Charge and spin diffusion on the metallic side of the metal-insulator transition: a self-consistent approach

Rodolfo A. Jalabert

IPCMS

UNIVERSITÉ DE STRASBOURG

Thomas Wellens (Freiburg)
Dietmar Weinmann (Strasbourg)
Pablo I. Tamborenea (Buenos Aires)
Guido Intronati (Buenos Aires & Strasbourg)

Outline

- Experimental motivation

- Spin-orbit coupling in the impurity-band

- Charge and spin diffusion: phenomenological approach

- Charge and spin diffusion: self-consistent approach

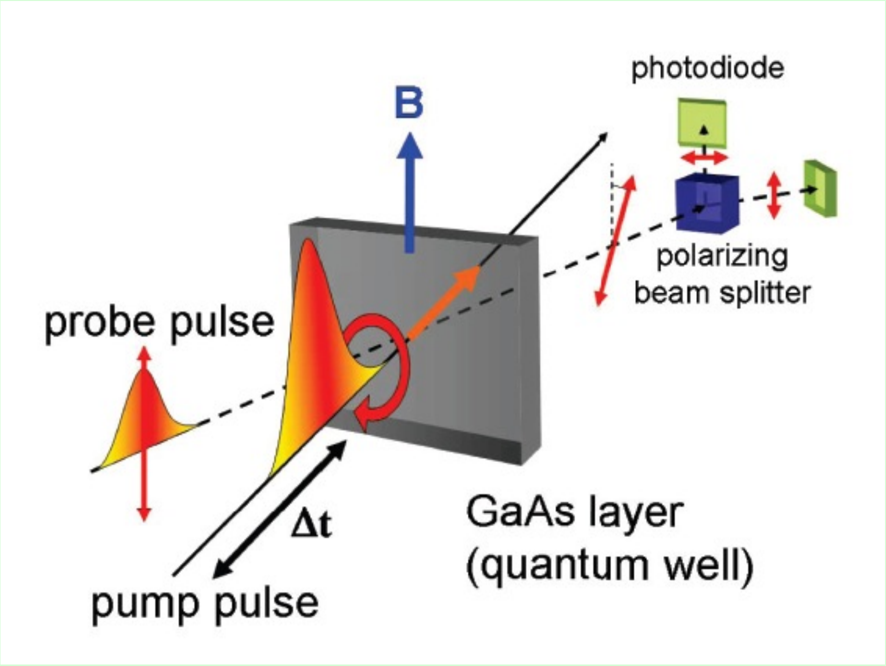
- Theory, numerics and experiments

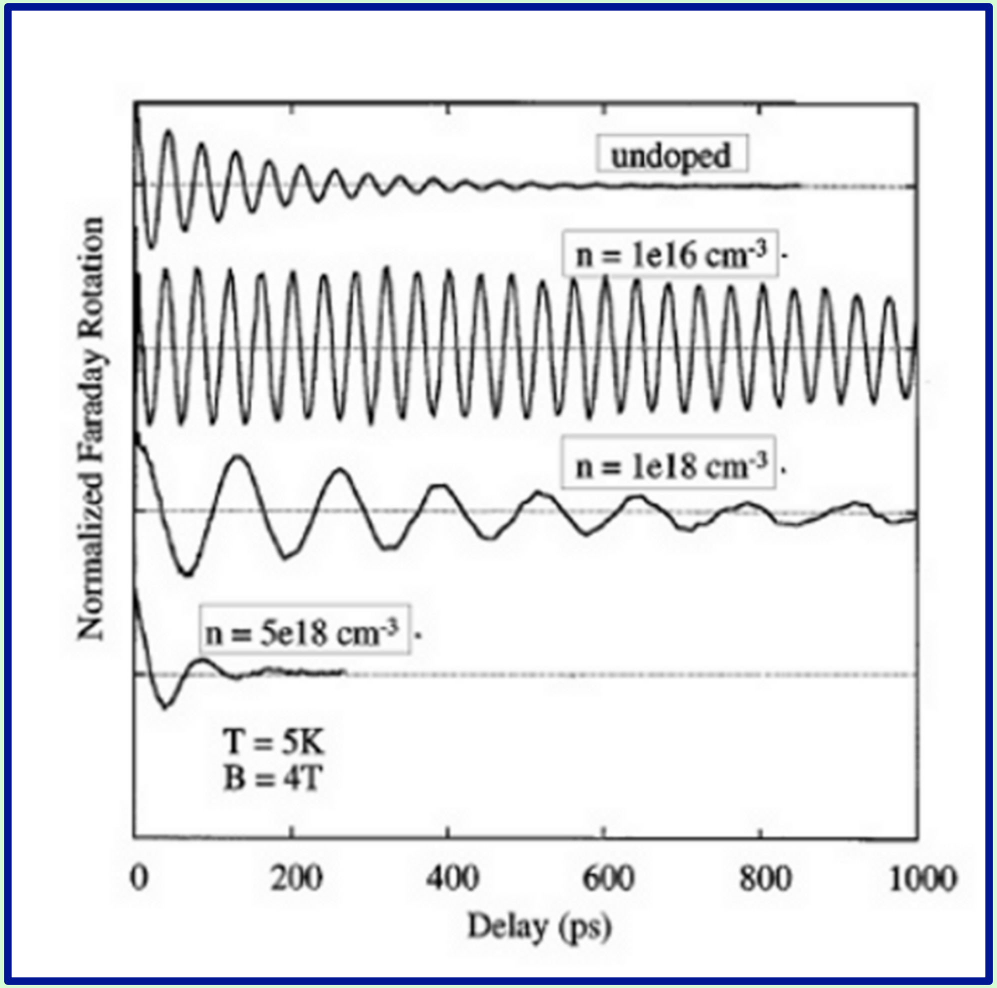
Experimental motivation

Minimum ESR linewidth for barely metallic n-type silicon

Zarifis, Castner, PRB 1987

Very long spin lifetimes for bulk n-doped GaAs





Kikkawa, Awschalom, PRL 1998

Maximum spin relaxation times around the MIT

Kikkawa, Awschalom, PRL 1998

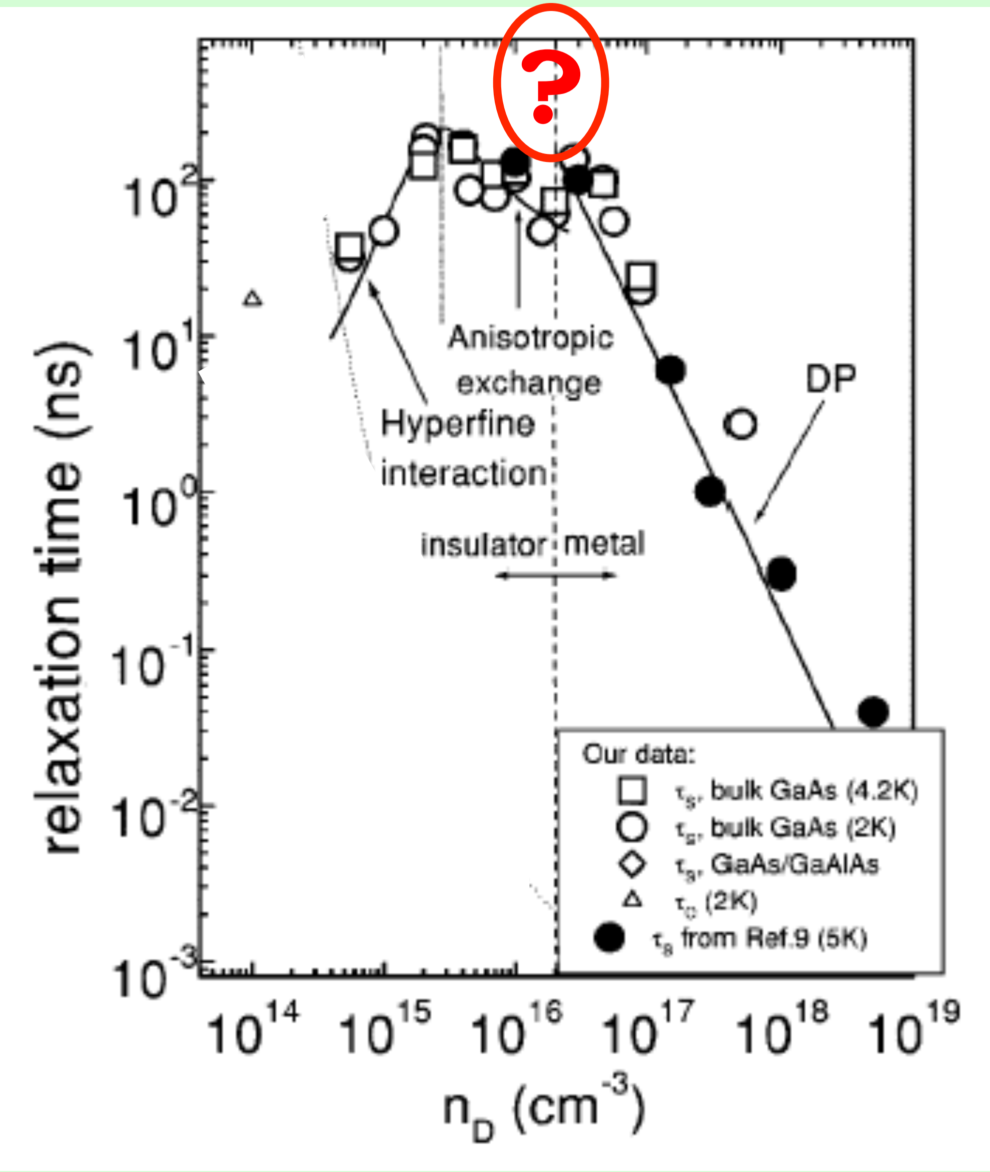
Very long spin lifetimes for bulk n-doped GaAs

larger than 100 ns

R.I. Dzhioev *et al.*, PRB 2002

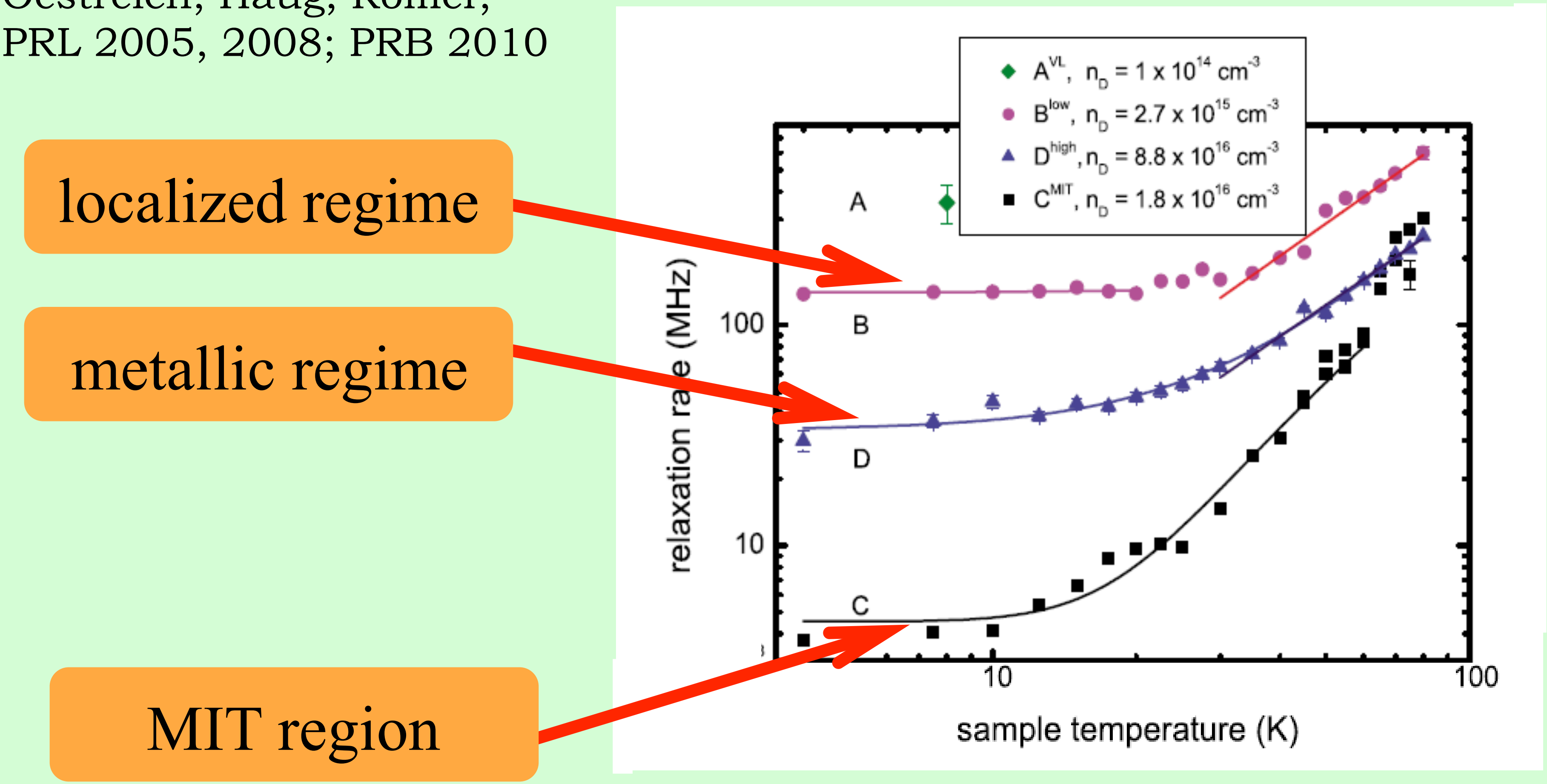
Dependence on Si dopant density in GaAs

Maximum relaxation times at MIT $n_c \approx 2\times10^{16}$ cm^{-3}



Spin noise spectroscopy for bulk n-doped GaAs in different transport regimes

Oestreich, Haug, Römer,
PRL 2005, 2008; PRB 2010

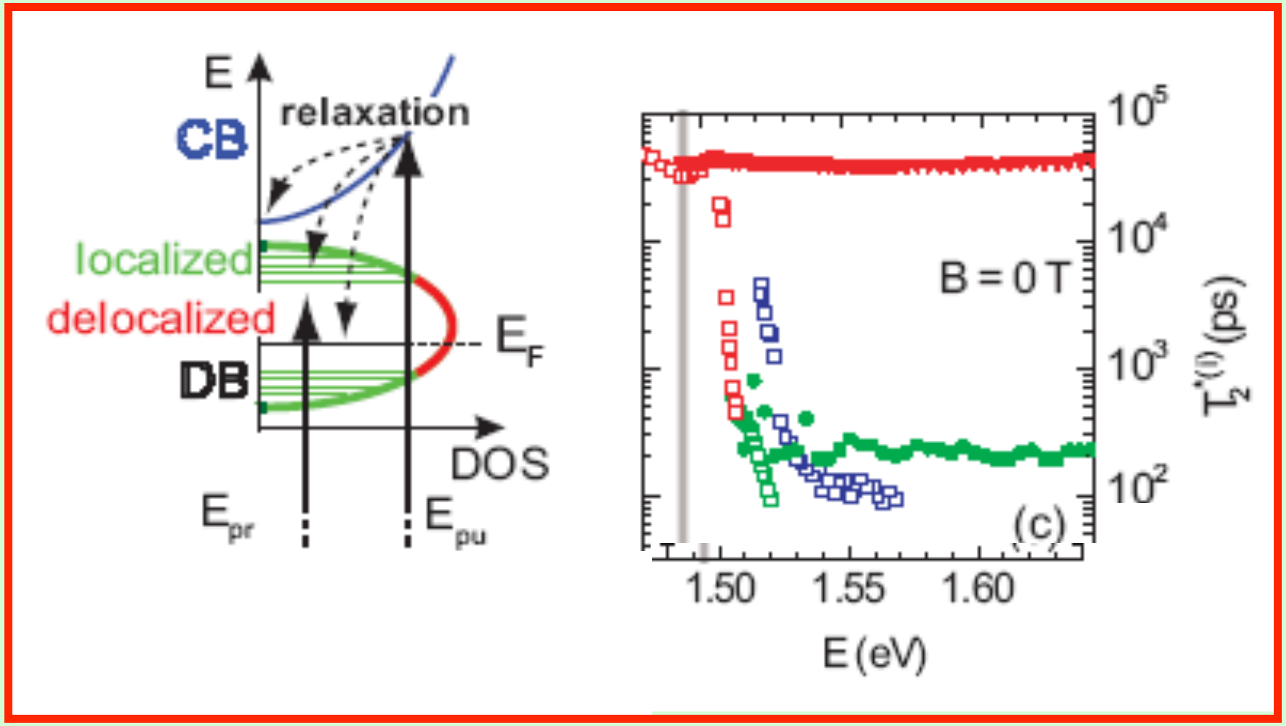


Very long spin lifetimes: 45 – 270 ns for $n_c \approx 1.8\times10^{16}$ cm^{-3} (MIT)

Connection between spin lifetime and transport properties

Mapping of spin lifetimes to electronic states in n-type GaAs near the metal-insulator transition

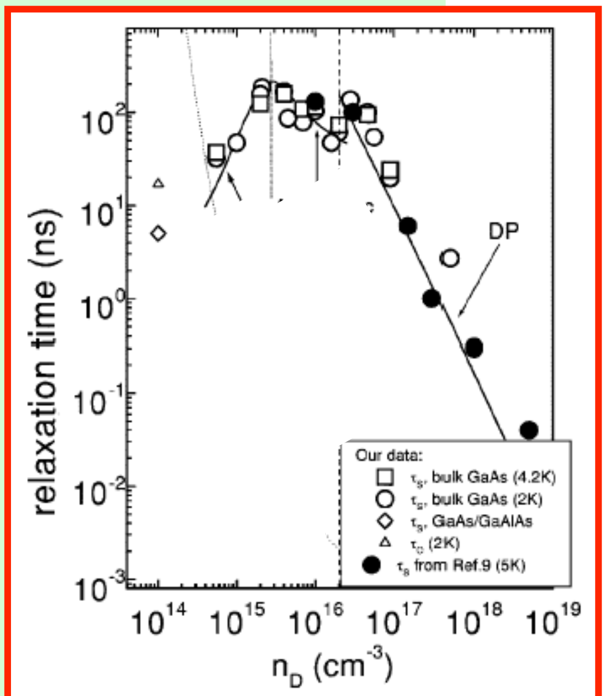
L. Schreiber, M. Heidkamp, T. Rohleder, B. Beschoten, and G. Güntherodt, arXiv: 0706.1884v1

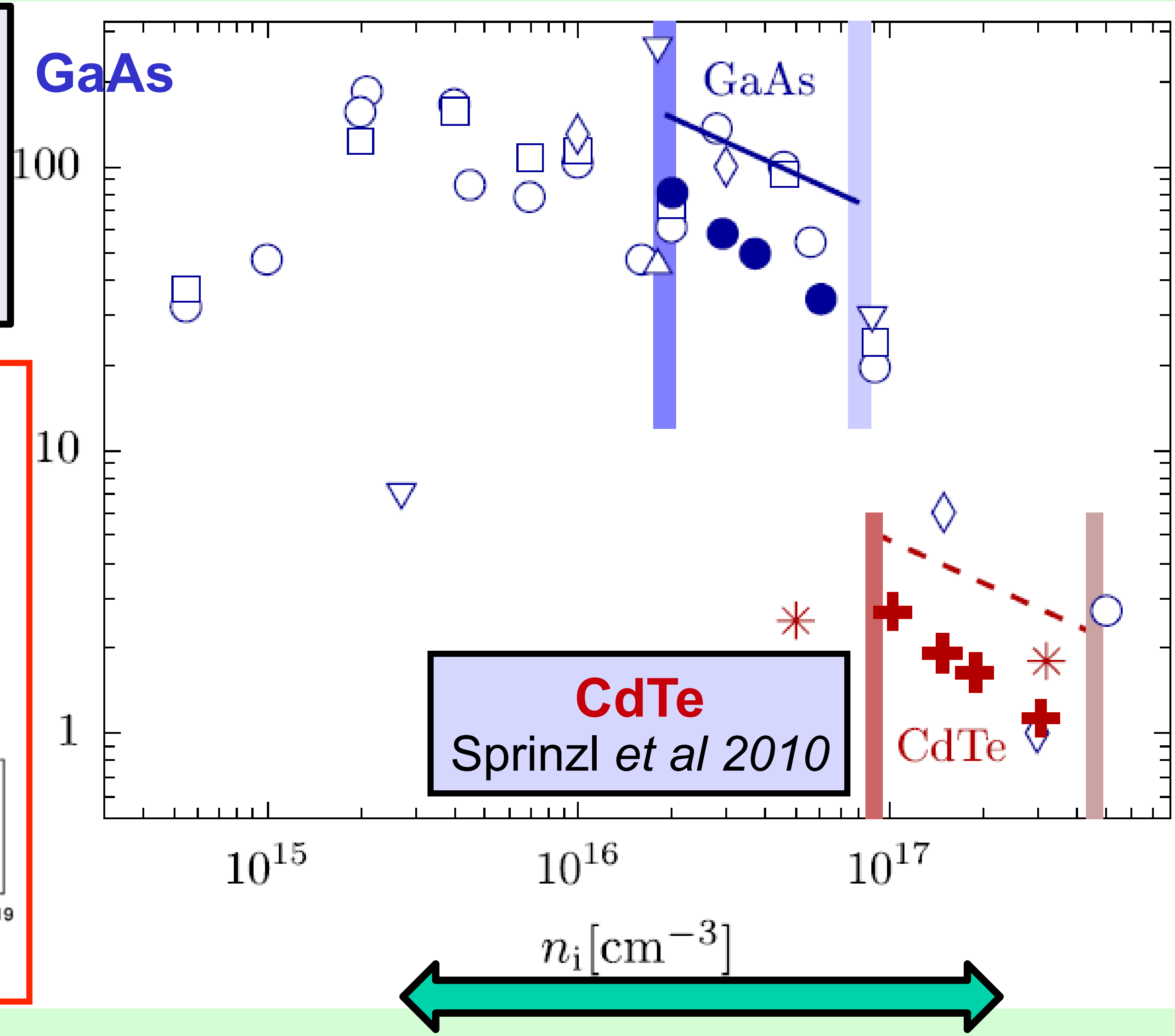


longest spin lifetimes for delocalized donor band states

Spin relaxation time for GaAs and CdTe: experiments, theory and numerics

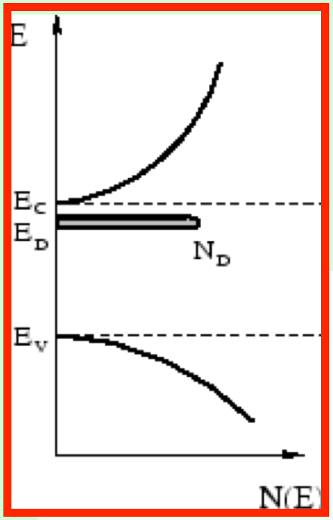
- △ Oestreich *et al* 2005
- ▽ Römer *et al* 2010
- ◇ Kikkawa & Awschalom 1998
- ○ Dzhioev *et al* 2002
- ● Our numerical result
- — Our analytical result



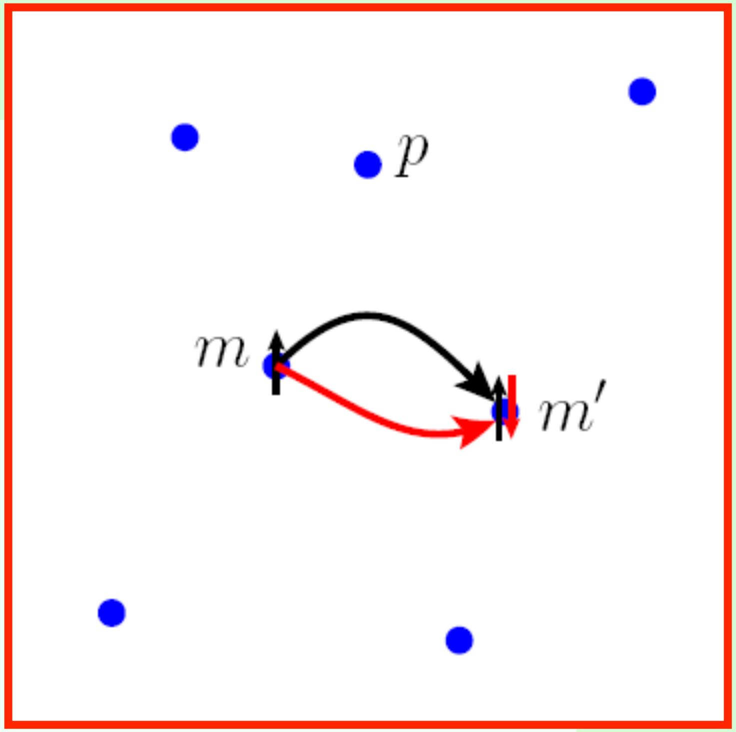


G. Intronati, P. Tamborenea, D. Weinmann, R.A.J., PRL 2012

Model for impurity-band conduction (Matsubara & Toyozawa)



$$H = T + \sum_p V_p$$



$V_p = -\frac{e^2}{\epsilon|\mathbf{r}-\mathbf{R}_p|}$ — impurity potential

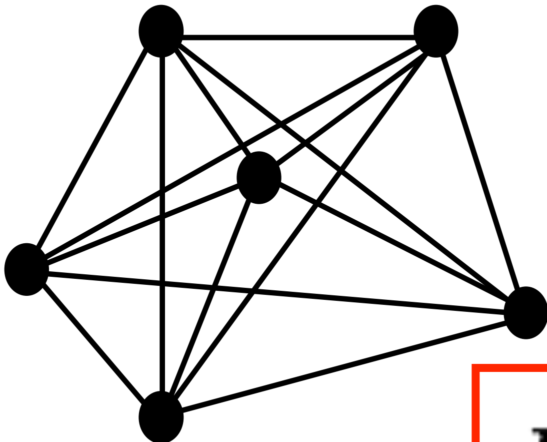
$(T + V_P)\,|p\rangle = \varepsilon_0\,|p\rangle$ — hydrogenic states

Working in the basis of impurity states

$$t_{mm'} = \langle m'|V_{m'}|m\rangle + \sum_{p\neq m,m'} \langle m'|V_p|m\rangle$$

$\langle m'|V_{m'}|m\rangle = -\frac{e^2}{\epsilon a}(1 + R^*_{mm'})\,e^{-R^*_{mm'}}$ — dominates transport

Problem of a random quantum network



$\mathbf{r}_m$, $\mathbf{r}_n$: random positions of sites m, n

$$H = \sum_{m \neq n} V(\mathbf{r}_m - \mathbf{r}_n) |m\rangle \langle n|$$

Diffusion given by the long-time behavior of the probability density

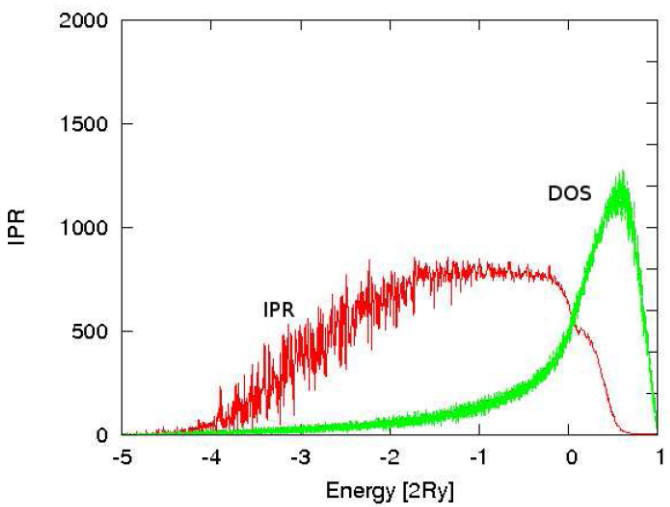
$$P(\mathbf{r}', \mathbf{r}, t) = \overline{\sum_{mn} \langle m | e^{-iHt/\hbar} | n \rangle \langle n | e^{iHt/\hbar} | m \rangle \delta(\mathbf{r}' - \mathbf{r}_m) \delta(\mathbf{r} - \mathbf{r}_n)}$$

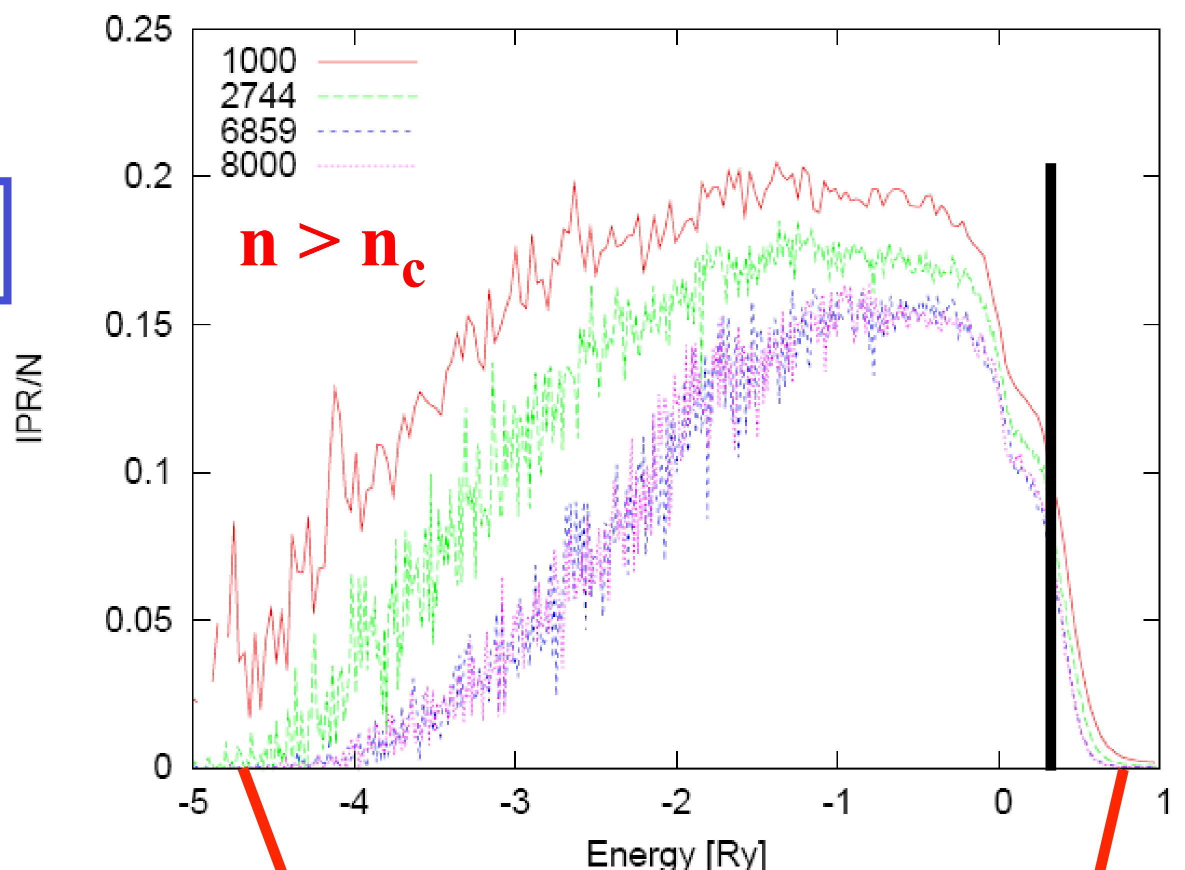
E. Akkermans and G. Montambaux, Mesoscopic Physics (2007)

Extended and localized states in the impurity-band

$n_c^{1/3} a_B \approx 0.25$

$\mathcal{N}_i = n_i a^3 \simeq 0.017$



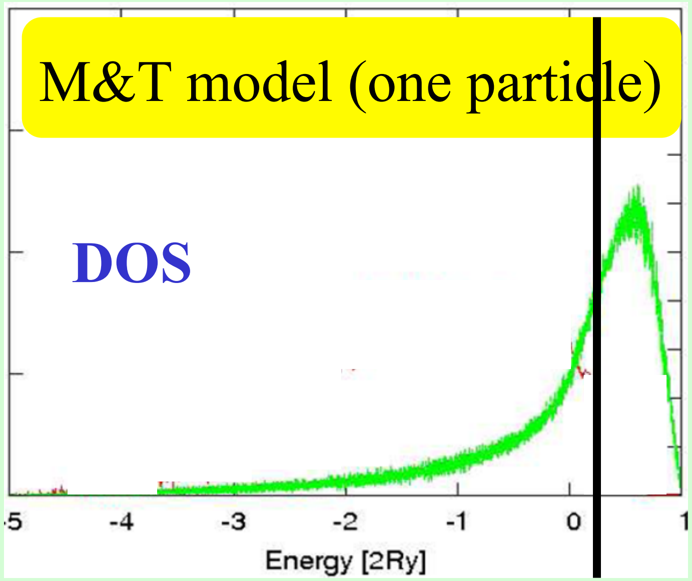


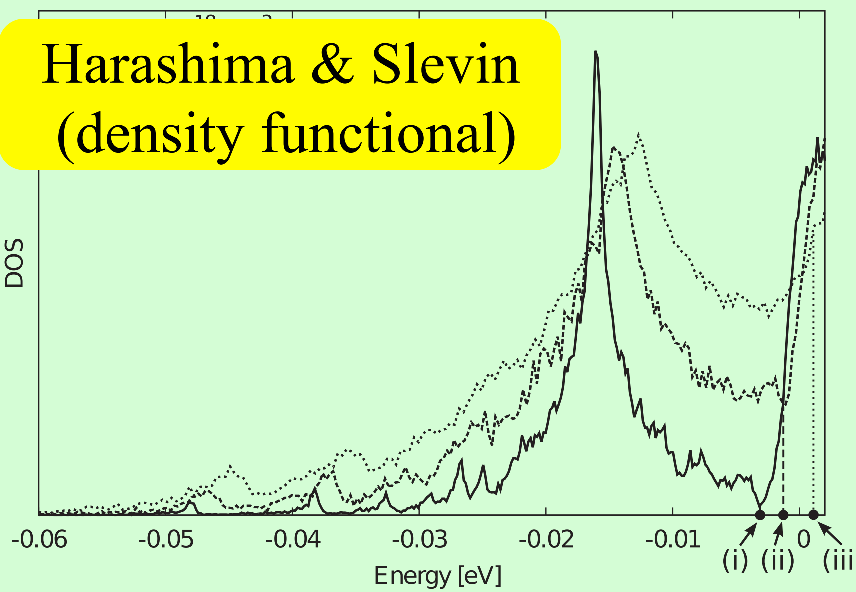
$$\mathrm{IPR} = \left[\frac{\sum_m |\langle \phi_m | \psi_i \rangle|^4}{(\sum_m |\langle \phi_m | \psi_i \rangle|^2)^2}\right]^{-1}$$

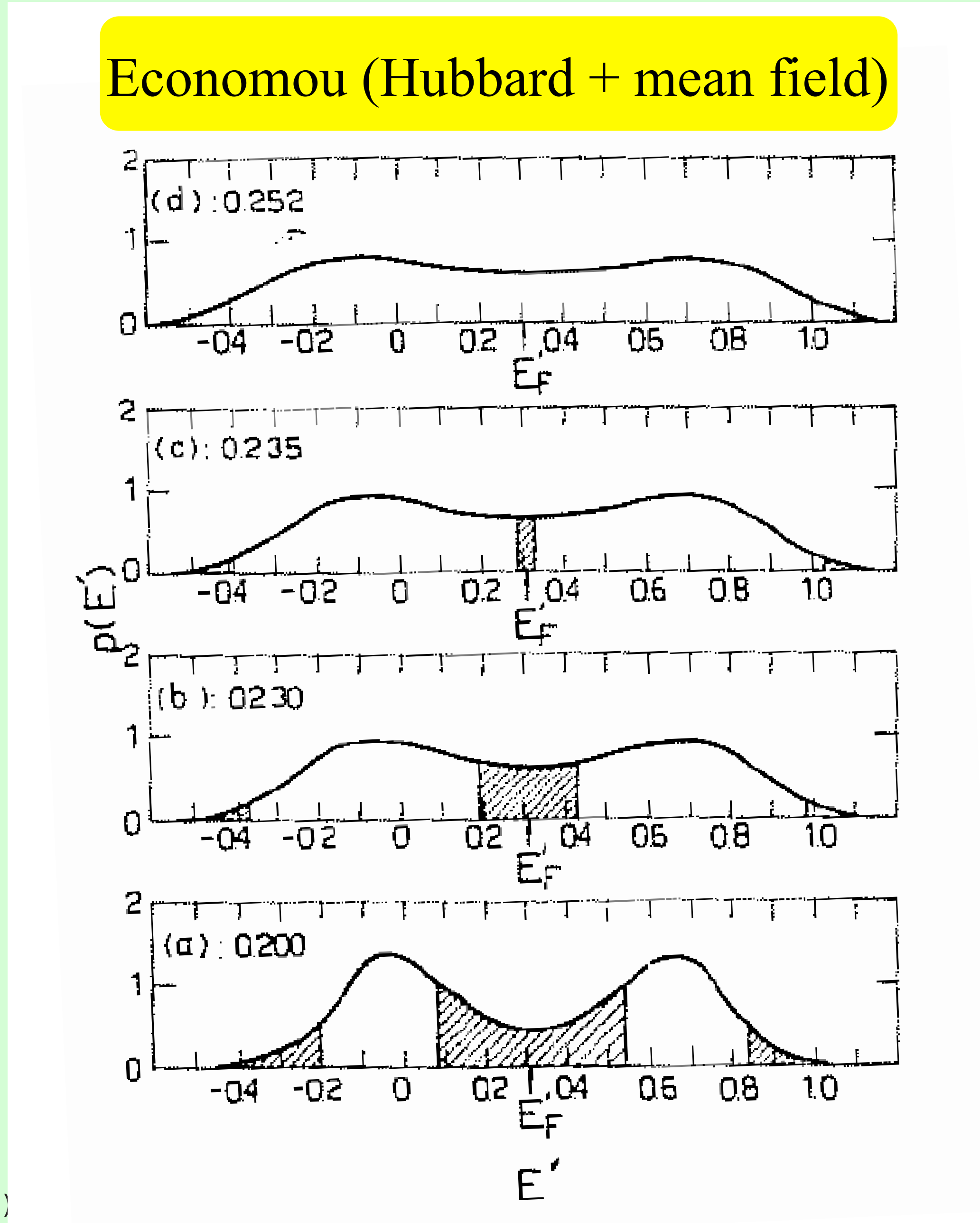
extended states

localized states

Impurity band: one vs. many particle models

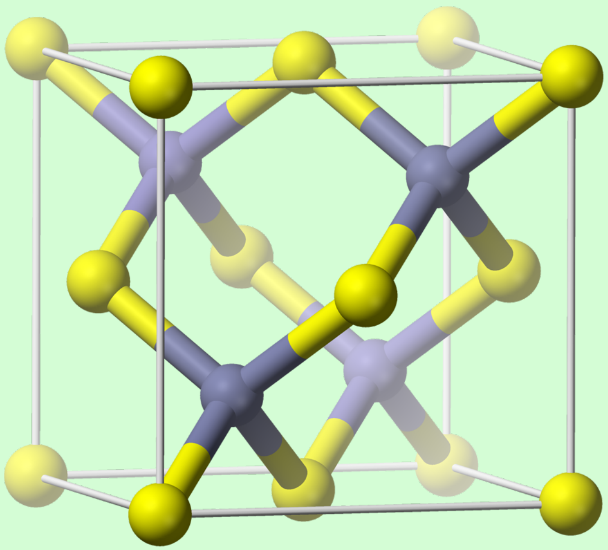






Effective Hamiltonian for zinc-blende semiconductors

$$H = H_0 + H_{SIA} + H_{BIA}$$



extrinsic coupling:

$$H_{SIA} = \lambda \boldsymbol{\sigma} \cdot \nabla V_{imp} \times \mathbf{k}$$

$\lambda \simeq -5.3\ \text{Å}^2$ **(GaAs)**

k : **momentum operator**

$\gamma = 27 eV\text{Å}^3$ **(GaAs)**

intrinsic coupling (Dresselhaus):

$$H_{BIA} = \gamma \left[\sigma_x k_x (k_y^2 - k_z^2) + \text{cyclic permutations}\right]$$

Impurity band with spin-orbit interaction

$$\mathcal{H} = \sum_{m' \neq m} \sum_{\sigma' \sigma} \mathcal{V}^{\sigma',\sigma}(\mathbf{r}_{m'} - \mathbf{r}_m) |m'\sigma'\rangle\langle m\sigma|$$

Hopping matrix: $\mathcal{V}(\mathbf{r}) = \begin{pmatrix} \mathcal{V}_0(\mathbf{r}) + i\mathcal{C}_z(\mathbf{r}) & i\,\mathcal{C}_x(\mathbf{r}) + \mathcal{C}_y(\mathbf{r}) \\ i\,\mathcal{C}_x(\mathbf{r}) - \mathcal{C}_y(\mathbf{r}) & \mathcal{V}_0(\mathbf{r}) - i\mathcal{C}_z(\mathbf{r}) \end{pmatrix}$

- spin-independent: $\mathcal{V}_0(\mathbf{r}) = -V_0 \left(1 + \frac{r}{a}\right) e^{-r/a}$
- spin-dependent: (Dresselhaus) $\mathcal{C}_x(\mathbf{r}) = -\frac{\gamma}{3a^5 r}\, x\, (y^2 - z^2)\, e^{-r/a}$

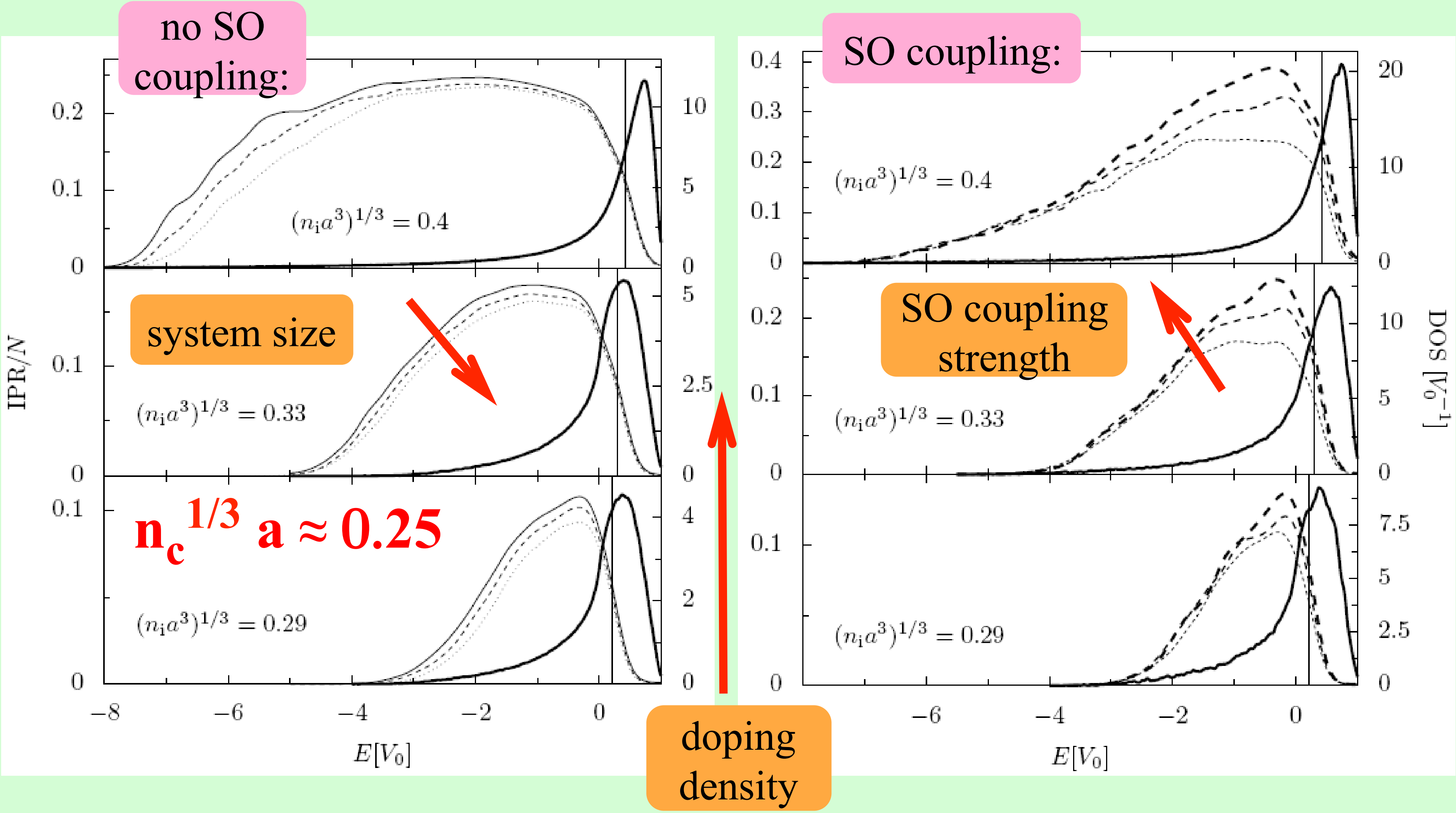
  $\mathcal{C}_{y,z}(\mathbf{r})$: cyclic permutations

$\mathbf{r} = (x, y, z)$

The hopping matrix is "para-odd" $\mathcal{V}(-\mathbf{r})\mathcal{V}(\mathbf{r}) = c(\mathbf{r})\,\mathbb{I}_2$

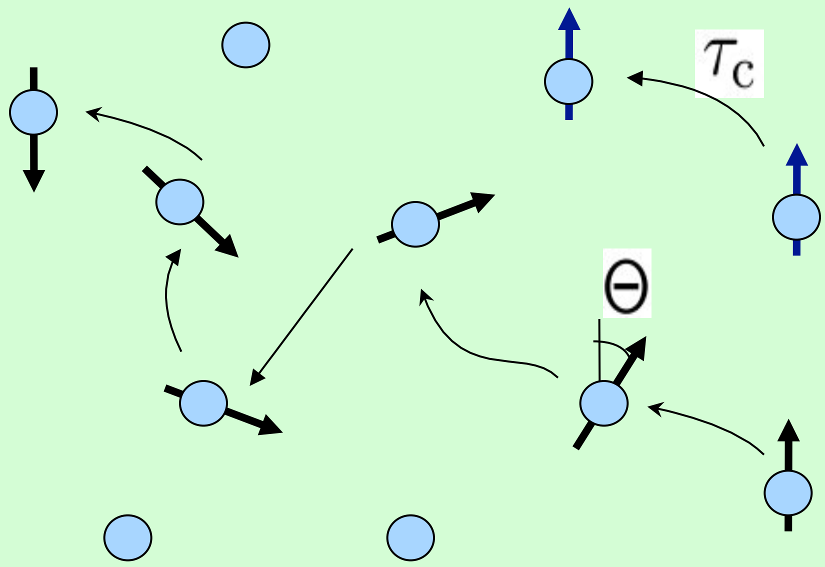
Dresselhaus spin-orbit coupling dominant for semiconductors with zinc-blende crystal structure *(G. Intronati, P. Tamborenea, D. Weinmann, R.A.J., PRL 2012)*

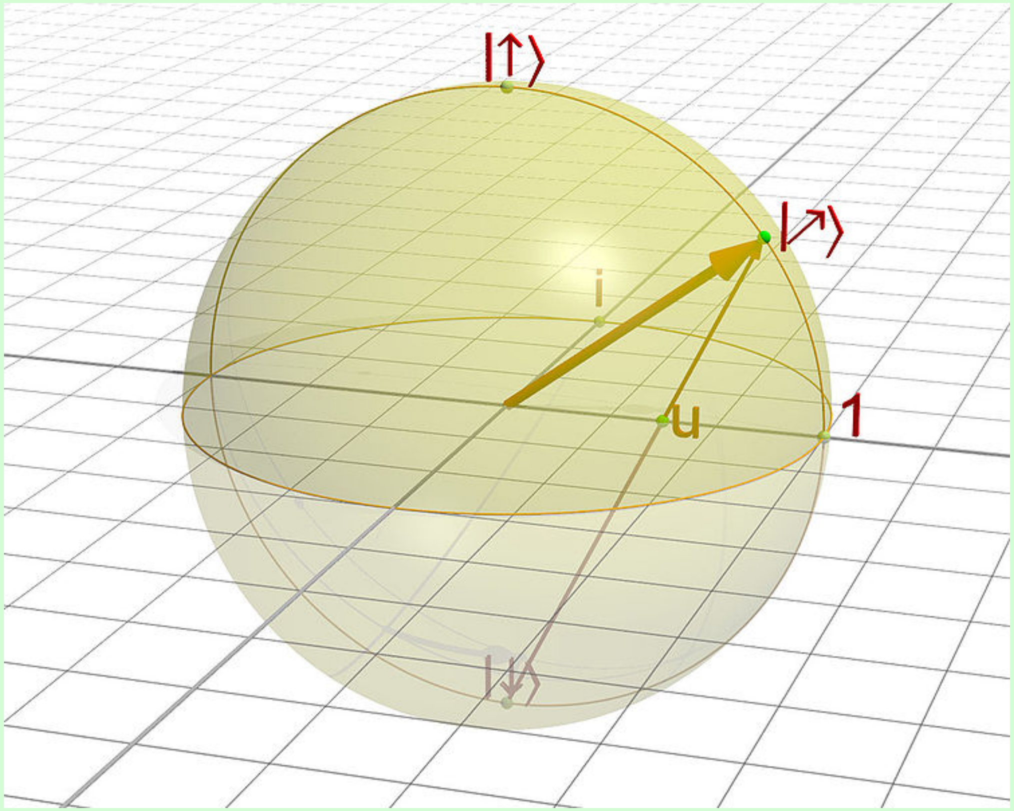
SO induced delocalization in the impurity-band



SO coupling { **does not considerably alter the DOS**
delocalizes by moving the mobility edge

Charge and spin diffusion: phenomenological approach





hopping rate:

$$\frac{1}{\tau_c} = \frac{\sqrt{2}}{\hbar}\left(\sum_{m\neq m'} |\langle m'\sigma|H_0|m\sigma\rangle|^2\right)^{1/2} \simeq \frac{\sqrt{14\pi}V_0}{\hbar}\mathcal{N}_i^{1/2}$$

$$\mathcal{N}_i = n_i a^3$$

Spin diffusion on the Bloch sphere

Evolution of the probability density for an initial condition at the north pole

$$\rho(\theta,t) = \sum_{n=0}^{\infty} \frac{2n+1}{4\pi} \exp\left[-\frac{1}{4}n(n+1)U(t)\right] P_n(\cos\theta)$$

$$U(t) = (t/\tau_c)\langle\alpha^2\rangle$$

Typical spin-rotation angle per hop:

$$\langle\alpha^2\rangle \propto \left(\frac{\gamma}{a^3 V_0}\right)^2$$

$$\langle S_z(t)\rangle = \int d\Omega \cos\theta \rho(\theta,t) = \exp\left[-\frac{1}{2}\frac{\langle\alpha^2\rangle t}{\tau_c}\right]$$

spin relaxation rate: ➡

$$\frac{1}{\tau_s} = \frac{1}{2}\frac{\langle\alpha^2\rangle}{\tau_c} = 0.36\,\frac{\gamma^2}{a^6 V_0 \hbar}\,\mathcal{N}_{\mathrm{i}}^{1/2}$$

G. Intronati, P. Tamborenea, D. Weinmann, R.A.J., PRL 2012

Charge and spin diffusion: self-consistent approach

- Green function:

$$g_{m',m}^{\sigma',\sigma(\pm)}(\varepsilon) = \langle m'\sigma' \left| \frac{1}{\varepsilon \pm i\eta - \mathcal{H}} \right| m\sigma \rangle$$

- Average local Green function:

$$G^{\sigma',\sigma(\pm)}(\varepsilon) = \overline{\left\langle m\sigma' \left| \frac{1}{\varepsilon \pm i\eta - \mathcal{H}} \right| m\sigma \right\rangle} = \delta_{\sigma',\sigma} G^{(\pm)}(\varepsilon)$$

$\rightsquigarrow$ density of states $\rho(\varepsilon) = -\frac{n_{\mathrm{i}}}{\pi} \, \mathrm{Im} \left\{ G^{(+)}(\varepsilon) \right\}$

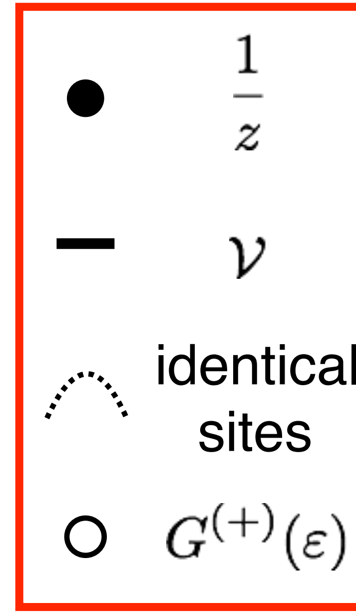
- Two-point Green function (intensity propagator):

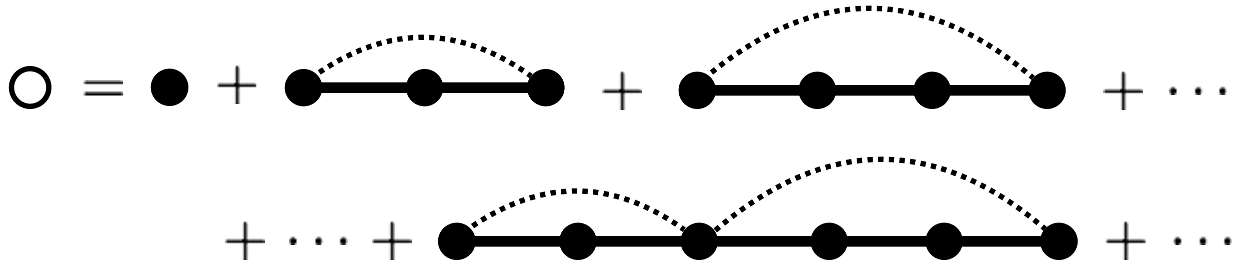
$$\Phi^{\sigma_1'\sigma_2',\sigma_1\sigma_2}(\varepsilon,\omega,\mathbf{r}) = \overline{\sum_{m'} g_{m',m}^{\sigma_1',\sigma_1(+)}\left(\varepsilon + \frac{\hbar\omega}{2}\right) g_{m,m'}^{\sigma_2,\sigma_2'(-)}\left(\varepsilon - \frac{\hbar\omega}{2}\right) \delta\left(\mathbf{r} - \mathbf{r}_{m'm}\right)}$$

$\rightsquigarrow$ charge and spin dynamics $\mathbf{r}_{m'm} = \mathbf{r}_{m'} - \mathbf{r}_m$

Average local Green function and self-energy

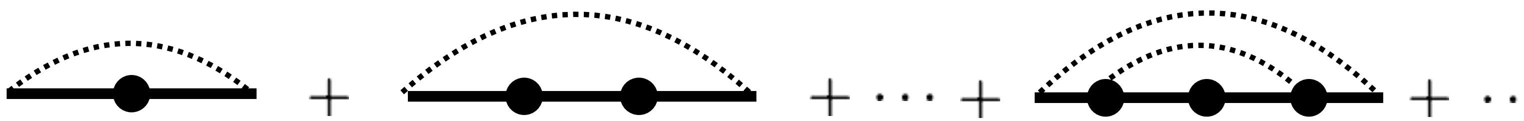
Locator expansion: $$\frac{1}{z-\mathcal{H}} = \frac{1}{z} + \frac{1}{z}\mathcal{H}\frac{1}{z} + \frac{1}{z}\mathcal{H}\frac{1}{z}\mathcal{H}\frac{1}{z} + \ldots$$ $z = \varepsilon + i\eta$



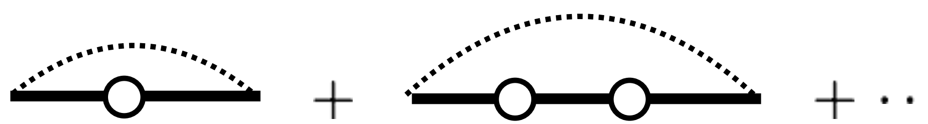


$$\circ = \frac{1}{\varepsilon - \Sigma^{(+)}(\varepsilon)}$$

Dyson equation

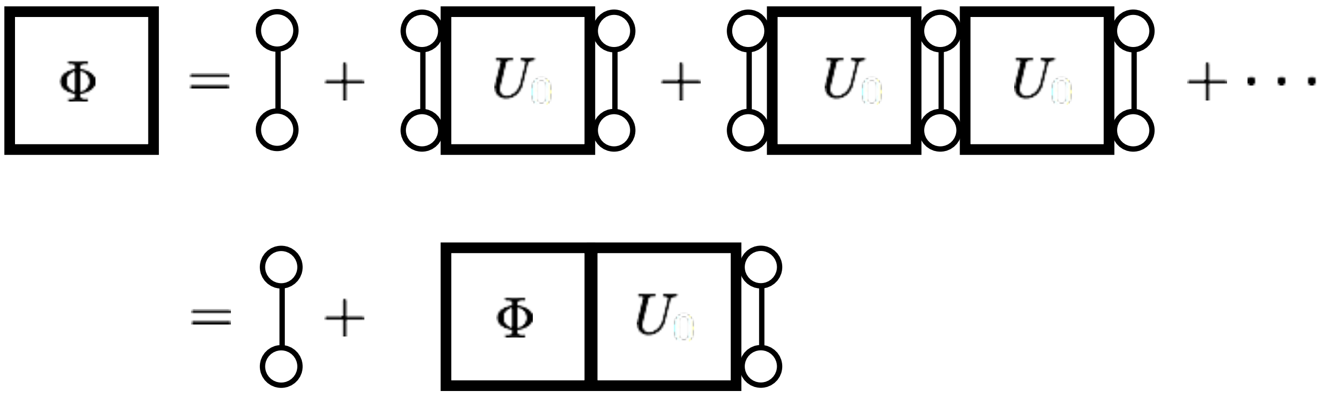
$\Sigma^{(+)}(\varepsilon) =$ 

Self-consistent approach: replace $\frac{1}{z}$ by $G^{(+)}(\varepsilon)$

$\Sigma^{(+)}(\varepsilon) =$ 

Average intensity propagator

Bethe-Salpeter equation for $\Phi = \overline{g^{(+)} g^{(-)}}$



upper line: $g^{(+)}$

lower line: $g^{(-)}$

$$\tilde{\Phi}(\varepsilon, \omega, \mathbf{q}) = \frac{1}{\left[G^{(+)}\left(\varepsilon + \hbar\omega/2\right)\; G^{(-)}\left(\varepsilon - \hbar\omega/2\right)\right]^{-1} - \tilde{U}(\varepsilon, \omega, \mathbf{q})}$$

Symmetry restrictions at $\mathbf{q}=0$

$$\tilde{U}(\varepsilon, \omega, 0) = \begin{pmatrix} \tilde{u}_1(\varepsilon,\omega) & 0 & 0 & \tilde{u}_2(\varepsilon,\omega) \\ 0 & \tilde{u}_1(\varepsilon,\omega) - \tilde{u}_2(\varepsilon,\omega) & 0 & 0 \\ 0 & 0 & \tilde{u}_1(\varepsilon,\omega) - \tilde{u}_2(\varepsilon,\omega) & 0 \\ \tilde{u}_2(\varepsilon,\omega) & 0 & 0 & \tilde{u}_1(\varepsilon,\omega) \end{pmatrix}$$

spin relaxation rate

Charge and spin dynamics

$$P^{\sigma'\sigma}(\varepsilon, t, \mathbf{r}) = \frac{n_{\mathrm{i}}}{\rho(\varepsilon)} \frac{\hbar}{2\pi} \int_{-\infty}^{+\infty} \mathrm{d}\omega \; e^{-i\omega t} \; \Phi^{\sigma'\sigma', \sigma\sigma}(\varepsilon, \omega, \mathbf{r})$$

- Spin relaxation: $$\int \mathrm{d}\mathbf{r} \; P^{\sigma\sigma}(\varepsilon, t, \mathbf{r}) \underset{t\to\infty}{\longrightarrow} \frac{1}{2}\left(1 + e^{-t/\tau_{\mathrm{s}}(\varepsilon)}\right)$$

with rate: $$\boxed{\frac{1}{\tau_{\mathrm{s}}(\varepsilon)} = \frac{4\pi\rho(\varepsilon)}{\hbar n_{\mathrm{i}}} \; \tilde{u}_2(\varepsilon, 0)}$$

- Spatial diffusion: $$\sum_{\sigma'} \tilde{P}^{\sigma'\sigma}(\varepsilon, \omega, \mathbf{q}) \underset{\omega, q\to 0}{\longrightarrow} \frac{1}{-i\omega + q^2 \mathcal{D}(\varepsilon)}$$

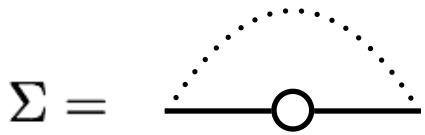
with diffusion constant:

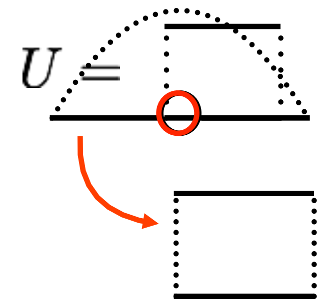
$$\boxed{\mathcal{D}(\varepsilon) = -\frac{\pi\rho(\varepsilon)}{\hbar n_{\mathrm{i}}} \partial^2_{q_\mu} \left(\tilde{U}^{++,++}(\varepsilon, 0, \mathbf{q}) + \tilde{U}^{++,--}(\varepsilon, 0, \mathbf{q}) \right)\Big|_{\mathbf{q}=0}}$$

Self-consistent scheme to get U from $\sum$

*D. Vollhardt and P. Wölfle, PRB **22**, 4666 (1980)*

Ward identity (ensures conservation of the norm)



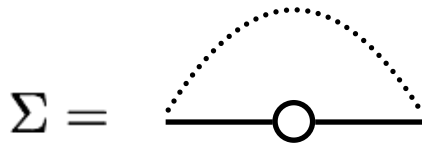


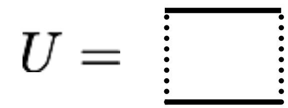
1) select and remove each Green function from Σ

2) fold left parts to lower line

Three levels of approximation i)-ii)

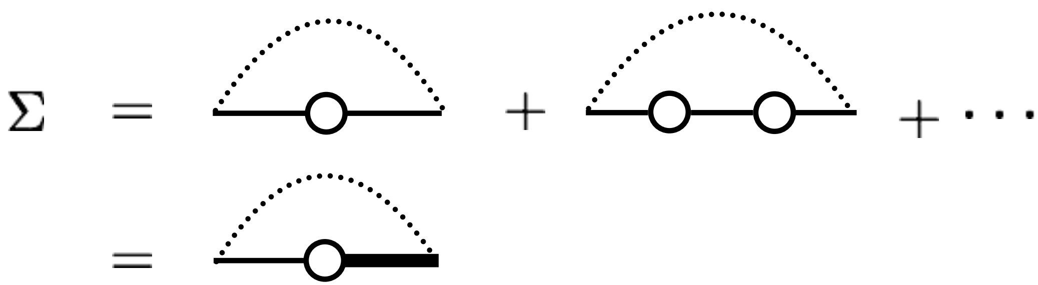
i) simplest (SSCA):

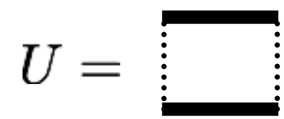




hop from one impurity to another one, and back again

ii) loop corrected (LCSCA):



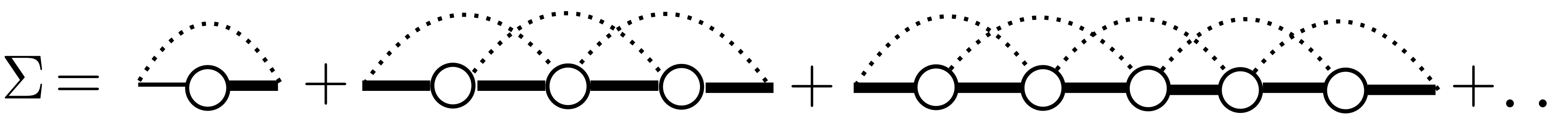


renormalised hopping amplitude:

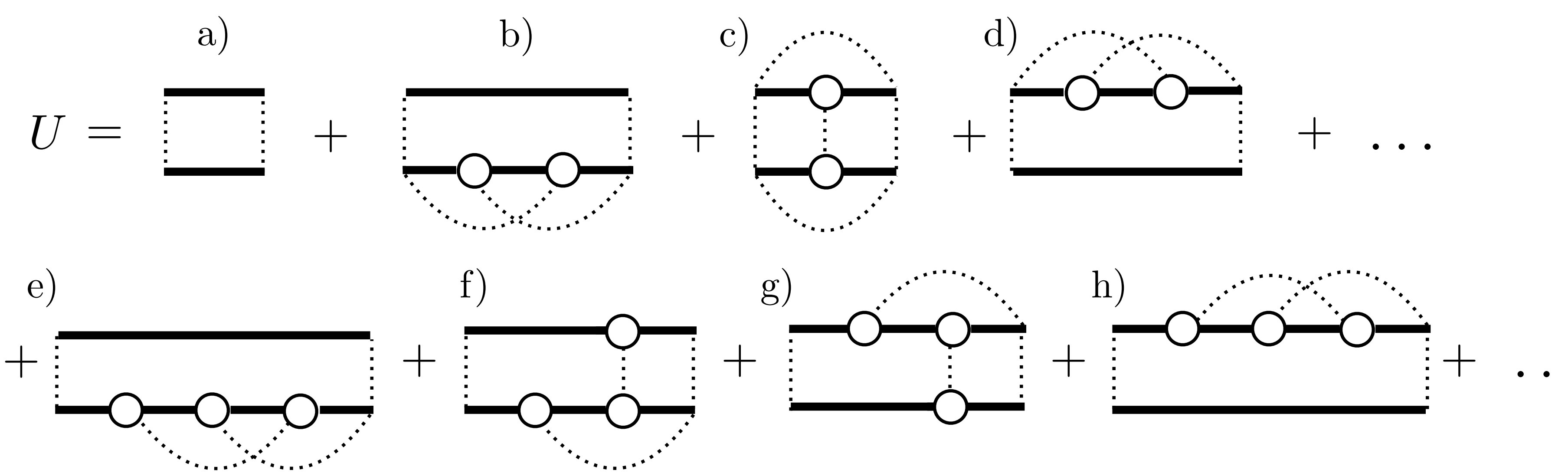
high-density limit: *T. Matsubara and Y. Toyozawa, Prog. Theoret. Phys. (1961)*

Three levels of approximation iii)

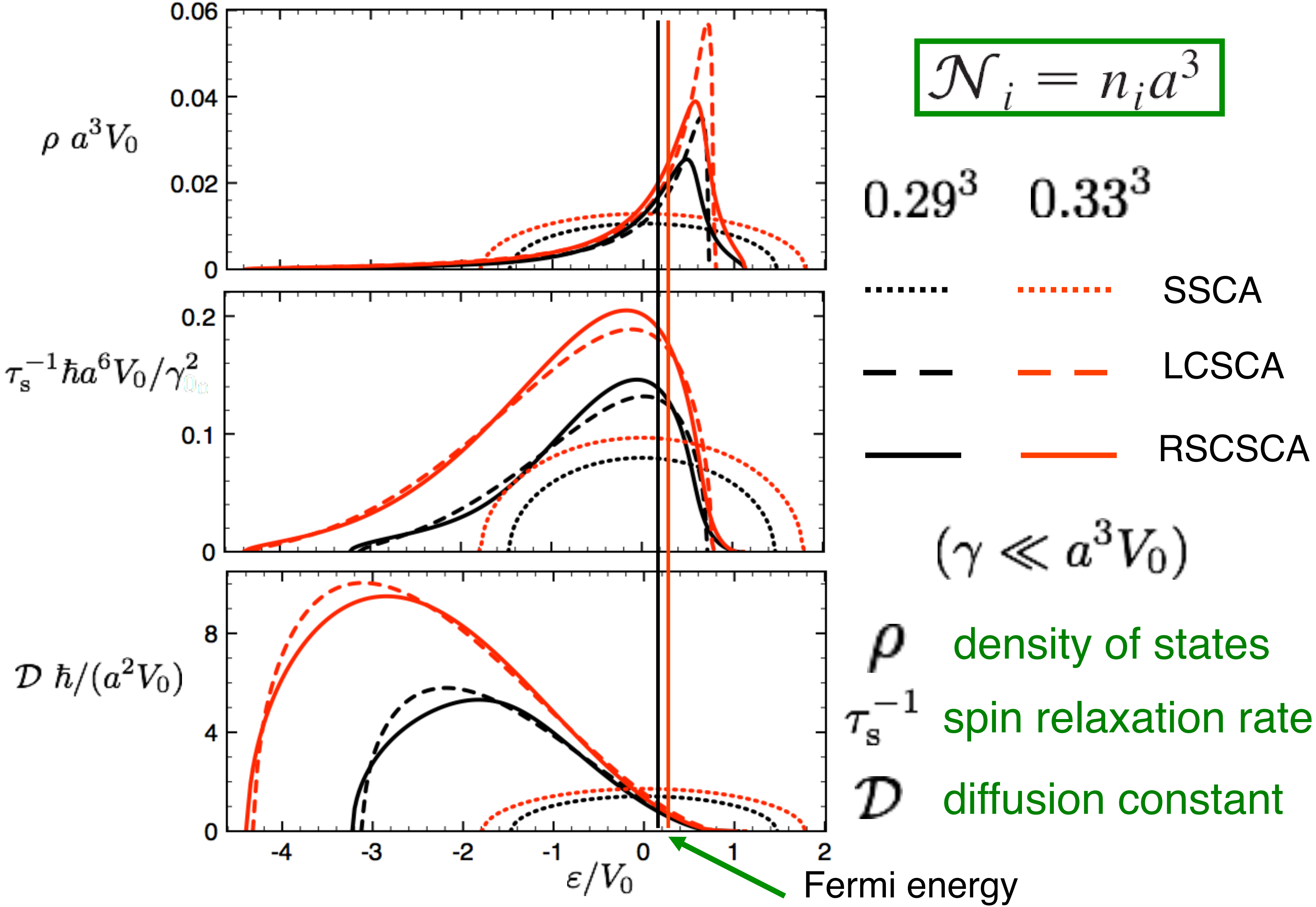
iii) repeated-scattering corrected (RSCSCA), crossed terms:



low-density limit: *P. V. Elyutin, J. Phys. C (1981)*



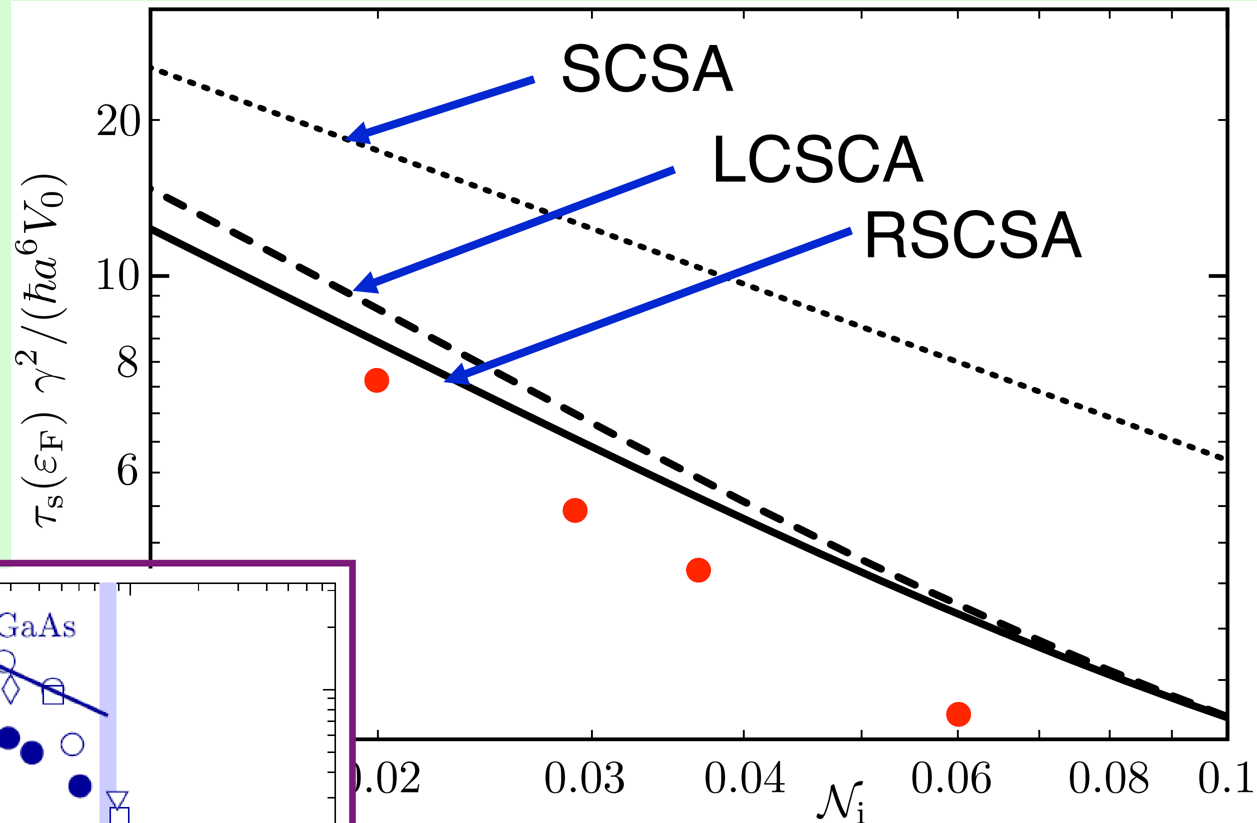
DOS, spin relaxation rate, diffusion coefficient

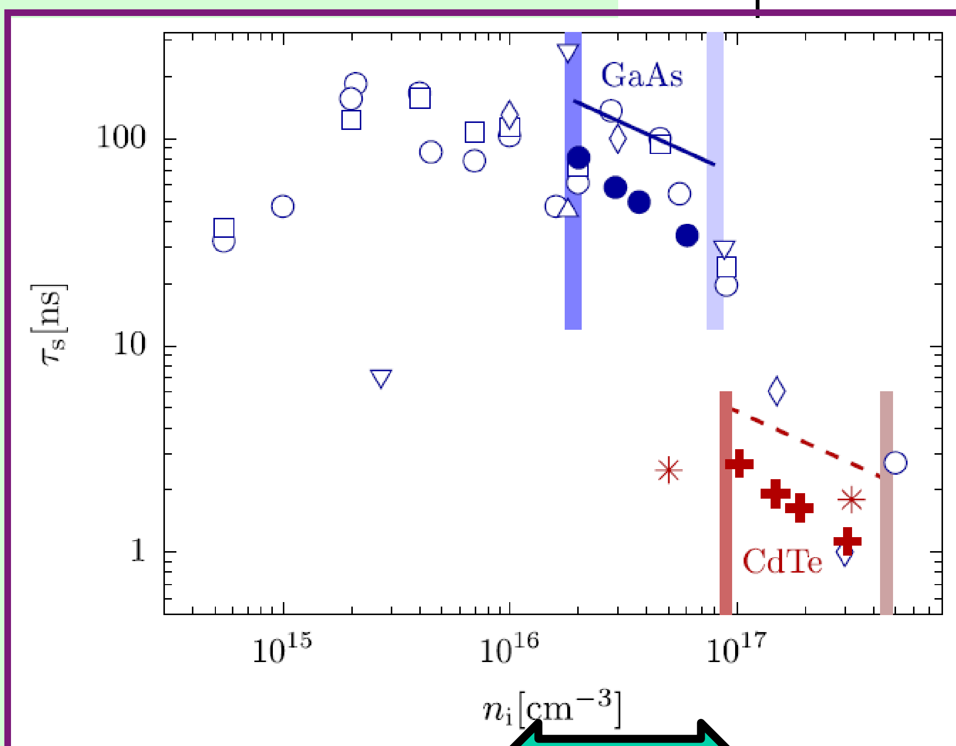


Spin relaxation time: self-consistent theory vs. numerics

● numerics

SCSA $\tau_s \propto \frac{1}{\sqrt{n_i}}$





T. Wellens & R.A.J.,
PRB 94, 144209 (2016)

Conclusions

• Spin-orbit interaction (**Dresselhaus**) in the impurity band of **n-doped zinc-blend** semiconductors.

• Numerics, phenomenological and self-consistent theories.

• Resulting spin relaxation times in **good agreement** with existing **experiments** in **GaAs** and **CdTe**.

• **Self-consistent theory** for the spin and spatial diffusion:
- average of one-particle and two-particle Green functions;
- **SSCA**: simplest self-consistent approximation scheme, reproduces the phenomenological results;
- **LCSCA** and **RSCSCA**: loop-corrected and crossed terms, good agreement with the numerical results.

G. Intronati, P. Tamborenea, D. Weinmann, R.A.J., PRL 2012
T. Wellens R.A.J., PRB 94, 144209 (2016)

Insulating regime (below n_c)

- Deeply localized regime: Hyperfine interaction

ω_p : spin precession frequency in the local field

τ_c : dwell time in the localization domain

$$< \varphi^2 > = \omega_p^2 \, \tau_c \, t$$ diffusion of the spin vector

Spin lifetime τ_s

$$\frac{1}{\tau_s} = \frac{2}{3}\langle\omega_p^2\rangle\tau_c$$

motional narrowing

- Localized regime: Anisotropic exchange

$$\frac{1}{\tau_s} = \frac{2}{3}\langle\gamma^2\rangle\frac{1}{\tau_c}$$

γ : spin rotation angle of total spin of two electrons during exchange process

Normal diffusion:

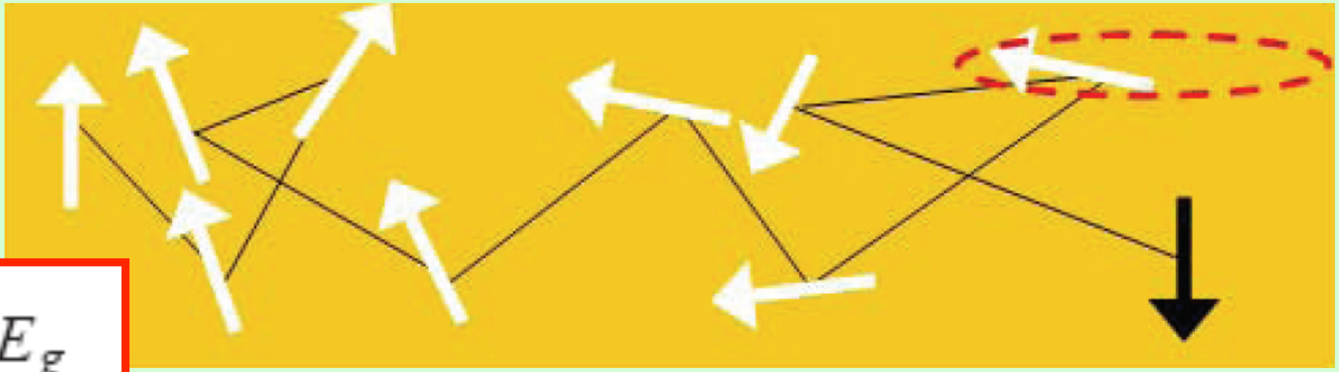
$$< R^2 > = \frac{l^2 \, t}{\tau_c} = v_F^2 \, \tau_c \quad t$$

Metallic regime (above n_c)

- D'yakonov-Perel mechanism:

spin-orbit interaction & absence of inversion symmetry

➡ electrons see **k**-dependent effective B field



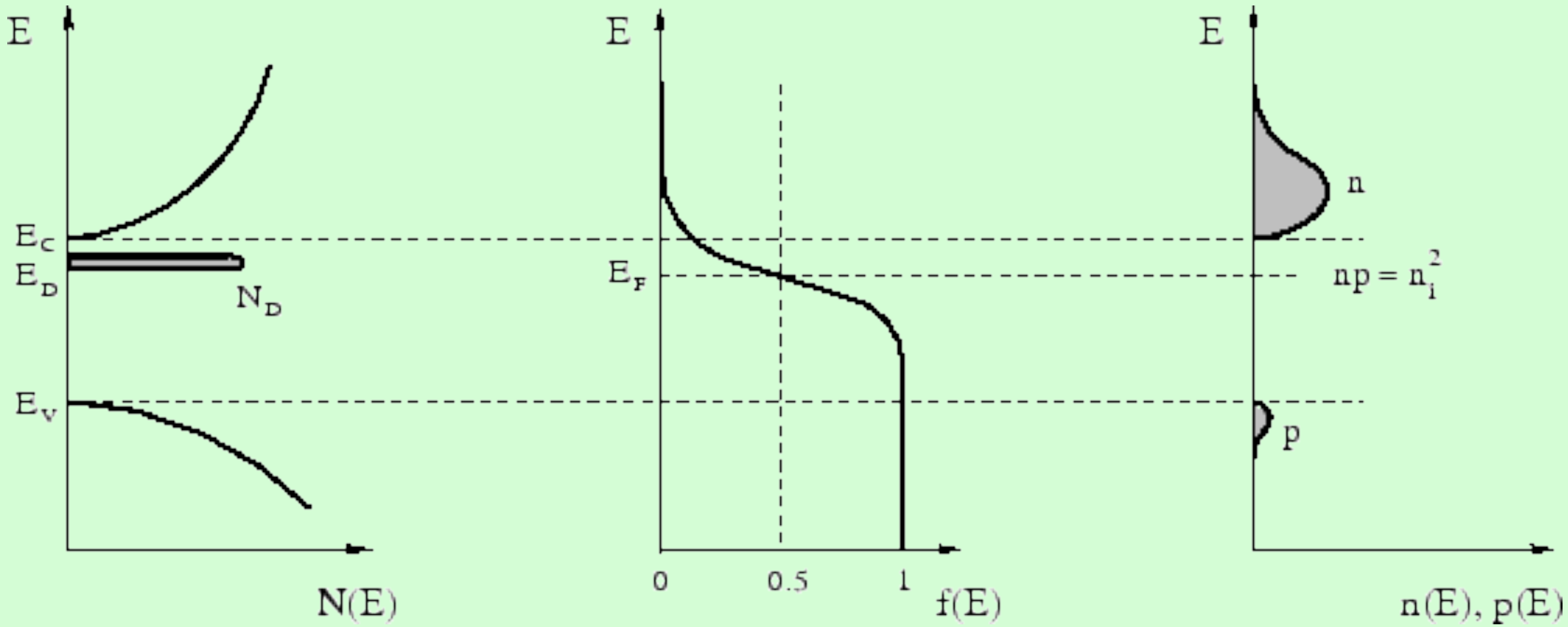
$$\tau_S = \frac{315}{16}\alpha^{-2}\frac{\hbar^2 E_g}{E^3 \tau_p(E)}$$

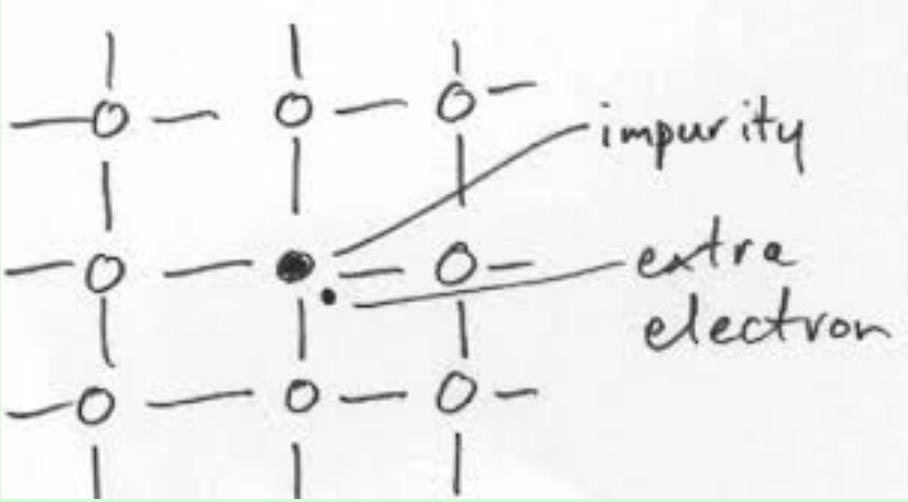
motional narrowing

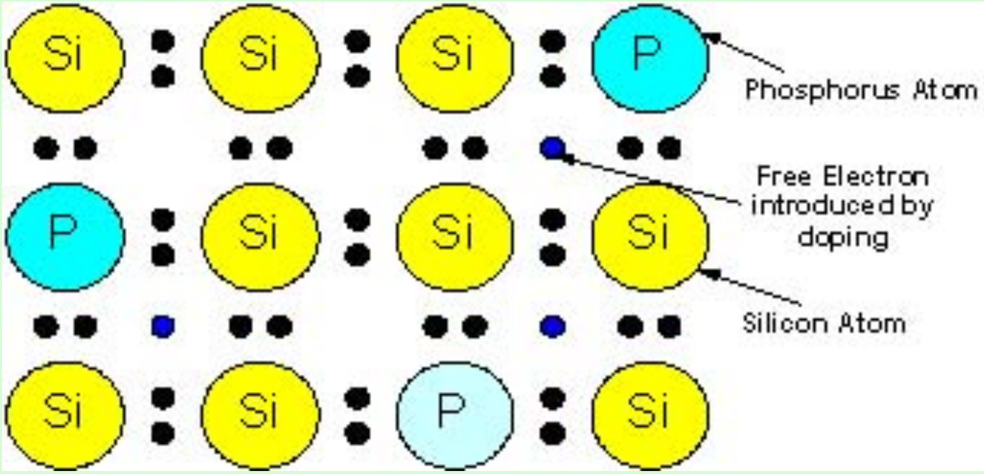
$\tau_p(E_F)$: momentum relaxation time **(impurity scattering)**

OK for electrons in the conduction band but
not for electrons in impurity band

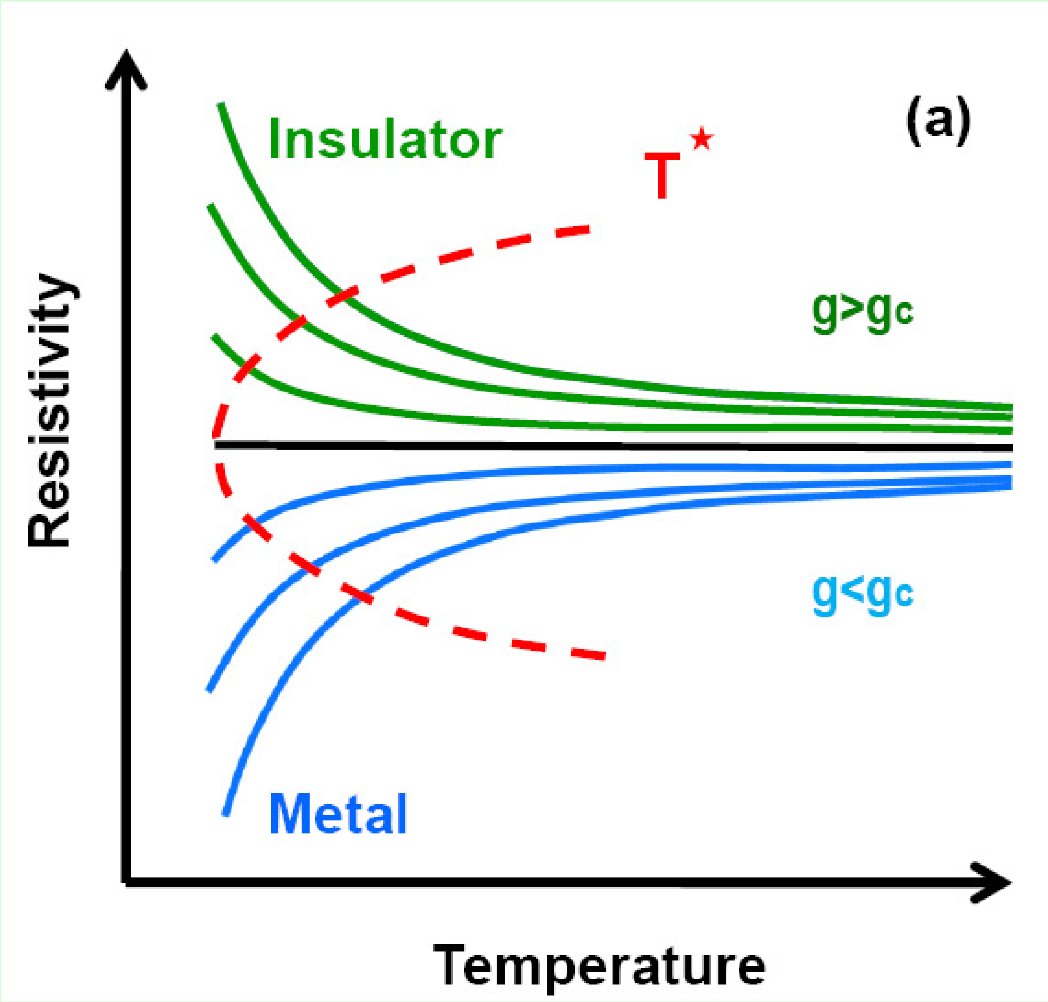
Impurity states and impurity band

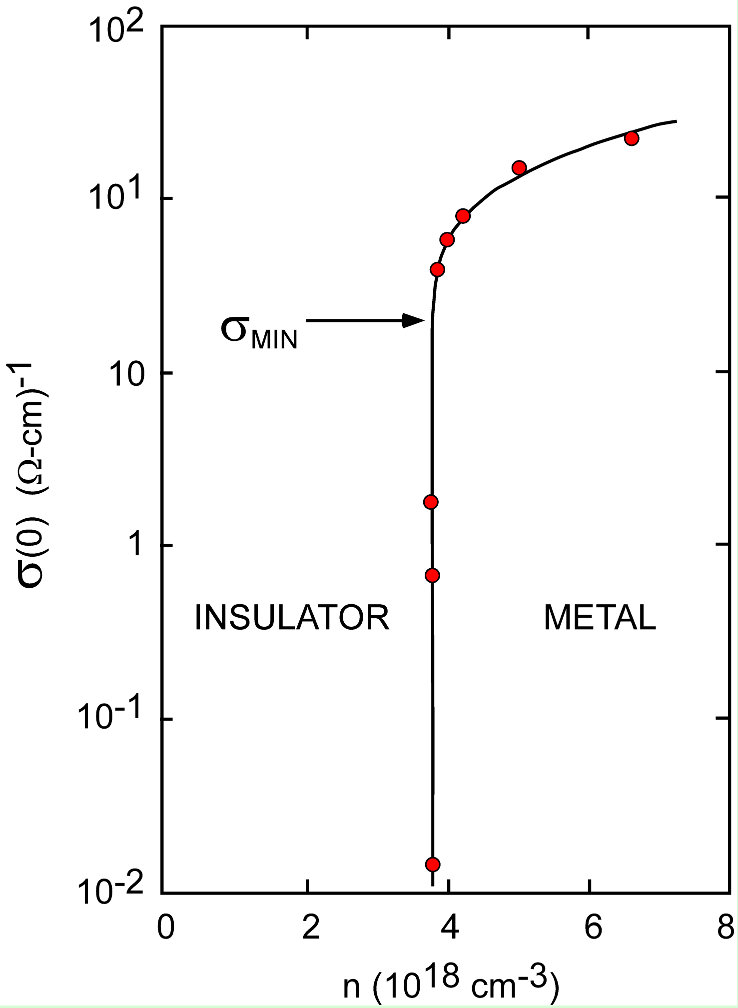






MIT in doped semiconductors





Impurity band conduction for GaAs: $n_c < n < n_h$

$n_c \approx 2\times10^{16}\ \mathrm{cm}^{-3}$ $n_h \approx 8\times10^{16}\ \mathrm{cm}^{-3}$

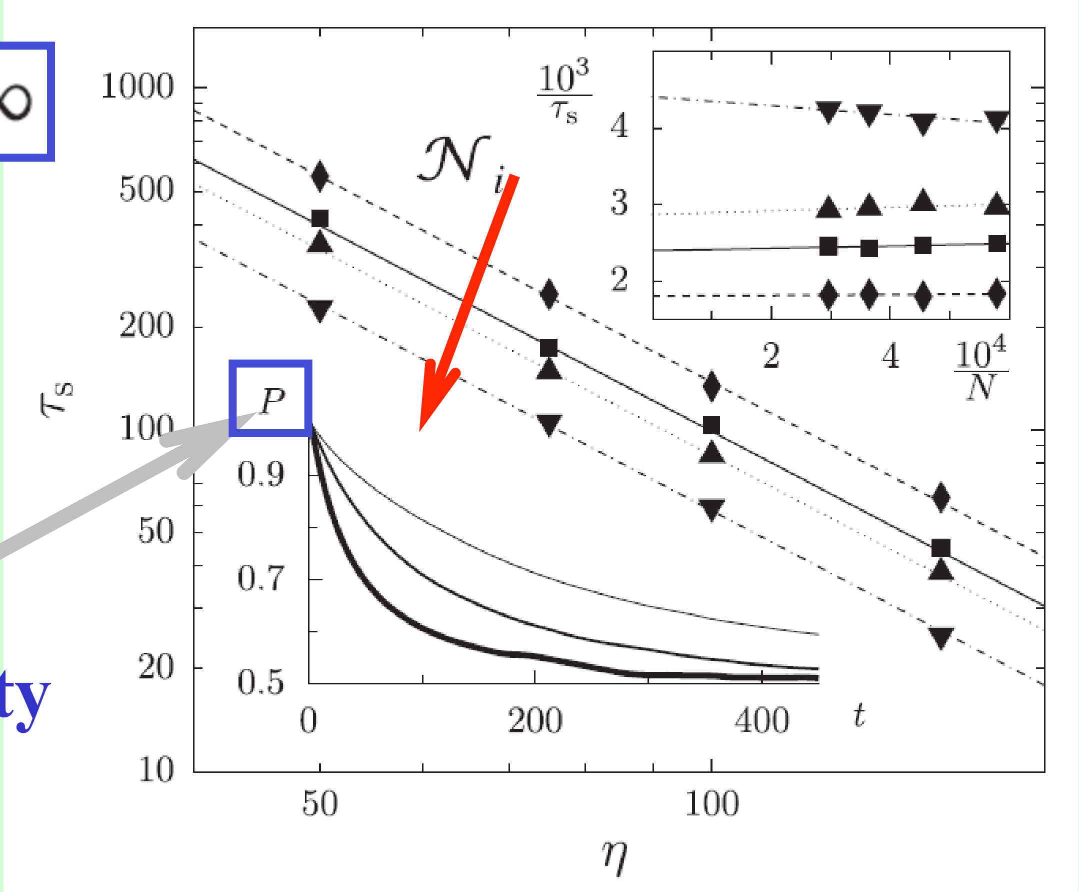
Numerical simulations

two difficulties:

- finite size effects
- small value of the SO coupling

our approach:

- extrapolate to $N \longrightarrow \infty$
- work with an enhanced coupling $\eta\gamma$



spin survival probability