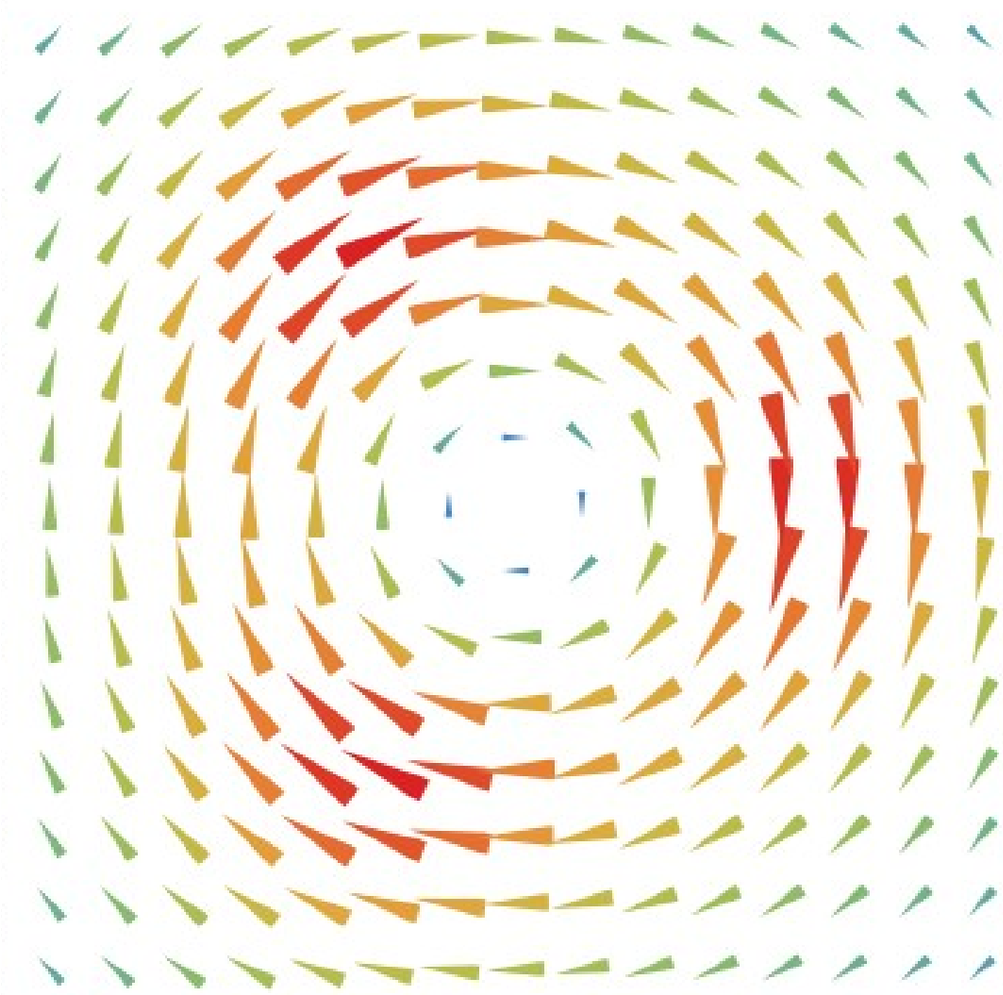
Spin coherence in graphene

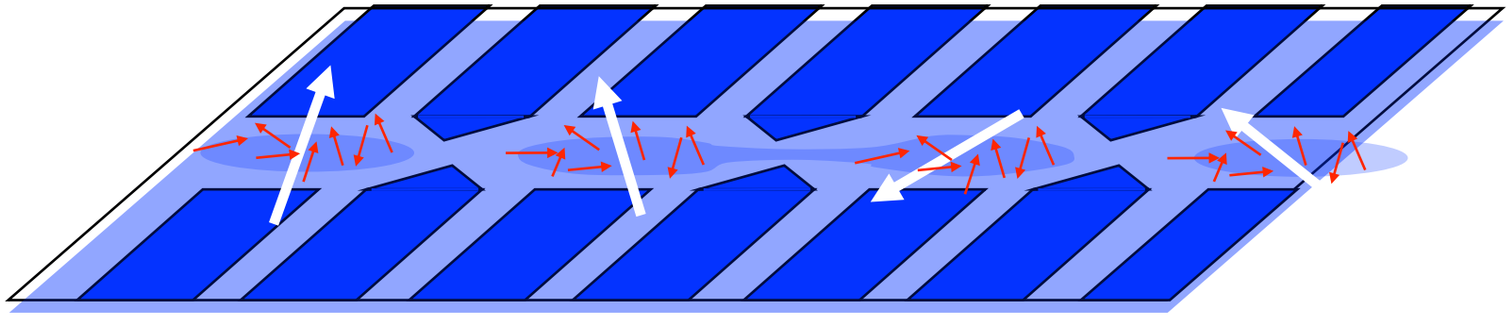


Guido Burkard

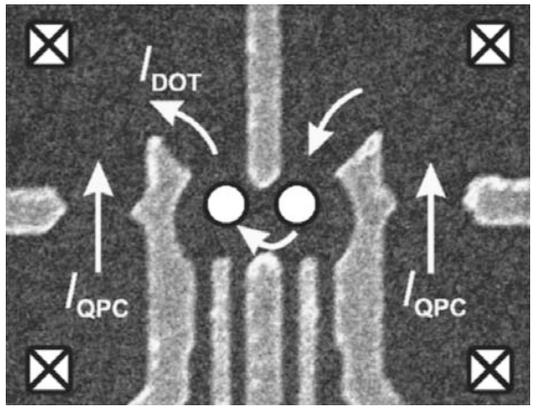
University of Konstanz, Germany

Universität Konstanz

Spin coherence & spin qubits

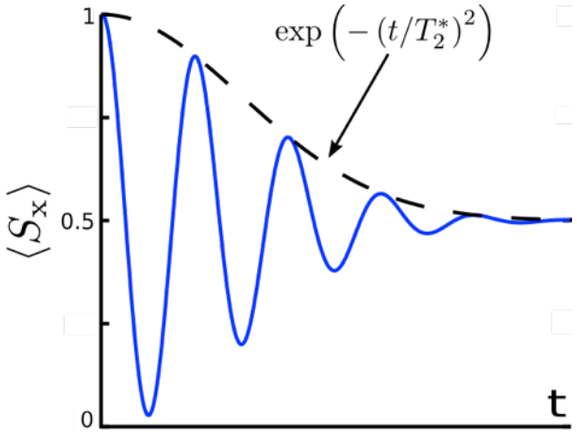


$$H_{\text{exch}} = \sum_{\langle i,j \rangle} J_{ij} \mathbf{s}_i \cdot \mathbf{s}_j$$



Elzermann *et al.*, PRB 2003
see also Hanson et al., RMP 2007

nuclear spins
spin decoherence

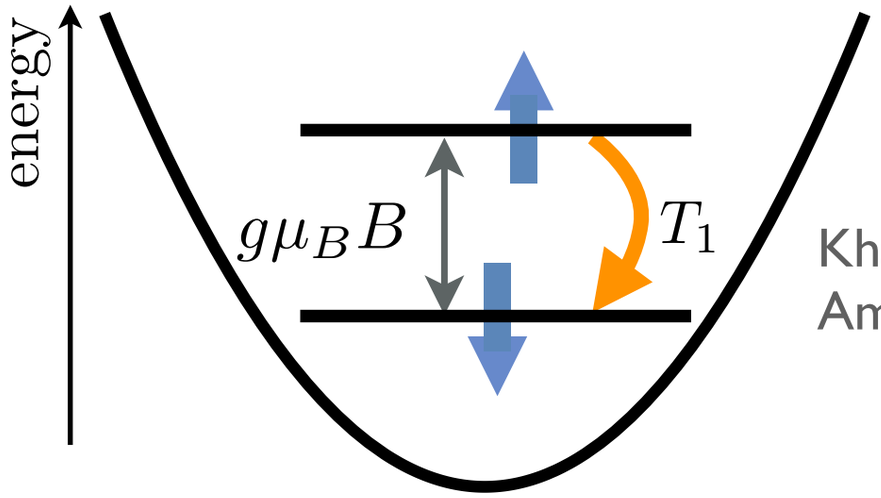


$$B \gg \delta B_{\text{nuc}}$$

$$T_2^* = \frac{\hbar}{\delta B_{\text{nuc}}}$$

Petta *et al.*, Science 2005

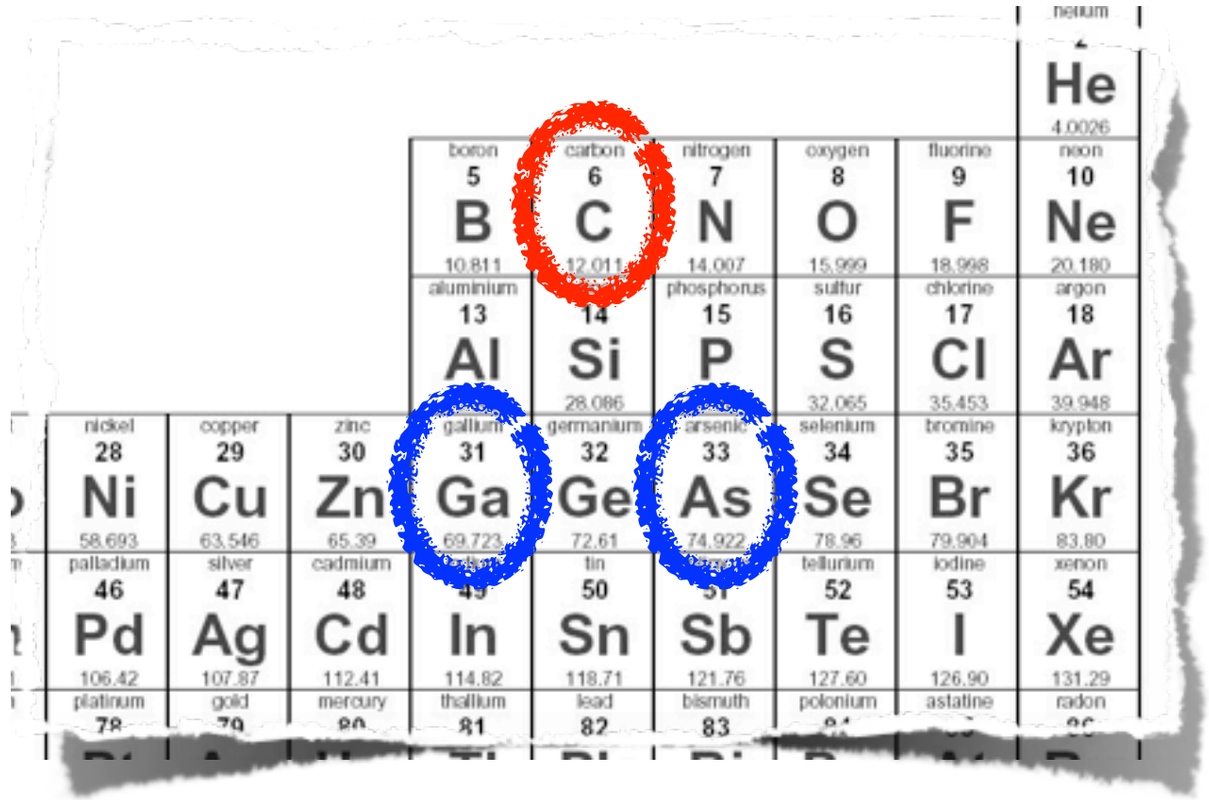
spin orbit interaction
electron phonon coupling
spin relaxation



$$T_1 \propto \frac{1}{B^5}$$

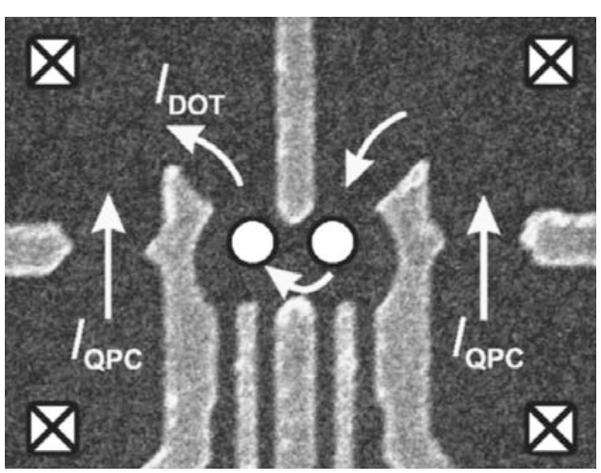
Khaetskii & Nazarov, PRB 2001
Amasha *et al.*, PRL 2008

Carbon as a material for spin qubits



99% ^{12}C I=0

1% ^{13}C I=1/2



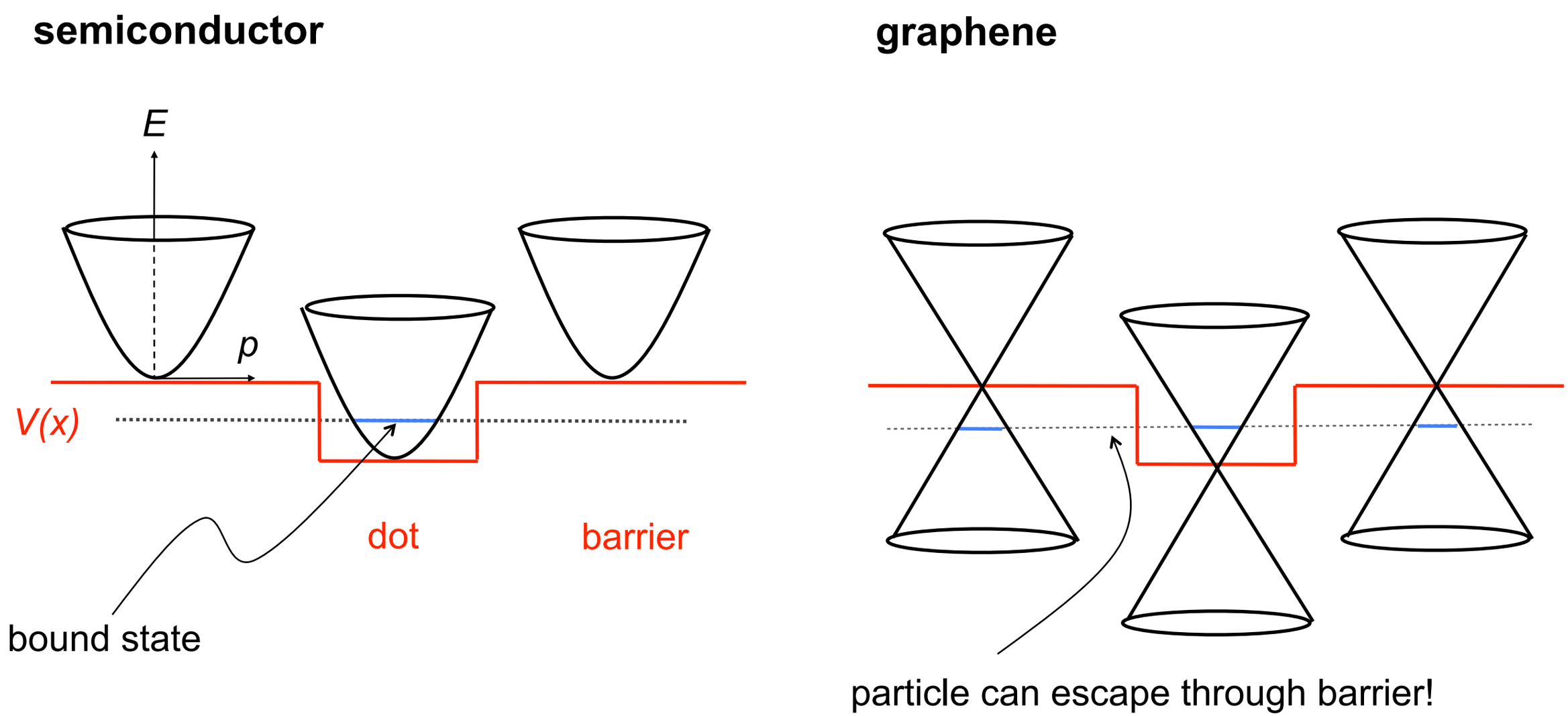
Elzermann *et al.*, PRB 2003

?

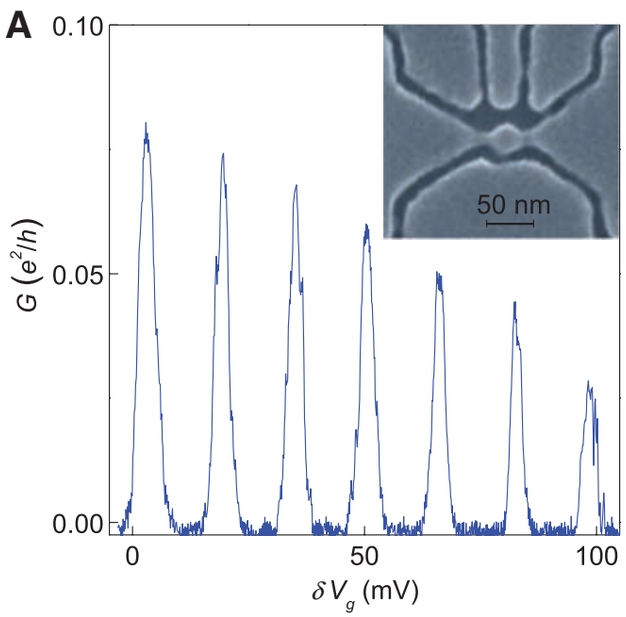
Outline

- quantum dots in graphene
- spin-valley hyperfine interaction in graphene
- spin relaxation of localized electrons
- spin relaxation of mobile electrons (spin transport)

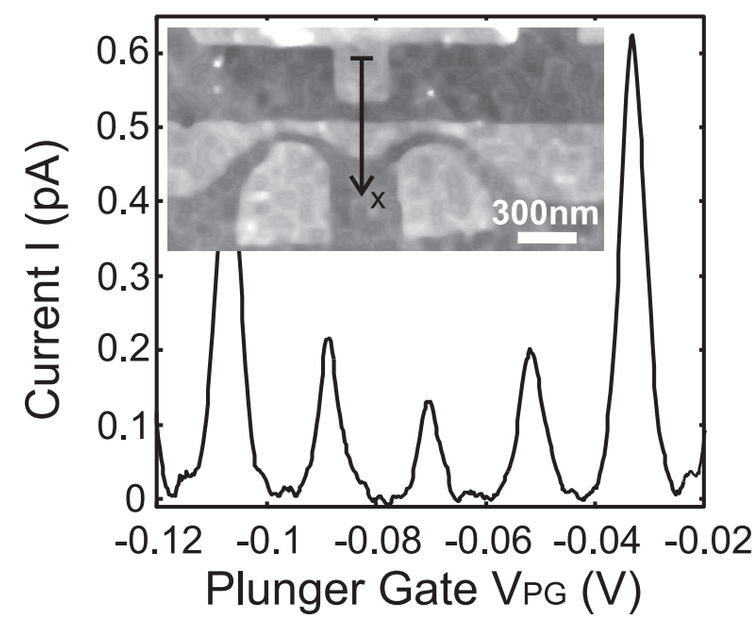
Quantum dots in graphene



Quantum dots in nanostructured graphene



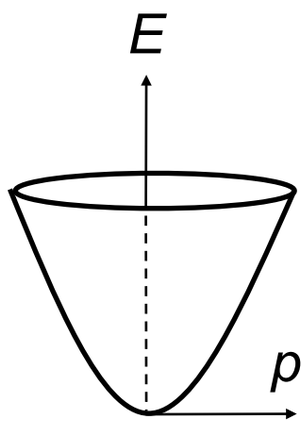
Ponomarenko *et al.*, Science 2008



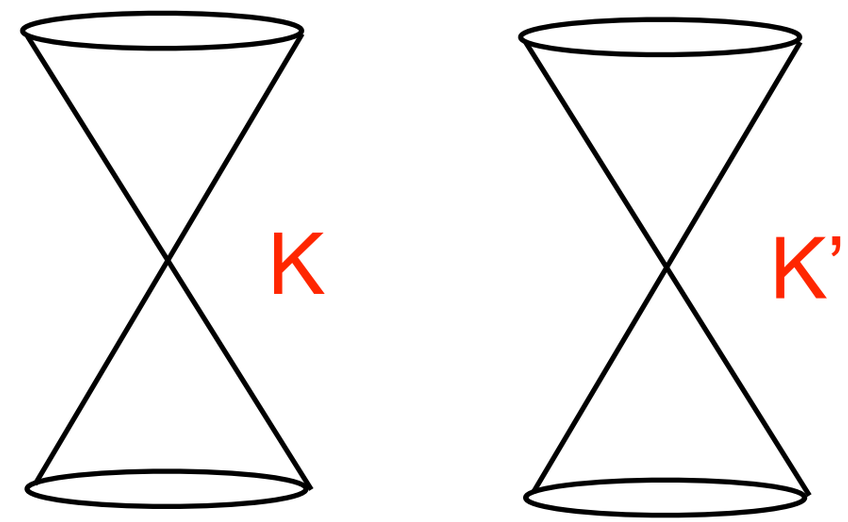
Stampfer *et al.*, APL 2008

Valley degeneracy and exchange

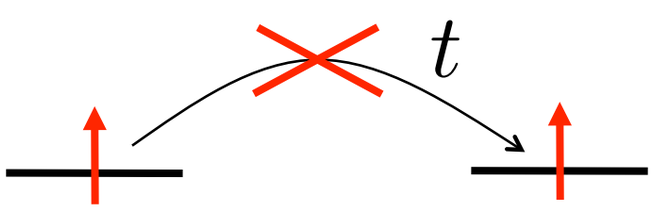
direct-gap semiconductor



graphene



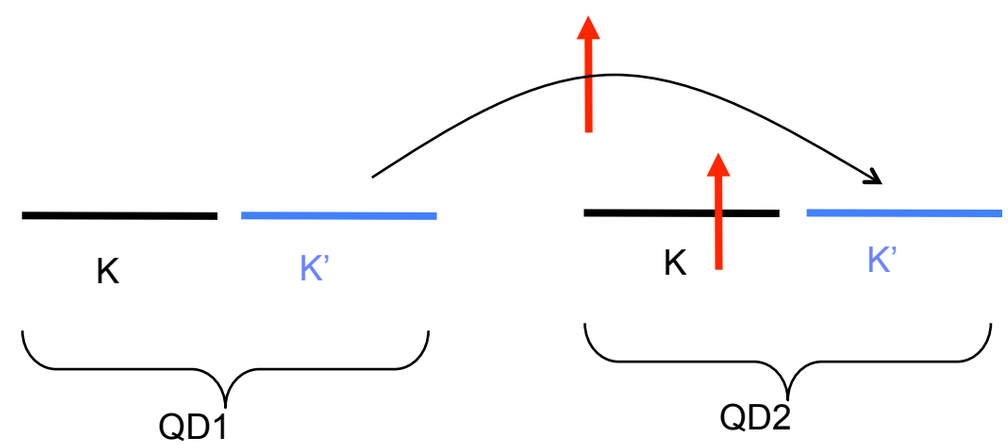
Pauli principle, exchange coupling



$J = t^2/U$

$$H_{\text{eff}} = \frac{J}{4}\vec{s}_1 \cdot \vec{s}_2$$

valley degeneracy



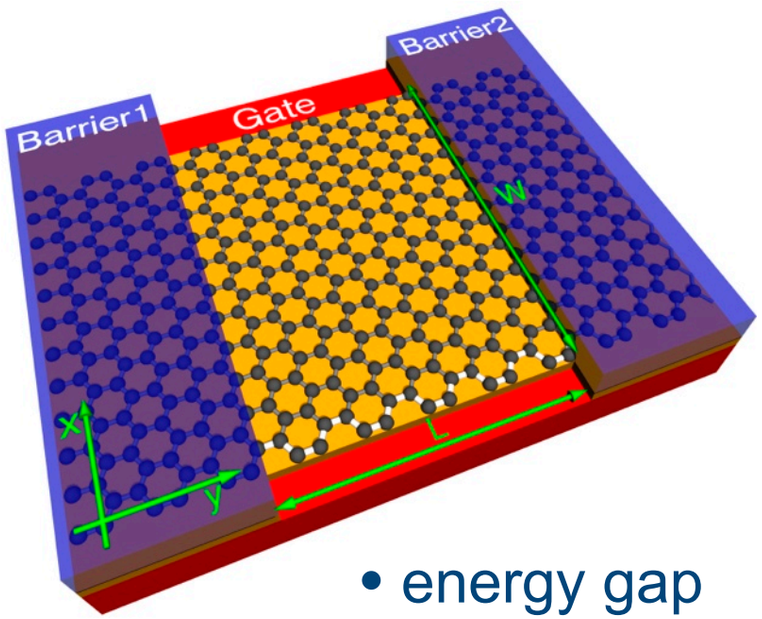
$$H_{\text{eff}} = \frac{J}{8}\left((\vec{s}_1 \cdot \vec{s}_2)(\vec{\tau}_1 \cdot \vec{\tau}_2) + \vec{s}_1 \cdot \vec{s}_2 + \vec{\tau}_1 \cdot \vec{\tau}_2 - 3\right)$$

spin-valley coupling

N. Rohling & GB, unpublished

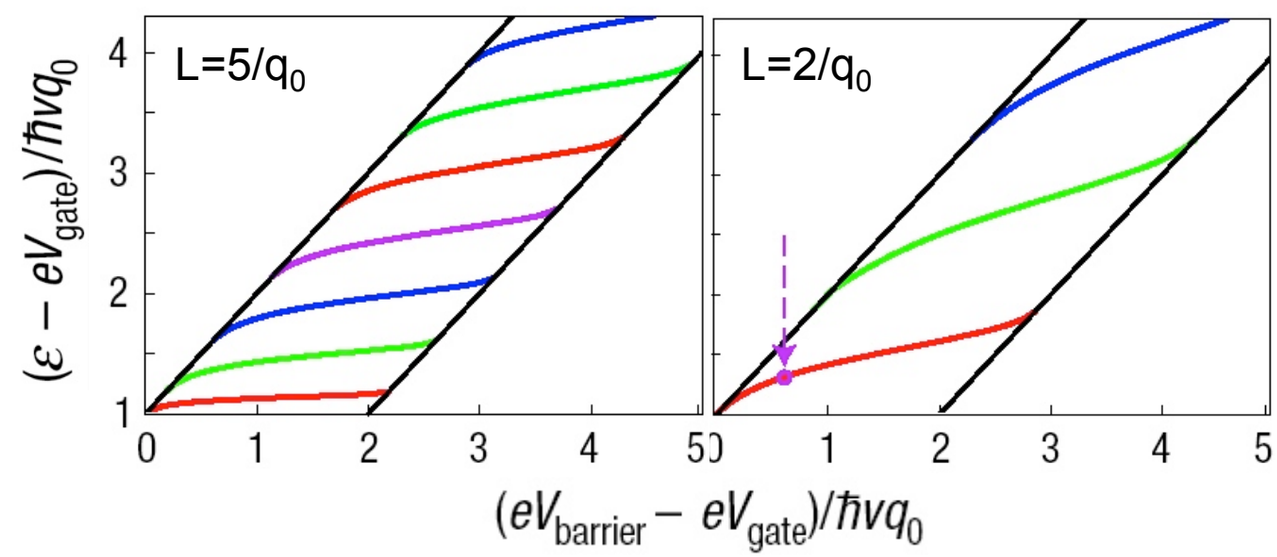
Graphene Nanoribbon QDs

Trauzettel, Bulaev, Loss & GB, Nature Physics (2007)

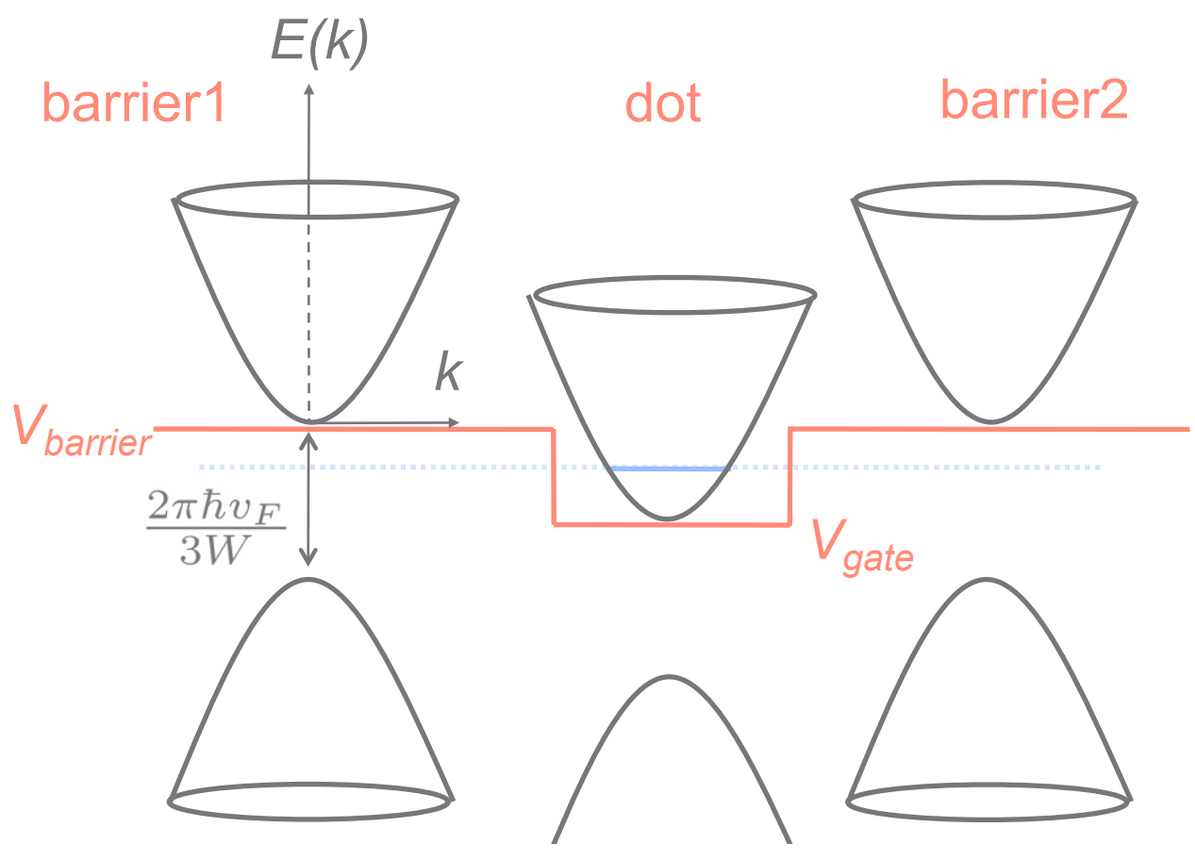


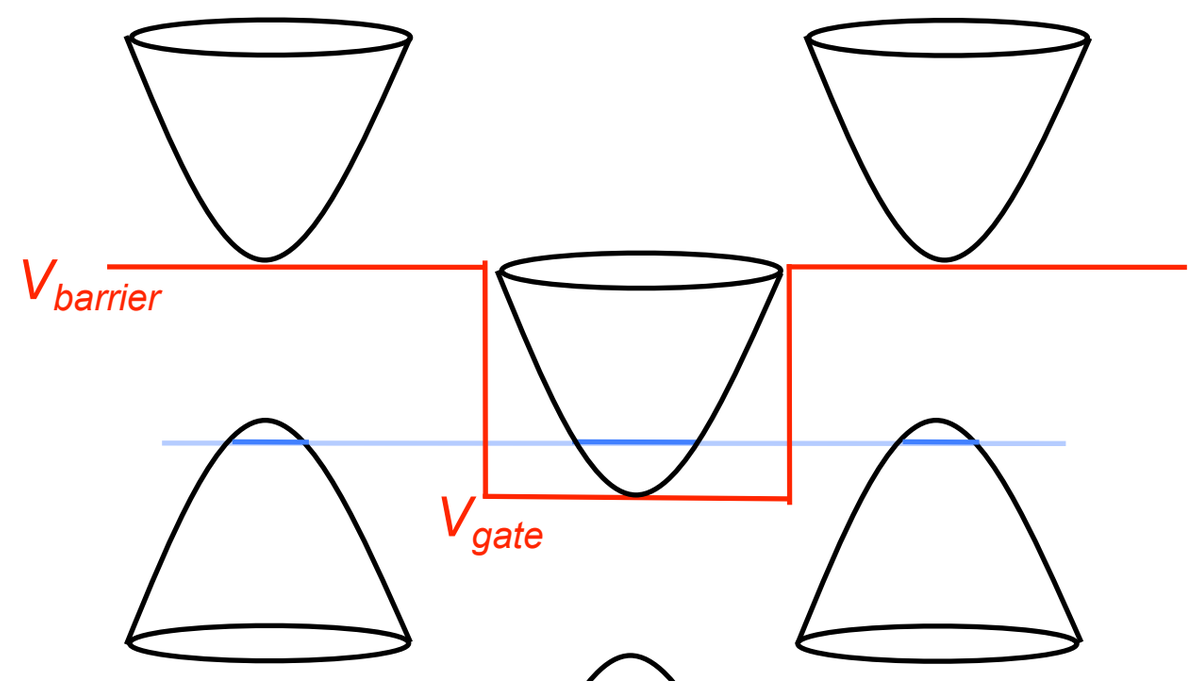
Dirac particle in a box: solve transcendental equation for ε

$$\tan(\tilde{k}L) = \frac{\hbar v \tilde{k}\sqrt{(\hbar v q_0)^2 - (\varepsilon - eV_{\text{barrier}})^2}}{(\varepsilon - eV_{\text{barrier}})(\varepsilon - eV_{\text{gate}}) - (\hbar v q_0)^2}$$

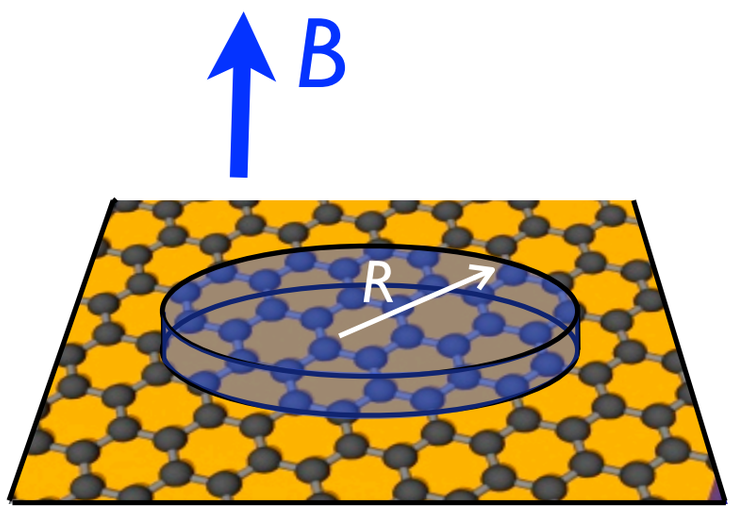


- energy gap
- broken valley degeneracy





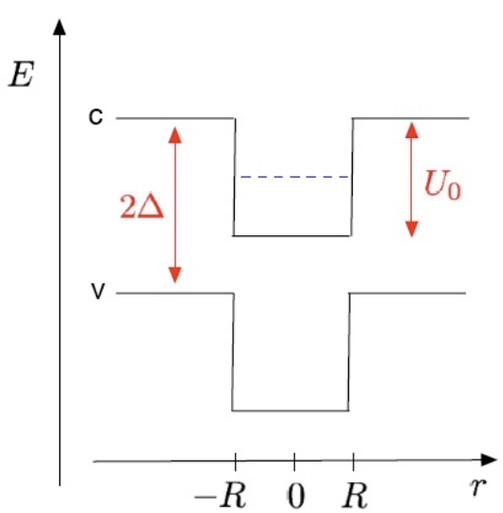
Quantum dots in gapped graphene



single layer: substrate-induced gap
Giovannetti *et al.*, Phys. Rev. B **76**, 073103 (2007).
Zhou *et al.*, Nature Mat. **6**, 770 (2007).

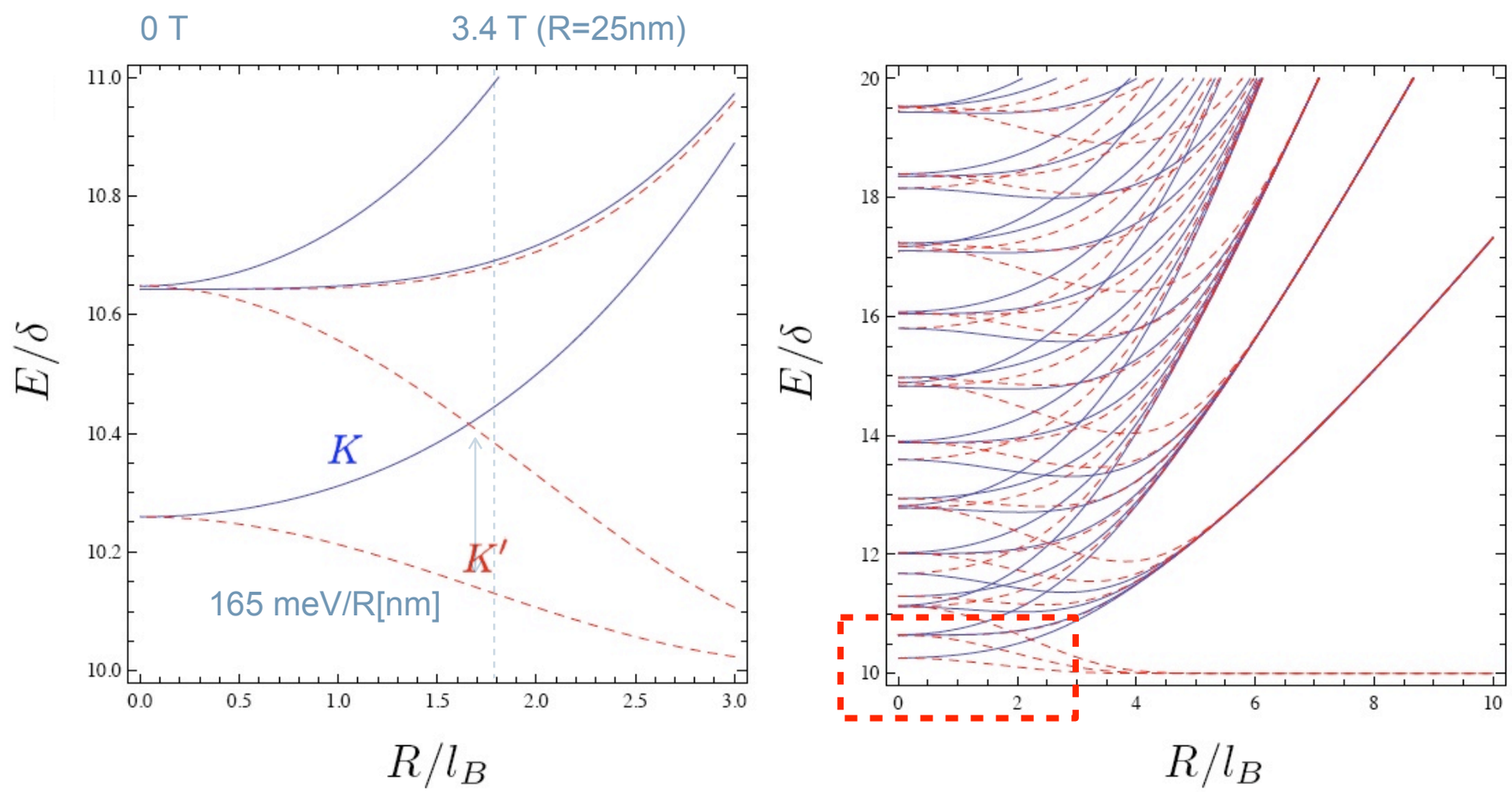
bilayer: electrically induced gap
McCann, PRB (2006).
Ohta *et al.*, Science (2006).

single layer case:



$$U_0 = \Delta$$
$$\Delta = 10\,\delta$$

$$l_B = (\hbar/eB)^{1/2}$$



Recher, Nilsson, GB & Trauzettel, PRB **79**, 085407 (2009)

Outline

- quantum dots in graphene
- **spin-valley hyperfine interaction in graphene**
- **spin relaxation of localized electrons**
- **spin relaxation of mobile electrons**

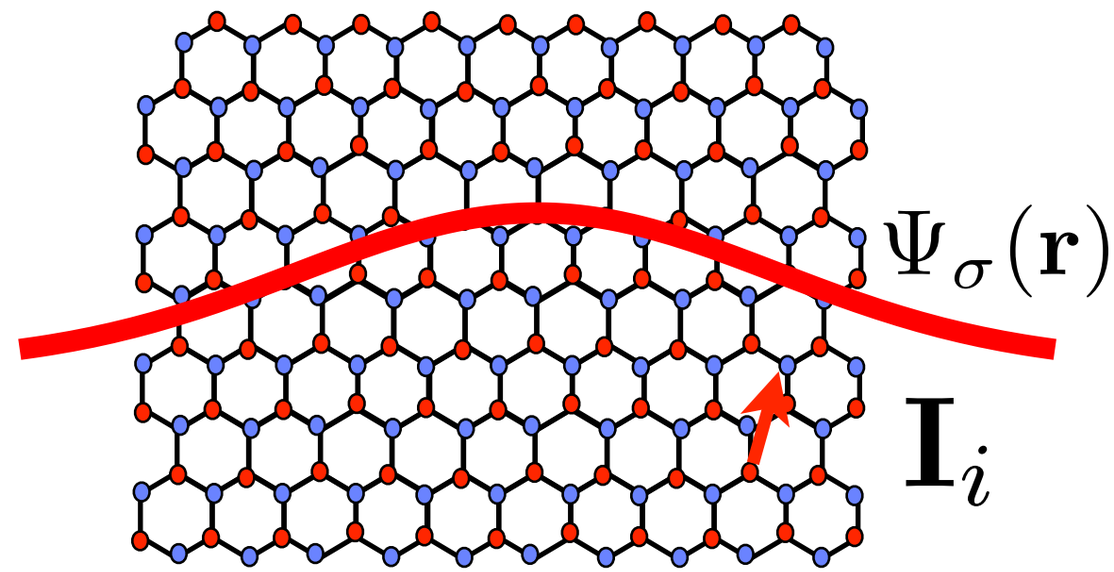
Hyperfine interaction in Graphene

main idea

- ^{13}C atom is an atomically sharp impurity

$$H_{\mathrm{hf},ij} \;=\; \delta_{ij}\boldsymbol{S}A\boldsymbol{I}_i.$$

- can take up momentum on the order of *2K*, thus leading to **inter-valley scattering**
- invisible to electron charge
- visible to spin via hyperfine coupling
- strength of coupling: same as hyperfine



Palyi & GB, PRB **80**, 201404(R) (2009)

Hyperfine interaction in Graphene

hyperfine Hamiltonian for Dirac wavefunctions:

valley mixing

K

$$H'_{\rm hf} = \Omega_{\rm cell} \boldsymbol{S} A \sum_i \boldsymbol{I}_i \begin{pmatrix} |\Psi_\sigma(\boldsymbol{r}_i)|^2 & e^{-i2Kx_i}\Psi^*_\sigma(\boldsymbol{r}_i)\Psi'_\sigma(\boldsymbol{r}_i) \\ e^{i2Kx_i}\Psi'^*_\sigma(\boldsymbol{r}_i)\Psi_\sigma(\boldsymbol{r}_i) & |\Psi'_\sigma(\boldsymbol{r}_i)|^2 \end{pmatrix}$$

valley mixing

K'

unit cell

sublattice

$$i = (l\sigma)$$

hyperfine Hamiltonian for Dirac electrons in graphene

Palyi & GB, PRB **80**, 201404(R) (2009)

$$H_{\rm hf} = \boldsymbol{S} \cdot \left(\boldsymbol{h}^{(0)} \tau_0 + \sum_{i=x,y,z} \boldsymbol{h}^{(i)} \tau_i \right)$$

valley-conserving terms
(same form as e.g. in GaAs)

valley-mixing terms (x,y)
valley dephasing term (z)

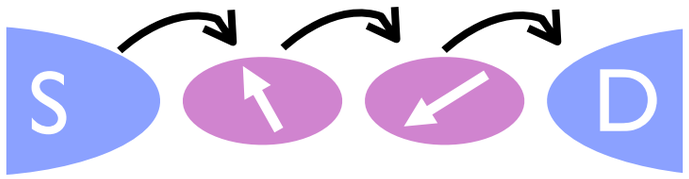
four 'nuclear fields' instead of one

$$\mathbf{h}^{(0)} = \frac{1}{2}\Omega_{\rm cell} A \sum_{l\sigma} \mathbf{I}_{l\sigma} \sum_v |\Psi^{(v)}_\sigma(\mathbf{r}_{l\sigma})|^2 \qquad \mathbf{h}^{(x)} = \Omega_{\rm cell} A \sum_{l\sigma} \mathbf{I}_{l\sigma} {\rm Re}\left[e^{2i\mathbf{K}\cdot\mathbf{r}_{l\sigma}} \Psi^*_\sigma(\mathbf{r}_{l\sigma})\Psi'_\sigma(\mathbf{r}_{l\sigma})\right]$$

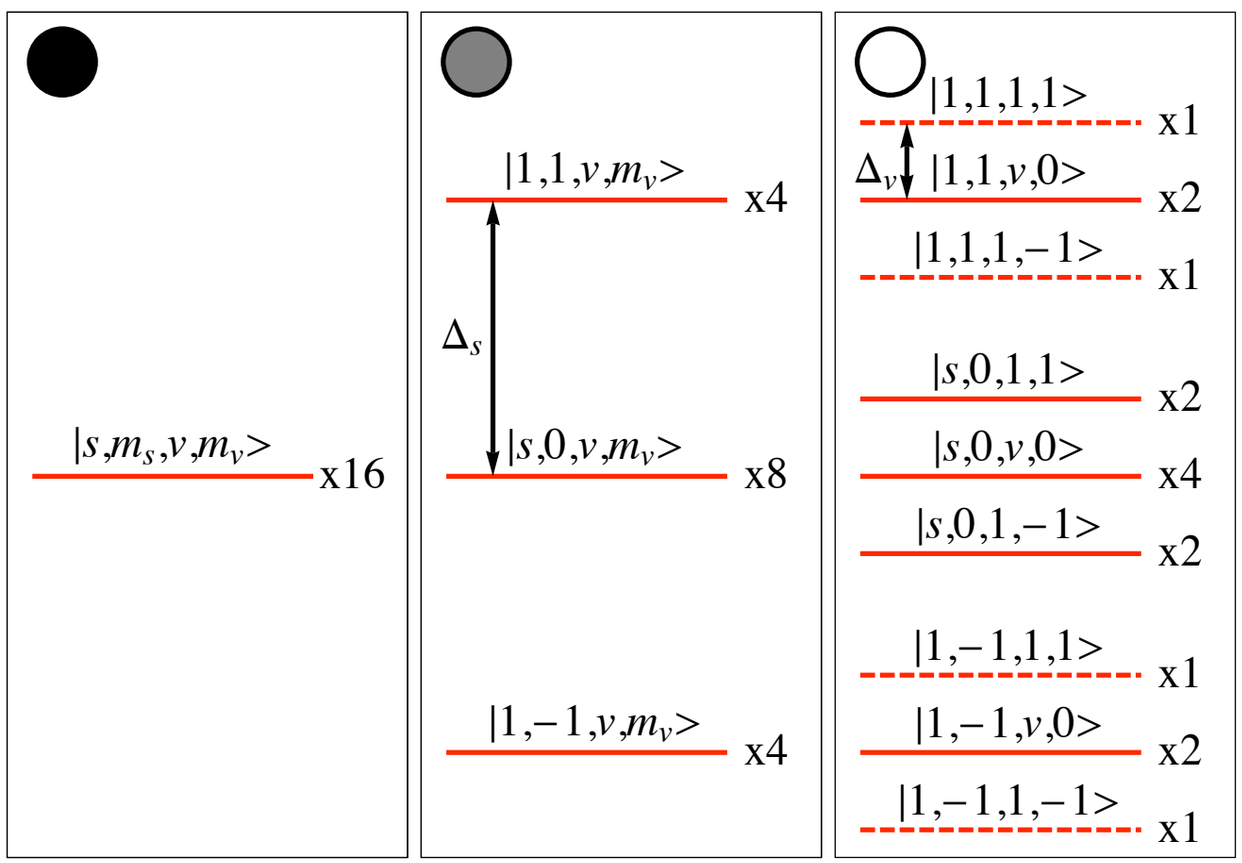
$$\mathbf{h}^{(z)} = \frac{1}{2}\Omega_{\rm cell} A \sum_{l\sigma} \mathbf{I}_{l\sigma} \sum_v v|\Psi^{(v)}_\sigma(\mathbf{r}_{l\sigma})|^2 \qquad \mathbf{h}^{(y)} = \Omega_{\rm cell} A \sum_{l\sigma} \mathbf{I}_{l\sigma} {\rm Im}\left[e^{2i\mathbf{K}\cdot\mathbf{r}_{l\sigma}} \Psi^*_\sigma(\mathbf{r}_{l\sigma})\Psi'_\sigma(\mathbf{r}_{l\sigma})\right]$$

Spin blockade in graphene dots

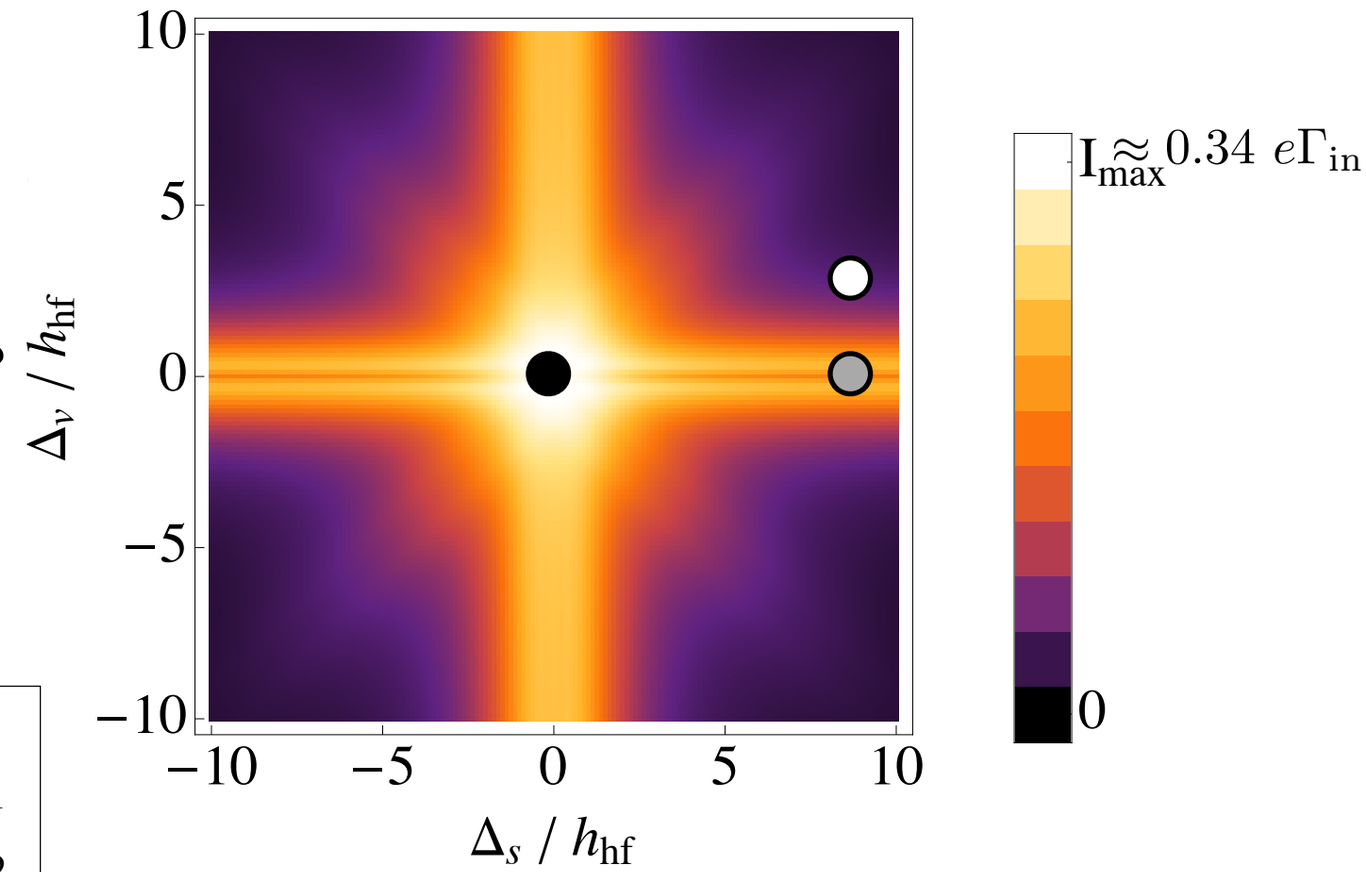
Pályi & GB, PRB 2009



- effect of **hyperfine** interaction on leakage current?
- no disorder and no spin-orbit interaction
- incoherent interdot tunneling



Energy level diagram in the (1,1) charge configuration

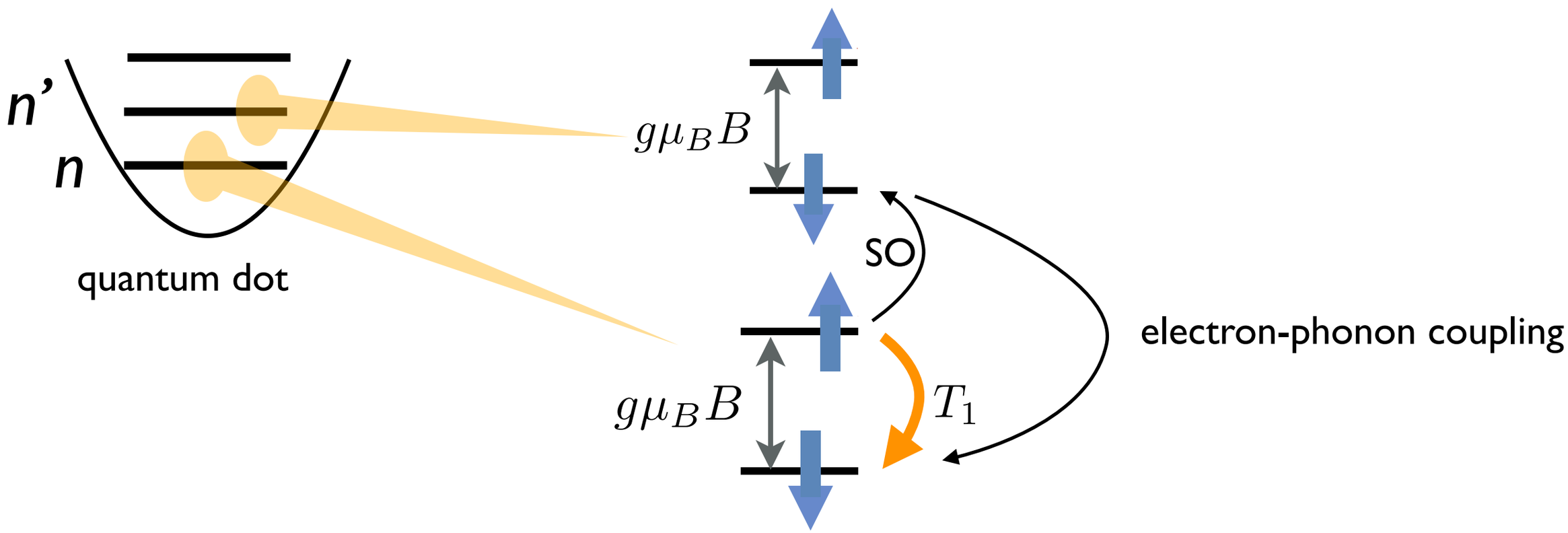


●	$\Delta_s = 0$, $\Delta_v = 0$	hf-induced **spin and valley** mixing no pure supertriplets **blockade lifted**
●	$\Delta_s \gg h_{\rm hf}$, $\Delta_v = 0$	hf-induced **valley** mixing no pure supertriplets **blockade lifted**
○	$\Delta_s \gg h_{\rm hf}$, $\Delta_v \gg h_{\rm hf}$	4 pure supertriplets (- - - - - - - -) **transport blocked**

Outline

- quantum dots in graphene
- spin-valley hyperfine interaction in graphene
- spin relaxation of localized electrons
- spin relaxation of mobile electrons

Spin relaxation of localized electrons



- spin-orbit (SO) interaction + electron phonon coupling (EPC), piezo-phonons
- van Vleck cancellation at B=0: qubit states form Kramers doublet van Vleck, Phys. Rev. 1940

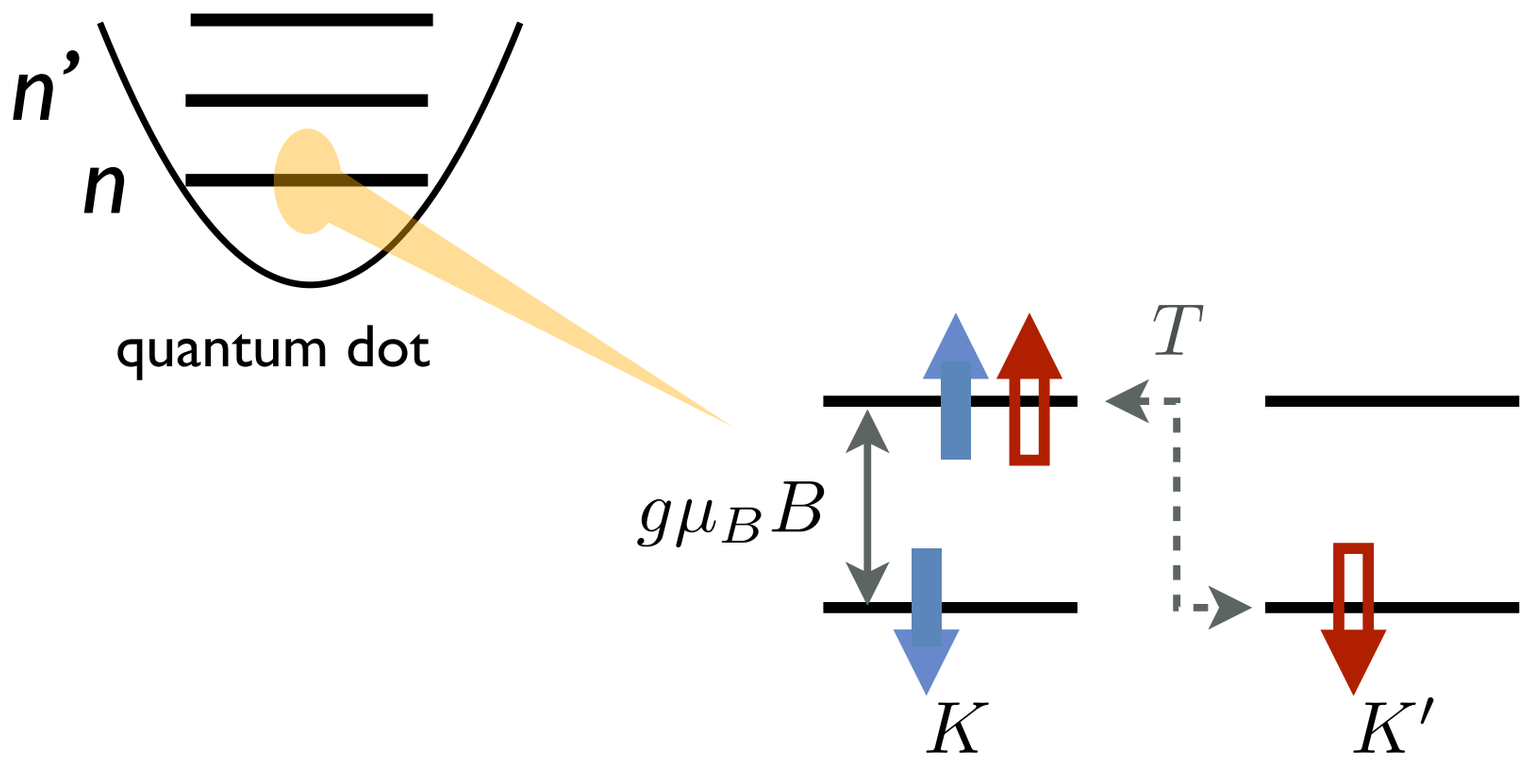
at B=0: $T_1 \to \infty$

B≠0: "at least" $T_1 \propto B^{-2}$ typical regime (GaAs): $T_1 \propto B^{-5}$

Khaetskii & Nazarov, PRB 2001
Amasha *et al.*, PRL 2008

graphene QDs?

Spin relaxation in graphene QDs



in some graphene QDs: valley degeneracy (*K, K'*):

(1) time reversal invariance at *B=0* intact,
but Kramers pair resides in different valleys

(2) either: Kramers qubit
or: pure spin qubit in one valley

spin qubit
there is **no van Vleck cancellation**

Spin-orbit interaction in graphene

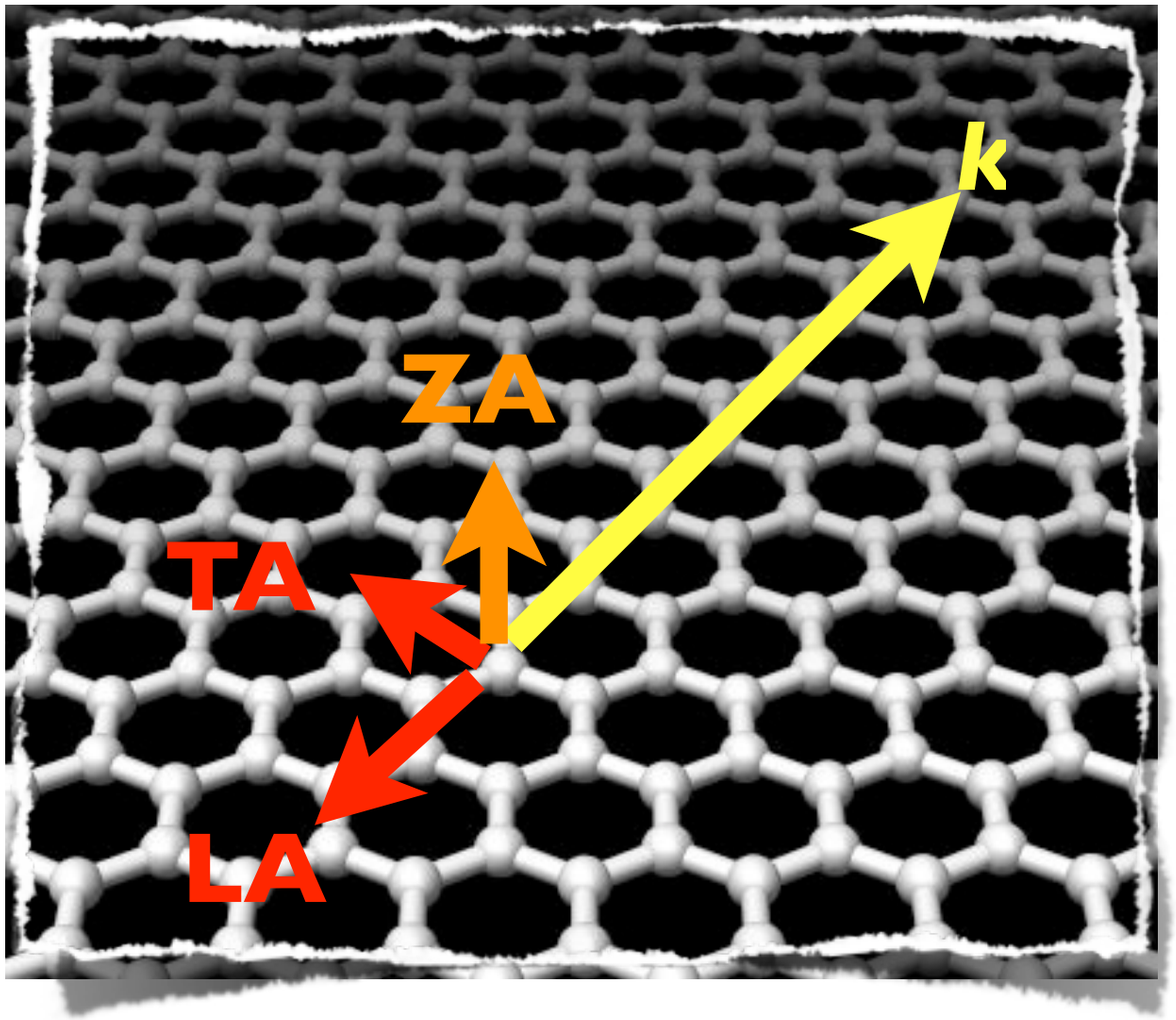
$$H_{\mathrm{SO}} = H_{\mathrm{i}} + H_{\mathrm{R}} = \Delta_{\mathrm{i}} \tau \sigma_z s_z + \Delta_{\mathrm{R}} (\tau \sigma_x s_y - \sigma_y s_x)$$

Kane & Mele, PRL 2005
Huertas-Hernando, Guinea & Brataas, PRB 2006
Castro Neto & Guinea PRL 2009
Gmitra *et al.* PRB 2009

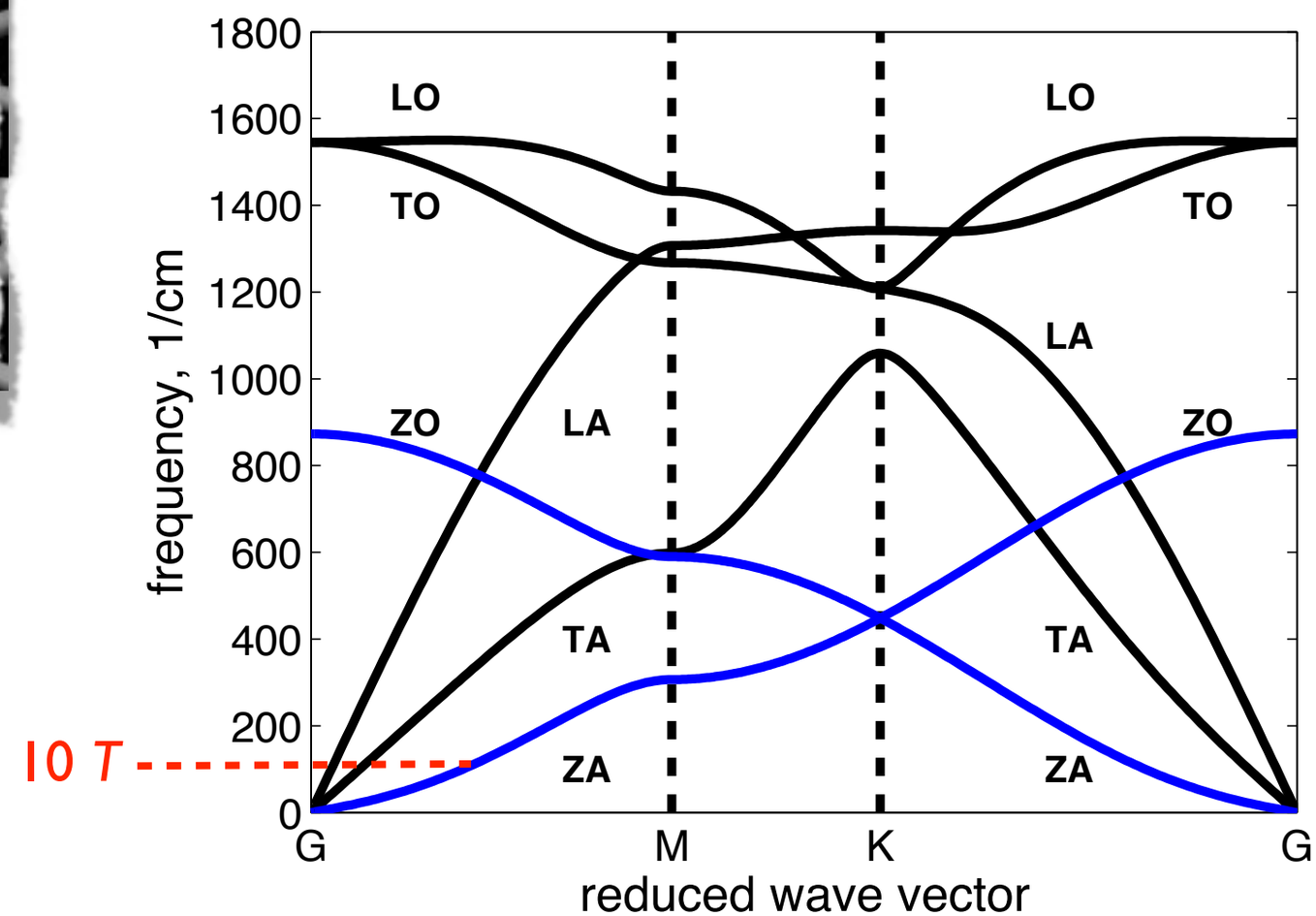
Rashba SOI
- tunable with external E-field
- selection rule $\Delta j = \pm 1$

intrinsic SOI
- selection rule $\Delta j = 0$

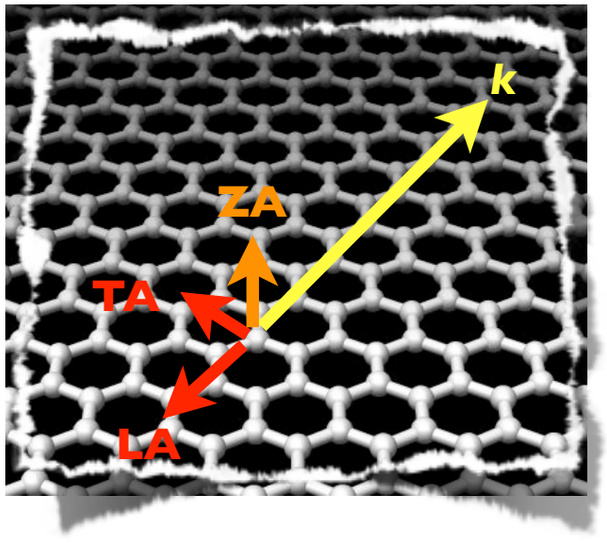
Phonons in Graphene



Falkovsky, JETP 2007 [cond-mat/0702409]



Electron-phonon coupling



(0) no piezo phonons!

(1) deformation potential
(only LA phonons, g_1~30 eV)

$$H_{\mathrm{EPC},1} = \frac{i g_1 q}{\sqrt{A\rho\omega_{\mathbf{q},\mu}}} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \left(e^{i\mathbf{qr}} b^\dagger - e^{-i\mathbf{qr}} b\right)$$

(2) bond-length change
(LA, and TA, g_2~1.5 eV)

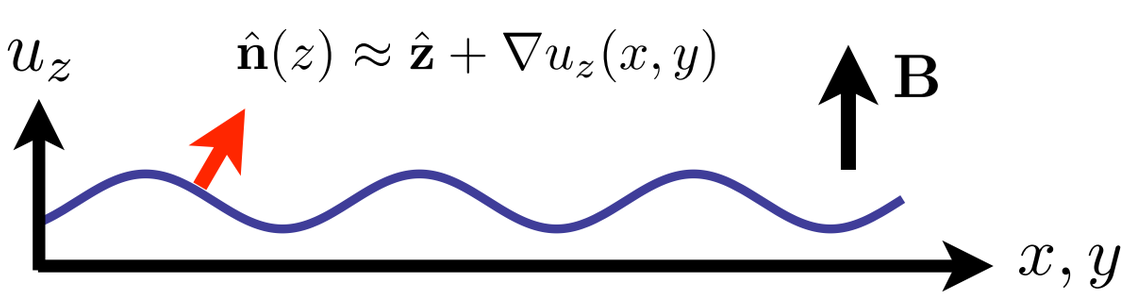
$$H_{\mathrm{EPC},2} = \frac{i g_2 q}{\sqrt{A\rho\omega_{\mathbf{q},\mu}}} \begin{pmatrix} 0 & e^{2i\phi_q} \\ e^{-2i\phi_q} & 0 \end{pmatrix} \left(e^{i\mathbf{qr}} b^\dagger - e^{-i\mathbf{qr}} b\right)$$

Ando, J. Phys. Soc. Jpn. 2005
Suzuura & Ando, PRB 2002
Mariani & von Oppen, PRL 2008.

(3) direct spin-phonon coupling (ZA phonons) Struck & GB, PRB 2010.

$$H_{\mathrm{SO}} = H_{\mathrm{i}} + H_{\mathrm{R}} = \Delta_{\mathrm{i}} \tau\sigma_z s_z + \Delta_{\mathrm{R}} (\tau\sigma_x s_y - \sigma_y s_x)$$

deflection coupling in CNTs: Rudner & Rashba, PRB 2010.



$$H_{\mathrm{i}} = H_{\mathrm{i}}^{(0)} + \Delta_{\mathrm{i}} \left(\partial_x u_z s_x + \partial_y u_z s_y\right) \sigma_z \tau$$

$$H_{\mathrm{R}} = H_{\mathrm{R}}^{(0)} + \Delta_{\mathrm{R}} \left(-\sigma_y \partial_x u_z + \tau\sigma_x \partial_y u_z\right) s_z$$

ZA phonons: $u_z = \sqrt{1/A\rho\omega_{\mathbf{q}}} \left(e^{i\mathbf{q}\cdot\mathbf{r}} b^\dagger + e^{-i\mathbf{q}\cdot\mathbf{r}} b\right)$

Relaxation rate

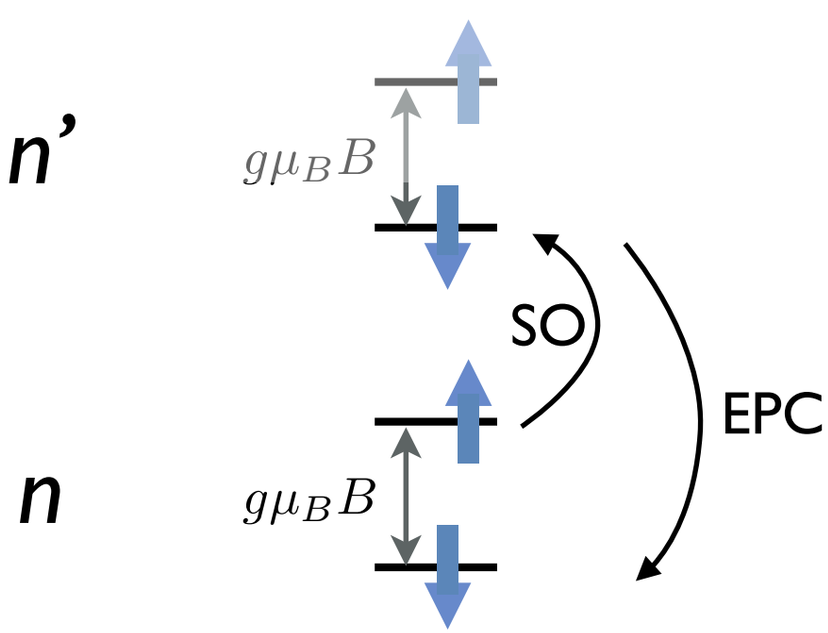
Fermi's Golden Rule $$\Gamma = 2\pi A \int \frac{\mathrm{d}^2 q}{(2\pi)^2} \left| (H_{\mathrm{EPC}})^{\uparrow\downarrow}_{nn} \right|^2 \delta(\omega_{\mathbf{q}} - g\mu_B B)$$

matrix elements $$(H_{\mathrm{EPC}})^{\uparrow\downarrow}_{nn} = \sum_{n' \neq n} \left[\frac{(H_{\mathrm{SO}})^{\uparrow\downarrow}_{nn'} (H_{\mathrm{EPC}})_{n'n}}{E_n - E_{n'} - \frac{1}{2} g\mu_B B} + \frac{(H_{\mathrm{EPC}})_{nn'} (H_{SO})^{\uparrow\downarrow}_{n'n}}{E_n - E_{n'} + \frac{1}{2} g\mu_B B} \right]$$

(LA, TA)

$$i\Delta_{\mathrm{i}} \sqrt{1/A\rho\omega_{\mathbf{q}}} \left(q_x \langle \uparrow | s_x | \downarrow \rangle + q_y \langle \uparrow | s_y | \downarrow \rangle \right) \langle n | \sigma_z e^{i\mathbf{q}\cdot\mathbf{r}} | n \rangle$$

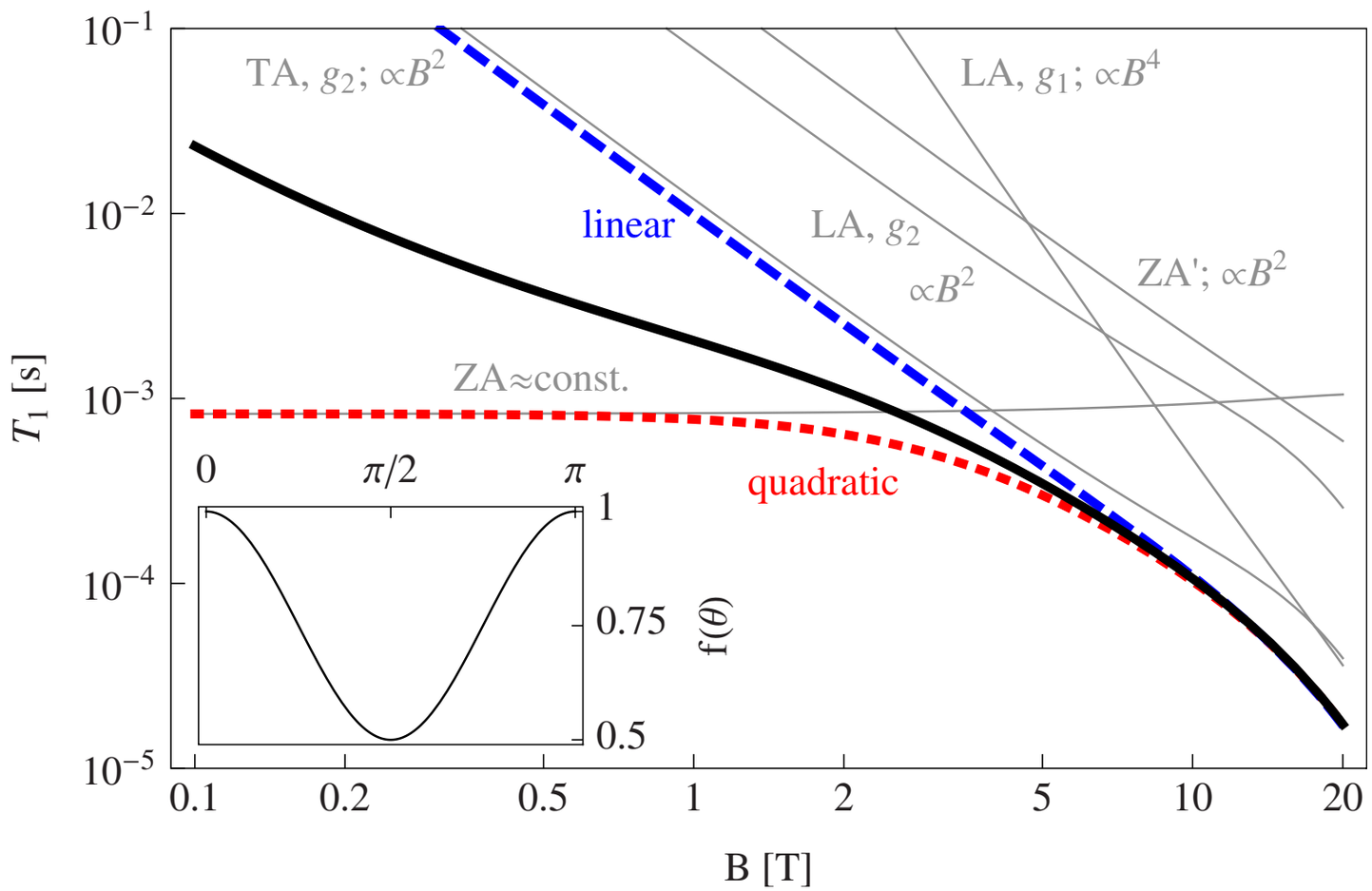
(ZA)



Spin relaxation in graphene QDs

Struck & GB, PRB (2010)

- spin relaxation T_1 in circular graphene QD
- no van Vleck cancellation
- B-independent T_1 at low B for quadratic dispersion of ZA mode, crossover to B^2 or B^4



$$U_0 = \Delta = 260\,\mathrm{meV}$$
$$R = 25\,\mathrm{nm}$$

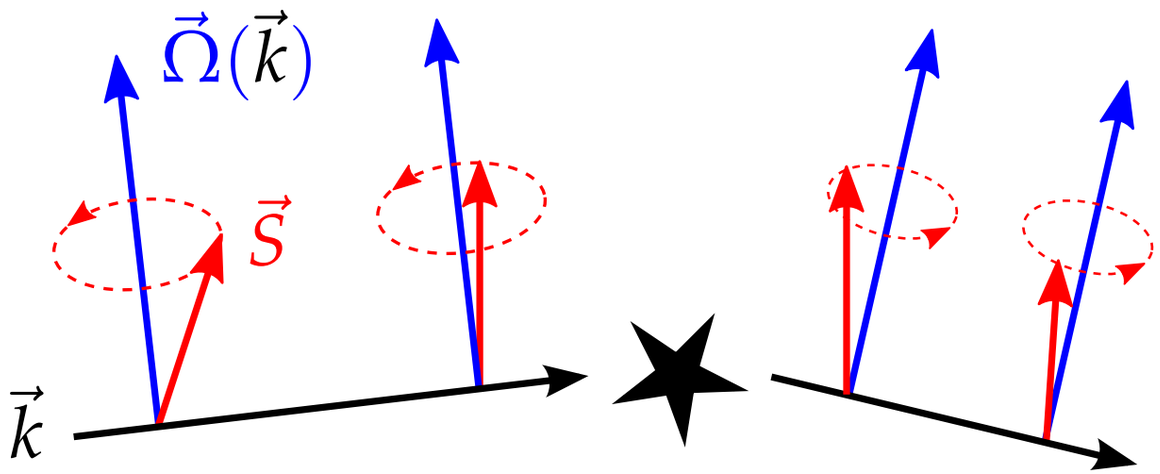
Outline

- quantum dots in graphene
- spin-valley hyperfine interaction in graphene
- spin relaxation of localized electrons
- spin relaxation of mobile electrons

Spin transport relaxation mechanisms

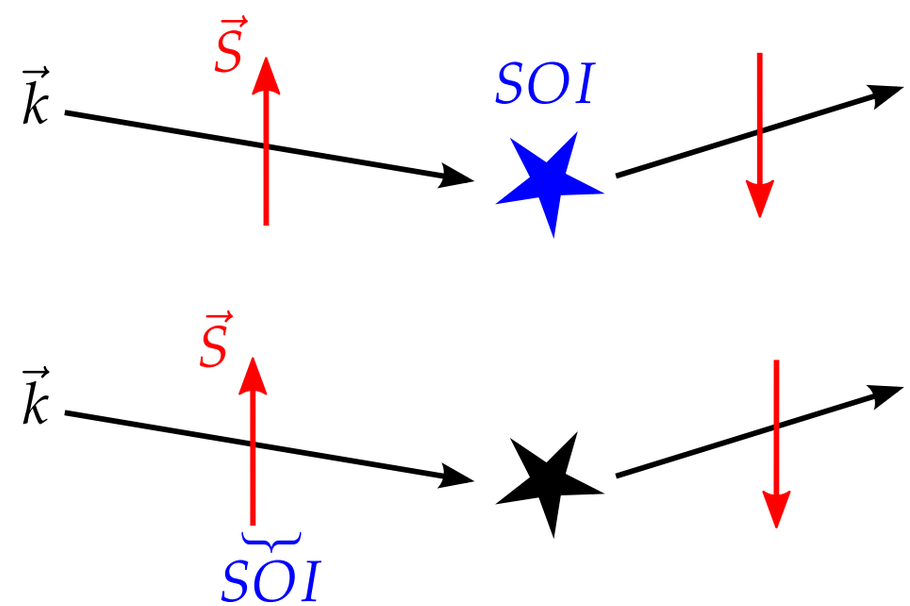
D'yakonov-Perel'

$\tau_S \propto 1/\tau_p$



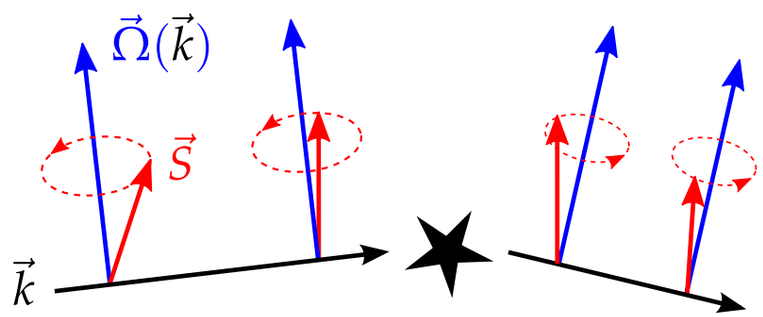
Elliot-Yafet

$\tau_S \propto \tau_p$

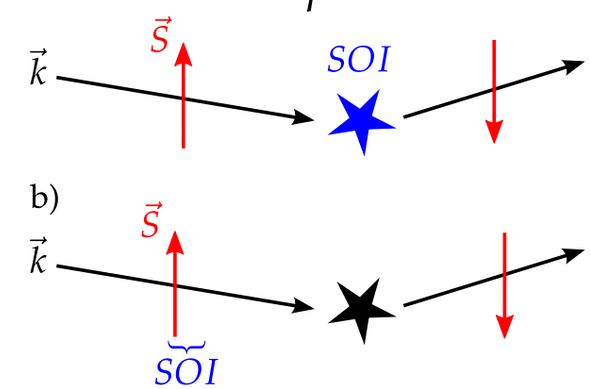


Summary of experimental observations

D'yakonov-Perel' $\tau_S \propto 1/\tau_p$

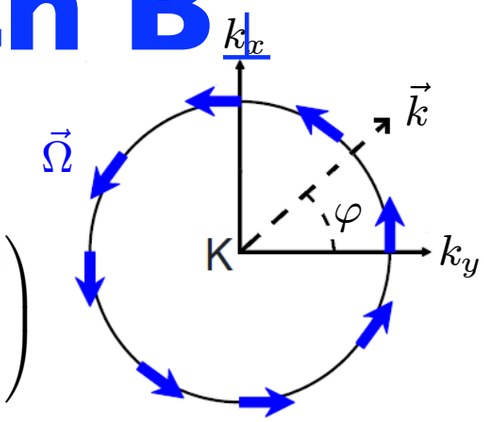


Elliot-Yafet $\tau_S \propto \tau_p$



N. Tombros *et al.*, Nature 2007	single layer	coherence time ~150 ps impurity limited Elliot-Yafet (EY) ?
N. Tombros *et al.*, PRL 2008	single layer	anisotropy
T.-J. Yang *et al.*, PRL 2011	bilayer	up to 2 ns D'yakonov-Perel' (DP)
W. Han and R. K. Kawakami, PRL 2011	single layer	~1ns
	bilayer	up to 6 ns D'yakonov-Perel' (DP)

DP Spin relaxation in SLG with $B_\perp$



spin-orbit: $H_{SO} = \lambda_I \tau \sigma_z s_z + \lambda_R(\tau \sigma_x s_y - \sigma_y s_x)$

$$\vec{\Omega}(\vec{k}) = m \underbrace{\frac{2\lambda_R}{\hbar}}_{\omega_R} \begin{pmatrix} -\sin(\phi_{\vec{k}}) \\ \cos(\phi_{\vec{k}}) \\ 0 \end{pmatrix}$$

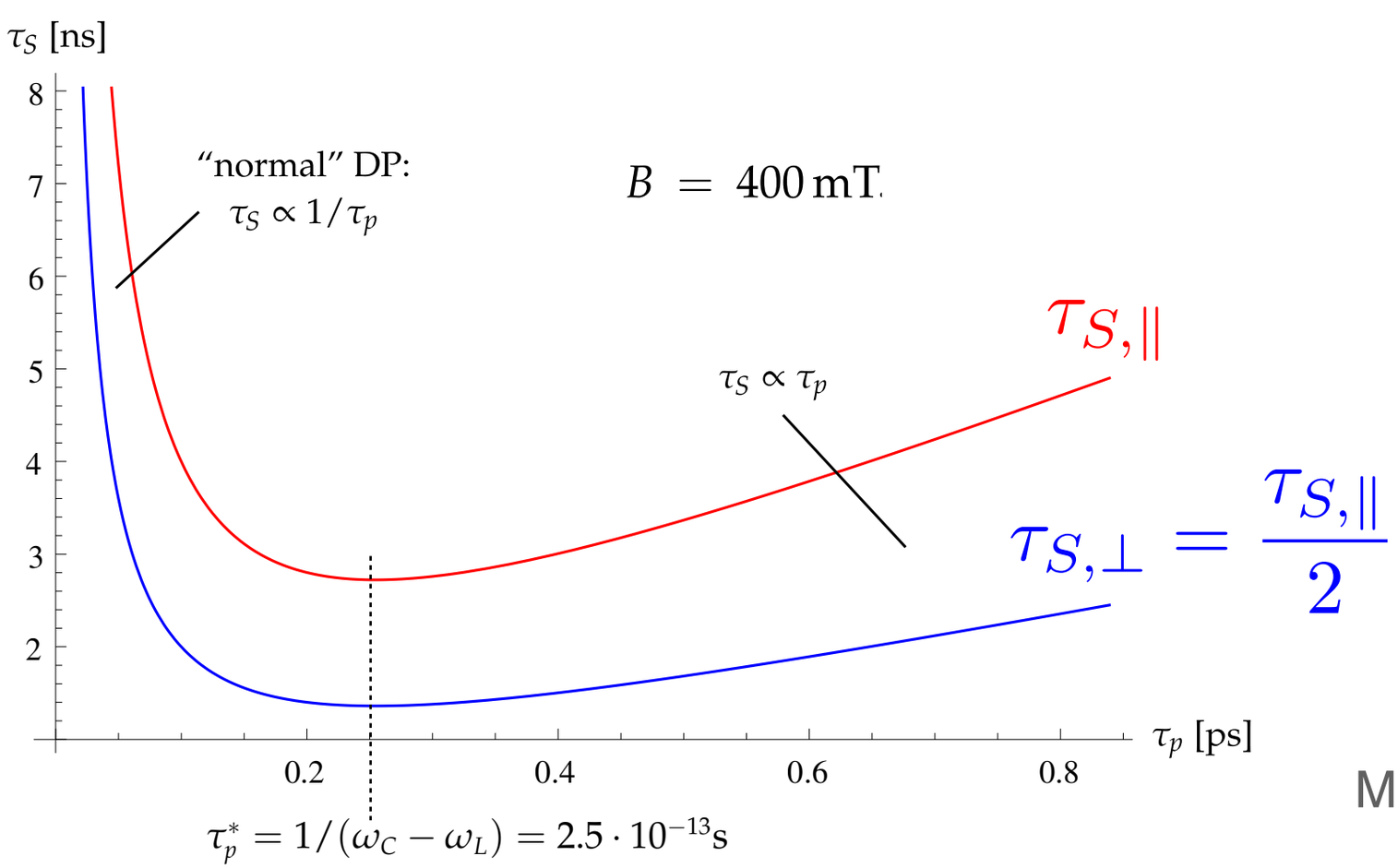
effective electron Hamiltonian: $H_{\text{eff}} = \vec{\Omega} \cdot \vec{s}$

kinetic spin Bloch equation:

$$\frac{\partial \vec{s}_{\vec{k}}}{\partial t} - \underbrace{\vec{\Omega}(\vec{k})}_{\text{SOI + Larmor}} \times \vec{s}_{\vec{k}} = -\int_0^{2\pi} \frac{d\phi'}{2\pi} W(\phi - \phi')\,(\vec{s}_{\vec{k}} - \vec{s}_{\vec{k}'})$$

longitudinal spin relaxation (decoherence):

$$\frac{1}{\tau_{S,\parallel}} = \frac{1}{T_2} = \frac{1}{2}\frac{\omega_R^2 \tau_p}{1 + (\omega_C - \omega_L)^2 \tau_p^2}$$



frequencies:

Rashba $\omega_R = \frac{2\lambda_R}{\hbar}$

cyclotron $\omega_C = \frac{e v_F^2}{E_F} B$

Larmor $\omega_L = 2\frac{\mu_B}{\hbar} B$

M. Diez & GB, manuscript in preparation

DP spin relaxation in BLG

interlayer potential

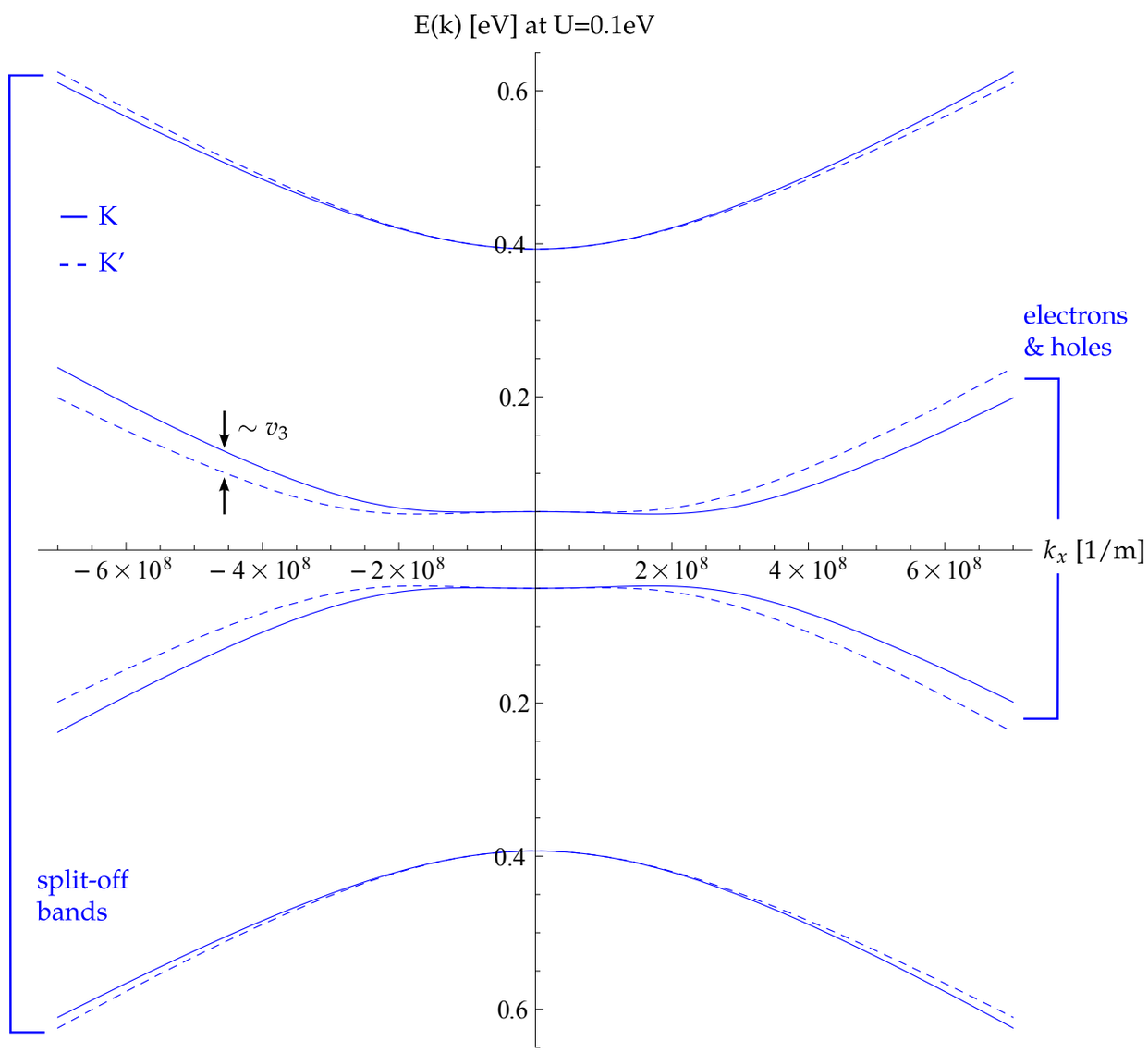
from skew interlayer hopping

interlayer hopping

$$H_0^\tau = \begin{pmatrix} \frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_3\, e^{-i\tau\phi} \\ \tau\,\hbar k v_F\, e^{-i\tau\phi} & \frac{U}{2} & \gamma_1 & 0 \\ 0 & \gamma_1 & -\frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} \\ \tau\,\hbar k v_3\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_F\, e^{-i\tau\phi} & -\frac{U}{2} \end{pmatrix}$$

A1 B1 A2 B2

McCann & Fal'ko, PRL 2006



DP spin relaxation in BLG

interlayer potential

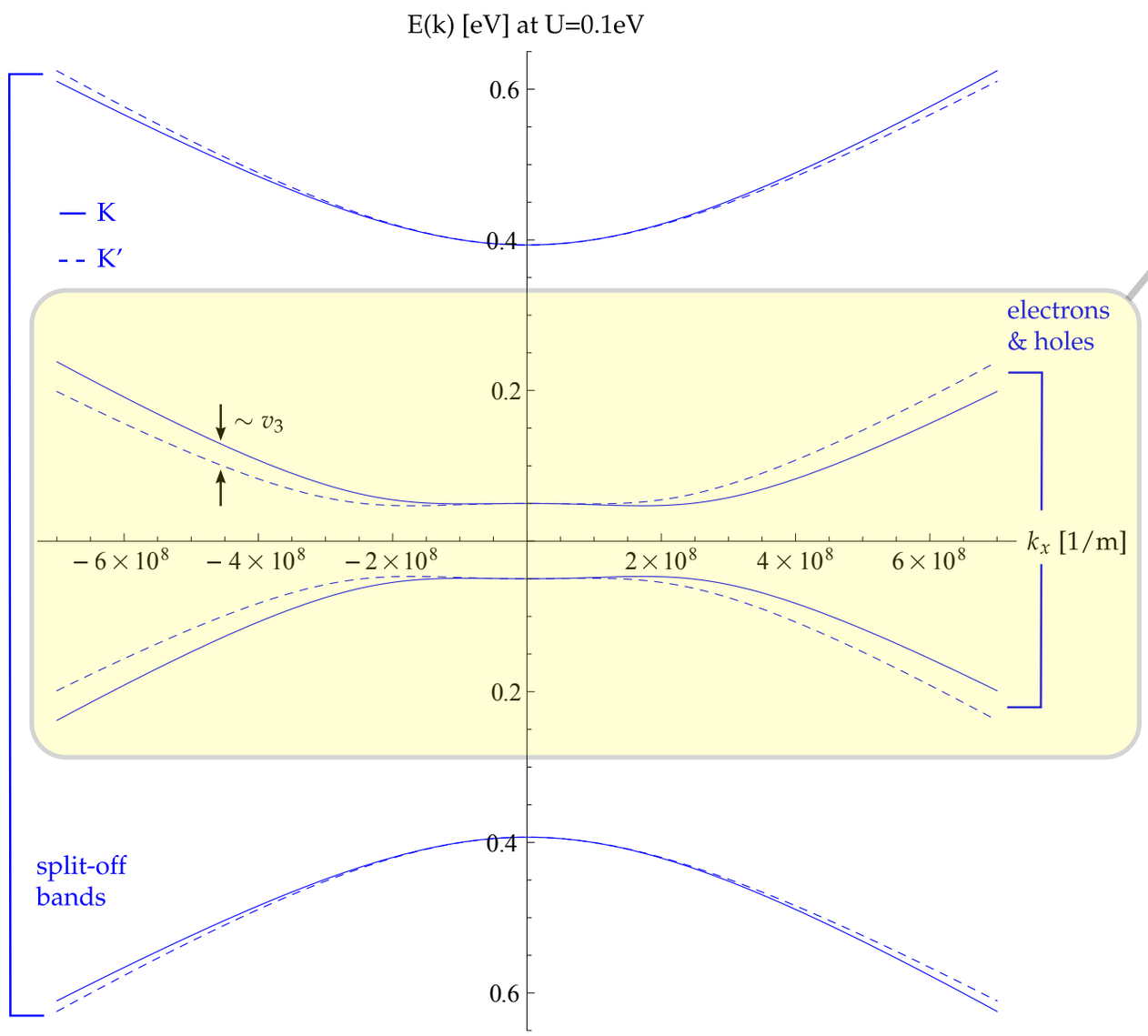
from skew interlayer hopping

interlayer hopping

$$H_0^\tau = \begin{pmatrix} \frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_3\, e^{-i\tau\phi} \\ \tau\,\hbar k v_F\, e^{-i\tau\phi} & \frac{U}{2} & \gamma_1 & 0 \\ 0 & \gamma_1 & -\frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} \\ \tau\,\hbar k v_3\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_F\, e^{-i\tau\phi} & -\frac{U}{2} \end{pmatrix}$$

A1 B1 A2 B2

McCann & Fal'ko, PRL 2006



$$H_0^{\text{eff}} = \begin{pmatrix} U\left(\frac{1}{2} - \frac{p^2 v_F^2}{\gamma_1^2}\right) & -\frac{p^2 v_F^2\, e^{i2\tau\phi}}{\gamma_1} + \tau\, p v_3\, e^{-i\tau\phi} \\ -\frac{p^2 v_F^2\, e^{-i2\tau\phi}}{\gamma_1} + \tau\, p v_3\, e^{i\tau\phi} & -U\left(\frac{1}{2} - \frac{p^2 v_F^2}{\gamma_1^2}\right) \end{pmatrix}$$

DP spin relaxation in BLG

interlayer potential

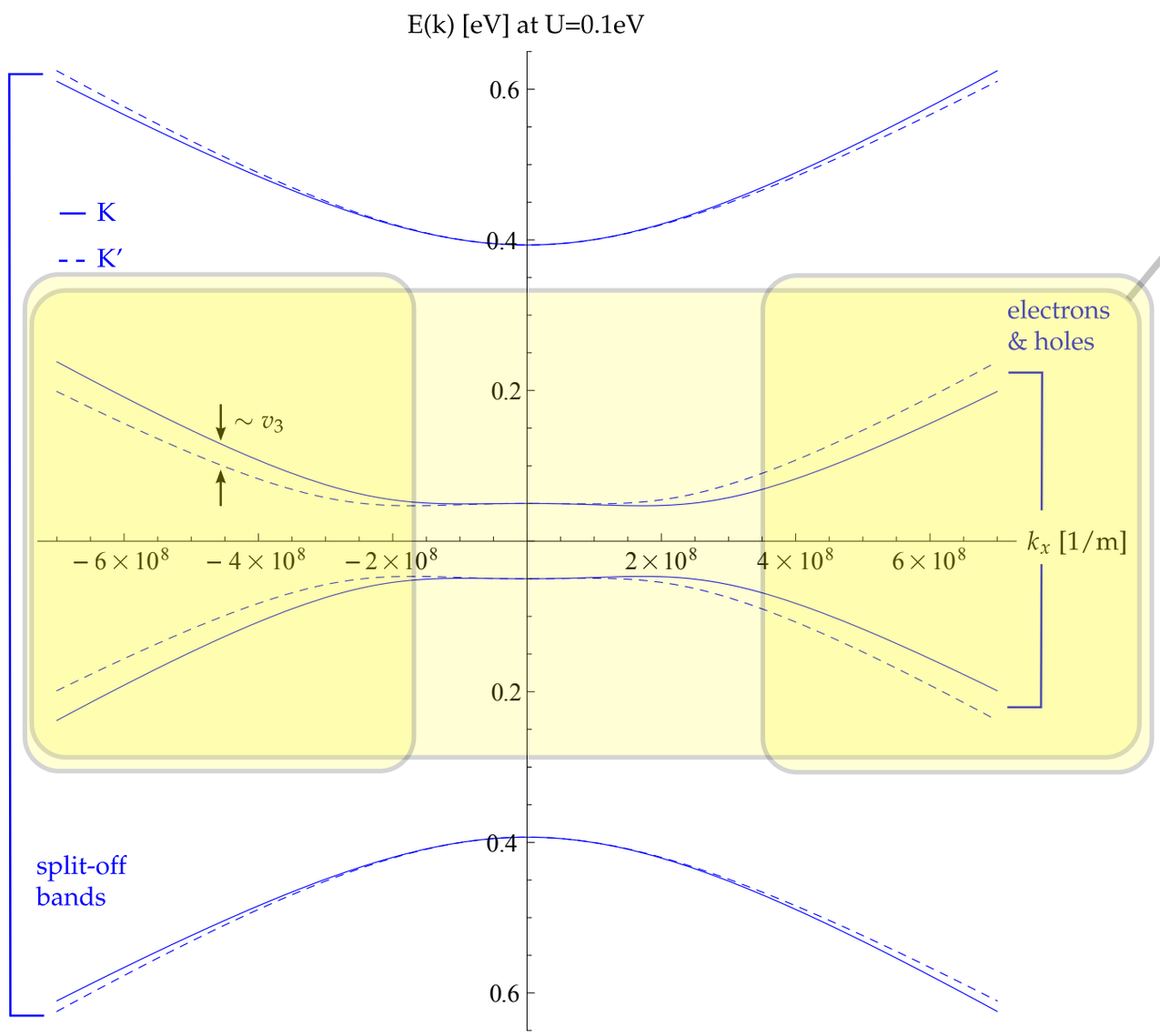
from skew interlayer hopping

interlayer hopping

$$H_0^\tau = \begin{pmatrix} \frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_3\, e^{-i\tau\phi} \\ \tau\,\hbar k v_F\, e^{-i\tau\phi} & \frac{U}{2} & \gamma_1 & 0 \\ 0 & \gamma_1 & -\frac{U}{2} & \tau\,\hbar k v_F\, e^{i\tau\phi} \\ \tau\,\hbar k v_3\, e^{i\tau\phi} & 0 & \tau\,\hbar k v_F\, e^{-i\tau\phi} & -\frac{U}{2} \end{pmatrix}$$

A1 B1 A2 B2

McCann & Fal'ko, PRL 2006



$$H_0^{\text{eff}} = \begin{pmatrix} U\left(\frac{1}{2} - \frac{p^2 v_F^2}{\gamma_1^2}\right) & -\frac{p^2 v_F^2\, e^{i2\tau\phi}}{\gamma_1} + \tau\, p v_3\, e^{-i\tau\phi} \\ -\frac{p^2 v_F^2\, e^{-i2\tau\phi}}{\gamma_1} + \tau\, p v_3\, e^{i\tau\phi} & -U\left(\frac{1}{2} - \frac{p^2 v_F^2}{\gamma_1^2}\right) \end{pmatrix}$$

$$\frac{p_F^2 v_F^2}{\gamma_1^2} = \frac{U^2 + \sqrt{4E_F^2(U^2+\gamma_1^2) - U^2\gamma_1^2}}{2(U^2+\gamma_1^2)} \approx \frac{E_F}{\gamma_1}$$

$$\gamma_1 \gg E_F > U/2$$

DP spin relaxation in BLG

$$H_{SO} = \lambda_1 \tau \sigma_z s_z + \lambda_2 \tau \mu_z s_z + \lambda_3 \mu_z \left(\sigma_y s_x - \tau \sigma_x s_y\right) + \lambda_4 \sigma_z \left(\mu_y s_x + \tau \mu_x s_y\right)$$

pseudospin spin layer valley

F. Guinea, N. J. Phys. **12**, 083063 (2010)

λ_1 [meV]	λ_2 [meV]	λ_3 [meV]	λ_4 [meV]
0.014	0.008	0.0055	0.48

largest contribution

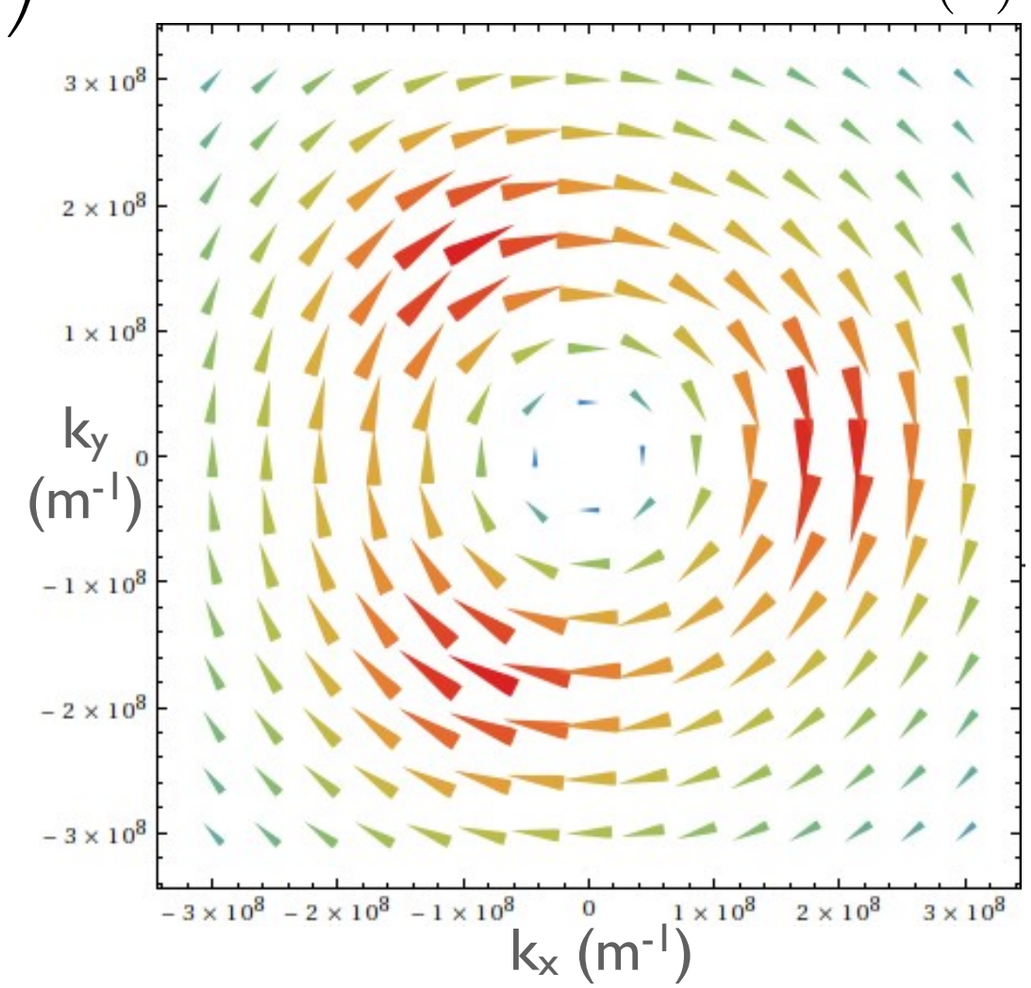
$$H^e_{\lambda_4} = \lambda_4 \frac{U}{E^0_{\text{eff}}} \frac{p v_F}{\gamma_1} \begin{pmatrix} 0 & i e^{-i\phi}\left(1 - \frac{p^2 v_F^2}{\gamma_1^2}\right) - \tau\, i e^{2i\phi} \frac{p v_3}{\gamma_1} \\ -i e^{i\phi}\left(1 - \frac{p^2 v_F^2}{\gamma_1^2}\right) + \tau\, i e^{-2i\phi} \frac{p v_3}{\gamma_1^2} & 0 \end{pmatrix} = \mathbf{\Omega}^e_{\lambda_4} \cdot \mathbf{s}$$

$$\Omega^e_{\lambda_4,x} = \frac{2\lambda_4}{\hbar} \frac{U}{E^0_{\text{eff}}} \frac{p v_F}{\gamma_1} \left[+ \left(1 - \frac{p^2 v_F^2}{\gamma_1^2}\right) \sin\phi + \tau \frac{p v_3}{\gamma_1} \sin 2\phi \right]$$

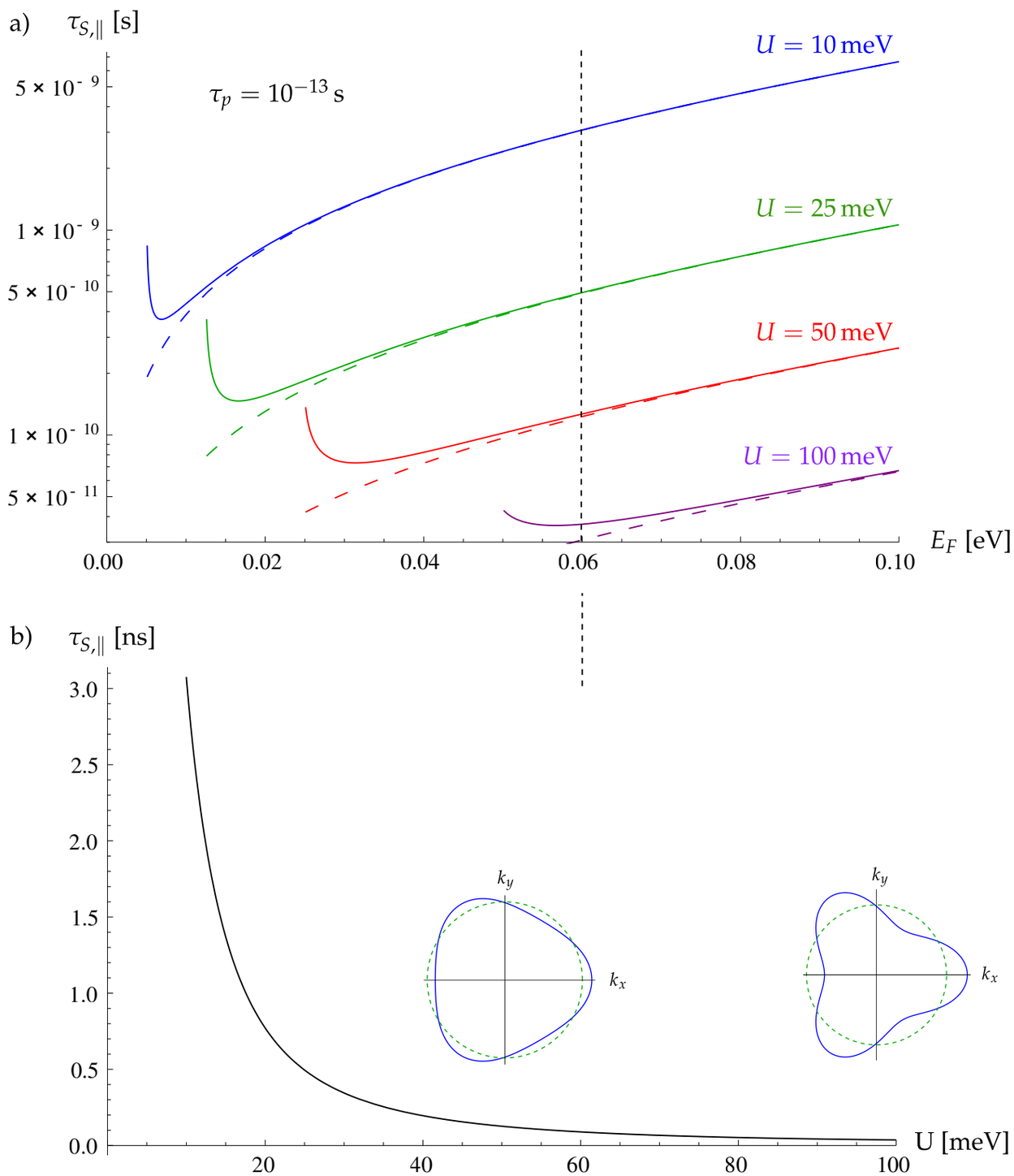
$$\Omega^e_{\lambda_4,y} = \frac{2\lambda_4}{\hbar} \frac{U}{E^0_{\text{eff}}} \frac{p v_F}{\gamma_1} \left[- \left(1 - \frac{p^2 v_F^2}{\gamma_1^2}\right) \cos\phi + \tau \frac{p v_3}{\gamma_1} \cos 2\phi \right]$$

$$\Omega^e_{\lambda_4,z} = 0 \,.$$

$\mathbf{\Omega}(\mathbf{k})$



DP spin relaxation in BLG



from kinetic spin Bloch equation:

$$\frac{1}{\tau^0_{S,\parallel}} = \frac{1}{2}\omega^2_{\lambda_4}\tau_p = \frac{2\lambda_4^2}{\hbar^2}\frac{U^2}{E_F^2}\frac{p_F^2 v_F^2}{\gamma_1^2}\left(1 - \frac{p_F^2 v_F^2}{\gamma_1^2}\right)^2 \tau_p$$

$$\frac{1}{\tau^0_{S,\perp}} = \omega^2_{\lambda_4}\tau_p = \frac{2}{\tau_{S,\parallel}},$$

in the regime $\gamma_1 \gg E_F > U/2$

$$\frac{p_F^2 v_F^2}{\gamma_1^2} = \frac{U^2 + \sqrt{4E_F^2(U^2+\gamma_1^2) - U^2\gamma_1^2}}{2(U^2+\gamma_1^2)} \approx \frac{E_F}{\gamma_1}$$

$$\frac{1}{\tau^0_{S,\parallel}} = \frac{1}{2\tau^0_{S,\perp}} \approx \frac{2\lambda_4^2}{\hbar^2}\frac{(\gamma_1 - E_F)^2 U^2}{E_F\gamma_1^3}\tau_p$$

M. Diez & GB, manuscript in preparation

Summary

- **spin-valley hyperfine interaction in graphene**
  couples nuclear spin to both electron spin and valley

- **spin relaxation of localized electrons**
  effect of flexural phonons saturates at low B fields

- **spin relaxation of mobile electrons**
  SLG: crossover $\tau_s \sim 1/\tau_p$ to $\tau_s \sim \tau_p$
  BLG: gate tunability of τ_s