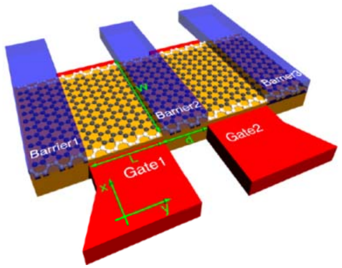
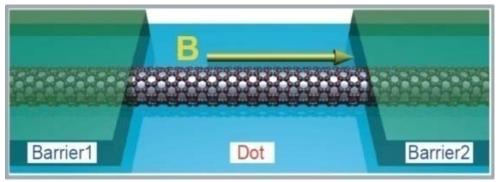
Spin qubits and decoherence in graphene and carbon nanotube quantum dots

School and conference on
Spin-based quantum information processing
16-20 August 2010
Konstanz





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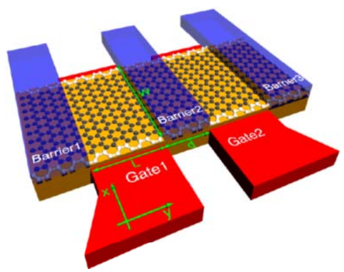
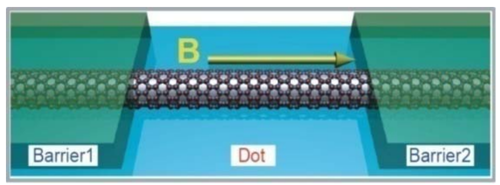
Daniel Loss (Basel)
Johan Nilsson (Gothenburg)
Patrik Recher (Würzburg)
Valentin Rychkov (Würzburg)

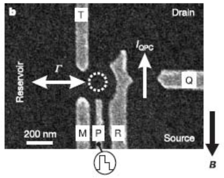
Why are spin qubits in carbon nanosystems interesting?

- … everything is different. → Exciting!
- … everything might be better. → Useful!
- …

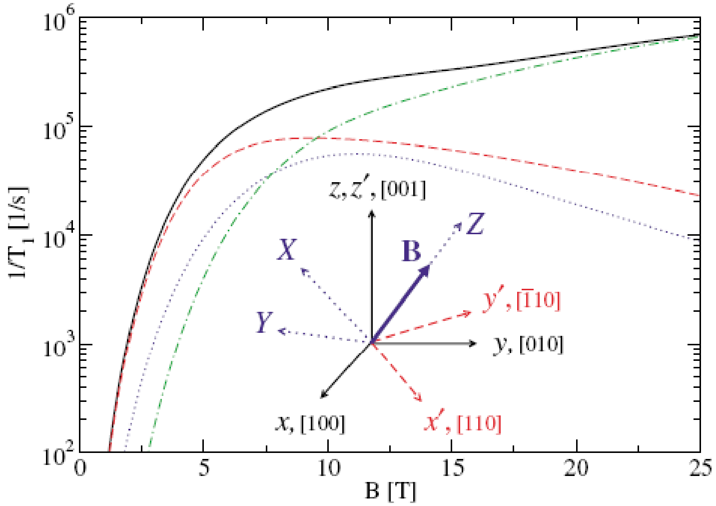




Why different?

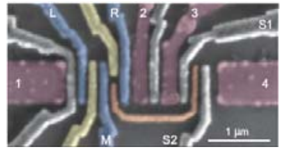


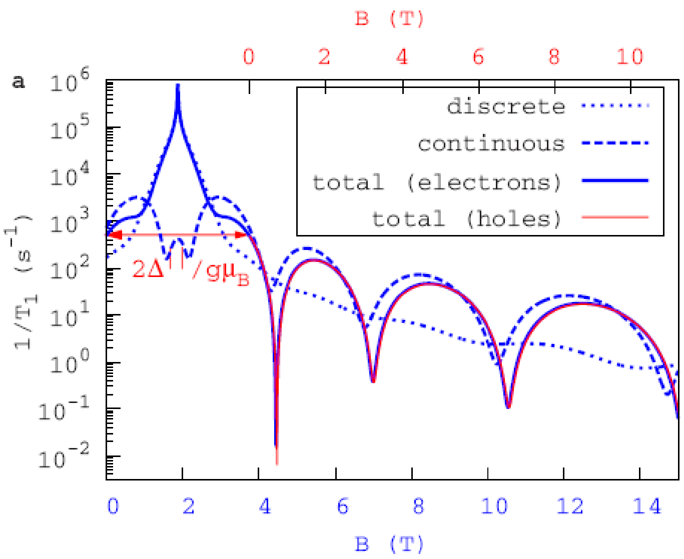
GaAs QDs



Golovach, Khaetskii & Loss PRL 2004

CNT QDs





Bulaev, BT & Loss PRB 2008

Why better?

- electron spin g-factor is 2 in carbon nanosystem, compared to $|g| \approx 0.4$ in GaAs $\rightarrow$ **faster spin rotations by ESR**

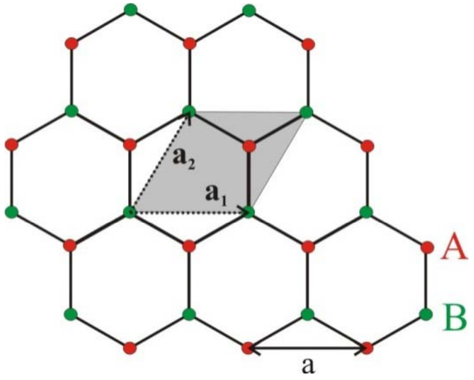
- spin-orbit coupling is weak in carbon (low atomic weight) $\longrightarrow$ **positive aspect for spin decoherence**

- natural carbon consists predominantly of the zero-spin isotope ^{12}C $\longrightarrow$ **spin decoherence** due to **hyperfine interaction** should be **weak**

Outline

- **spin qubits** in graphene QDs → peculiarities and visions
- **spin relaxation and dephasing** in carbon nanotube QDs due to SOI and electron-phonon coupling
- **spin decoherence** in carbon nanotube and graphene QDs due to hyperfine interaction

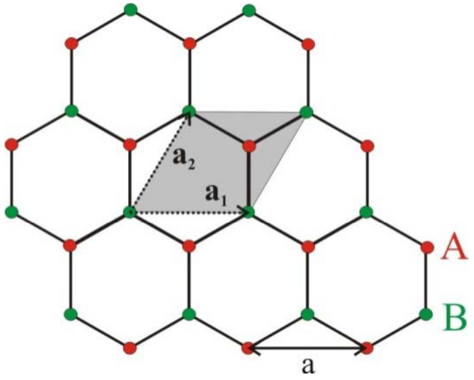
Tight binding model
→ Dirac fermions



$$H = -t \sum_{\langle i,j \rangle,\sigma} a^{\dagger}_{i,\sigma} b_{j,\sigma} + b^{\dagger}_{j,\sigma} a_{i,\sigma}$$

Wallace Phys. Rev. 1947

Tight binding model
→ Dirac fermions



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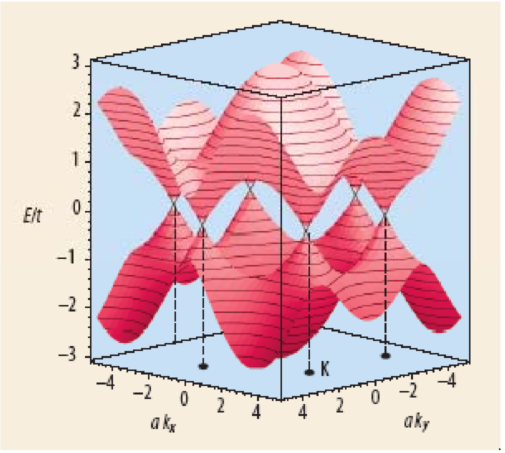
Wallace Phys. Rev. 1947

$$H|\psi\rangle = E(\vec{k})|\psi\rangle$$

→



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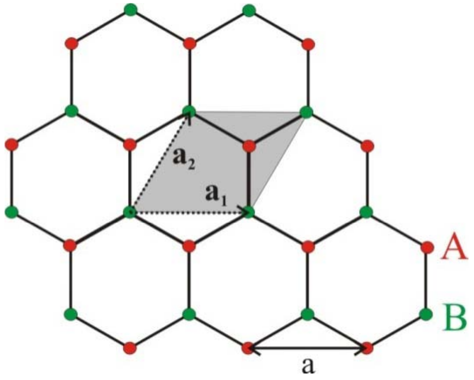

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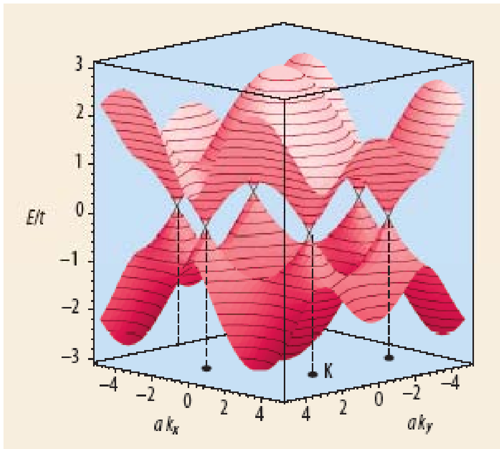
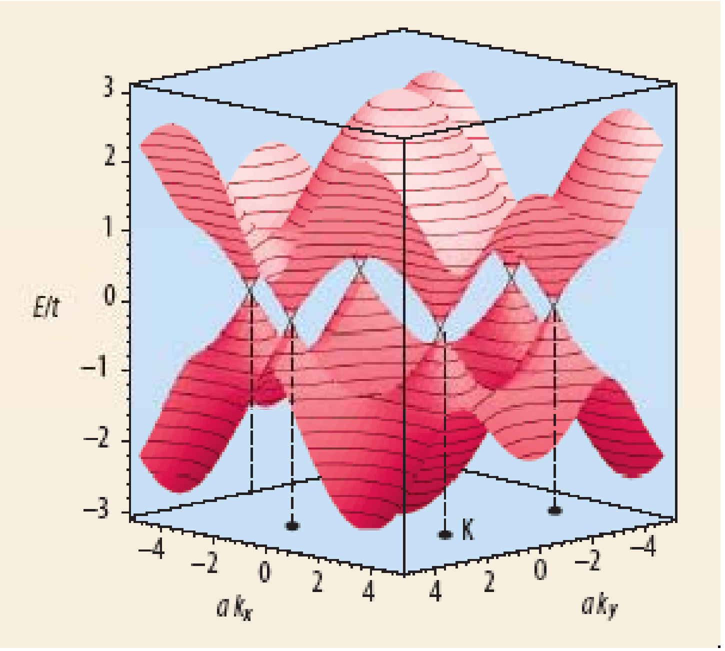


Expansion around K-points ⇒ $$\hbar v_F \vec{k}\vec{\sigma} \begin{pmatrix} \alpha_k \\ \beta_k \end{pmatrix} = E(\vec{k}) \begin{pmatrix} \alpha_k \\ \beta_k \end{pmatrix}$$

Semenoff PRL 1984

Why is it difficult to form spin qubits in graphene?

- Problem (i): difficult to create a tunable quantum dot in graphene - *keyword*: Klein tunneling



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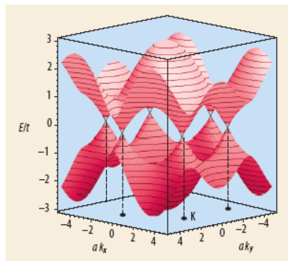
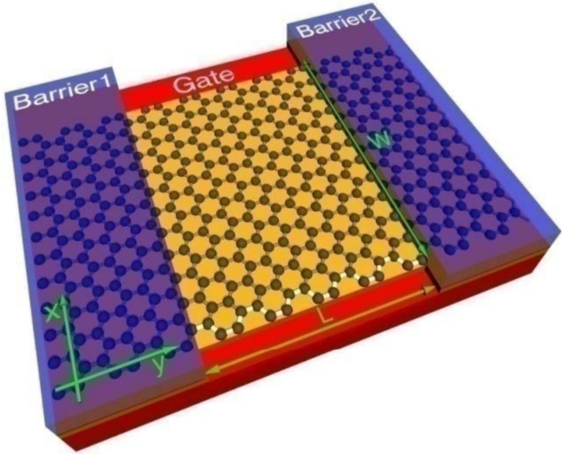


- Problem (ii): difficult to lift of the valley degeneracy (K and K') → crucial to do two-qubit operations using Heisenberg exchange

$$H_{exch} = J\vec{S}_1 \cdot \vec{S}_2$$

Solutions to confinement problem

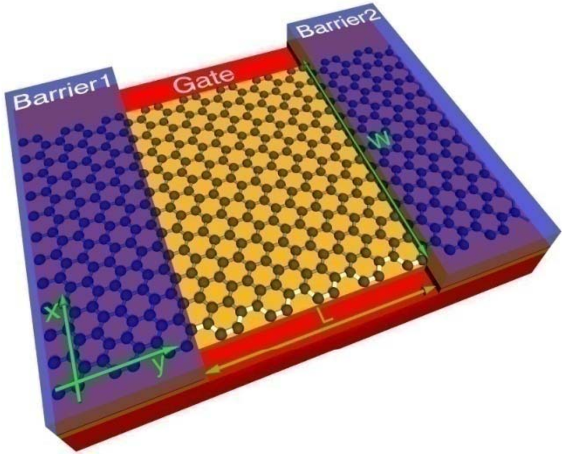


suitable boundary conditions

Silvestrov & Efetov PRL 2007

BT, Bulaev, Loss & Burkard NP 2007

Solutions to confinement problem



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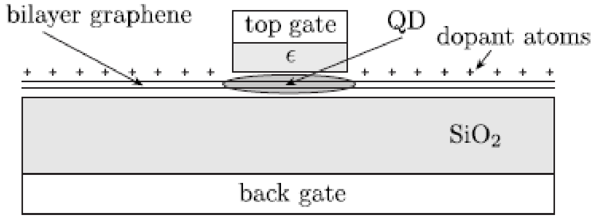

biased bilayer graphene

Pereira, Vasilopoulos & Peeters Nano Lett. 2007

Recher, Nilsson, Burkard & BT PRB 2007

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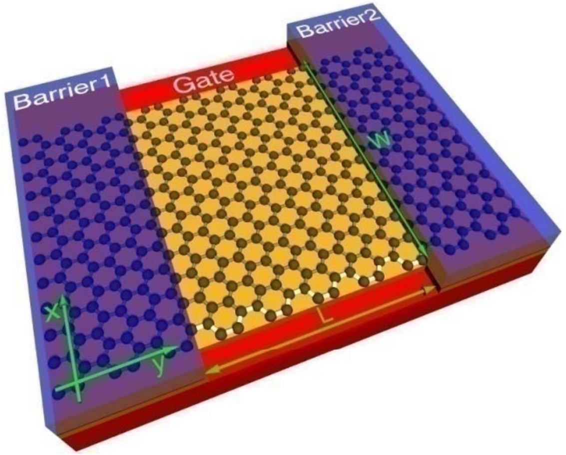

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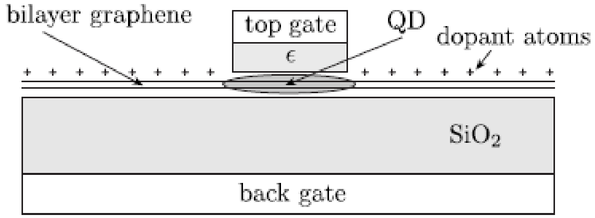

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$$\boxed{v\left[\mathbf{p}+\frac{e}{c}\mathbf{A}(x,y)\right]\cdot\vec{\sigma}\psi(x,y)=\varepsilon\psi(x,y)}$$

magnetic confinement

De Martino, Dell'Anna & Egger PRL 2007

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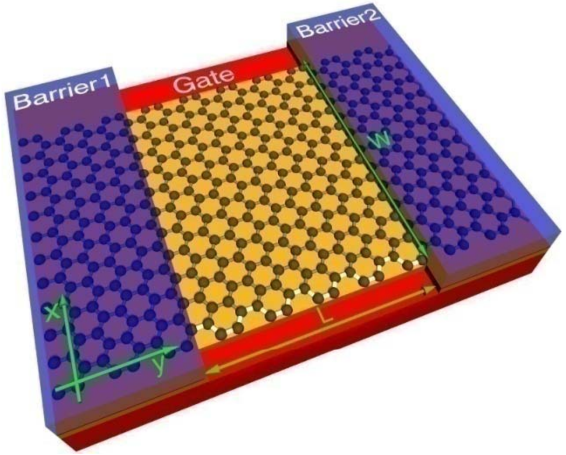

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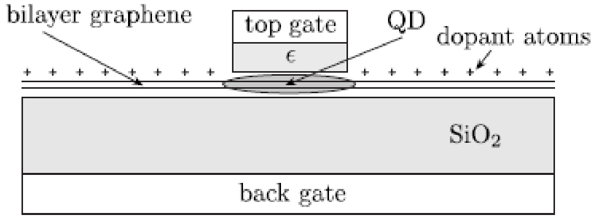

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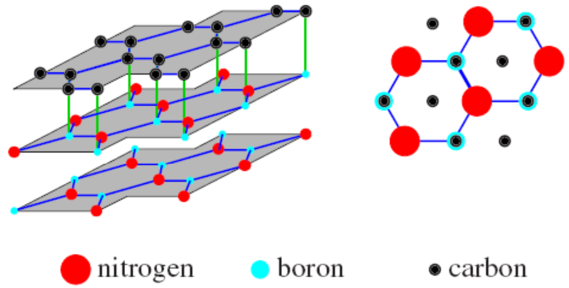
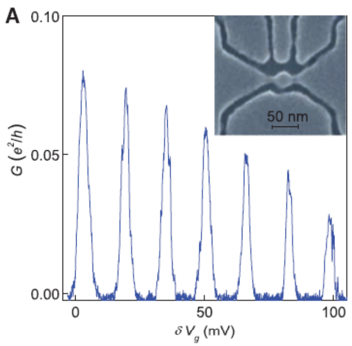


substrate-induced band gap

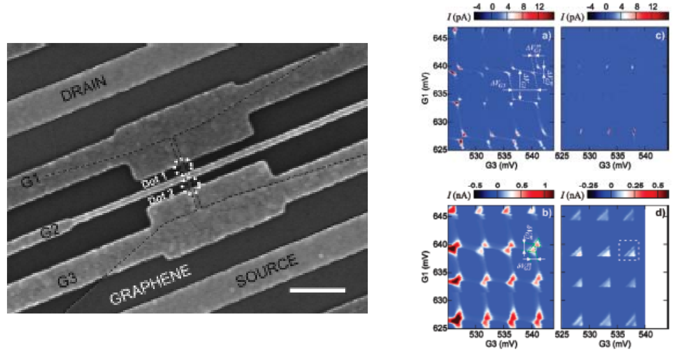
Giovannetti, … & van den Brink PRB 2007

Recher, Nilsson, Burkard & BT PRB 2007 remember Klaus Ensslin's talk

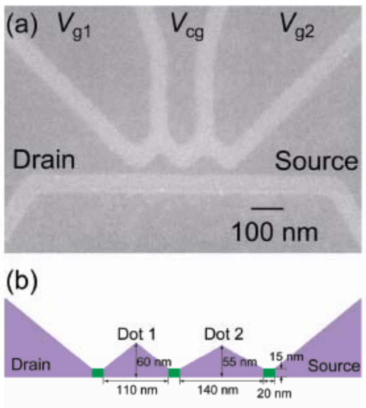
Experimental achievements in graphene quantum dots



Ponomarenko, … & Geim Science 2008

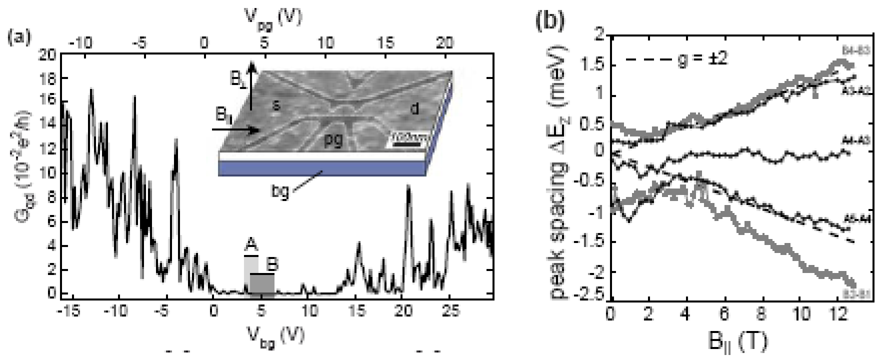


Liu, Oostinga, Morpurgo & Vandersypen PRB 2009
Liu, Hug & Vandersypen Nano Lett. 2010



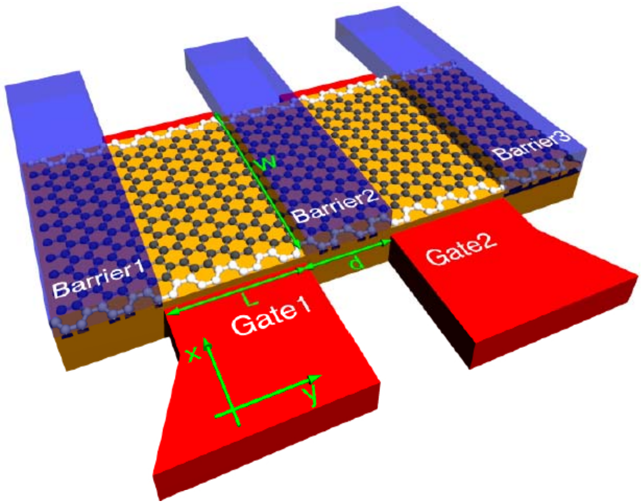
Moriyama, … & Ishibashi Nano Lett. 2009

+ many papers of the Ensslin group, e.g.



Güttinger, Frey, Stampfer, Ihn & Ensslin arXiv 2010

Solution to valley degeneracy problem



graphene nanoribbon with semiconducting armchair boundary conditions

BT, Bulaev, Loss & Burkard Nature Phys. 2007

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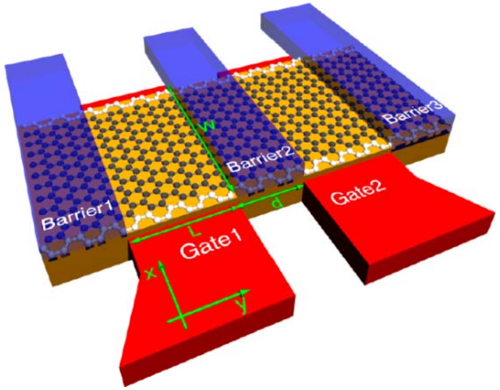

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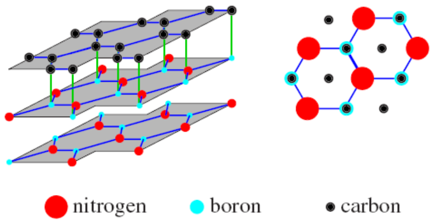
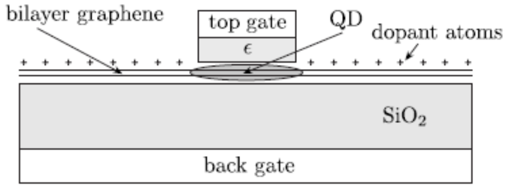
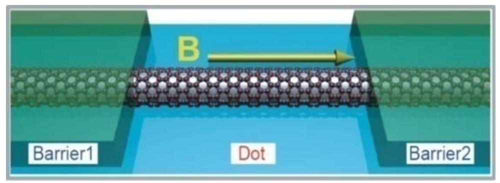
BT, Bulaev, Loss & Burkard Nature Phys. 2007

lifting of valley degeneracy with external magnetic field





Recher, Nilsson, Burkard & BT PRB 2007



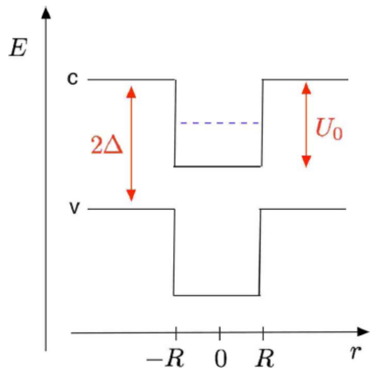
Bulaev, BT & Loss PRB 2008

QD in single-layer graphene on hexagonal boron nitride

$$\Delta \approx 50\ \mathrm{meV}$$

$$\boxed{H_{\tau=\pm} = v\left[\mathbf{p} + \frac{e}{c}\mathbf{A}\right]\cdot\vec{\sigma} + \tau\Delta\sigma_z + U(r)}$$

$$U(r) = U_0\theta(r-R)$$



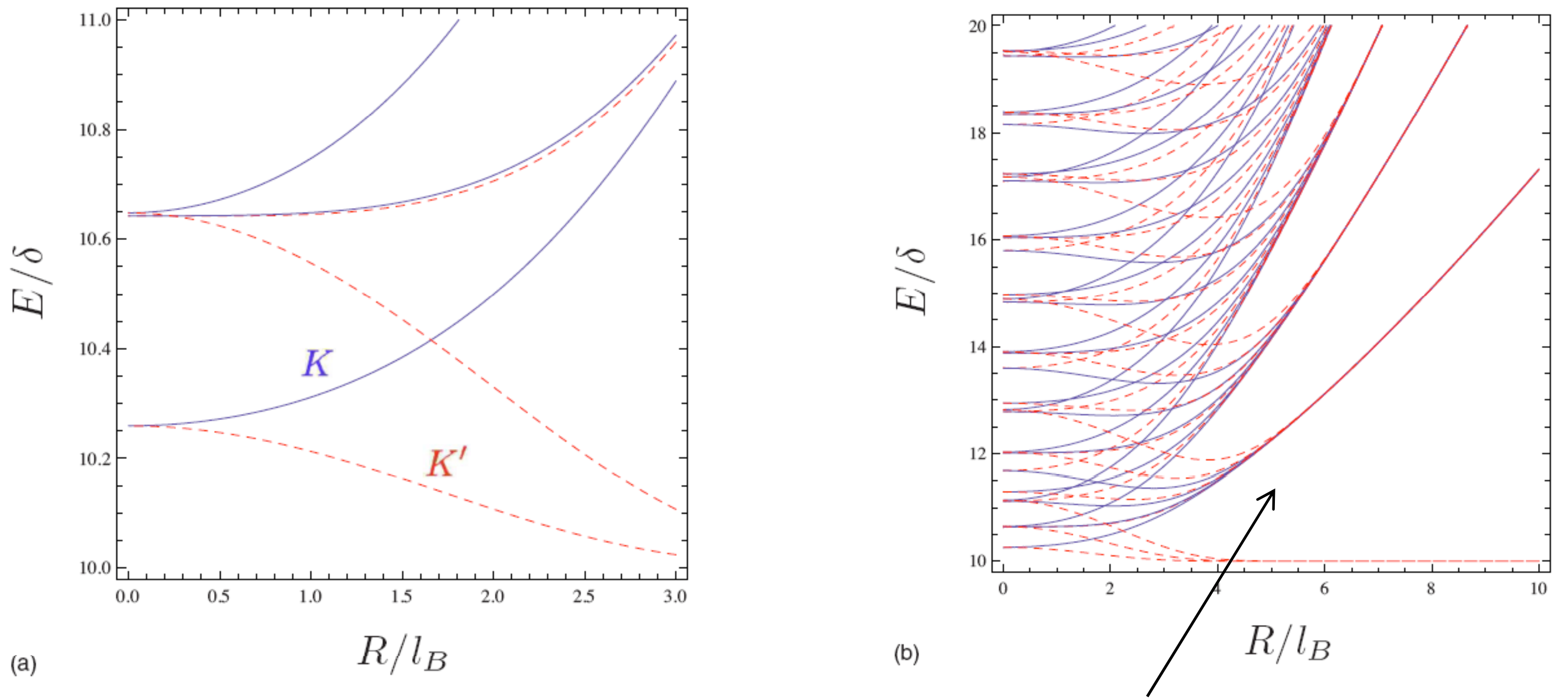
The eigenvalue problem $\boxed{H_\tau\Psi^\tau(r,\varphi) = E\Psi^\tau(r,\varphi)}$
is solved by:

$$\boxed{\Psi^\tau(r,\varphi) = e^{i(j-1/2)\varphi}\begin{pmatrix} \mathrm{X}_A^\tau(r) \\ \mathrm{X}_B^\tau(r)e^{i\varphi} \end{pmatrix}}$$

$$\boxed{\mathrm{X}_\sigma^\tau(r) = 2^{(1+n_\sigma)/2}e^{-br^2/2}r^{n_\sigma}\begin{cases} \alpha_\sigma U(q_\sigma, 1+n_\sigma, br^2), & \mathrm{r>R} \\ \beta_\sigma M(q_\sigma, 1+n_\sigma, br^2), & \mathrm{r<R} \end{cases}}$$

confluent hypergeometric functions

Lifting of valley degeneracy



merging into bulk Landau levels: $\boxed{E_n = \pm\delta\sqrt{\left(\Delta / \delta\right)^2 + 2n\left(R / l_B\right)^2},\ n = 1,2,3,\ldots}$

magnetic length $l_B = \left(\hbar / eB\right)^{1/2}$

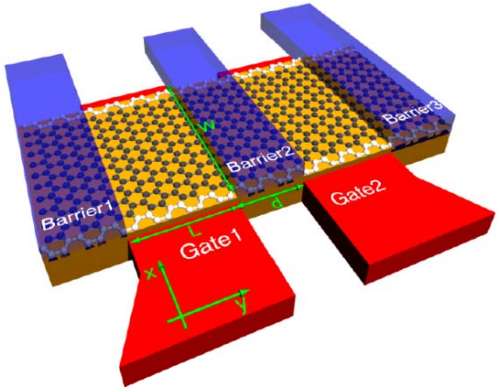
QD level spacing $\delta = \hbar v / R$

Relation to symmetry breaking

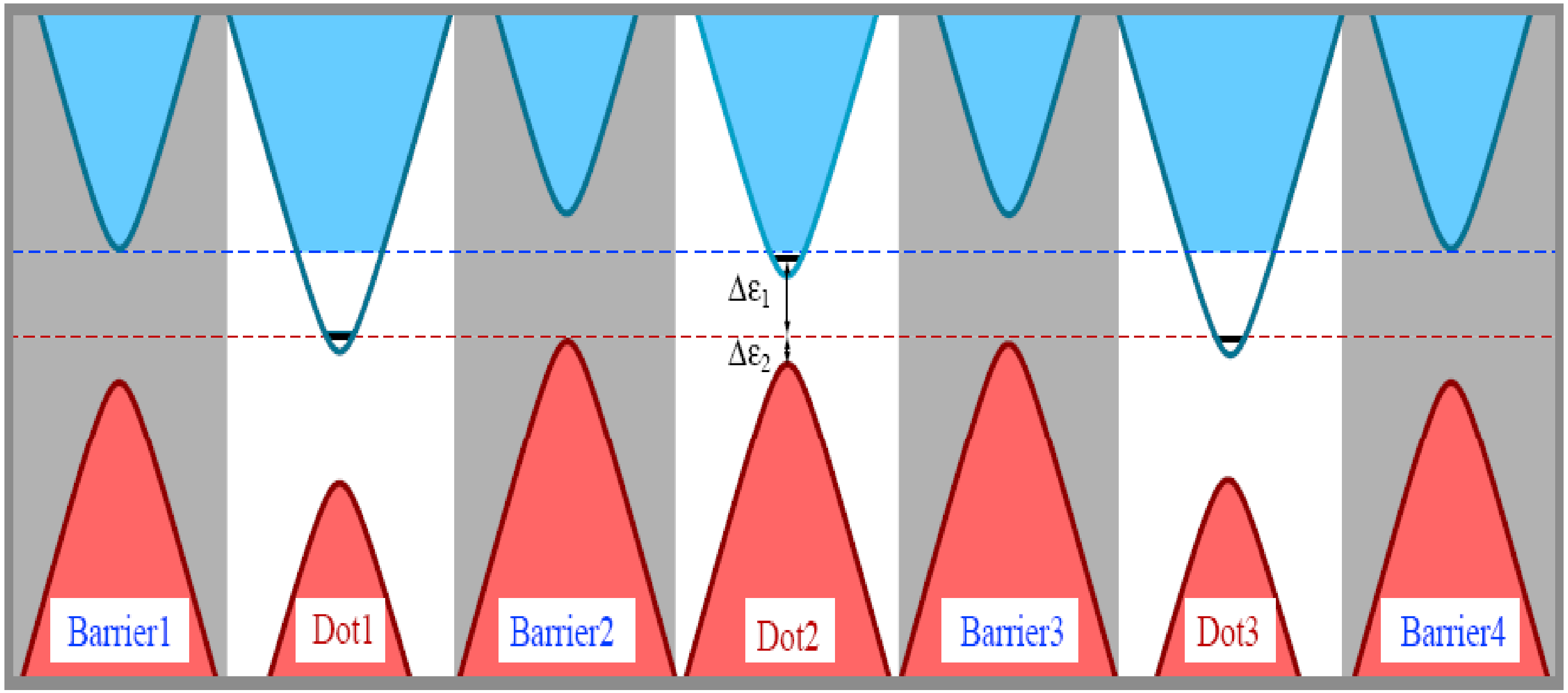
$$H_{\tau=\pm} = v\left[\mathbf{p} + \frac{e}{c}\mathbf{A}\right]\cdot\vec{\sigma} + \tau\Delta\sigma_z + U(r)$$

- breaks effective TRS p → -p and σ → -σ by mass term
- breaks inversion symmetry x → -x of the graphene lattice by mass term
- breaks true TRS by finite magnetic field $T = -(\tau_y \otimes \sigma_y)C$

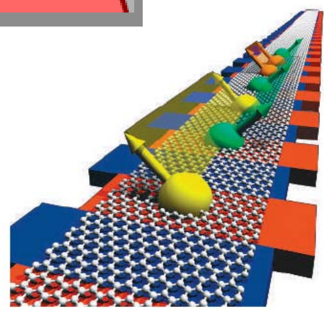
Only the combination of **A** and Δ yields lifting of valley degeneracy without mixing the valleys



The Qubit Piano

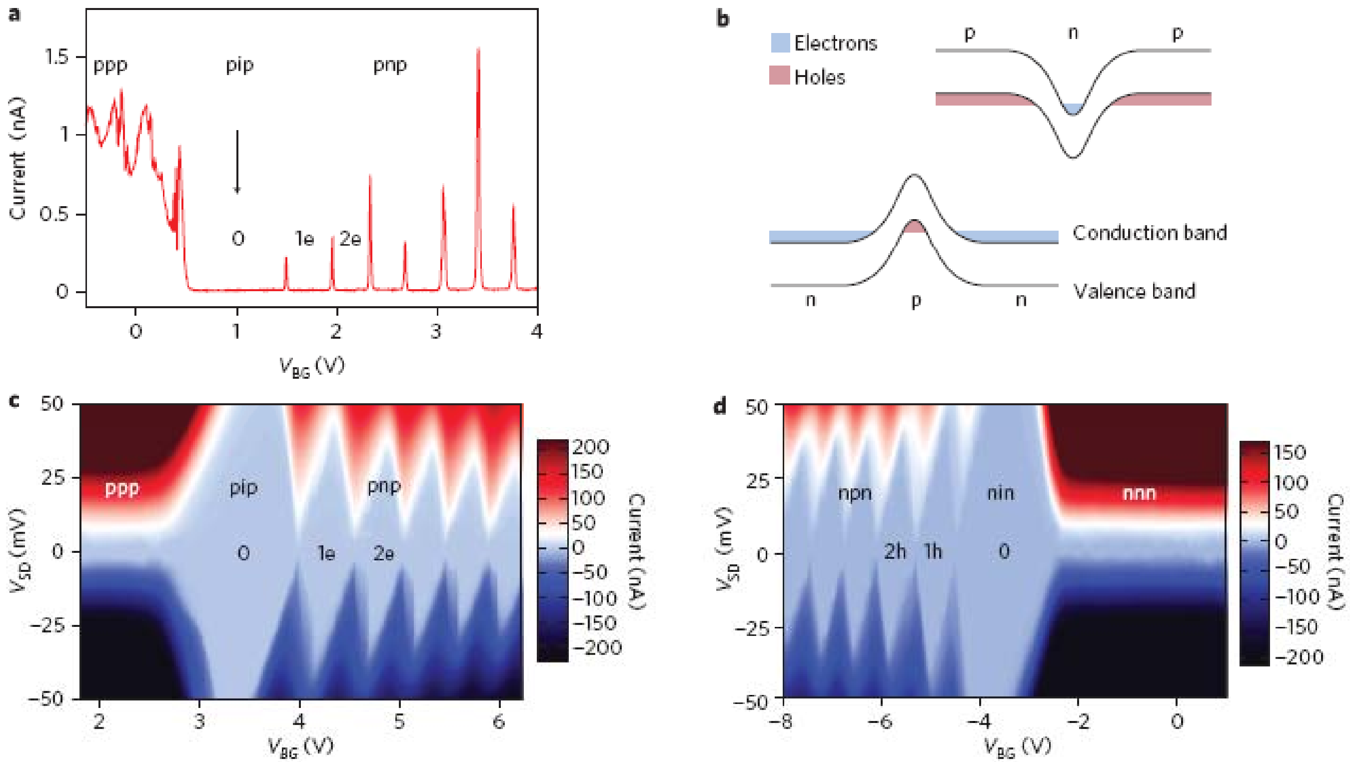


BT, Bulaev, Loss & Burkard Nature Phys. 2007



remember Gary Steele's talk

Proof of principle: experiments on SWNTs



Steele, Götz & Kouwenhoven Nature Nano. 2009

Outline

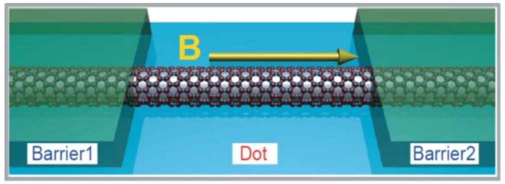
- **spin qubits** in graphene QDs → peculiarities and visions

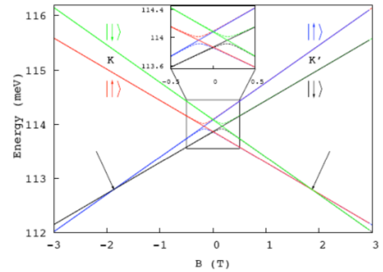
- **spin relaxation and dephasing** in carbon nanotube QDs due to SOI and electron-phonon coupling

- **spin decoherence** in carbon nanotube and graphene QDs due to hyperfine interaction

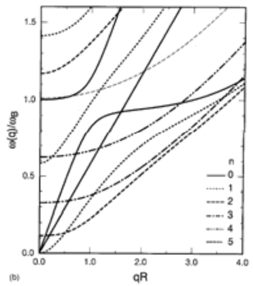
What do we need to understand to solve this problem?

- electronic spectrum





- influence of spin-orbit coupling



- low-energy phonons

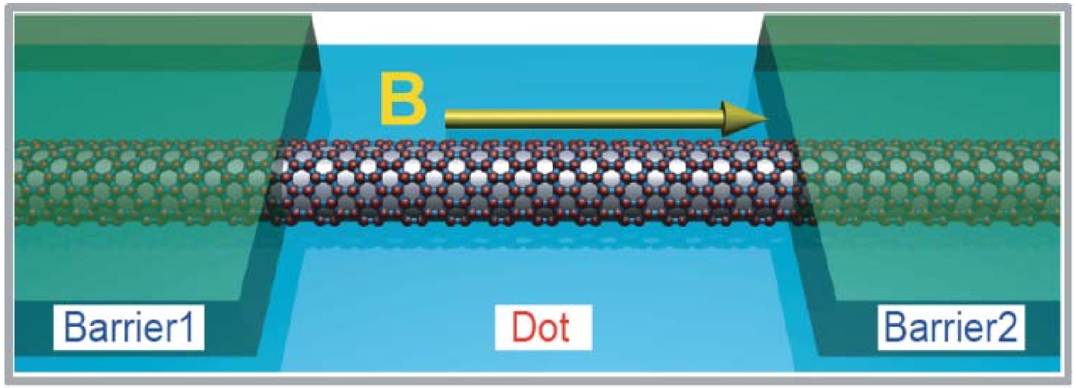
- electron-phonon coupling

$$V_{el-ph} = \begin{pmatrix} V_1 & V_2 \\ V_2^* & V_1 \end{pmatrix} + H.c.$$

- Bloch-Redfield equations

$$\frac{1}{T_1} = \frac{2\pi}{\hbar}\sum_q (2N_q + 1)\left|M_{\omega_q}\right|^2 \delta\left(\left|E_{\kappa_0^+,k_0,1/2} - E_{\kappa_0^-,k_0,-1/2}\right| - \hbar\omega_q\right)$$

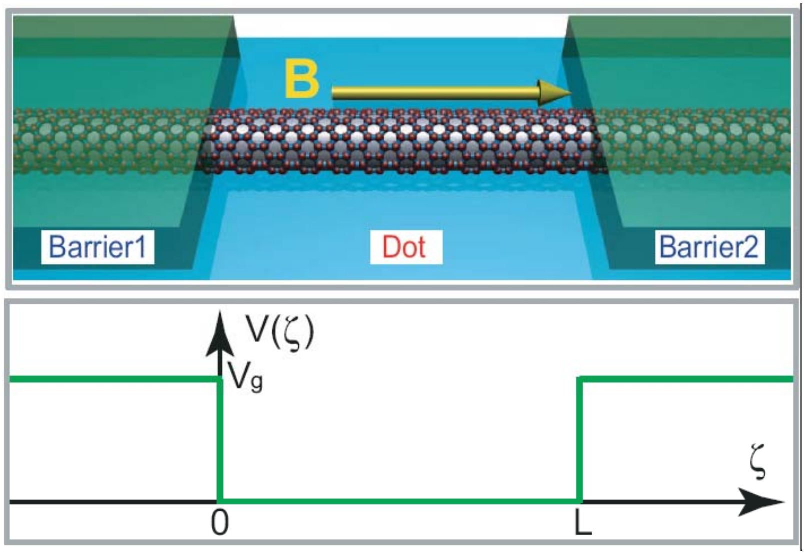
System: SWNT quantum dot



- semi-conducting SWNT
- B-field along tube axis ($\rightarrow$ valley degeneracy lifted)
- electrostatic confinement

Wave function

$$\Psi_{\kappa_m,k,S_\zeta}(\varphi,\zeta)=\frac{e^{i\mathbf{Kr}}}{\sqrt{2\pi}}e^{i(m-\tau_3\nu/3+\Phi_{AB}/\Phi_0)\varphi}\Phi_{m,k}(\xi)\left|S_\zeta\right\rangle$$



$$\Phi_{m,k}(\xi)=\begin{cases}\Phi^L_{m,k}(\xi), & \xi<0\\ \Phi^D_{m,k}(\xi), & 0\le\xi\le L\\ \Phi^R_{m,k}(\xi), & \xi>L\end{cases}$$

see also Karsten Flensberg's talk

Spin-orbit interaction in SWNTs

intrinsic SOI (Dresselhaus-like):

$$H_{SO}^{\mathrm{int}} = \Delta_{\mathrm{int}} \tau_3 \sigma_3 \left(S_+ e^{i\varphi} + S_- e^{-i\varphi} \right)$$

extrinsic SOI (Bychkov-Rashba-like):

$$H_{SO}^{E} = \Delta_E \left[i \tau_3 \sigma_1 \left(-S_+ e^{i\varphi} + S_- e^{-i\varphi} \right) + 2 \sigma_2 S_\zeta \right]$$

extrinsic SOI (due to curvature):

$$H_{SO}^{curv} = i \Delta_{curv}^{\perp} \sigma_2 \left(-S_+ e^{i\varphi} + S_- e^{-i\varphi} \right) + \Delta_{curv}^{\parallel} \tau_3 \sigma_1 2 S_\zeta$$

yields spin mixing

yields zero-field splitting

Ando JPSJ 2000; *Kane & Mele* PRL 2005; *Huertas-Hernando, Guinea & Brataas* PRB 2006; *Jeong & Lee* PRB 2009; *Izumida, Sato & Saito* JPSJ 2009; *Jespersen et al.* arXiv2010

Hierarchy of energy scales

- For moderate electric fields ($E < 0.1$ V/nm):

$$\boxed{\Delta_E < \Delta_{\mathrm{int}} < \Delta_{curv}}$$

$\Delta_{\mathrm{int}} \approx 1\mu\mathrm{eV}$

- Curvature-induced SOI is dominant:

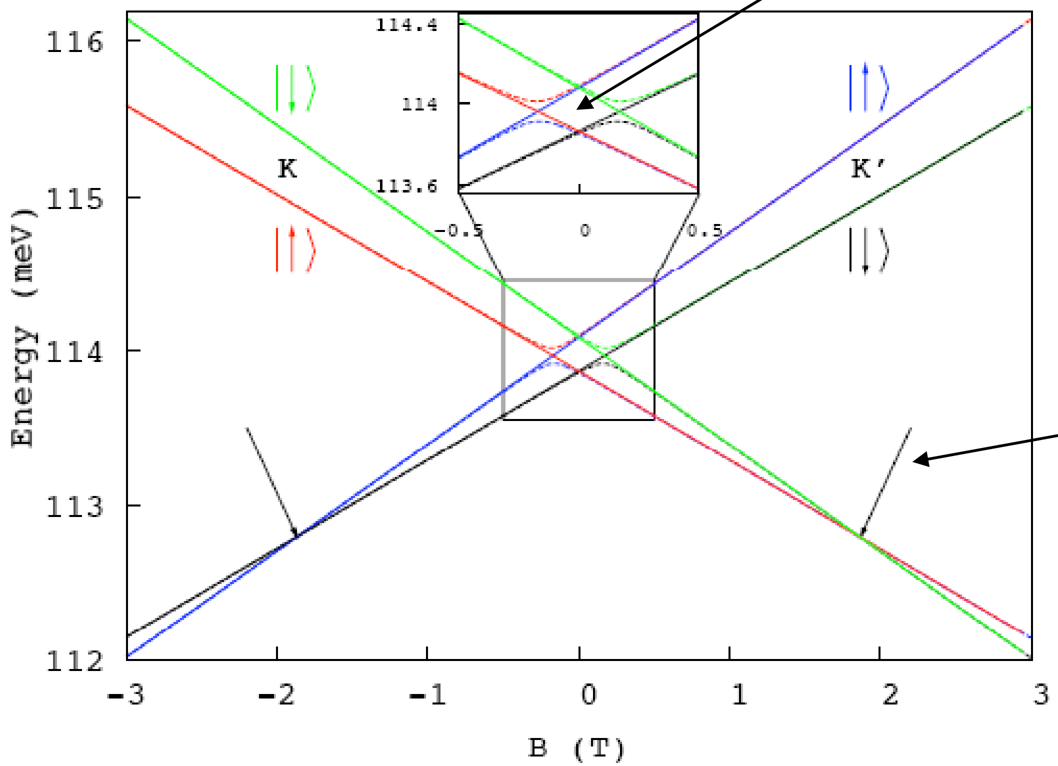
$$\boxed{\Delta^{\parallel}_{curv} \approx 0.17\mathrm{meV}/\mathrm{R[nm]}}$$

$$\boxed{\Delta^{\perp}_{curv} \approx -0.26\mathrm{meV}/\mathrm{R[nm]}}$$

absolute values based on first-principle calculations for graphite

Energy spectrum with SOI

ZFS ≈ 0.22 meV for R ≈ 1.6 nm



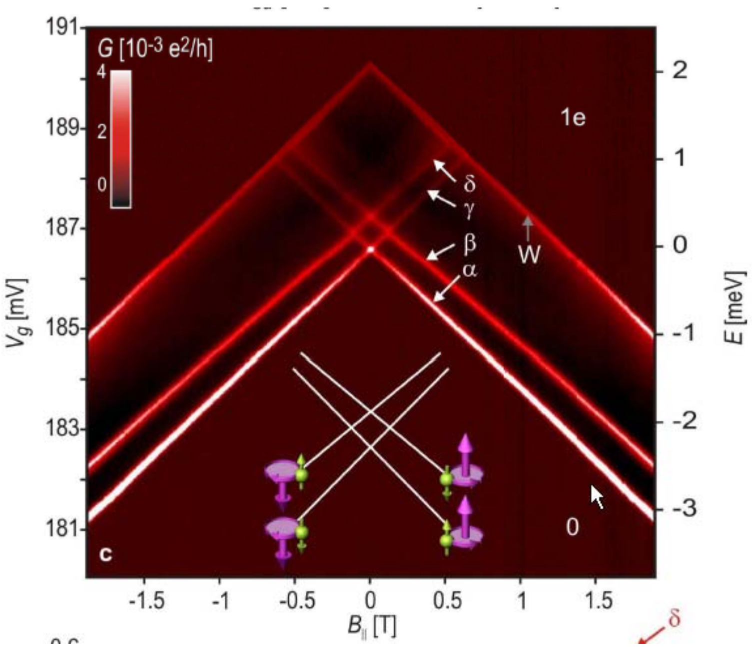
crossings of spin up and spin down

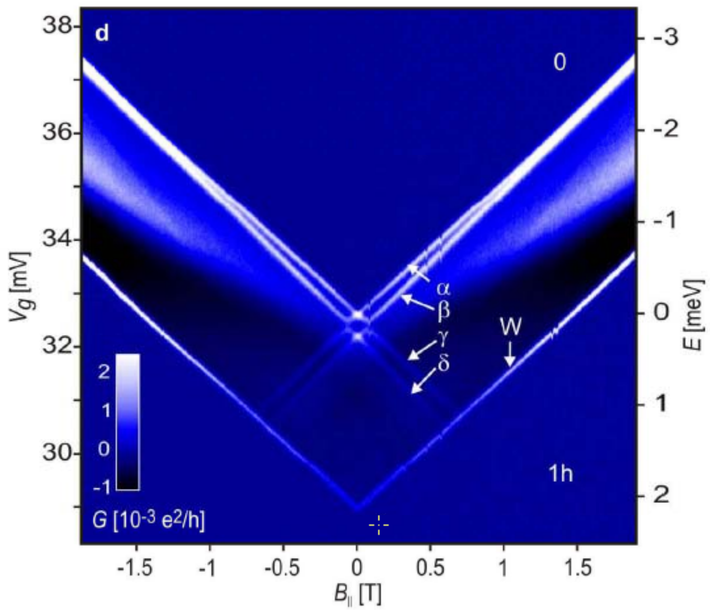
$$H = H_0 + H_Z + H_{SO}^{curv} + H_{K-K'}$$

$$H_{K-K'} = \Delta_{K-K'} \tau_1$$

remember Shahal Ilani's talk

Measurement by Cornell group





electron spectrum

hole spectrum

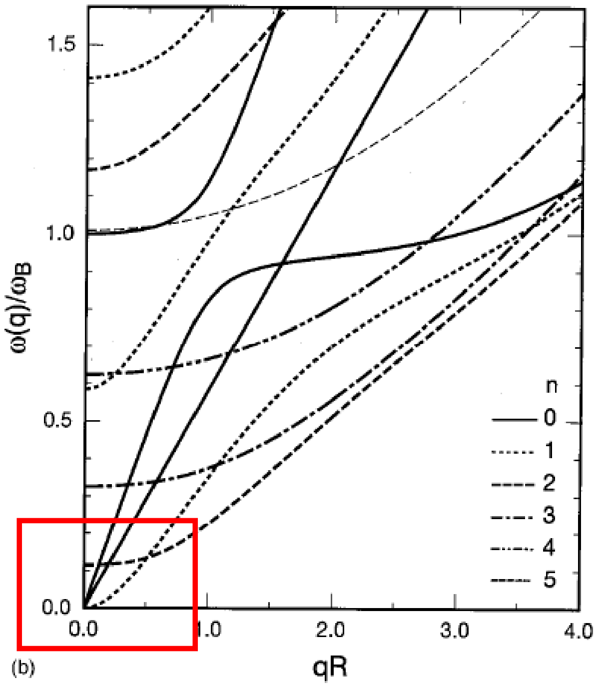
$\boxed{2\Delta_{SO} \approx 0.37\text{meV}}$ $\boxed{2\Delta_{K-K'} \approx 65\mu\text{eV}}$

$\boxed{2\Delta_{SO} \approx 0.21\text{meV}}$ $\boxed{2\Delta_{K-K'} \approx 0.1\text{meV}}$

Kuemmeth, Ilani, Ralph & McEuen, Nature 2008

Low-energy phonons in SWNTs

- twisting mode (TM)
- stretching mode (SM)
- bending mode (BM)



Suzuura & Ando PRB 2002

Low-energy phonons in SWNTs

- twisting mode (TM)
- stretching mode (SM)
- bending mode (BM)

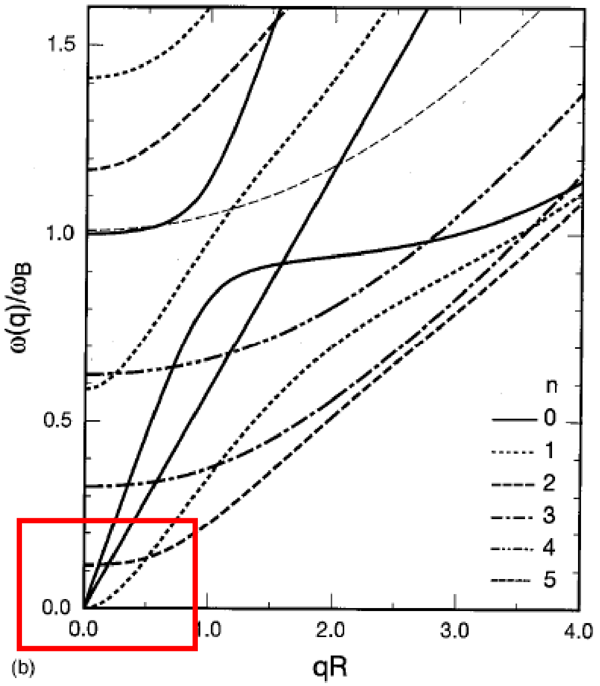
continuum model (with force-constant tensor Λ):

$$\boxed{\ddot{\mathbf{u}}(\mathbf{r},t) = \Lambda \mathbf{u}(\mathbf{r},t)} \qquad \boxed{\mathbf{u}(\mathbf{r},t) = (u_\varphi, u_\zeta, u_r)}$$

solutions take the form:

$$\boxed{\mathbf{u}(\mathbf{r},t) = \mathbf{A}\exp[i(m\varphi + q\zeta - \omega t)]}$$

only BM has m=1; other modes m=0



Electron-phonon coupling

$$\boxed{V_{el-ph} = \begin{pmatrix} V_1 & V_2 \\ V_2^* & V_1 \end{pmatrix} + H.c.}$$

Deformation potential:

$$\boxed{V_1 = g_1 \left(u_{\varphi\varphi} + u_{\zeta\zeta} \right)}$$

$$u_{\varphi\varphi} = \frac{1}{R}\frac{\partial u_\varphi}{\partial \varphi} + \frac{u_r}{R} \qquad u_{\zeta\zeta} = \frac{\partial u_\zeta}{\partial \zeta}$$

Bond-length change:

$$\boxed{V_2 = g_2 e^{3i\theta} \left(u_{\varphi\varphi} - u_{\zeta\zeta} + 2i u_{\varphi\zeta} \right)}$$

$$2u_{\varphi\zeta} = \frac{\partial u_\varphi}{\partial \zeta} + \frac{1}{R}\frac{\partial u_\zeta}{\partial \varphi}$$

electron-phonon coupling in SWNTs:
$g_1 \approx 30\text{eV} \qquad g_2 \approx 1.5\text{eV}$

Lowest energy levels

$$\boxed{\Psi_{0,0,\pm 1/2} \approx \Psi_{\kappa_0^\pm, k_0, \pm 1/2} + \sum_{n \neq 0} \lambda_{k_n}^\pm \Psi_{\kappa_{\mp 1}^\mp, k_n, \mp 1/2} + \frac{L}{2\pi} \int_{\pm k_c}^{\infty} dk \lambda_k^\mp \Psi_{\kappa_{\mp 1}^\mp, k, \mp 1/2}}$$

level mixing:
discrete spectrum

level mixing:
continuous spectrum

$$\boxed{\lambda_{k_n}^\pm = \pm i \Delta_{curv}^\perp \frac{\left\langle \Phi_{\kappa_{\mp 1}^\mp, k_n}(\zeta) \middle| \sigma_2 \middle| \Phi_{\kappa_0^\pm, k_0}(\zeta) \right\rangle}{E_{\kappa_{\mp 1}^\mp, k_n, \mp 1/2} - E_{\kappa_0^\pm, k_0, \pm 1/2}}}$$

$$\boxed{\kappa_m^\pm = \kappa_m \pm \Delta_{curv}^\parallel / \hbar v}$$

1st order perturbation theory in H_{SO}^{curv}

Spin relaxation rate I

Using Bloch-Redfield theory:

$$\boxed{\frac{1}{T_1} = \frac{2\pi}{\hbar}\sum_q \left(2N_q + 1\right)\left|M_{\omega_q}\right|^2 \delta\left(\left|E_{\kappa_0^+,k_0,1/2} - E_{\kappa_0^-,k_0,-1/2}\right| - \hbar\omega_q\right)}$$

$$\boxed{\frac{1}{T_1} = \frac{2^{5/4}\pi L}{\hbar^2\sqrt{c_S R\omega_0}}\left(2N_{\omega_0} + 1\right)\left|M_{\omega_0}\right|^2}$$

$$\omega_0 = \left|E_{\kappa_0^+,k_0,1/2} - E_{\kappa_0^-,k_0,-1/2}\right|/\hbar \approx \left|\omega_Z - 2\tau_3\Delta_{curv}^{\parallel}\right|$$

Matrix element of spin-flip transitions:

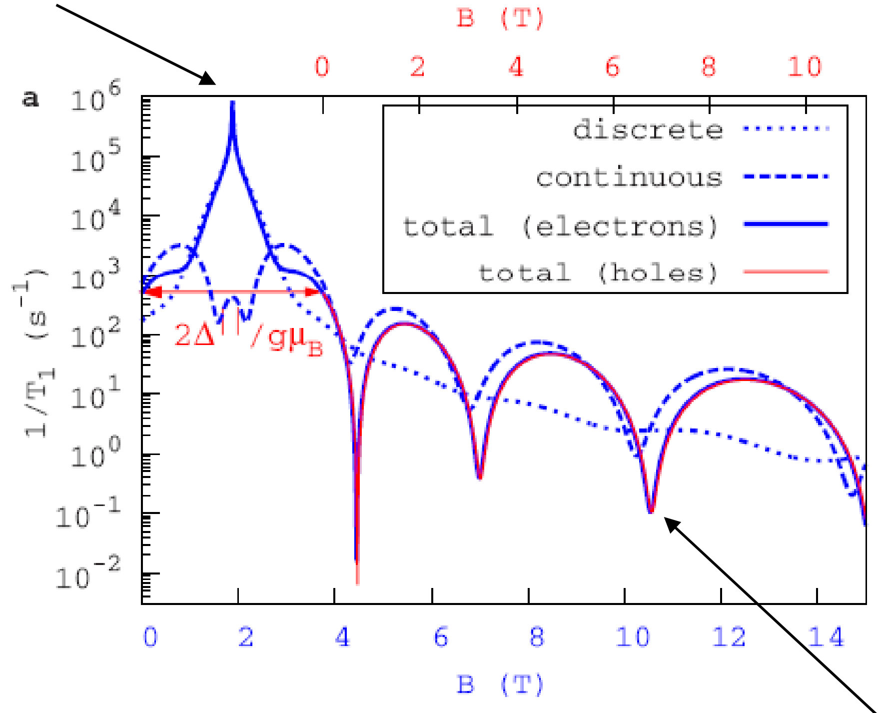
$$\boxed{M_\omega = \left\langle \Psi_{0,0,-1/2}\middle| V_{el-ph}(\omega) \middle| \Psi_{0,0,1/2}\right\rangle}$$

For BM phonons: $\boxed{T_2 = 2T_1}$

Spin relaxation rate II

$$\boxed{\frac{1}{T_1} \propto \frac{1}{\sqrt{\omega_0}}}$$

$$\boxed{\omega_0 \approx \left|\omega_Z - 2\tau_3\Delta^{\|}_{curv}\right|}$$

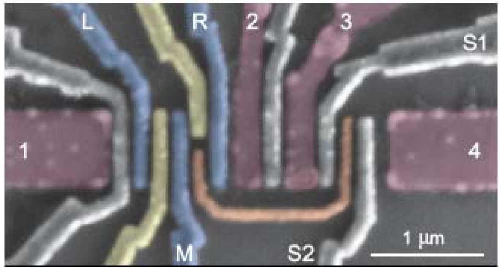


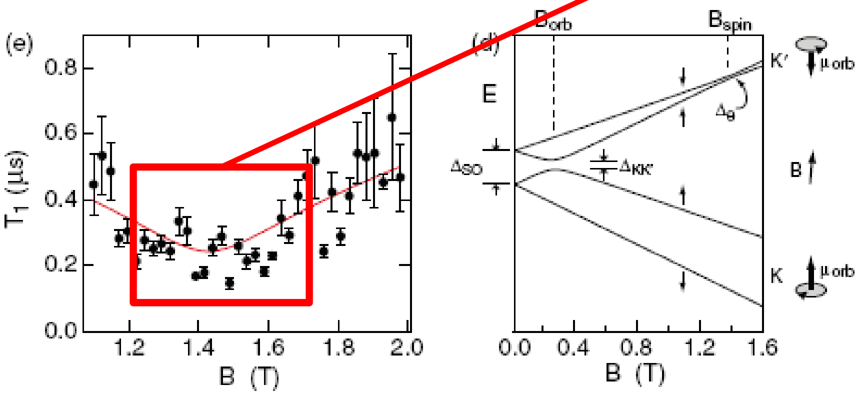
Bulaev, BT & Loss PRB 2008

resonances at magic B-field values

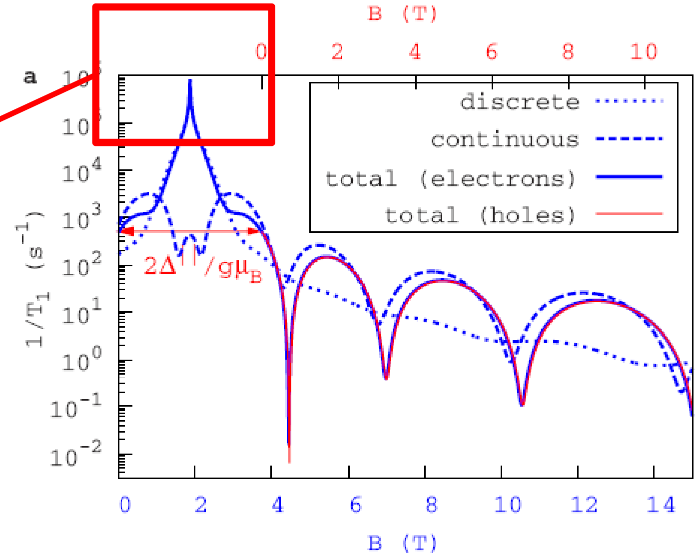
similar analysis in graphene quantum dots: *Struck & Burkard* arXiv 2010

Comparison with NT-experiment





Churchill … & Marcus PRL 2009



Bulaev, BT & Loss PRB 2008

Outline

- **spin qubits** in graphene QDs → peculiarities and visions
- **spin relaxation and dephasing** in carbon nanotube QDs due to SOI and electron-phonon coupling
- **spin decoherence** in carbon nanotube and graphene QDs due to hyperfine interaction

remember Bill Coish's talk

Nuclear-spin interactions

Fermi contact interaction $\longrightarrow$ $h_1^k = \frac{2\mu_0}{3}\gamma_S \gamma_{j_k} \delta(\mathbf{r}_k)\mathbf{S}\cdot\mathbf{I}_k$

anisotropic hyperfine interaction $\longrightarrow$ $h_2^k = \frac{\mu_0}{4\pi}\gamma_S \gamma_{j_k} \frac{3(\mathbf{n}_k\cdot\mathbf{S})(\mathbf{n}_k\cdot\mathbf{I}_k)-\mathbf{S}\cdot\mathbf{I}_k}{r_k^3(1+d/r_k)}$

coupling of electron angular momentum to nuclear spins $\longrightarrow$ $h_3^k = \frac{\mu_0}{4\pi}\gamma_S \gamma_{j_k} \frac{\mathbf{L}_k\cdot\mathbf{I}_k}{r_k^3(1+d/r_k)}$

carbon specialty: ^{12}C vs ^{13}C abundance

Effective Spin Hamiltonian I

Electron states: $\left|\Psi_\sigma\right\rangle = \left|\Phi;u\right\rangle\left|\sigma\right\rangle$ where $\left\langle \mathbf{r}\middle|\Phi;u\right\rangle = \Phi(\mathbf{r})u(\mathbf{r})$

use hydrogenic orbital approximation
for Bloch amplitudes at K and K':

$$u(\mathbf{r}) = \sum_{\mathbf{R}} \pi(\mathbf{r}-\mathbf{R})$$

$$\pi(\mathbf{r}) = N_{nm}\left\{\psi_{2p_\perp}(\mathbf{r}) + \frac{\pi}{\sqrt{12L}}\left(\psi_{2s}(\mathbf{r}) + \sin(3\theta_{nm})\psi_{2p_t}(\mathbf{r}) + \cos(3\theta_{nm})\psi_{2p_c}(\mathbf{r})\right)\right\}$$

see also (for sp-hybridization in NTs): *Kleiner & Eggert* PRB 2001

Effective Spin Hamiltonian I

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calculate: $\left\langle \Psi_\sigma \middle| h_1^k \middle| \Psi_{\sigma'}\right\rangle = \frac{2\mu_0\gamma_S\gamma_{13C}}{3}\sum_k \left|u(\mathbf{r}_k)\right|^2 \left|\Phi(\mathbf{r}_k)\right|^2 \left\langle \sigma \middle| \mathbf{S}\cdot\mathbf{I}_k \middle| \sigma'\right\rangle$

$\Rightarrow$ effective spin Hamiltonian: $\boxed{H_1 = \sum_k A_k^{(1)}\mathbf{S}\cdot\mathbf{I}_k}$

$$A_k^{(1)} = A_1 v_0 \left|\Phi(\mathbf{r}_k)\right|^2$$

Effective Spin Hamiltonian II

$$\boxed{H_{hf} = \mathbf{h} \cdot \mathbf{S}, \quad h_j = \sum_k A_k^j I_k^j}$$

Fischer, BT & Loss PRB 2009

$$A_k^j = A_j v_0 \left| \Phi\left(\mathbf{r}_k\right) \right|^2 \text{ and } A_j = A_1 + A_2^j$$

A_1: coupling strength for Fermi contact interaction

A_2^j: coupling strength for anisotropic hyperfine interaction in j direction

Effective Spin Hamiltonian II

$$\boxed{H_{hf} = \mathbf{h}\cdot\mathbf{S}, \quad h_j = \sum_k A_k^j I_k^j}$$

Fischer, BT & Loss PRB 2009

$$A_k^j = A_j v_0 \left|\Phi\left(\mathbf{r}_k\right)\right|^2 \text{ and } A_j = A_1 + A_2^j$$

A_1: coupling strength for Fermi contact interaction

A_2^j: coupling strength for anisotropic hyperfine interaction in j direction

For flat graphene:

$$\boxed{A_1 = 0 \text{ and } A_2^z = A_2^y = -\frac{A_2^x}{2} = -\frac{\mu_0 \gamma_S \gamma_{13C} Z_{eff}^3}{240\pi a_0^3}}$$

Effective Spin Hamiltonian II

$$\boxed{H_{hf} = \mathbf{h}\cdot\mathbf{S}, \quad h_j = \sum_k A_k^j I_k^j}$$

Fischer, BT & Loss PRB 2009

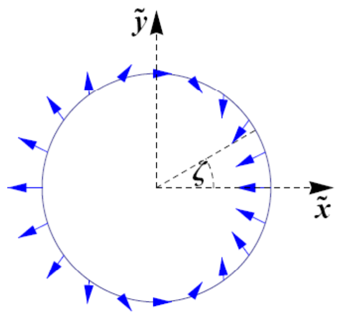
$$A_k^j = A_j v_0 \left|\Phi\left(\mathbf{r}_k\right)\right|^2 \text{ and } A_j = A_1 + A_2^j$$

A_1: coupling strength for Fermi contact interaction

A_2^j: coupling strength for anisotropic hyperfine interaction in j direction

For nanotubes or rippled graphene:

$$\boxed{A_1 \neq A_2^x \neq A_2^y \approx A_2^z \text{ all finite}}$$

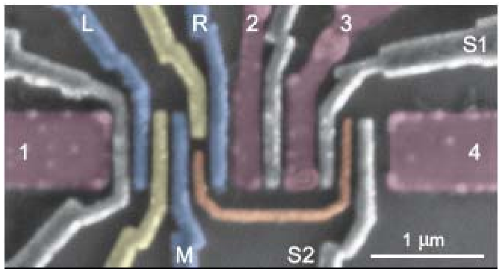


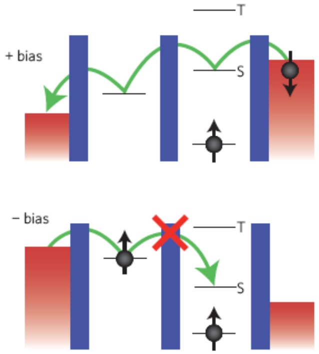
anisotropic Knight shift

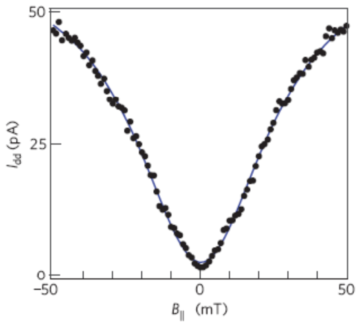
Hyperfine coupling strengths

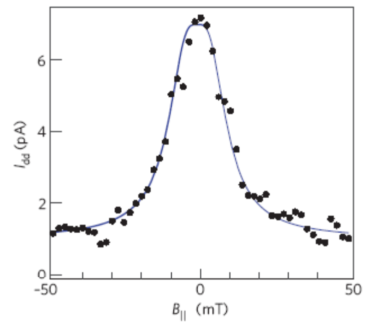
	A_1 [μeV]	A_2^x [μeV]	$A_2^{y,z}$ [μeV]
our values (CNT)	0.05	0.6	-0.3
our values (Graphene)	0	0.6	-0.3
Yazyev[38] (Graphene flakes)	-0.2	0.6	-0.3
Pennington, Stenger[37] (C_{60})	0.1	0.9	-0.5

comparison to (first) experiments: $\boxed{A \sim 100\ \mu\text{eV}}$



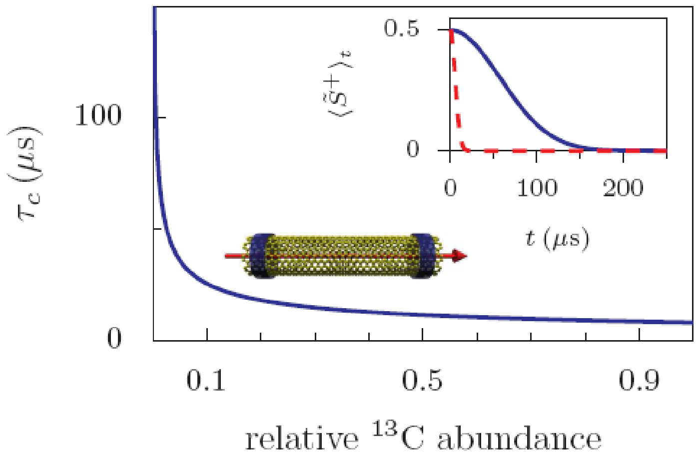


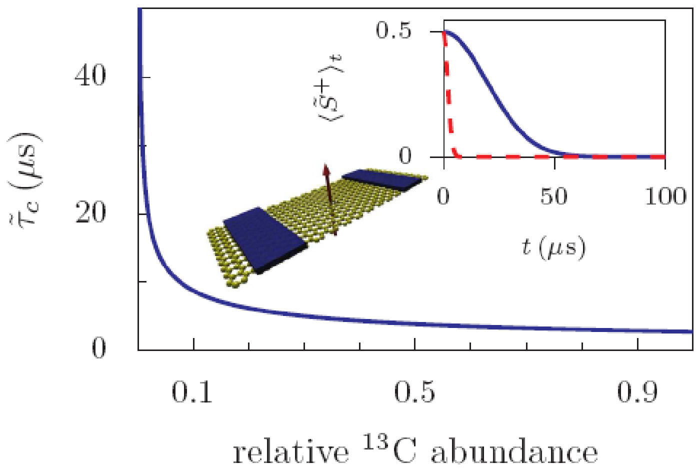




Churchill … & Marcus Nature Phys. 2009

Electron-spin decoherence time: Gaussian decay





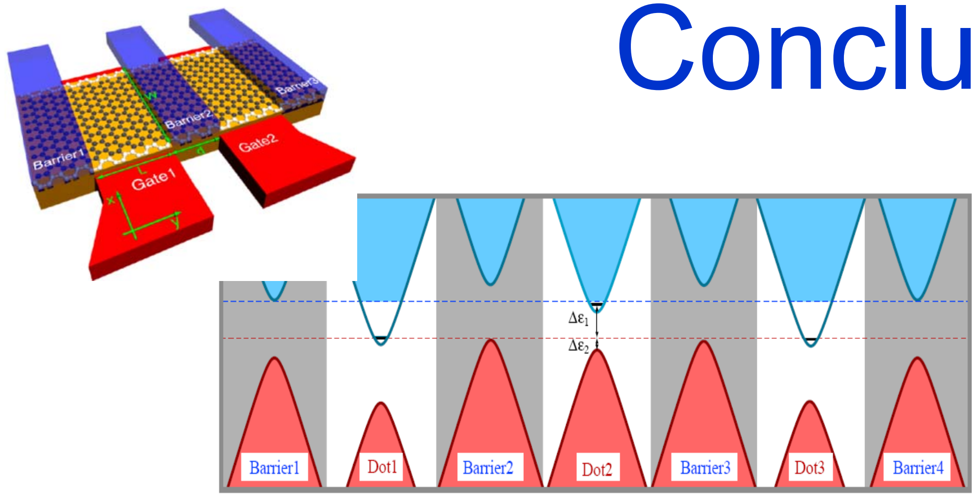
For $N_{13} \gg 1$: $\langle S^+\rangle_t = \langle S^+\rangle_0 \, e^{-t^2/\tau_c^2}$

Fischer, BT & Loss PRB 2009

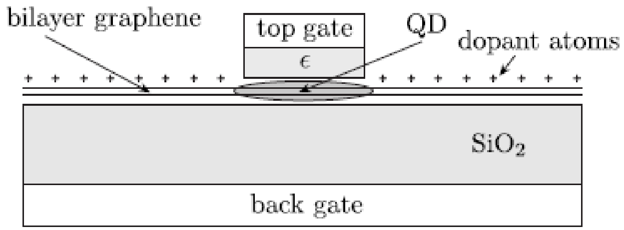
$$\boxed{\tau_c = \frac{2\hbar}{\sqrt{1-p^2}} \frac{N}{\sqrt{N_{13}} A_z}}$$

polarization of nuclear bath

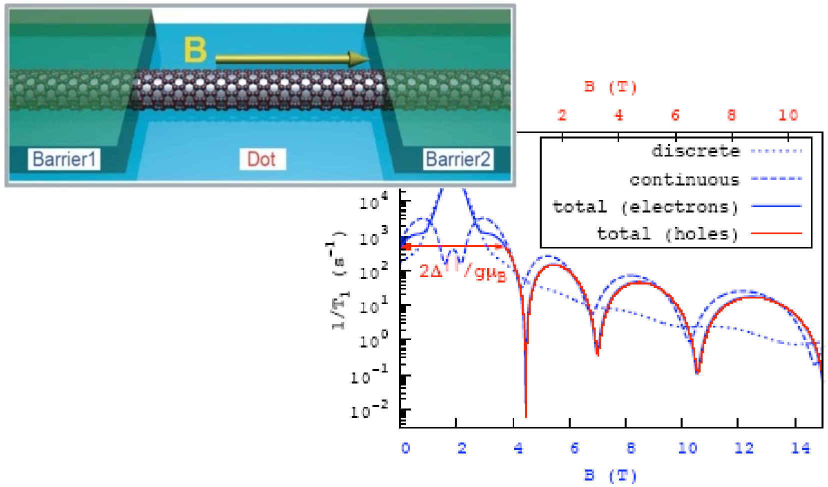
Conclusions



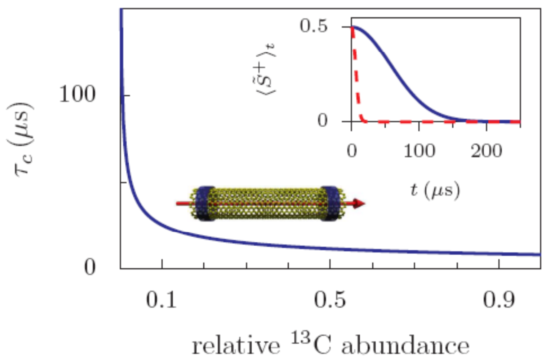
BT, Bulaev, Loss & Burkard Nature Phys. 2007



Recher, Nilsson, Burkard & BT PRB 2009



Bulaev, BT & Loss PRB 2008



Fischer, BT & Loss PRB 2009

<u>Topical Review:</u> *Recher & BT* Nanotechology **21**, 302001 (2010)