Advances in the theory and experiments of spin spectroscopy

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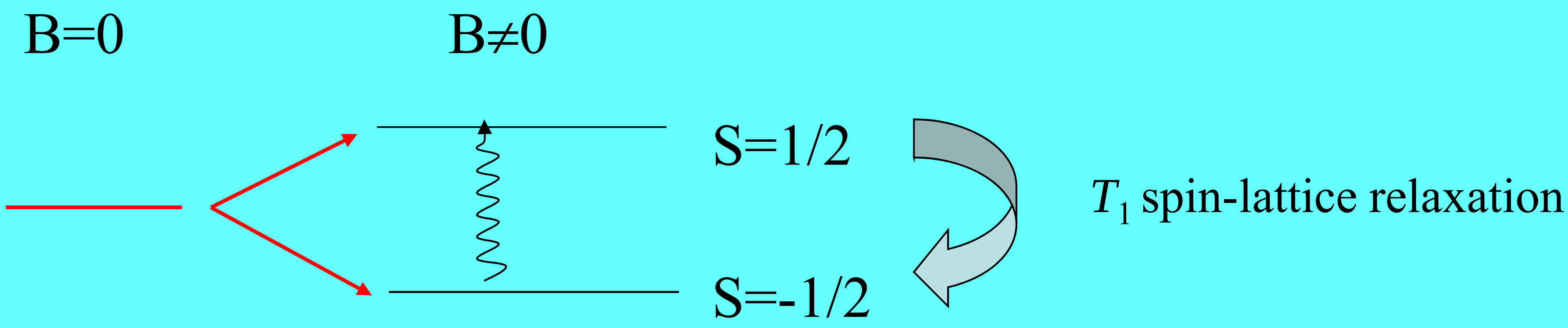
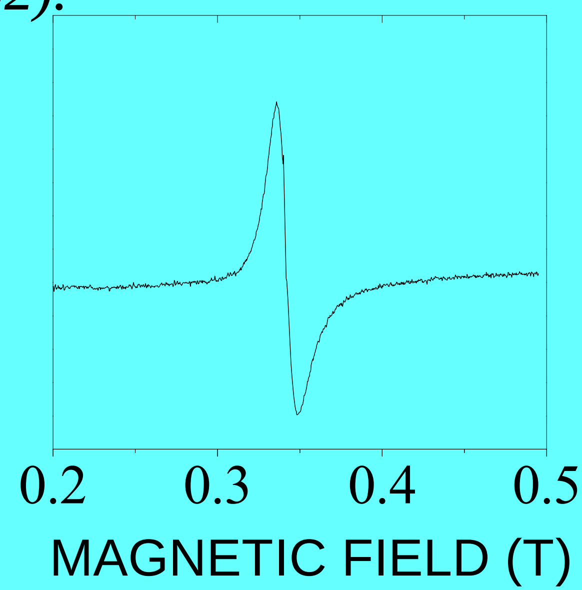
BME PHYSICS

Spin spectroscopy-Electron spin resonance



$$H_Z = g\mu_B BS \rightarrow h\nu = g\mu_B B$$

First CESR on Na: *Griswold, Kip, Kittel, PR 88, 951 (1952).*



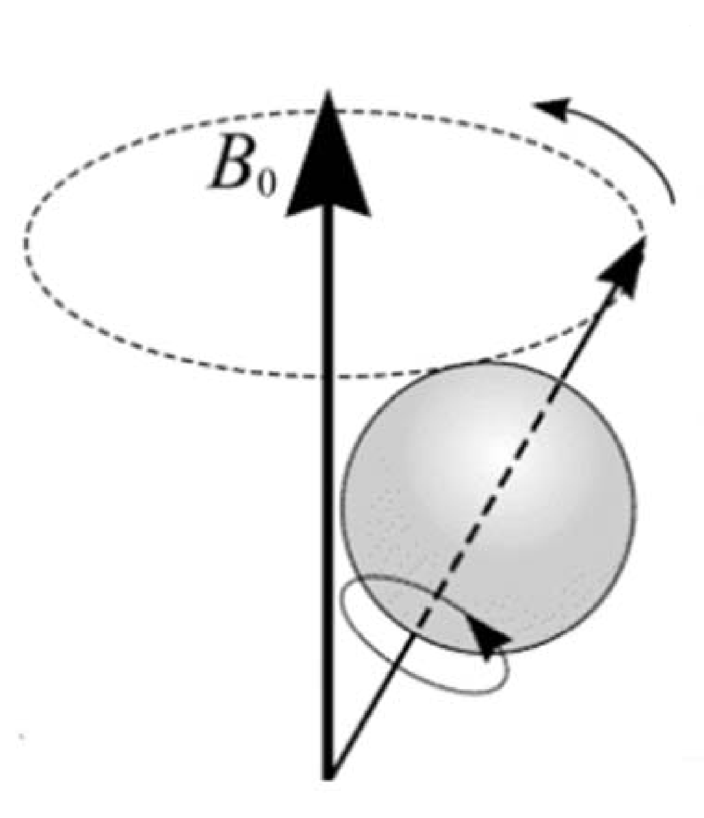
<u>Measurables:</u>

Intensity $\chi_{\mathrm{Pauli}} = \frac{1}{4}\mu_0 g^2 \mu_{\mathrm{B}}^2 D(\epsilon_{\mathrm{F}})$

Width $\Delta B = 1/\gamma T_1$

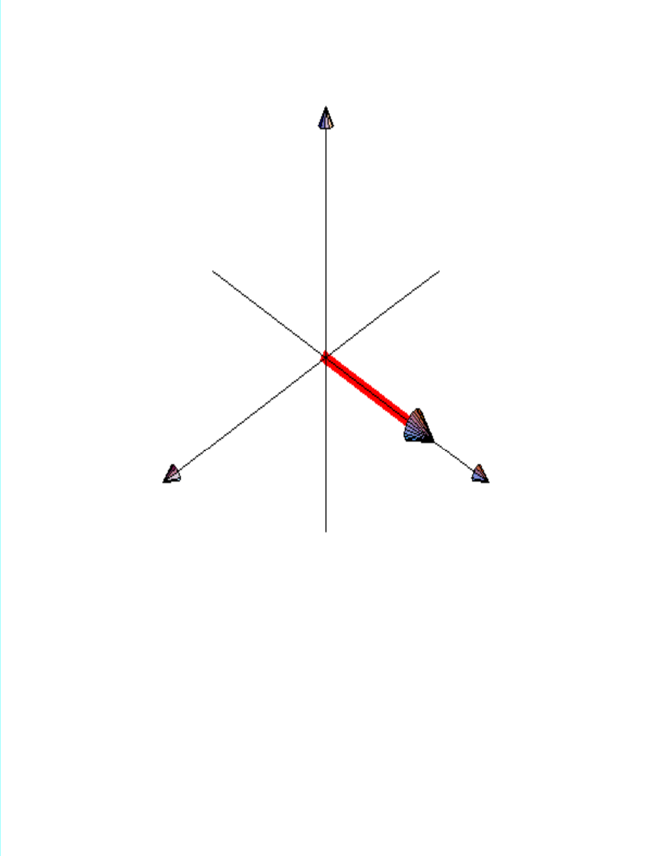
Resonance field/frequency $\Delta g = g - g_0,\ g_0 = 2.0023$

Phenomenology, Bloch Equations



Larmor-precession
$\omega_L = \gamma B_0$

B_1: small in-plane field



$$\frac{dM_{x,y}}{dt} = \gamma[\boldsymbol{\mu} \times \mathbf{B}_0]_{x,y} - \frac{M_{x,y}}{T_2}$$

$$\frac{dM_z}{dt} = \gamma[\boldsymbol{\mu} \times \mathbf{B}_0]_z + \frac{M_0 - M_z}{T_1}$$

T_2: Spin-spin Rel.time

T_1: Spin-lattice Rel.time

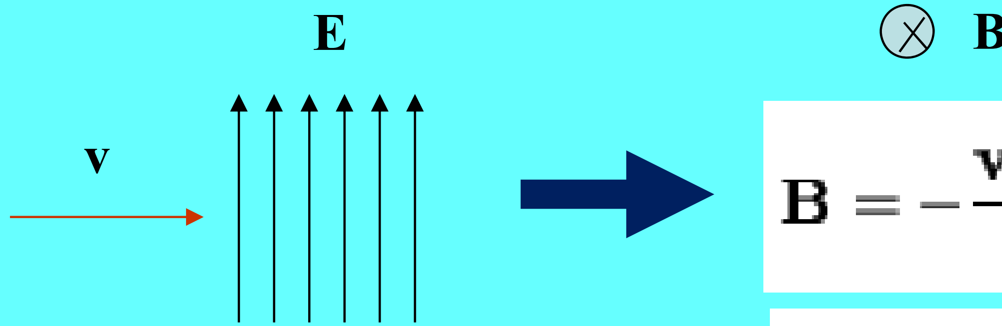
What is relaxation time?

	Bloch notation	Historic name	Modern name	Physical origin
T_1	Longitudinal	spin-lattice	Spin-relaxation	Energy transfer/ Fluctuating SOC
T_2	Transversal	spin-spin	Decoherence Irreversible decoherence	Like-spin change/ Motional narrowing+ Scattering/ Fluctuating SOC
T_2^*	non-Gaussian	Inhomogeneous broadening	Dephasing Reversible decoherence	Inhomogeneities, defects

Hierarchy: $T_2^* < T_2 \leq T_1$ in isotropic systems: $T_1 = T_2 = \tau_s$

Spin-orbit coupling

Electrodynamics:



$$\mathbf{B} = -\frac{\mathbf{v} \times \mathbf{E}}{c^2}$$

Interacts with electrons' magnetic moment

$$\hat{\mu} = -\frac{e}{2m} g \hat{\mathbf{S}}, \quad g = 2$$

$$-\mathbf{v} \times \mathbf{E} = \mathbf{v} \times \nabla \frac{Ze}{4\pi\varepsilon_0 r} = \mathbf{v} \times \frac{\mathbf{r}}{r} \frac{d}{dr} \frac{Ze}{4\pi\varepsilon_0 r} = \frac{1}{m} \hat{\mathbf{L}} \frac{Ze}{4\pi\varepsilon_0 r^3}$$

$$\hat{H}_{SO} = -\hat{\mu}\mathbf{B} = \frac{Ze^2}{4\pi\varepsilon_0} \frac{1}{2m^2c^2} \frac{\hat{\mathbf{S}}\hat{\mathbf{L}}}{r^3}$$

Types of internal *E* field:

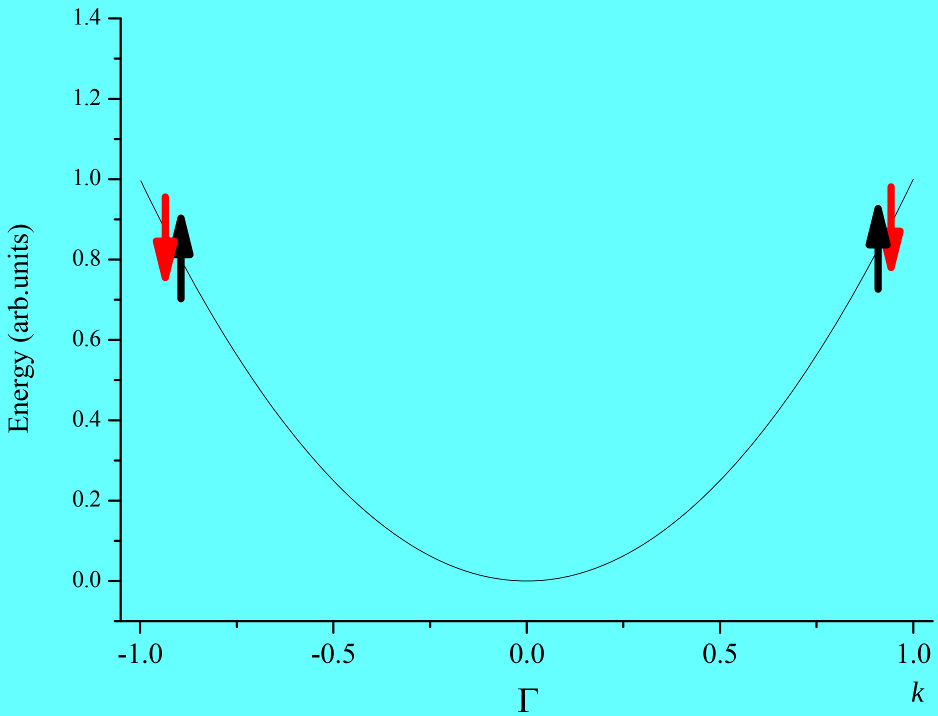
$$E_{n,l+\frac{1}{2}} - E_{n,l-\frac{1}{2}} = \frac{mc^2}{2} \frac{\alpha^4 Z^4}{n^3 l(l+1)}$$

Intrinsic (atomic)	
Dresselhaus	e.g. GaAs
Bychkov-Rashba	gating field
Proximity	heterolayers

Role of inversion symmetry

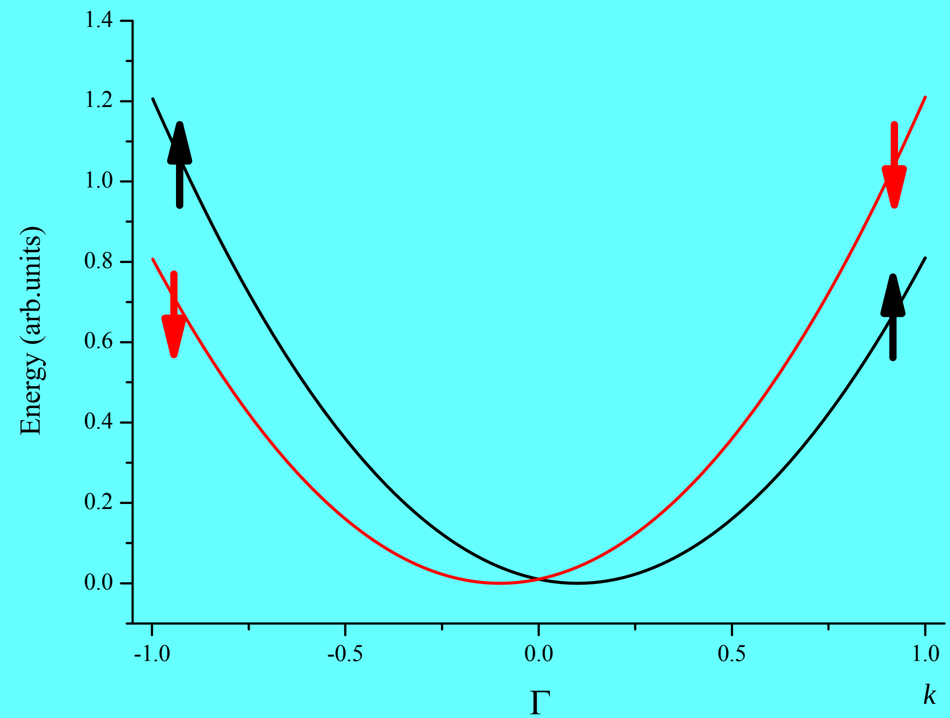
Time reversal: k,↑> and –k,↓> are always degenerate *even with SOC*

inversion



k,↑> and k,↓> are degenerate

broken (GaAs)



Effective, *k*-dep. internal magnetic field

Phenomenology of spin-relaxation

inversion

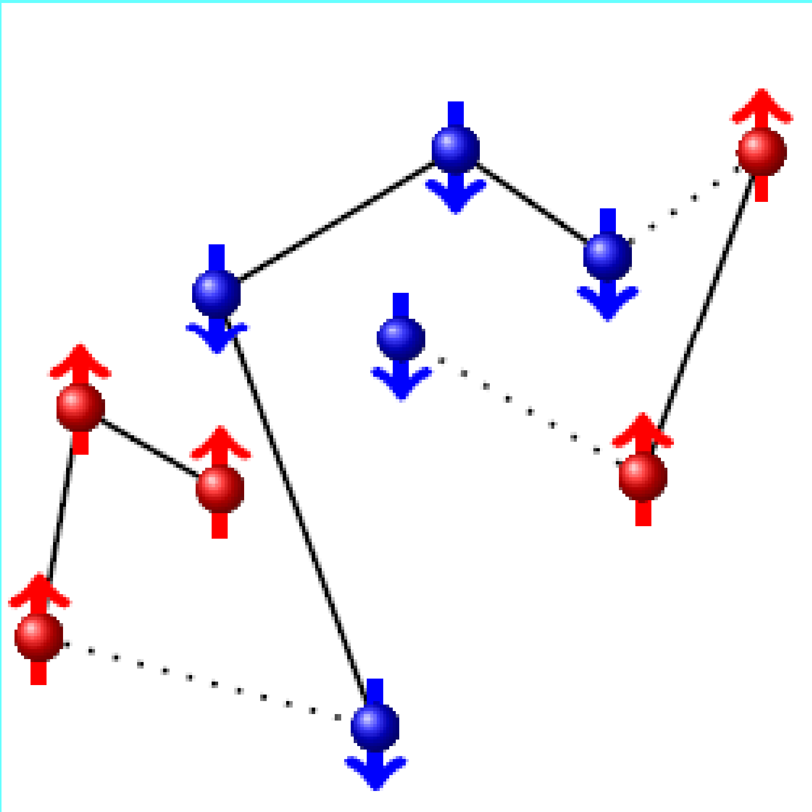
$1/\tau_s$ α $1/\tau$

Γ_s α Γ

broken

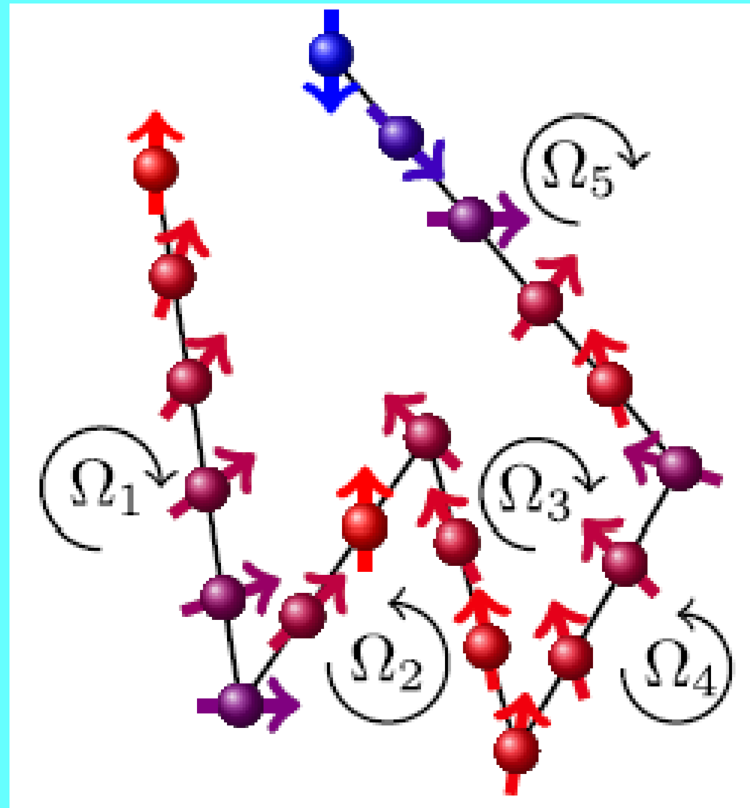
$1/\tau_s$ α τ

Γ_s α $1/\Gamma$



Elliott-Yafet

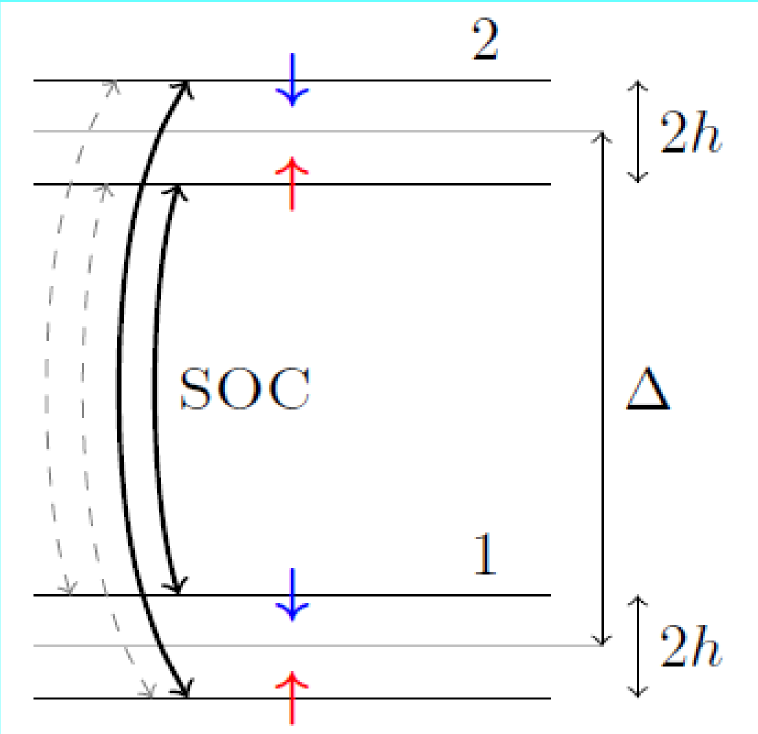
1st order pert. theory



Dy'akonov Perel'

"motional narrowing"

The Elliott-Yafet theory: Without SOC pure spin up/down states



$$\hat{\mathcal{H}} = \begin{pmatrix} h & 0 & 0 & L_{\boldsymbol{k}} \\ 0 & -h & L_{\boldsymbol{k}}^* & 0 \\ 0 & L_{\boldsymbol{k}} & \Delta + h & 0 \\ L_{\boldsymbol{k}}^* & 0 & 0 & \Delta - h \end{pmatrix}$$

$$|\widetilde{\mp}\rangle_{\mathbf{k}} = \left[a_{\mathbf{k}}\left(\mathbf{r}\right)|+\rangle + b_{\mathbf{k}}\left(\mathbf{r}\right)|-\rangle\right] e^{i\mathbf{kr}}$$

$$\frac{|b_{\mathbf{k}}|}{|a_{\mathbf{k}}|} \propto \frac{L}{\Delta E}$$

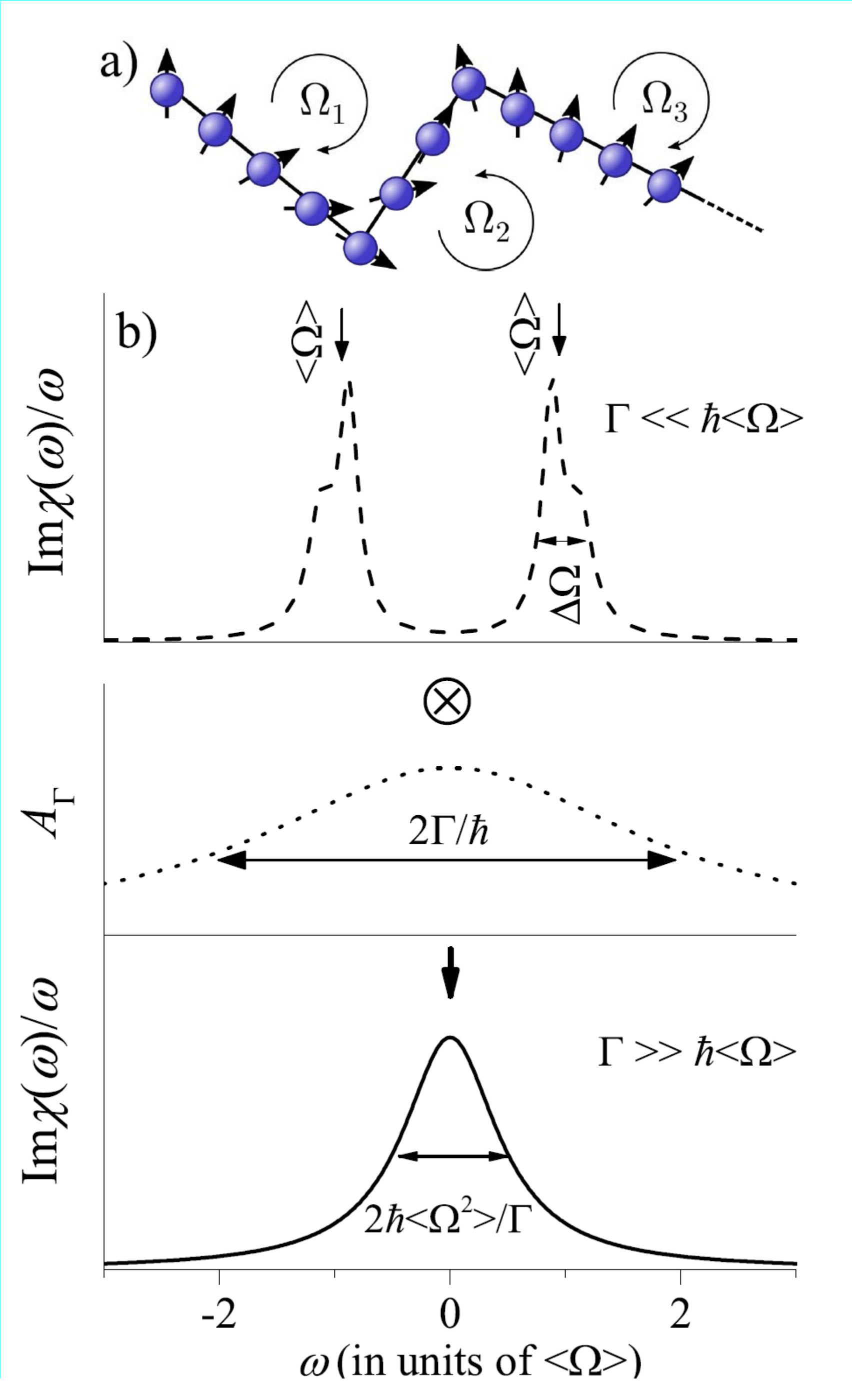
SOC mixes spin up/down states

Time dependent perturbation theory:

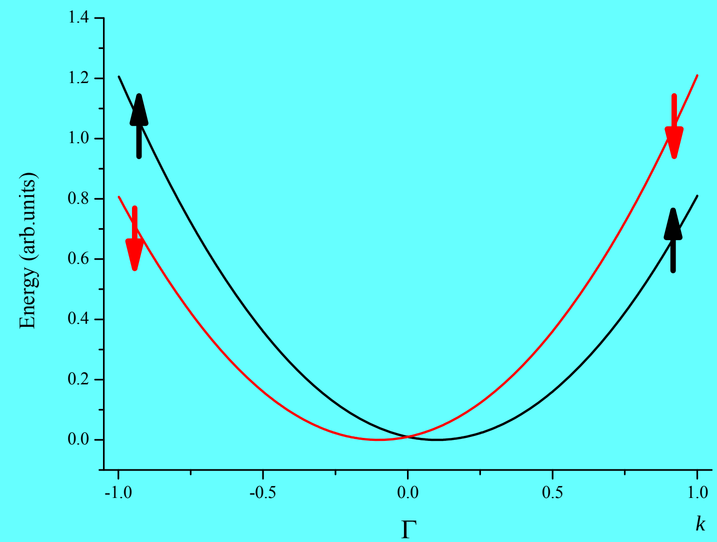
$$\Gamma_{\mathrm{s}} = \alpha \frac{L^2}{\Delta^2}\Gamma$$

Proportional resistivity and ESR width!

The Dy'akonov-Perel' theory: internal fields



SOC ($\mathcal{L}$) → ε(k)→B(k) →Ω(k)



Condition: <Ω>τ << 1

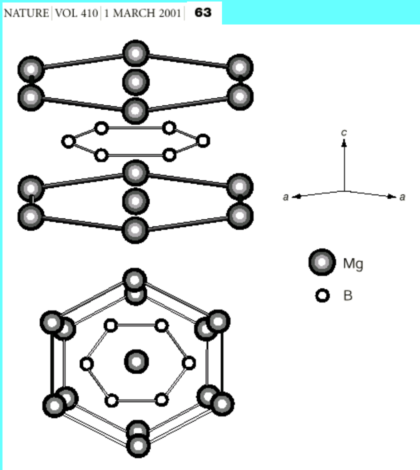
or

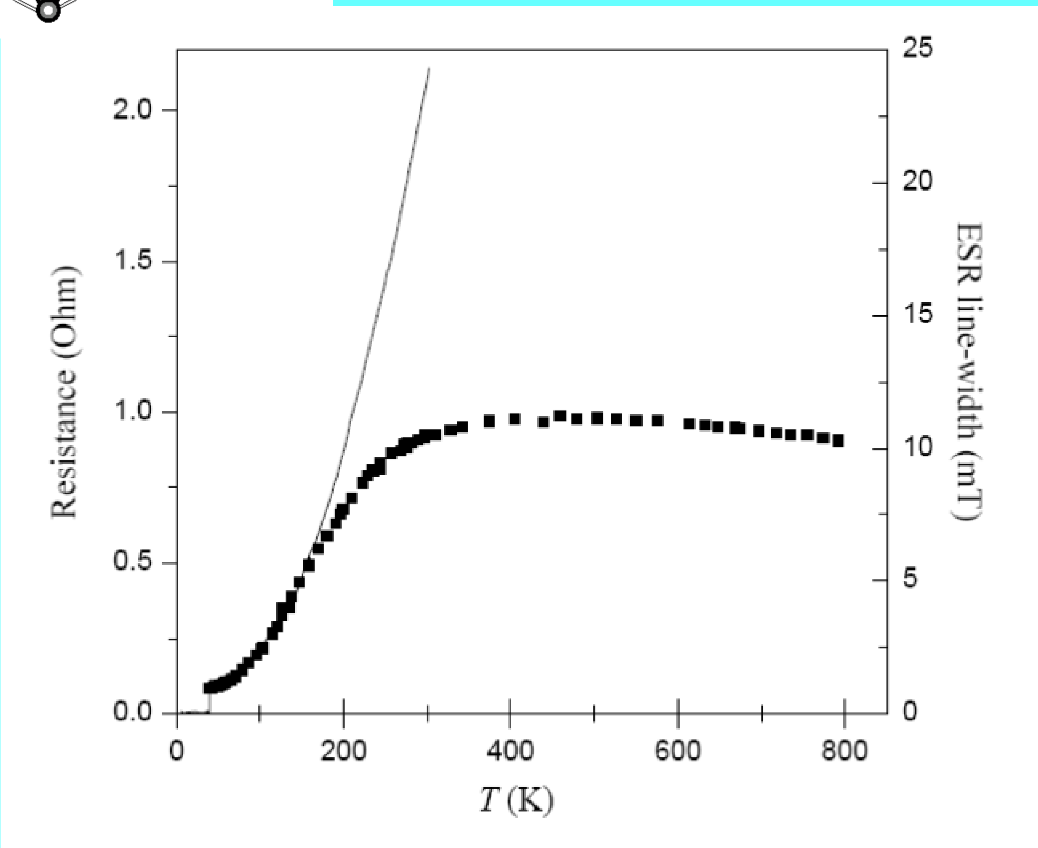
$\mathcal{L} \ll \Gamma$

Result:

$$\Gamma_s = \alpha \frac{\mathcal{L}^2}{\Gamma}$$

Anomalous spin-lattice relaxation (or line-width) in MgB_2





F. Simon et al. PRL 87, 047002 (2001).

Anomaly appears above 150 K

No magnetic field-

No thermal history dependence

No purity, no isotope,
No sample type dependence

It is a true electronic effect

Reproduced by
Rettori et al. 2001
Monod et al. 2001

The generalized Elliott-Yafet theory

FS et al. PRL ***101****, 177003 (2008).*

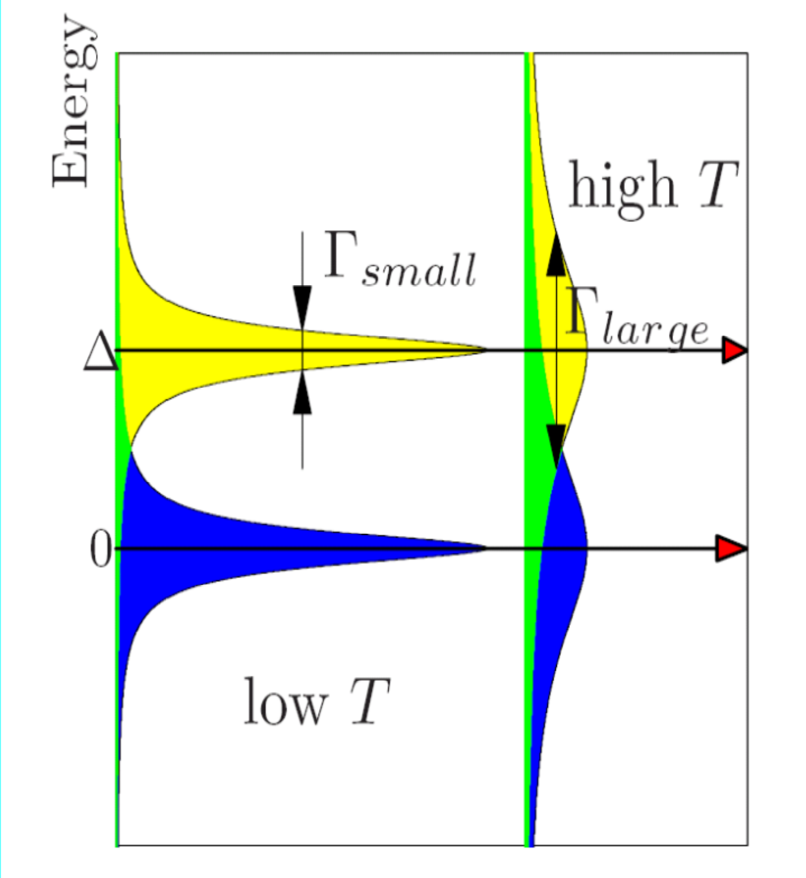
In EY: τ does not play a role, treated to lowest order

For elemental metals Δ≈10 eV $\Gamma = 2\pi k_B T \lambda \approx 6$ meV

$\lambda = 0.1$ electron-phonon coupling at $T = 100$ K

$$\Gamma_{\text{spin}} = \left\langle \frac{L^2}{\Delta^2} \Gamma \right\rangle_{\text{FS}}$$

$$\Gamma_{\text{spin}} = \left\langle \frac{L^2}{\Delta^2 + \Gamma^2} \Gamma \right\rangle_{\text{FS}}$$

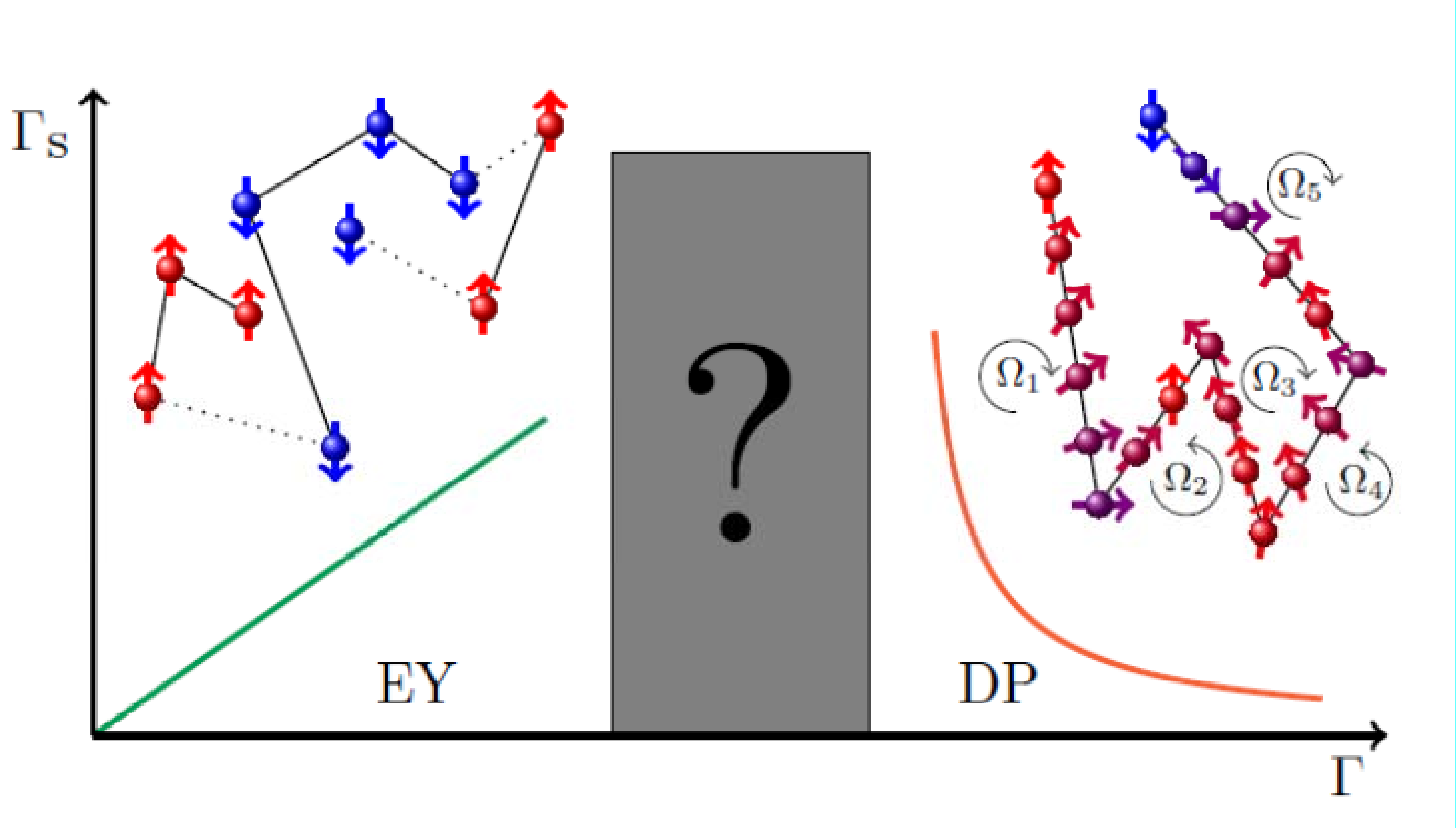


400 K-en, Γ=0.24 eV

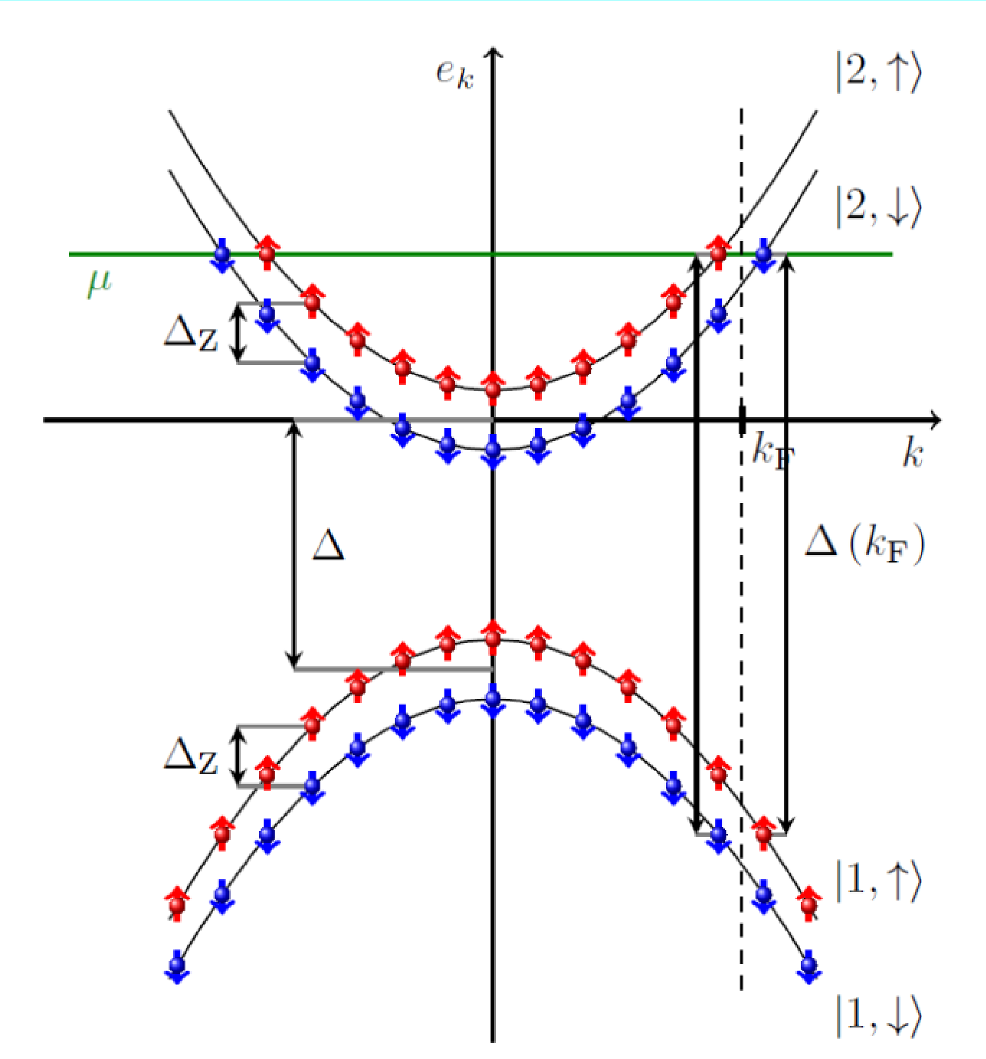
Large EPC!

Where is the small gap?

The generalized treatment of EY and DP



The model: 2 DEG



$$L_{\alpha,\alpha',s,s'}(k) = \begin{pmatrix} 0 & \mathscr{L} & 0 & L \\ \mathscr{L}^\dagger & 0 & L^\dagger & 0 \\ 0 & L & 0 & \mathscr{L} \\ L^\dagger & 0 & \mathscr{L}^\dagger & 0 \end{pmatrix}$$

L: interband matrix elements
$\mathscr{L}$: intraband

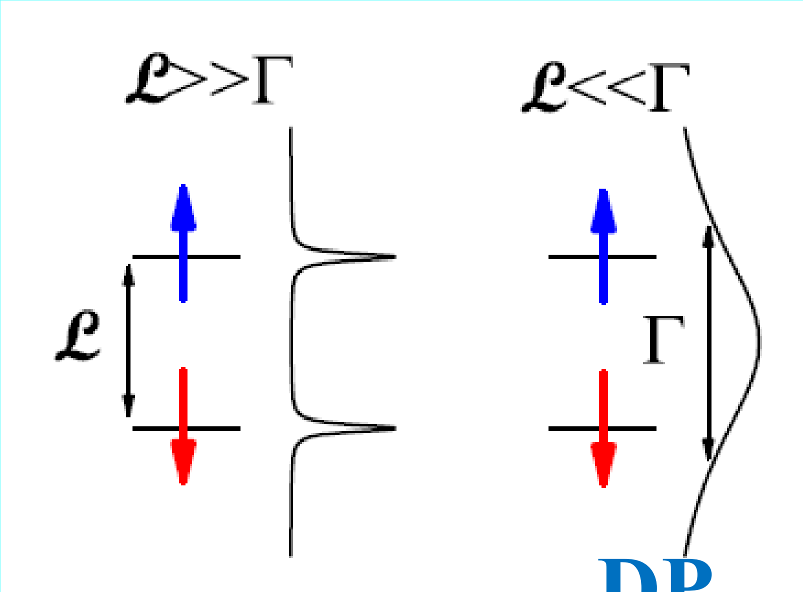
Dynamic spin susceptibility:

$$I(\omega) = \frac{B_\perp^2 \omega}{2\mu_0} \chi''_\perp (q = 0, \omega) V$$

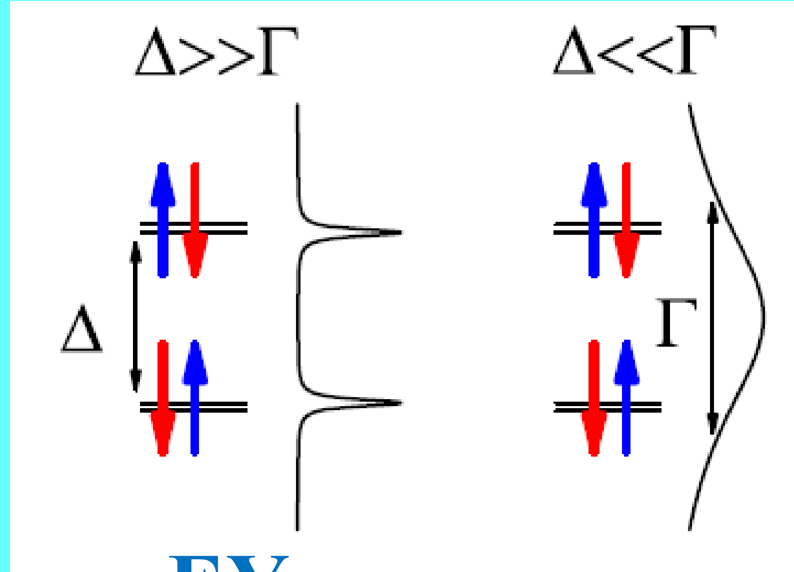
$$\Gamma_s = \frac{4\Gamma |\mathscr{L}(k_F)|^2}{4\Gamma^2 + \Delta_Z^2} + \frac{4\Gamma |L(k_F)|^2}{4\Gamma^2 + \Delta^2(k_F)}$$

Boross, Dóra, Kiss, Simon,
Sci. Rep. 3, 3233 (2013).

The intuitive unification I.



DP



EY

$$H_{\mathrm{DP}} = \begin{bmatrix} 0 & \mathcal{L}_k \\ \mathcal{L}_k^* & 0 \end{bmatrix}$$

$$H_{\mathrm{EY}} = \begin{array}{c} \\ 1\uparrow \\ 1\downarrow \\ 2\uparrow \\ 2\downarrow \end{array} \begin{array}{c} \begin{array}{cccc} 1\uparrow & 1\downarrow & 2\uparrow & 2\downarrow \end{array} \\ \begin{bmatrix} 0 & 0 & 0 & L_k \\ 0 & 0 & L_k^* & 0 \\ 0 & L_k & \Delta_k & 0 \\ L_k^* & 0 & 0 & \Delta_k \end{bmatrix} \end{array}$$

Add 2 virtual states+magnetic field

$$H_{\mathrm{DP,amended}} = \begin{bmatrix} 0 & \mathcal{L}_k & 0 & 0 \\ \mathcal{L}_k{}^* & \Delta_{Z,k} & 0 & 0 \\ 0 & 0 & \Delta_{Z,k} & \mathcal{L}_k \\ 0 & 0 & \mathcal{L}_k{}^* & 0 \end{bmatrix}$$

Transform the states

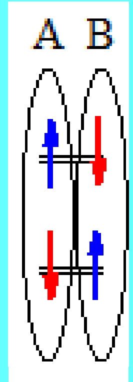
$$H_{\mathrm{EY,renamed}} = \begin{array}{c} \\ A\uparrow \\ A\downarrow \\ B\uparrow \\ B\downarrow \end{array} \begin{array}{c} \begin{array}{cccc} A\uparrow & A\downarrow & B\uparrow & B\downarrow \end{array} \\ \left[\begin{array}{cc|cc} 0 & L_k & 0 & 0 \\ L_k{}^* & \Delta_k & 0 & 0 \\ \hline 0 & 0 & \Delta_k & L_k \\ 0 & 0 & L_k{}^* & 0 \end{array}\right] \end{array}$$

The intuitive unification II.

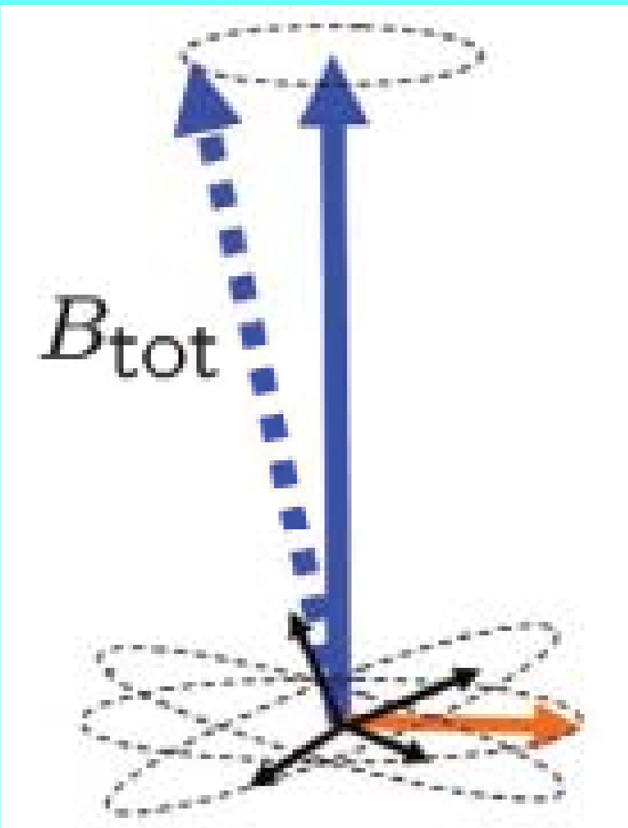
How to obtain the EY result in the DP language?

$$H_{\rm EY} = \begin{array}{c} \\ 1\uparrow \\ 1\downarrow \\ 2\uparrow \\ 2\downarrow \end{array} \begin{array}{c} \begin{array}{cccc} 1\uparrow & 1\downarrow & 2\uparrow & 2\downarrow \end{array} \\ \left[\begin{array}{cccc} 0 & 0 & 0 & L_k \\ 0 & 0 & L_k^* & 0 \\ 0 & L_k & \Delta_k & 0 \\ L_k^* & 0 & 0 & \Delta_k \end{array}\right] \end{array}$$

$$|A\uparrow\rangle = |1\uparrow\rangle$$
$$|A\downarrow\rangle = |2\downarrow\rangle$$
$$|B\uparrow\rangle = |2\uparrow\rangle$$
$$|B\downarrow\rangle = |1\downarrow\rangle$$



$$H_{\rm EY,renamed} = \begin{array}{c} \\ A\uparrow \\ A\downarrow \\ B\uparrow \\ B\downarrow \end{array} \begin{array}{c} \begin{array}{cccc} A\uparrow & A\downarrow & B\uparrow & B\downarrow \end{array} \\ \left[\begin{array}{cccc} 0 & L_k & 0 & 0 \\ L_k{}^* & \Delta_k & 0 & 0 \\ 0 & 0 & \Delta_k & L_k \\ 0 & 0 & L_k{}^* & 0 \end{array}\right] \end{array}$$



$$\frac{1}{T_1} = (\overline{\omega_x^2} + \overline{\omega_y^2}) \frac{\tau_c}{\omega_0^2 \tau_c^2 + 1} \qquad \text{(IV.36)}$$

J. Fabian *et al.* Acta Physica Slovaka 2007

$$\Gamma_s = \alpha \frac{L^2}{\Delta^2 + \Gamma^2} \Gamma$$

Same as the quantum kinetics approach!

A Monte Carlo approach to Spin-Relaxation

A Monte Carlo approach to spin-relaxation

Szolnoki, Sci. Rep. 2017

The model:

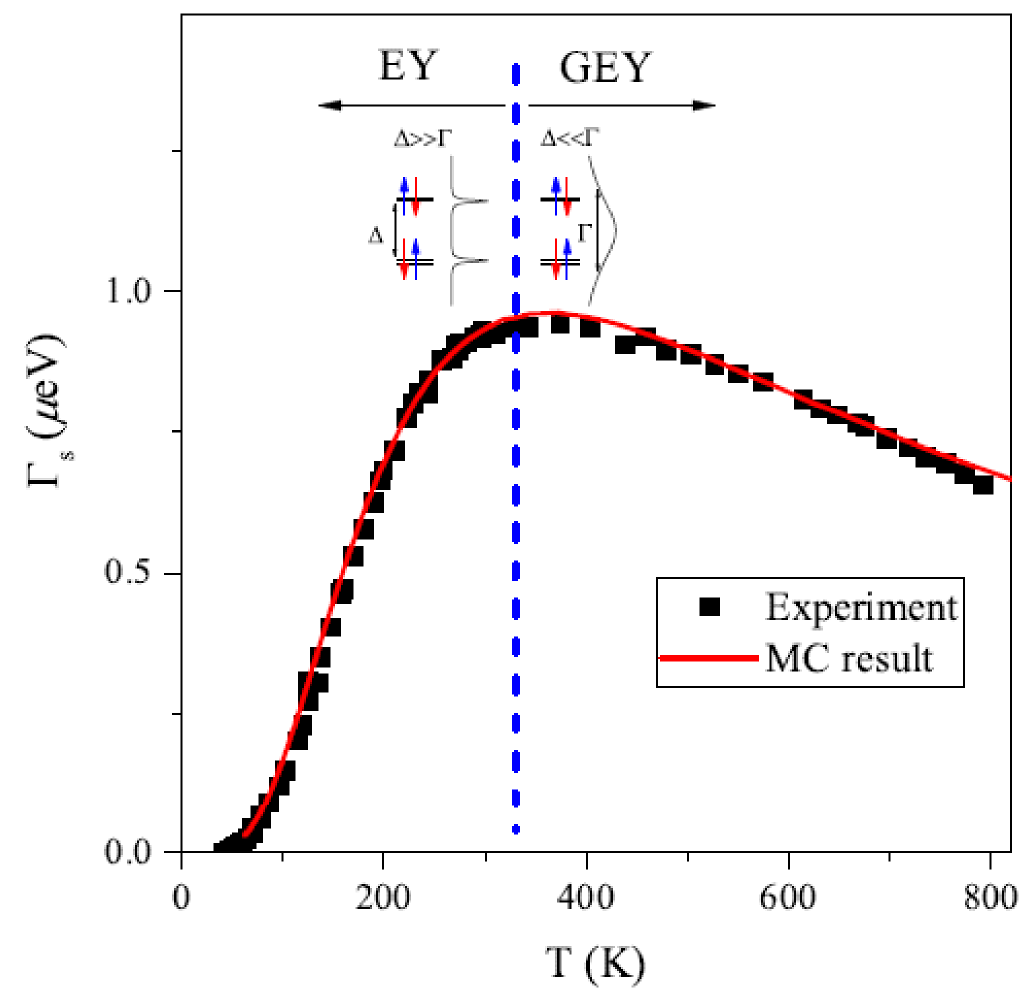
1. Polarized spin ensemble

↓

2. Evolution due to Ω(k)

↓

3. New random k



Conclusions

Forget the simple classifying phenomenology of EY and DP

Think about EY spin-flip as continuous precession

Think about DP as a half of an inversionally symmetric system

The MC is useful to tackle SR for both DP and EY for arbitrary Γ

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