Entanglement of electron and nuclear spins in solids: why and how?

Viktor Ivády & Ádám Gali

Entanglement Day 2015
September 25, 2015 @ Wigner RCP

Outline

- Brief introduction
- Color centers in solids
  - Optical polarization of the electron spin
  - Optical polarization of the nuclear spin: entangled states
- Theory for quantitative description
- Recent results
- Summary

Important feature of nuclear spins

- Nuclear gyromagnetic constants are very small
  1. Drawback: The Zeeman splitting of the nuclear spin states are very small even in huge magnetic fields
     - At non-zero temperatures, the thermal occupation of the spin states is almost identical
     - Very small “static nuclear spin polarization” (<<<1%)
     - NMR sensitivity is low
  2. Advantage: The nuclear spins are weakly interact with their environment (compared with electron spin)
     - They have long spin coherence time
     - Attractive for quantum information application

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- DNP is important in many areas
  - e.g. for sensitivity-enhanced NMR and MRI measurements

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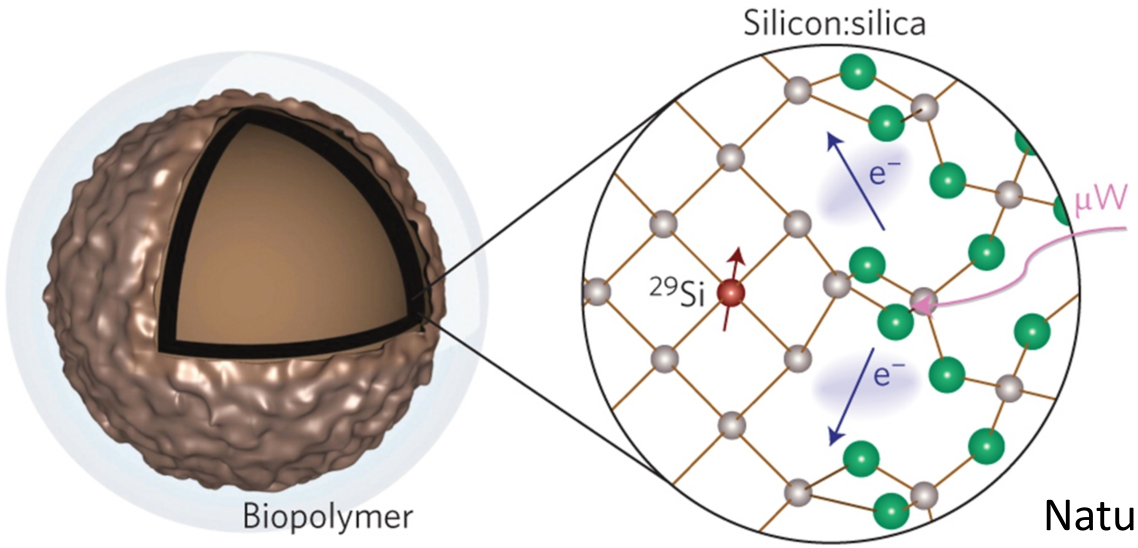

Nature Nano. 8, 363–368 (2013)

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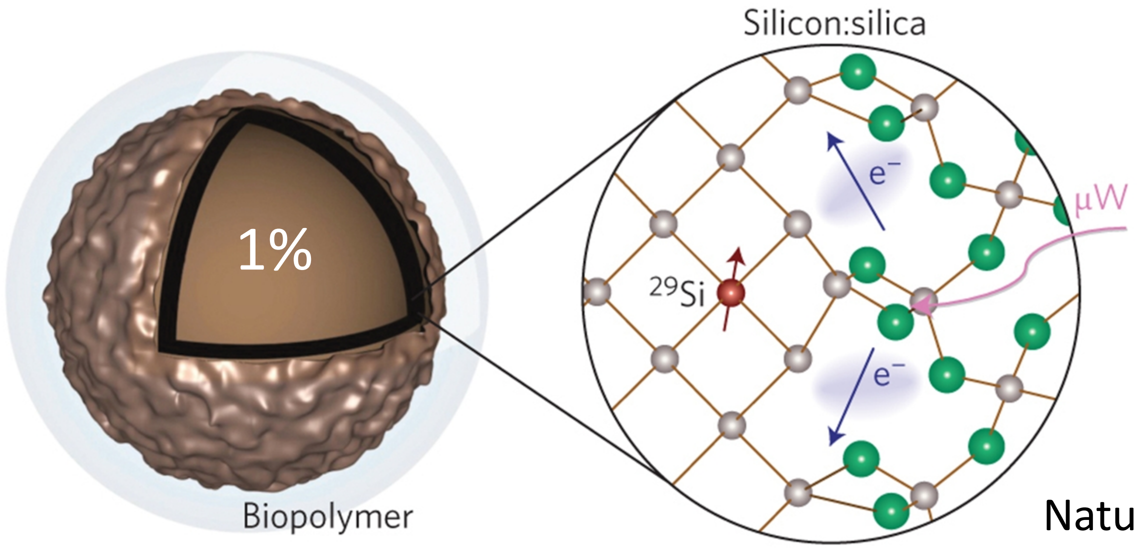

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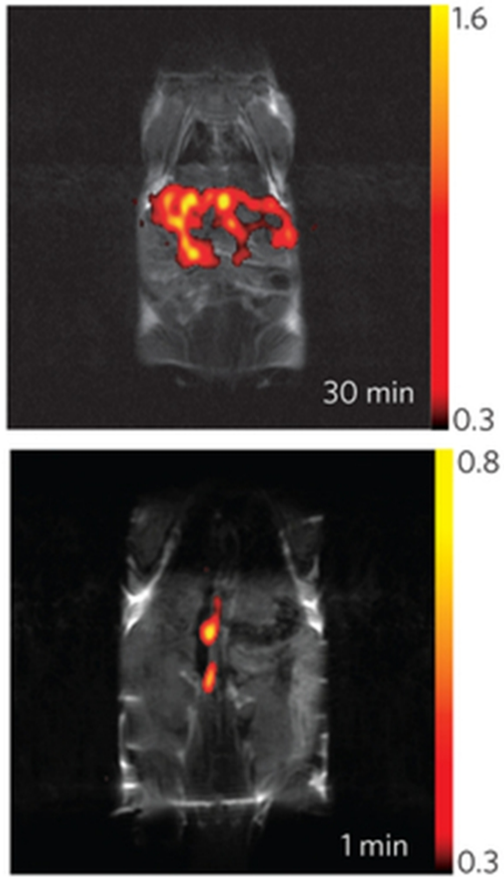
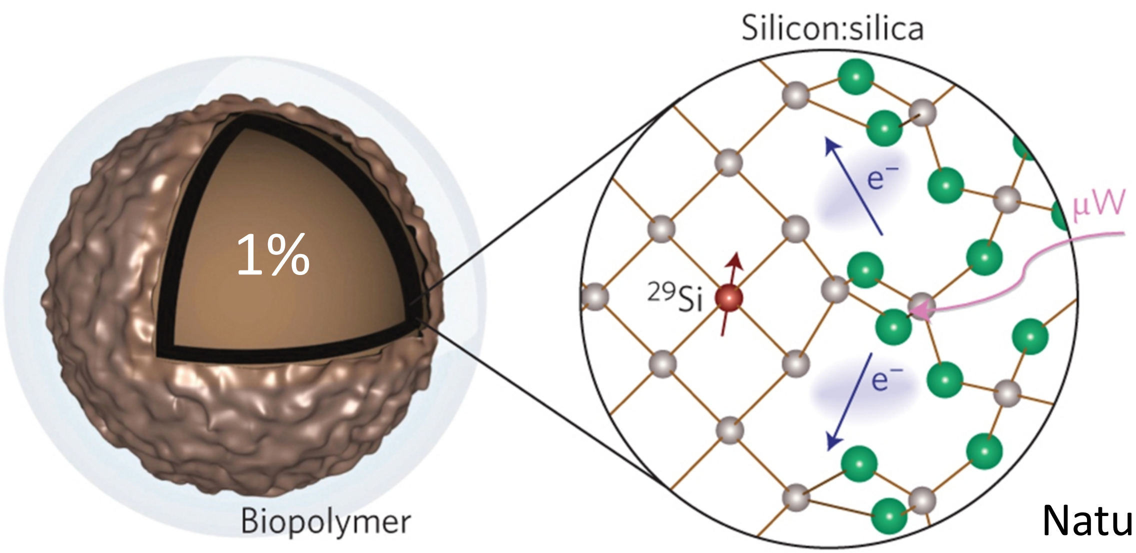
In vivo MRI





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How

- Basic phenomena: spin polarization transfer
  - e.g.: from accessible electron spins to the nuclear spins
  - For the transfer, entangled electron-nuclear spin states are used
- There are several DNP techniques
  - Via the Overhauser-effect, the solid-effect, the cross-effect, and the thermal-mixing
  - A recent new approach is to use the spins of controllable high spin state point defects

DNP with point defects

- One direction to realize DNP:

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  – room temperature DNP, etc.

  → **hyperpolarized sample holders for sensitivity-enhanced NMR**

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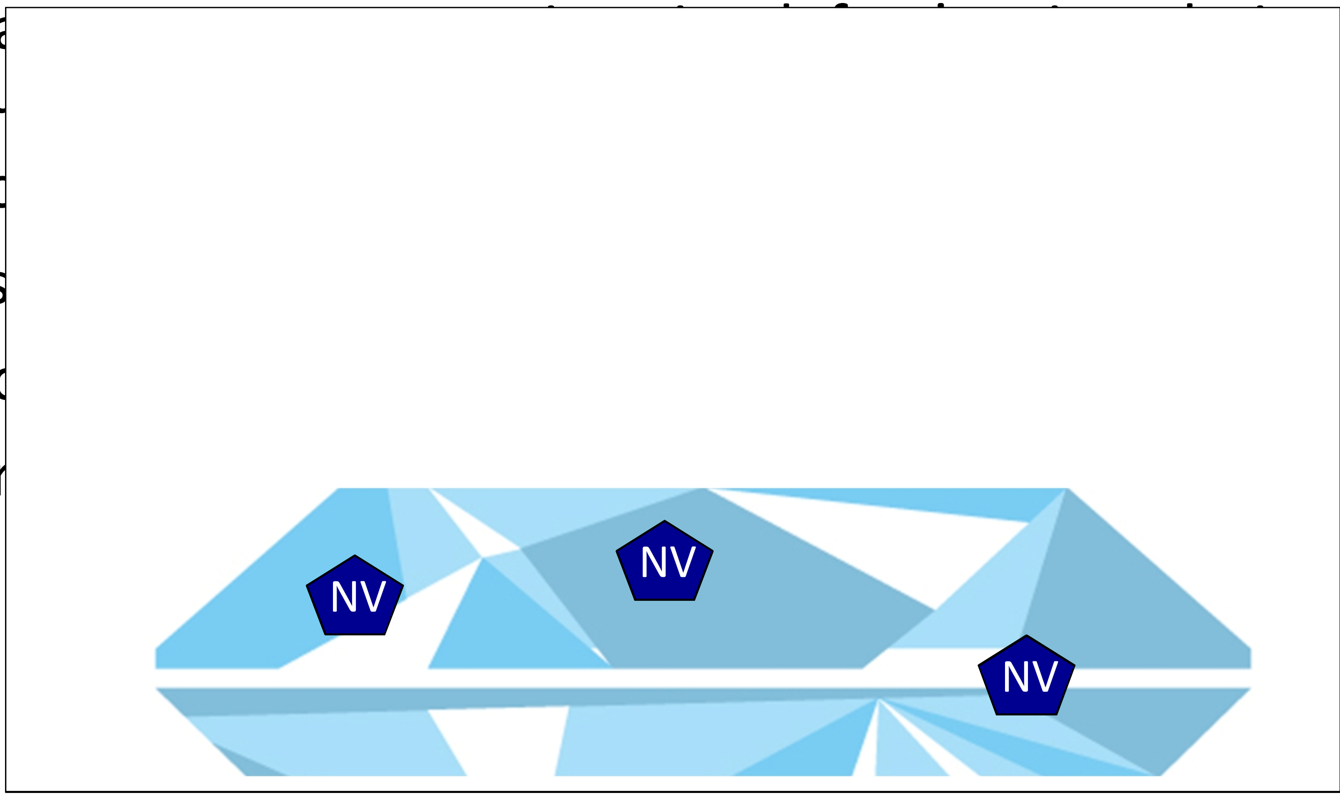
- Re…

  Us…

  – …

  – …

  → … **…d NMR**



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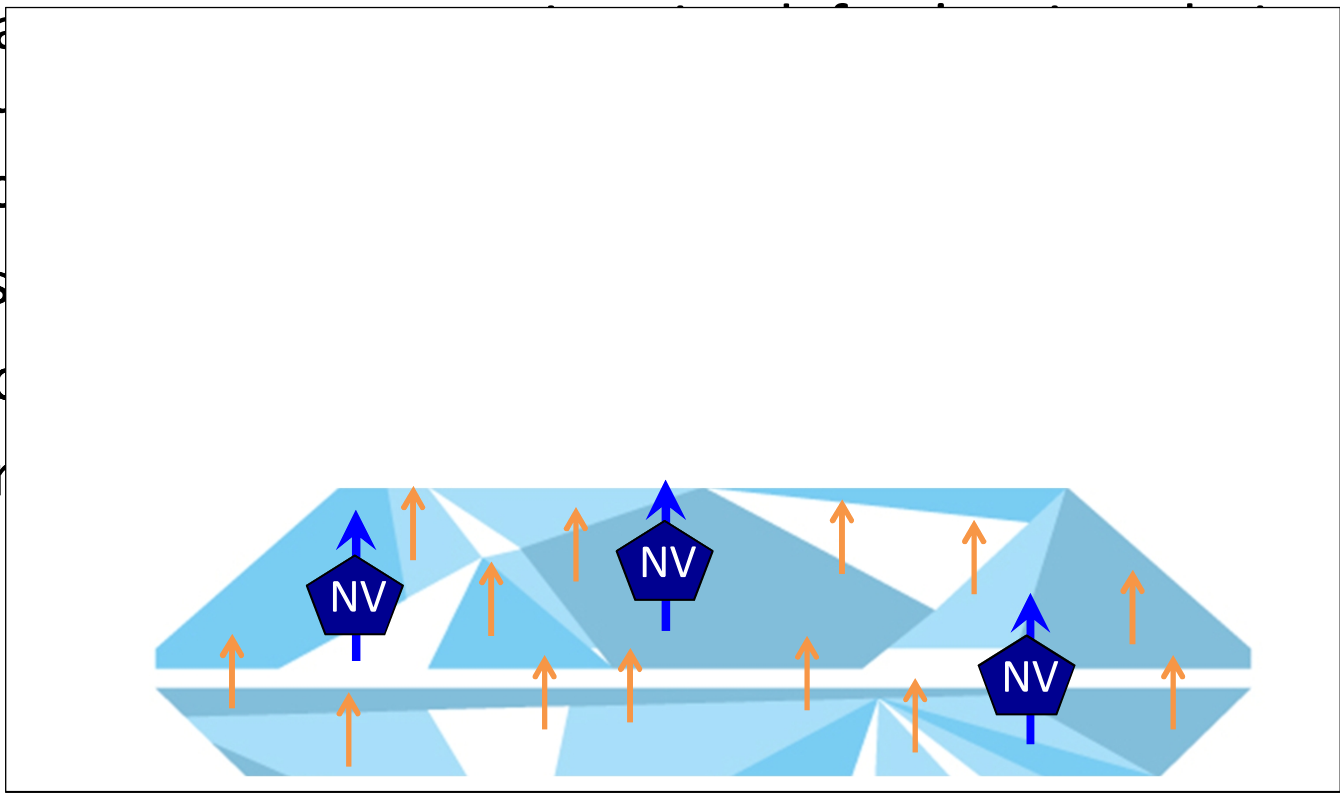

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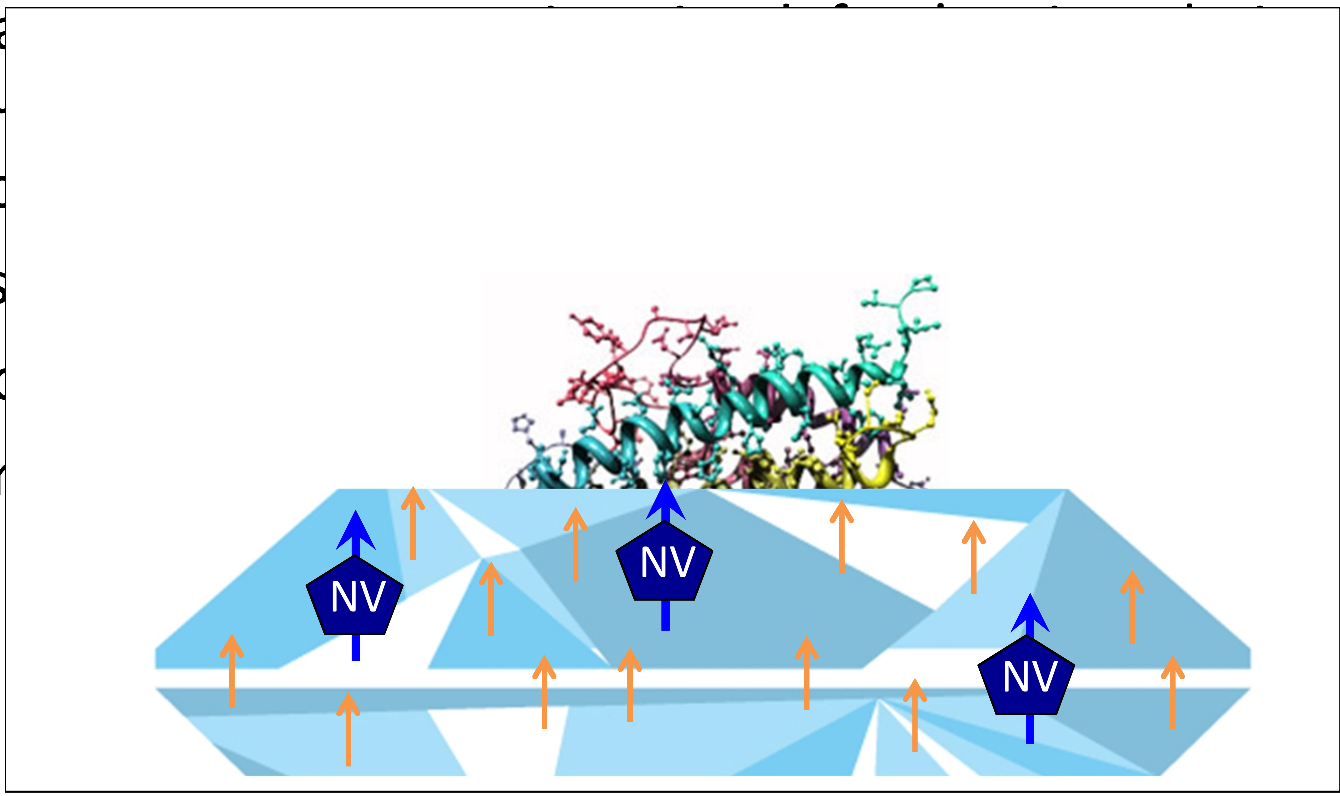

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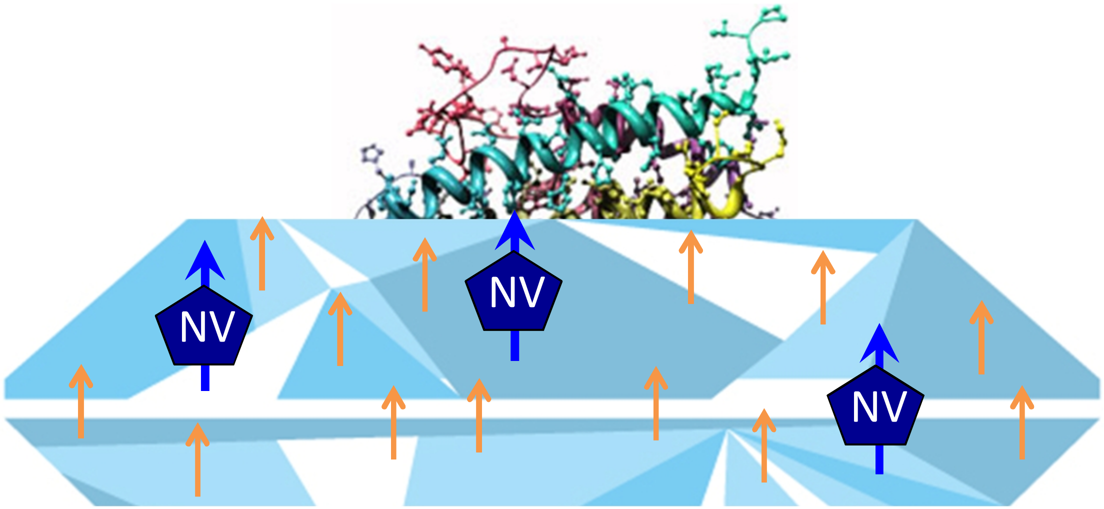

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  U

  –

  –

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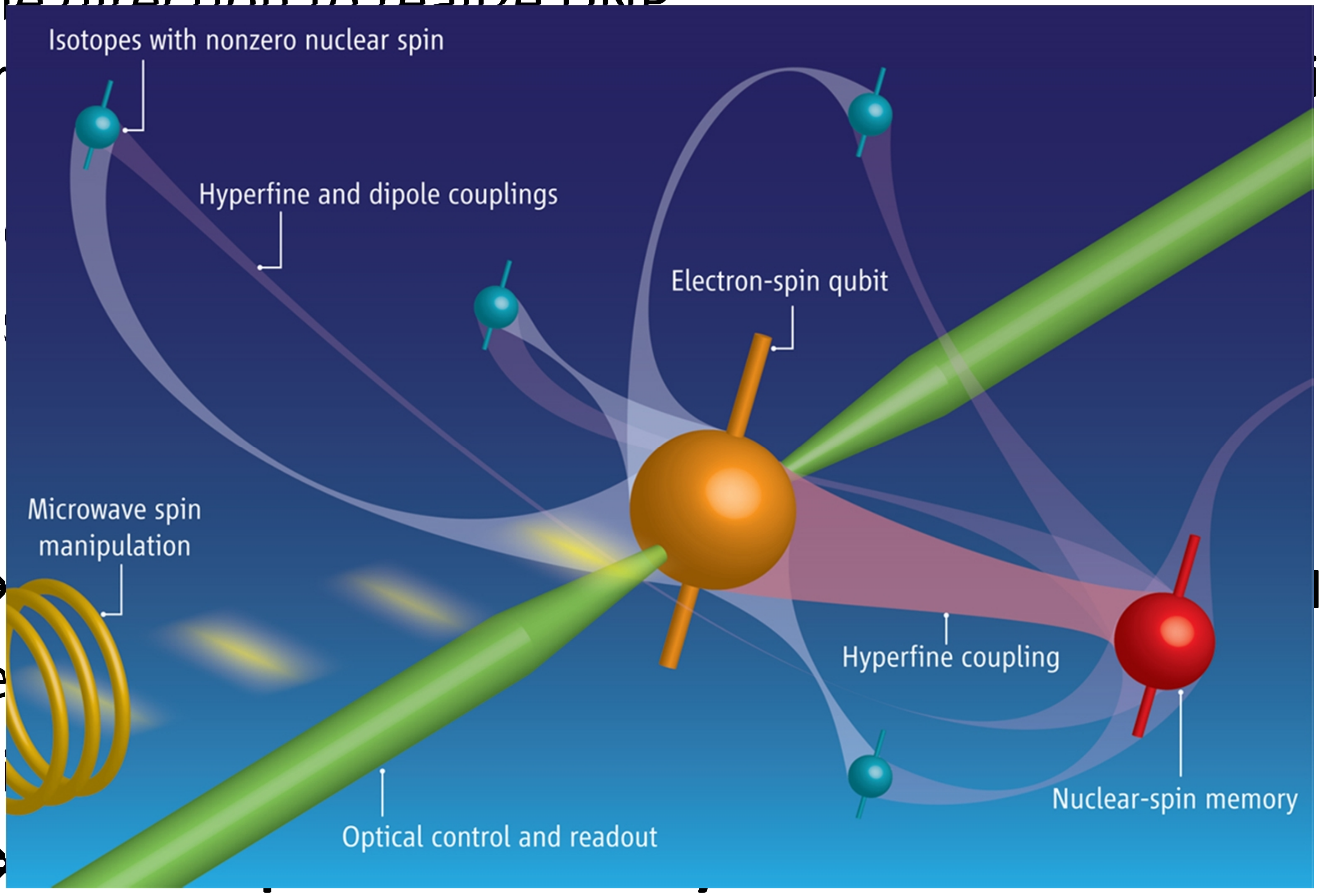
- Ne

  H

  ⇒



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Not for all configurations!

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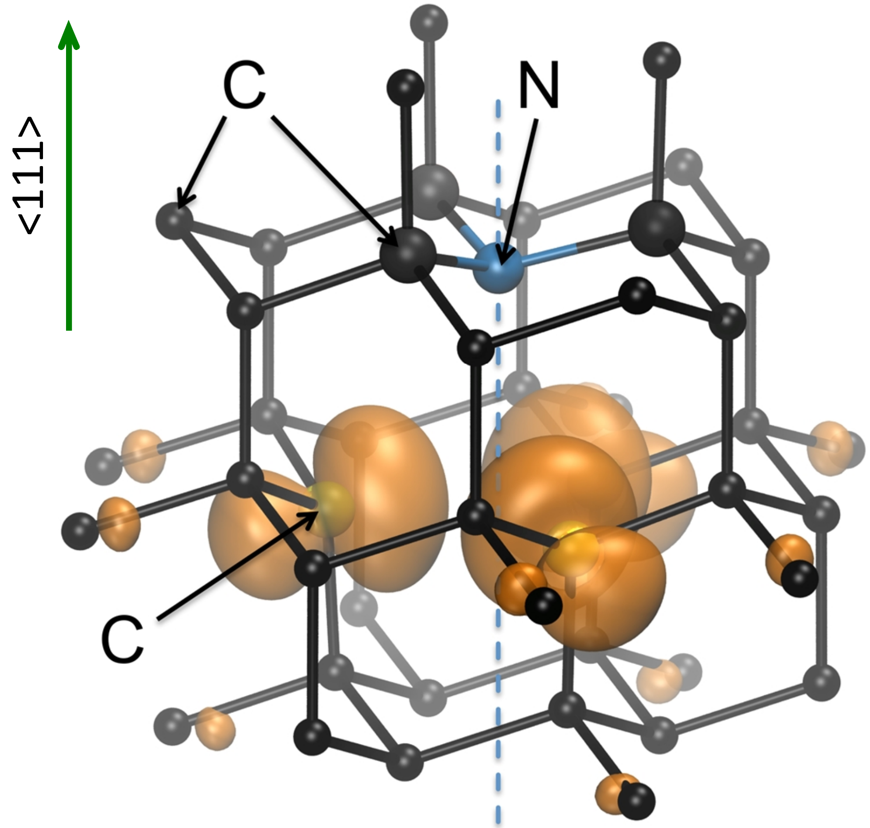
NV center in diamond



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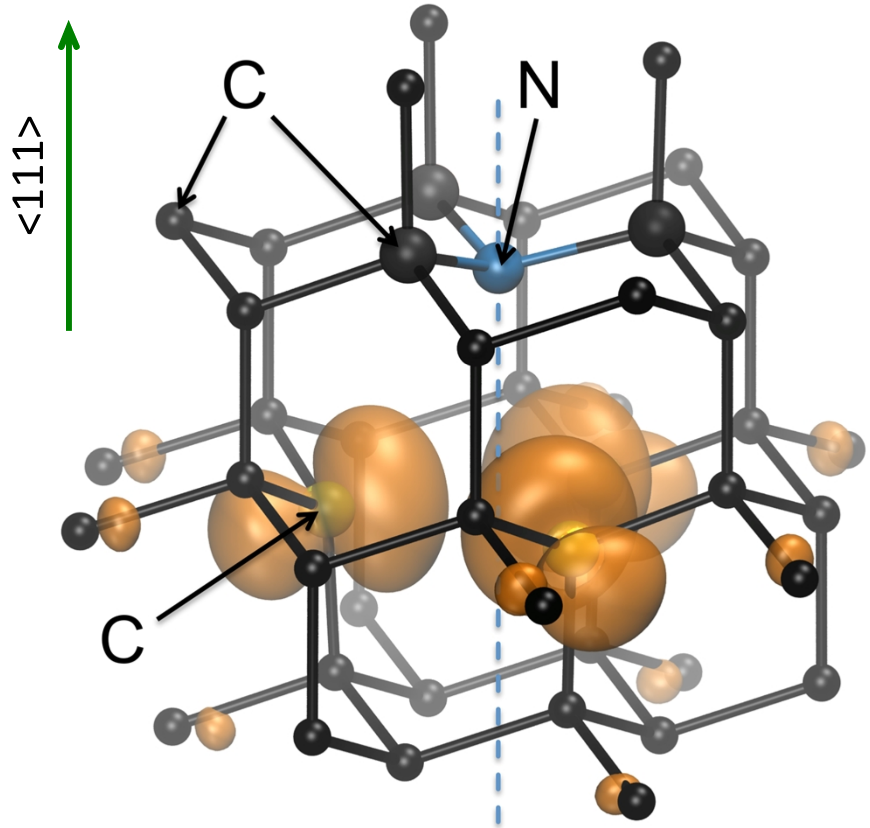
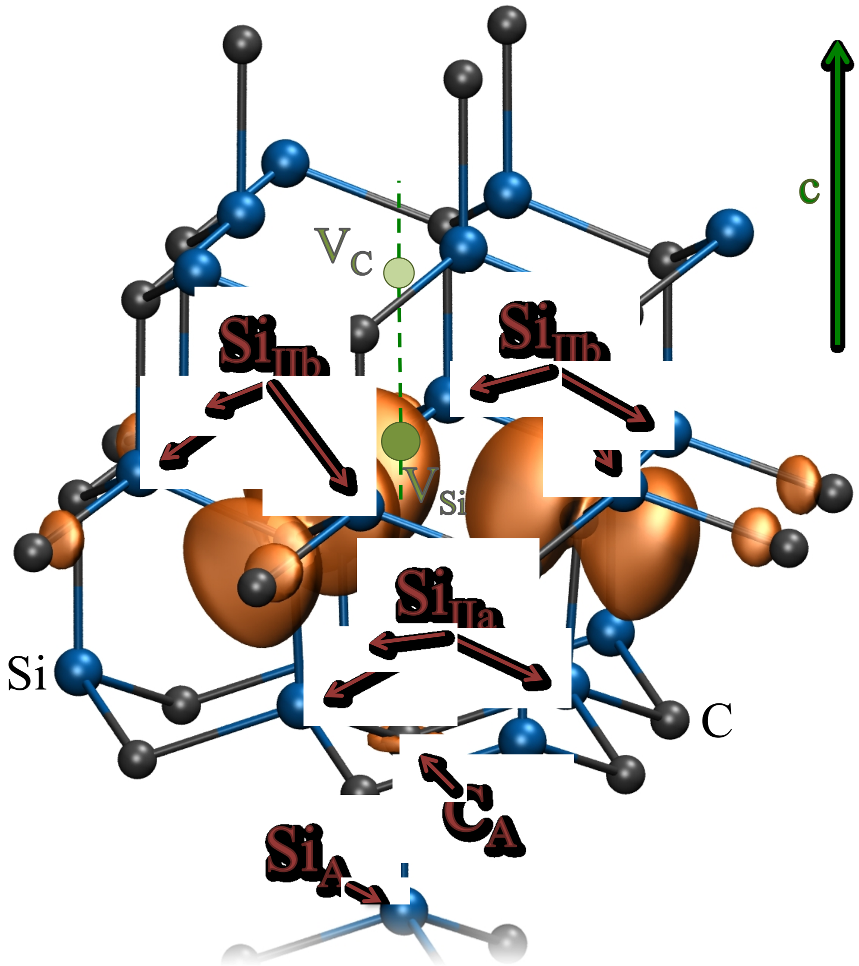


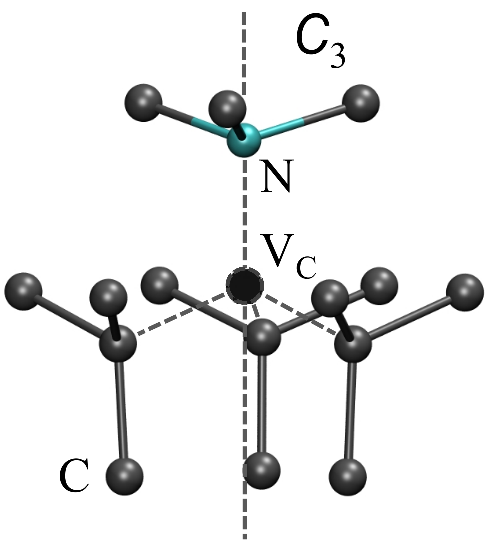
Divacancy in SiC



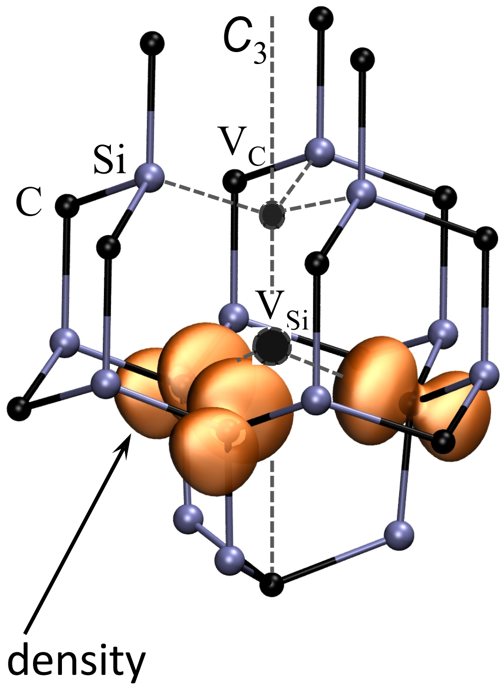
NV-center diamond and divacany in SiC

Structure

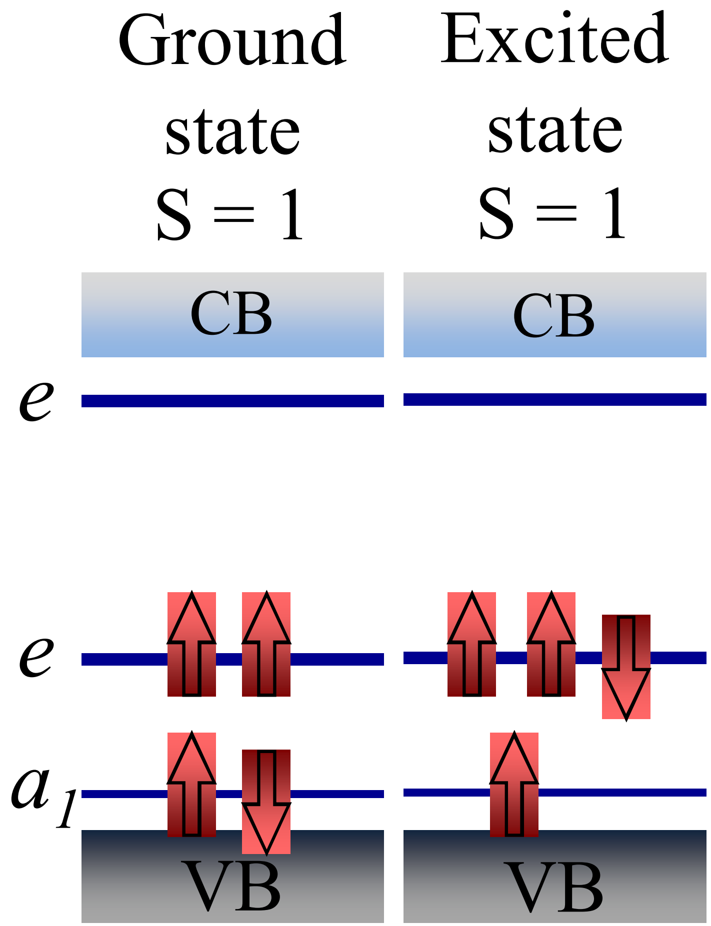
NV center



divacany

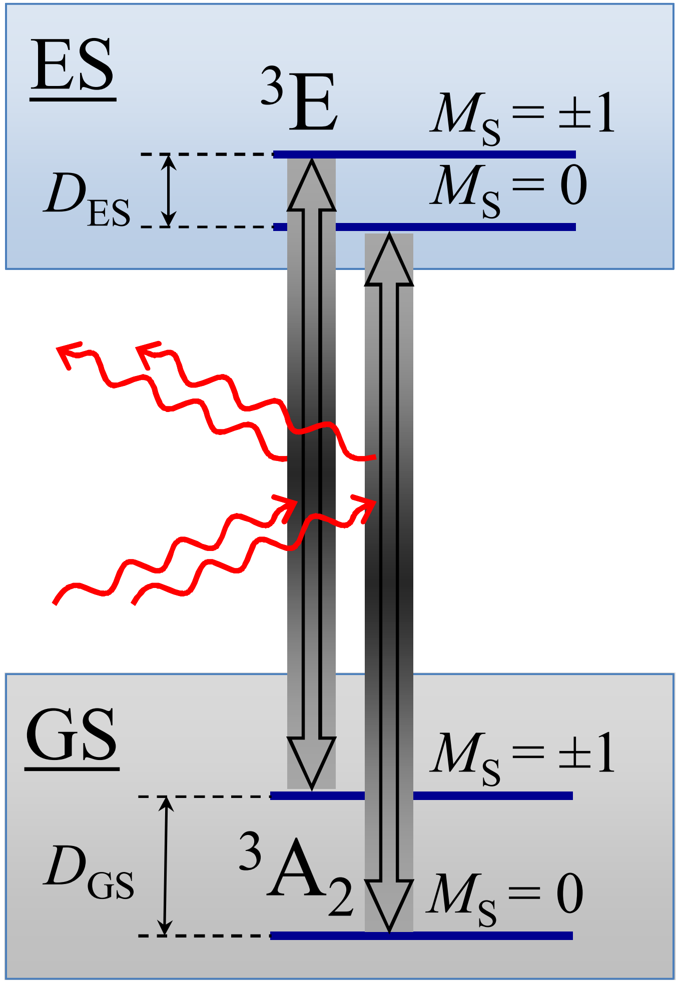


Electron configuration



Optical electron spin polarization and the ODMR measurement

- Due to the S = 1 spin state and the localized spin density, the spin states split in the GS and the ES (zero-field splitting, D)



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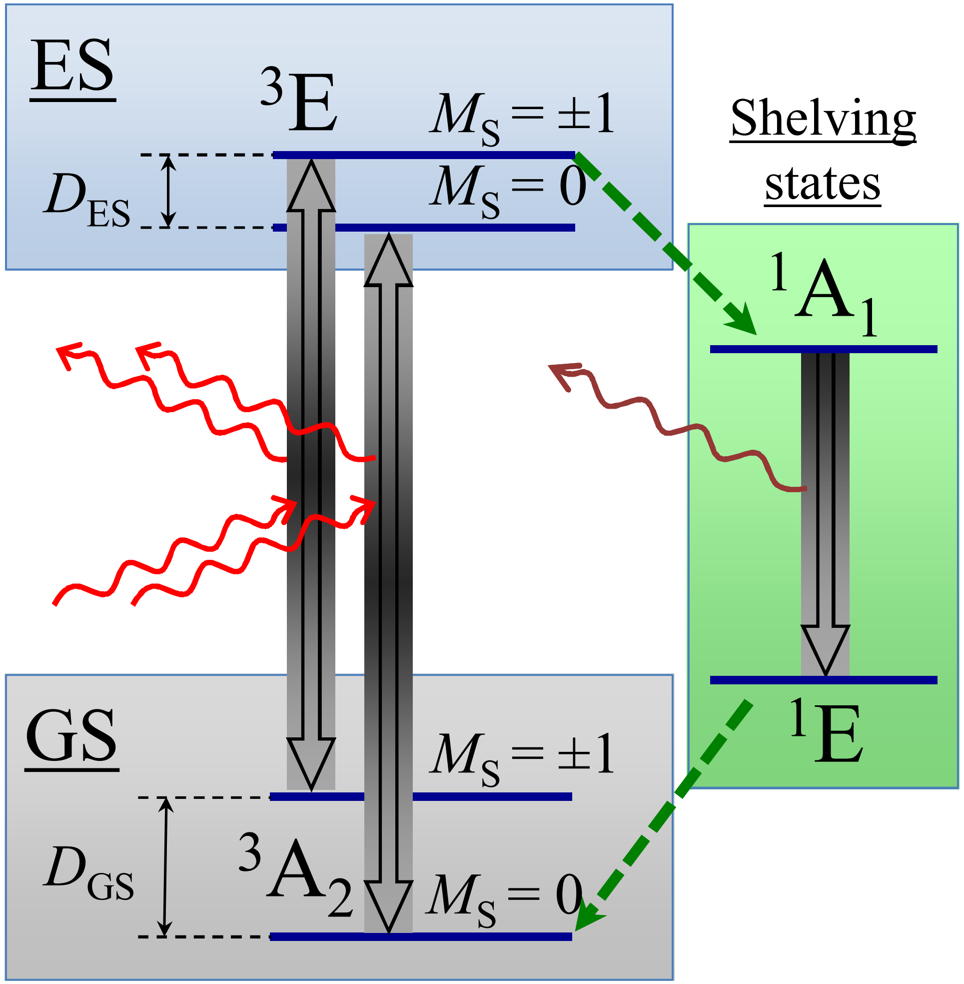
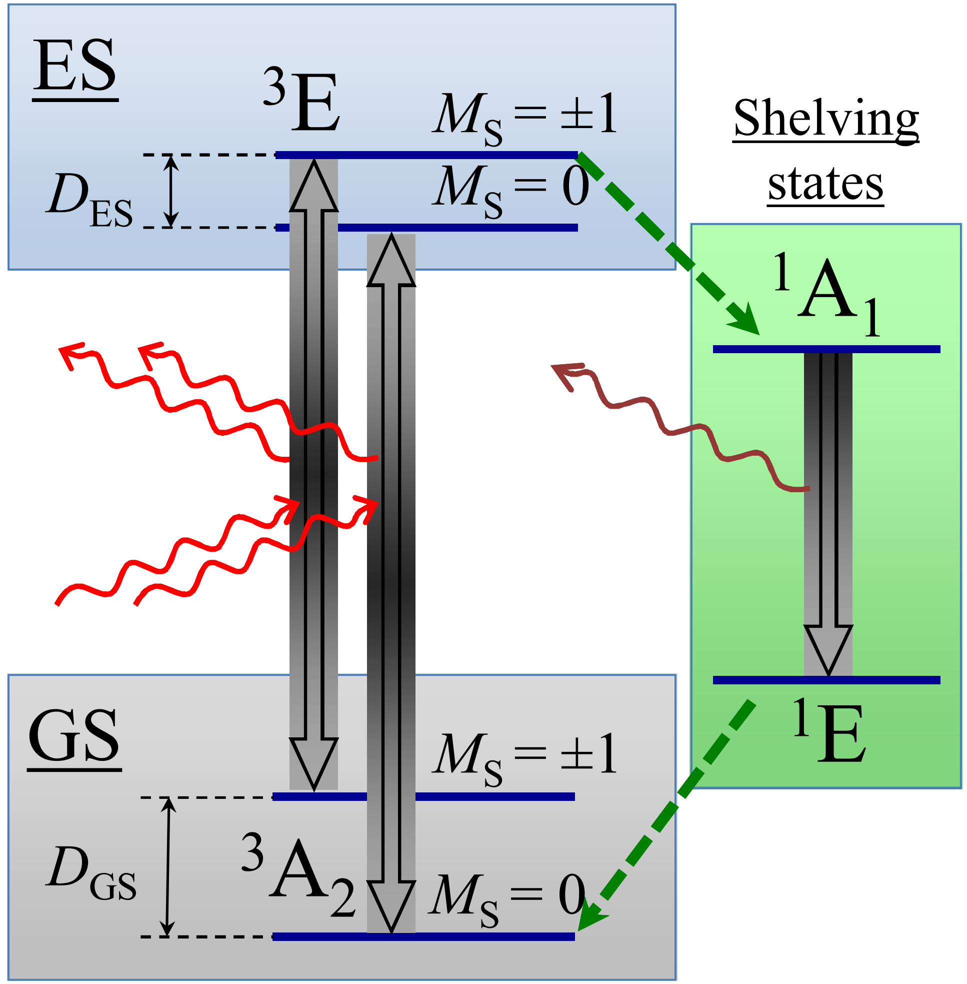


Optical electron spin polarization and the ODMR measurement

- Due to the S = 1 spin state and the localized spin density, the spin states split in the GS and the ES (zero-field splitting, D)
- A spin selective non-radiative decay path allows spin polarization of the defect
- ODMR measurement:
  - Continuous optical excitation polarizes the spin in $M_S = 0$
    - **maximal luminescence**
  - Resonant microwave field can flip the spin to $M_S = \pm 1$ state.
    - from the $M_S = \pm 1$ states a non-radiative path is



Spin Hamiltonian

The general case for one adjacent nuclei:

$$\hat{H} = \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{D}\hat{\mathbf{S}} + \mu_{\mathrm{B}}\mathbf{B}^{\mathrm{T}}\mathbf{g}_{\mathrm{e}}\hat{\mathbf{S}} + \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{A}\hat{\mathbf{I}} + \mu_{\mathrm{N}}\mathbf{B}^{\mathrm{T}}\mathbf{g}_{\mathrm{N}}\hat{\mathbf{I}}$$

For the considered defects, magnetic field along the symmetry axis

$$\hat{H} = D\hat{S}_z^2 + g_{\mathrm{e}}\mu_{\mathrm{B}}B\hat{S}_z + \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{A}\hat{\mathbf{I}}$$

where

$$\hat{H}_{\mathrm{hyp}} = \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{A}\hat{\mathbf{I}} = \frac{A_{\perp}}{2}\left(\hat{S}_{+}\hat{I}_{-} + \hat{S}_{-}\hat{I}_{+}\right) + A_{\parallel}\hat{S}_z\hat{I}_z$$

In the basis of

M_S = {0,-1}, M_I = {±1/2}

$|0\uparrow\rangle,\ |0\downarrow\rangle,\ |-1\uparrow\rangle,\ |-1\downarrow\rangle$

$$H = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & d & 0 \\ 0 & d & \varepsilon_{-1}^{\uparrow} & 0 \\ 0 & 0 & 0 & \varepsilon_{-1}^{\downarrow} \end{pmatrix}$$

$$\varepsilon^{\uparrow\downarrow} = (D - g_{\mathrm{c}}\mu_{\mathrm{B}}B) \mp \frac{A_{\parallel}}{2}$$

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$$|+\rangle = \alpha|0,\downarrow\rangle + \beta|-1,\uparrow\rangle \quad |-\rangle = \beta|0,\downarrow\rangle - \alpha|-1,\uparrow\rangle$$

Hyperfine term

Hyperfine interaction term in the basis of $|0\uparrow\rangle$, $|0\downarrow\rangle$, $|-1\uparrow\rangle$, and $|-1\downarrow\rangle$ states.

$$\hat{H}_{\text{hyp}}(A_{xx}, A_{yy}, A_{zz}, \theta) = \frac{1}{2}\begin{pmatrix} 0 & 0 & \frac{1}{\sqrt{2}}b & \frac{1}{\sqrt{2}}c_- \\ 0 & 0 & \frac{1}{\sqrt{2}}c_+ & -\frac{1}{\sqrt{2}}b \\ \frac{1}{\sqrt{2}}b & \frac{1}{\sqrt{2}}c_+ & -a & -b \\ \frac{1}{\sqrt{2}}c_- & -\frac{1}{\sqrt{2}}b & -b & a \end{pmatrix},$$

where

$$a = A_{zz}\cos^2\theta + A_{xx}\sin^2\theta$$

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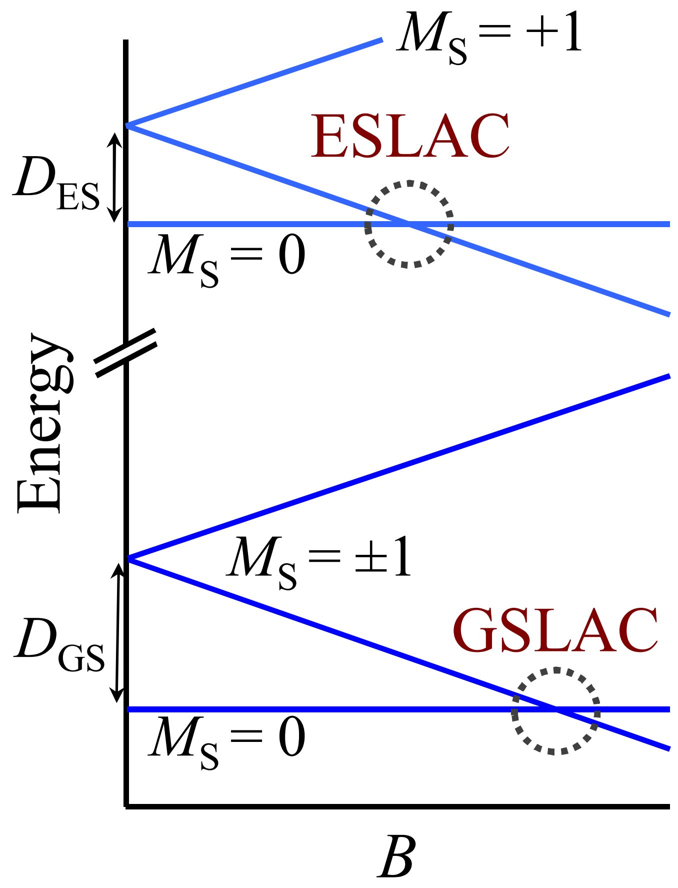
Level anti-crossing (LAC)

- To realize polarization transfer the nuclear and electron spin states should be coupled effectively

- For the NV and the divacancy, the large electron spin-electron spin interaction (zero field splitting) hinder the coupling, D >> A

- By applying a magnetic field, the M_S = 0 and M_S = -1 states approach each other
  - **At LAC, the hyperfine interaction can flip both the electron and nuclear spin states**



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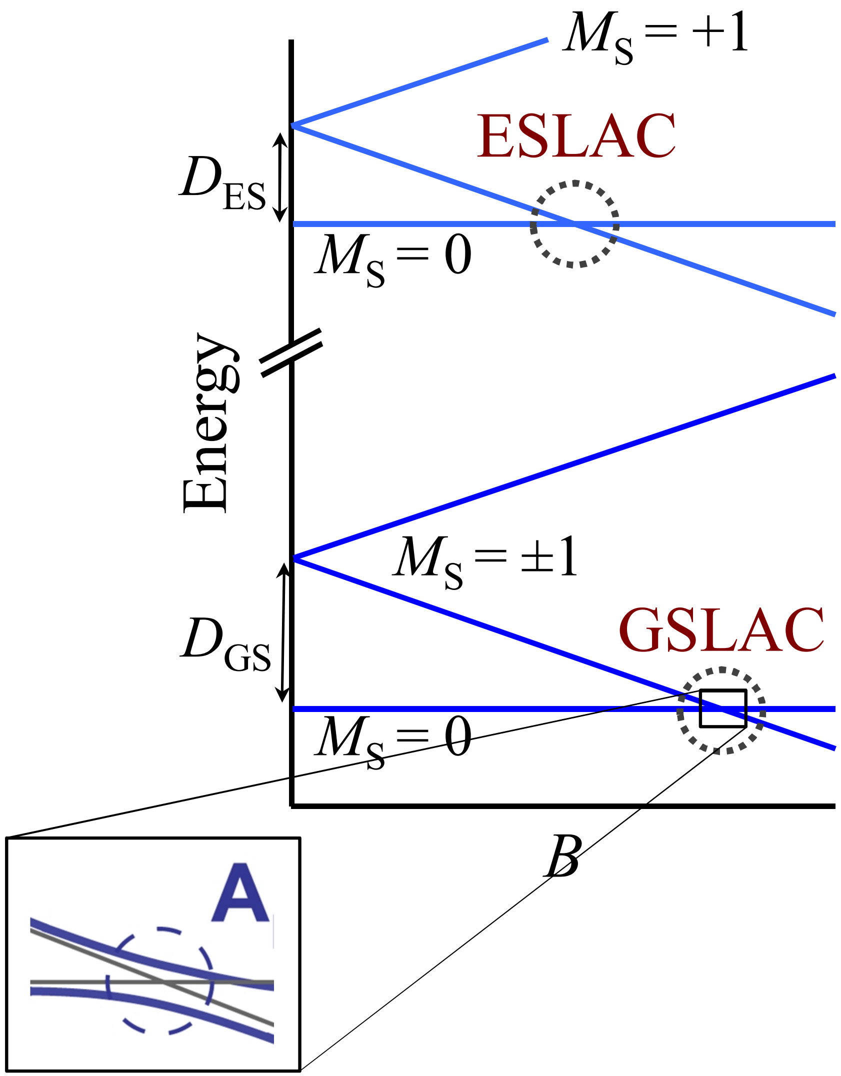
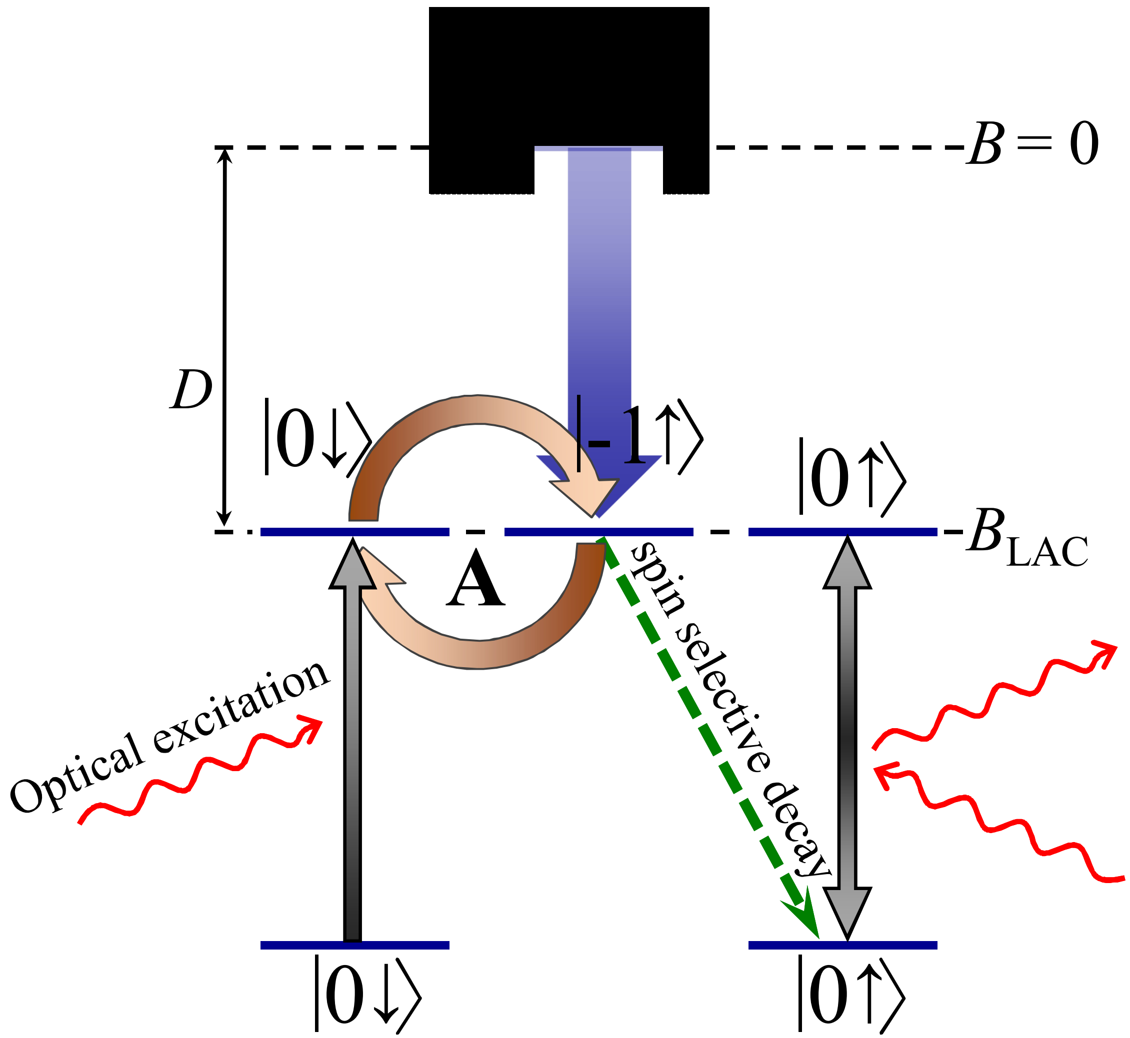


The DNP cycle

- Consider one adjacent nuclear spin (I = 1/2) to a NV center or divacay in SiC

- At LAC, continuous optical excitation and the subsequent non-radiative decay polarizes both the electron and nuclear spins



DNP cycle

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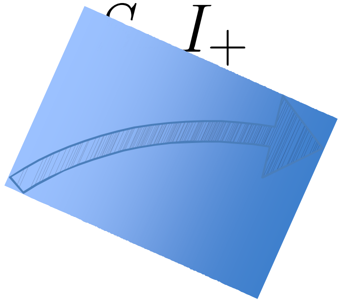
$$\hat{S}_{-}\,\hat{I}_{+}$$



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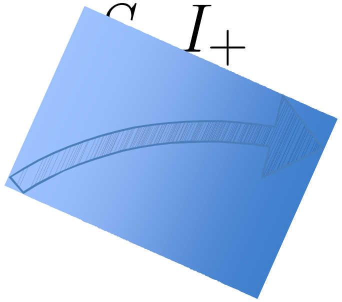

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n-radiative decay

the electron

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Nuclear spin re effects try to the equilibrium non- spin polarized state

Our model – full cycle – single nucleus

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GS time evolution

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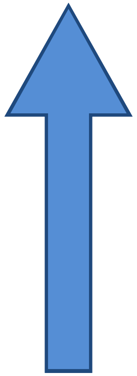
Optical excitation

Spin conserving



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Our model – full cycle – single nucleus

ES time evolution

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Optical excitation

Spin conserving

GS time evolution

$$\hat{H}_{\mathrm{GS}} = \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{D}_{\mathrm{GS}}\hat{\mathbf{S}} + \mu_{\mathrm{B}}\mathbf{B}^{\mathrm{T}}\mathbf{g}_{\mathrm{e}}\hat{\mathbf{S}} + \hat{\mathbf{S}}^{\mathrm{T}}\mathbf{A}_{\mathrm{GS}}\hat{\mathbf{I}} + \mu_{\mathrm{N}}\mathbf{B}^{\mathrm{T}}\mathbf{g}_{\mathrm{N}}\hat{\mathbf{I}},$$

Our model – full cycle – single nucleus

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Optical excitation

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Decay

May flip the electron spin.

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Steady state nuclear spin polarization:

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Optical excitation
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No simplifications here!

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Steady state nuclear spin polarization: $P = \dfrac{p_+ - p_-}{p_+ + p_- + \kappa}$

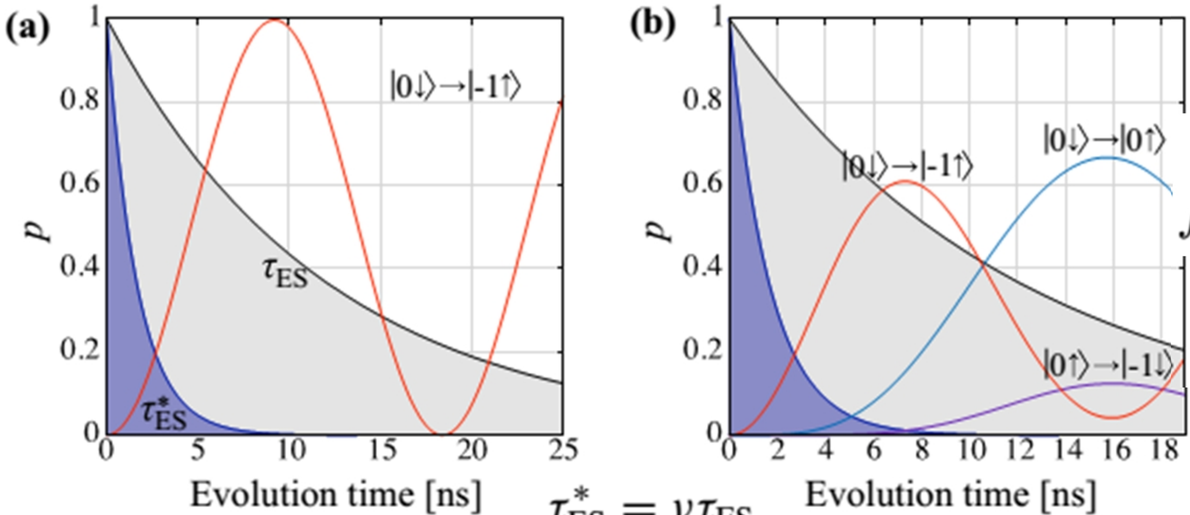
κ: relaxation time of nuclear spins per number of optical cycles per unit time

Parameters

- time does matter, particularly, in ES

  A is relatively small → spin rotation is slow → ≈ lifetime and/or coherence time of e-spin

NV & ^{15}N (I=1/2)



$$\tau_{\mathrm{ES}}^* = \nu \tau_{\mathrm{ES}}$$

$$p^{\mathrm{ES}}(\chi_{\mathrm{Initial}}|\chi_{\mathrm{Final}}) = \int_0^{\tau_{\mathrm{ES}}} \varrho(t) \left| \langle \chi_{\mathrm{Final}} | e^{-i\hat{H}^{\mathrm{ES}} t/\hbar} | \chi_{\mathrm{Initial}} \rangle \right|^2 dt$$

$$\varrho(t) = \frac{1}{\tau_{\mathrm{ES}}^*} e^{-t/\tau_{\mathrm{ES}}^*}$$

- finite coherence time of e-spin in GS reduces the nuclear spin rotation probability: μ

$$A_{xx}^{\mathrm{GS}}, A_{yy}^{\mathrm{GS}}, A_{zz}^{\mathrm{GS}}, \theta_{\mathrm{GS}}, A_{xx}^{\mathrm{ES}}, A_{yy}^{\mathrm{ES}}, A_{zz}^{\mathrm{ES}}, \theta_{\mathrm{ES}}, D_{\mathrm{GS}}, E_{\mathrm{GS}}, D_{\mathrm{ES}}, E_{\mathrm{ES}}, \Gamma, \tau_{\mathrm{ES}}; \mu, \nu, \kappa; B, \theta_{\mathrm{B}}$$

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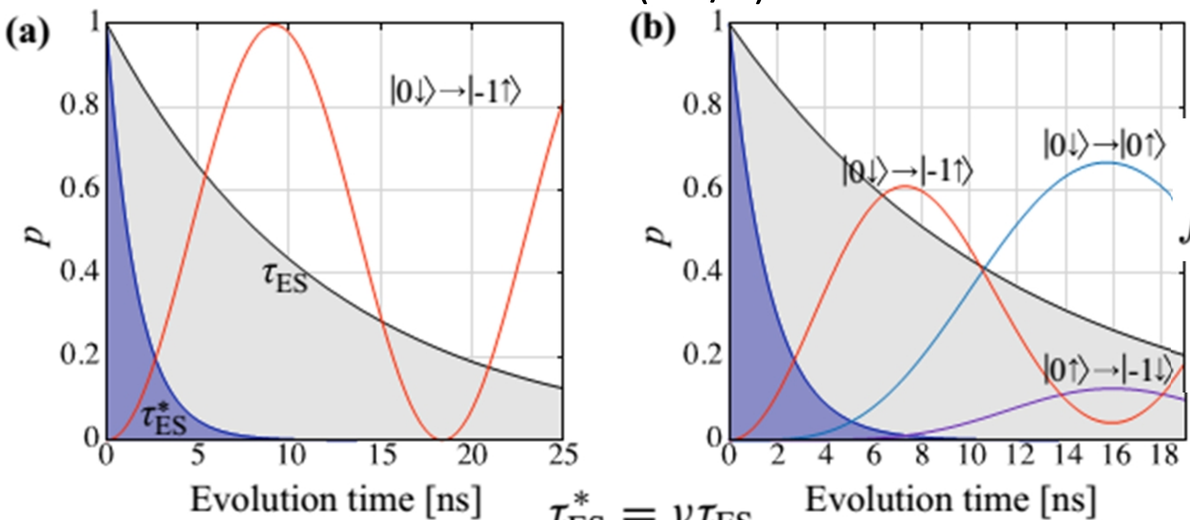
NV & ^{15}N (I=1/2)



$$\tau^*_{ES} = \nu \tau_{ES}$$

$$p^{ES}(\chi_{Initial}|\chi_{Final}) = \int_0^{T_{ES}} \varrho(t) \left| \langle \chi_{Final} | e^{-i\hat{H}^{ES}t/\hbar} | \chi_{Initial} \rangle \right|^2 dt$$

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Determined either by calculations or experiment

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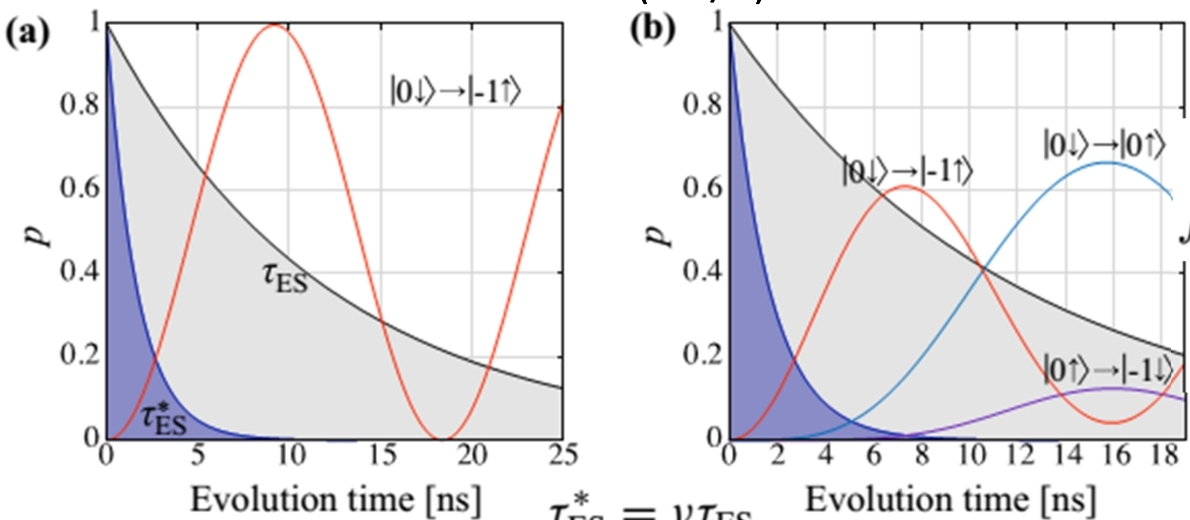

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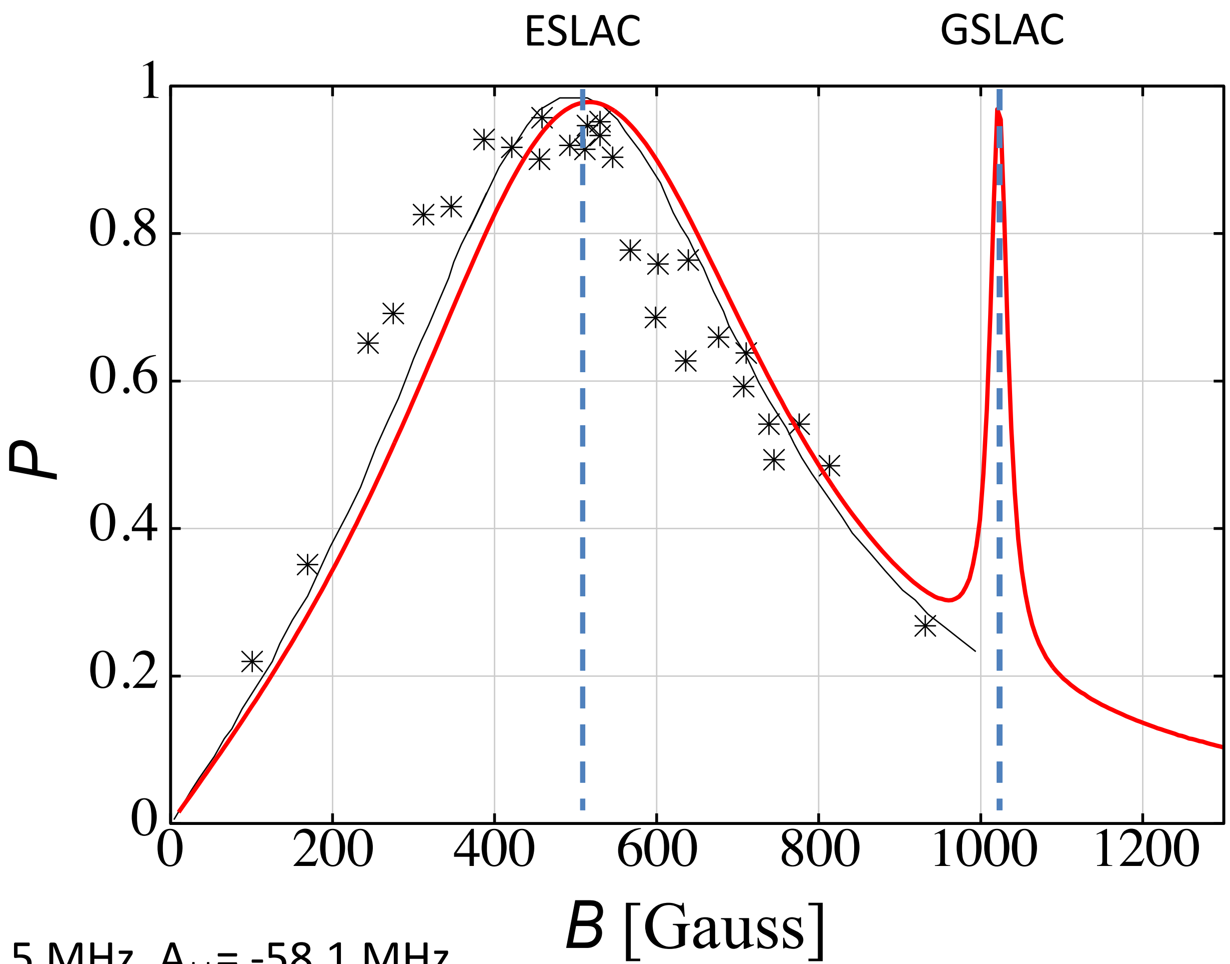
Typical results, NV & ^{15}N (I=1/2)



ES: $A_\perp$= -38.5 MHz, $A_{||}$= -58.1 MHz

GS: $A_\perp$= 3.9 MHz, $A_{||}$= 3.4 MHz

What determines the polarizability?

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- There are theories
  - Directions cosines of the hyperfine interactions are important
    - High polarizability for nuclei on the axis of the defect
  - No satisfactory consensus has been achieved.

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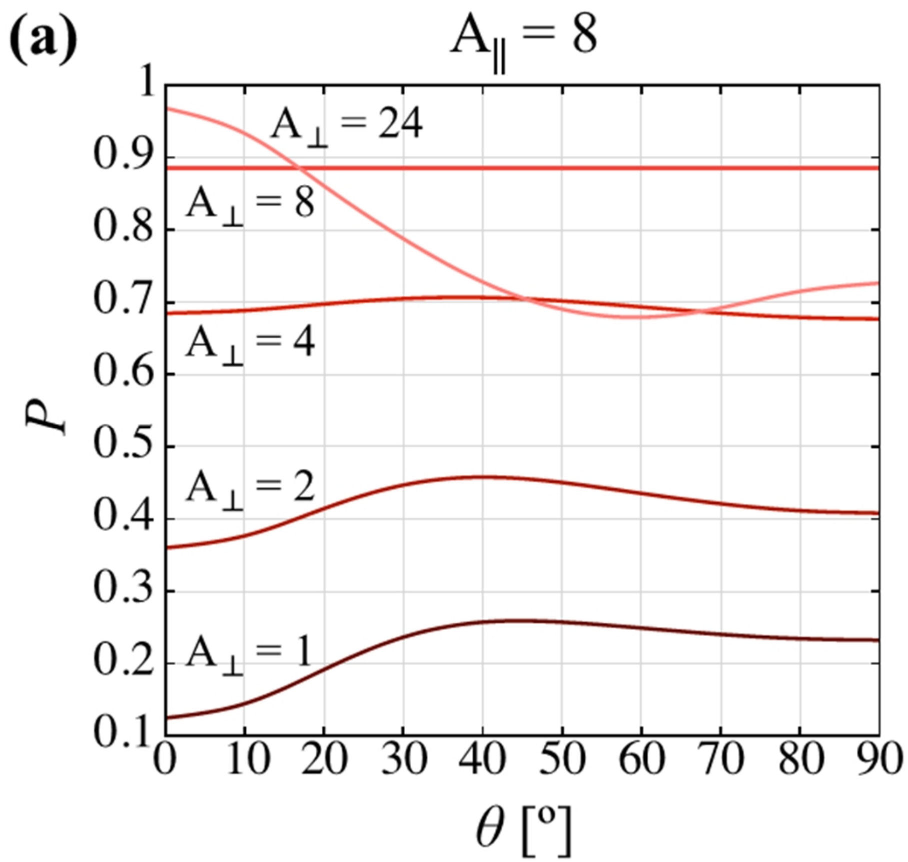

Excited state, I = ½, quantities are in MHz unit.

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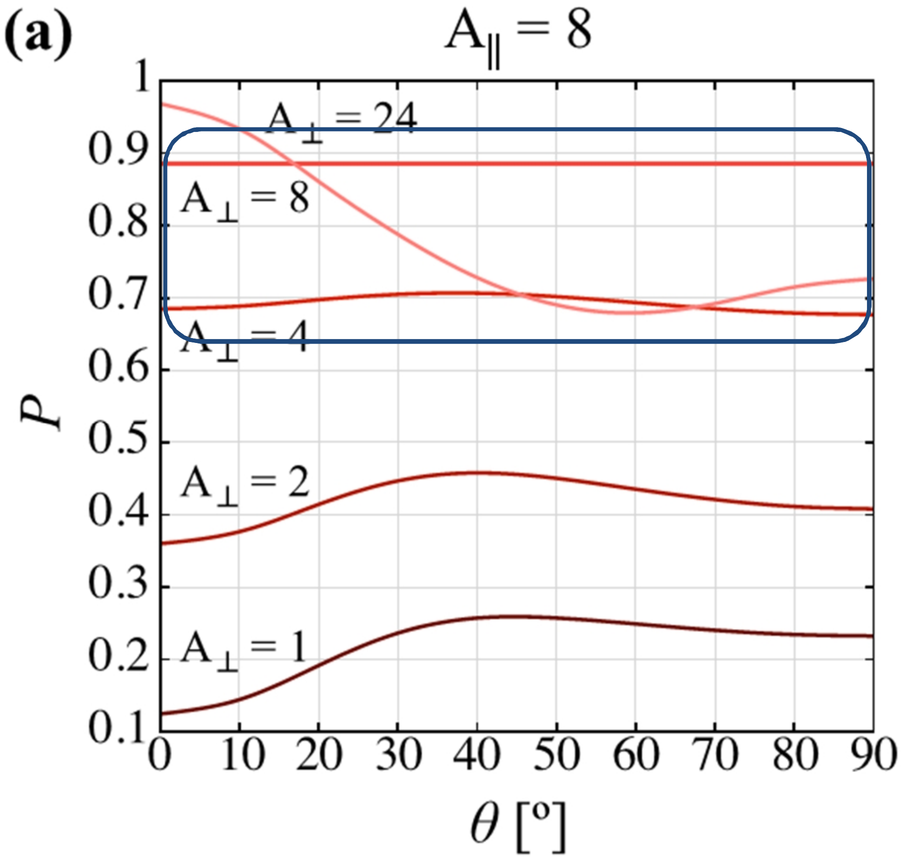
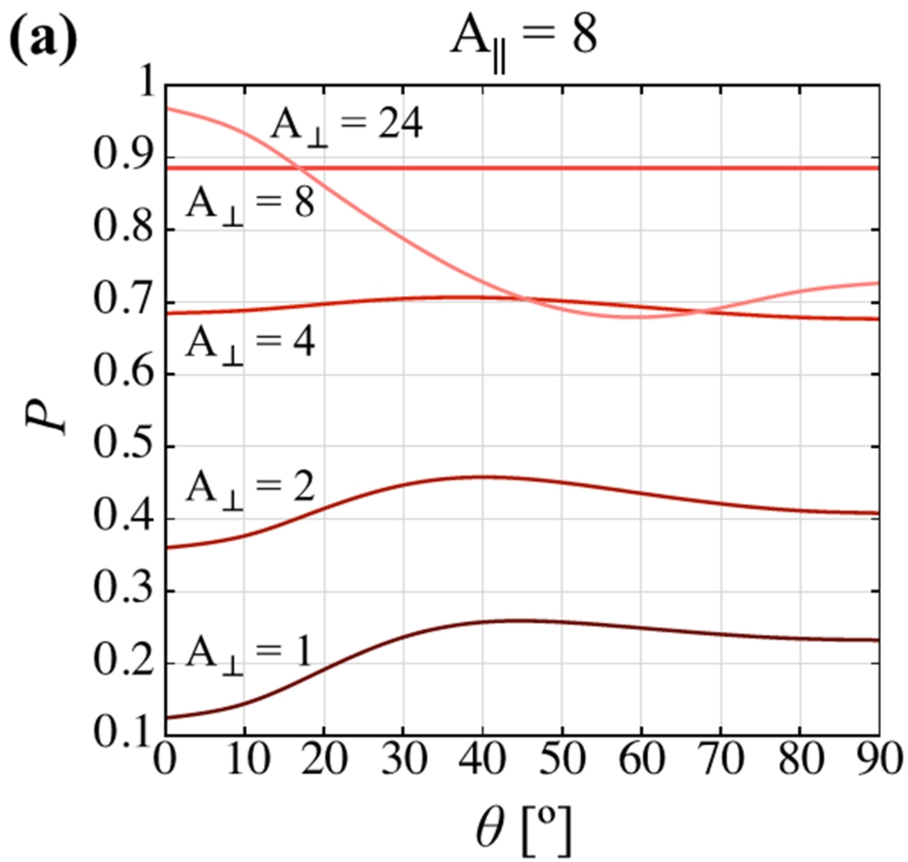


General cases

Excited state, I = ½, quantities are in MHz unit.

What determines the polarizability?



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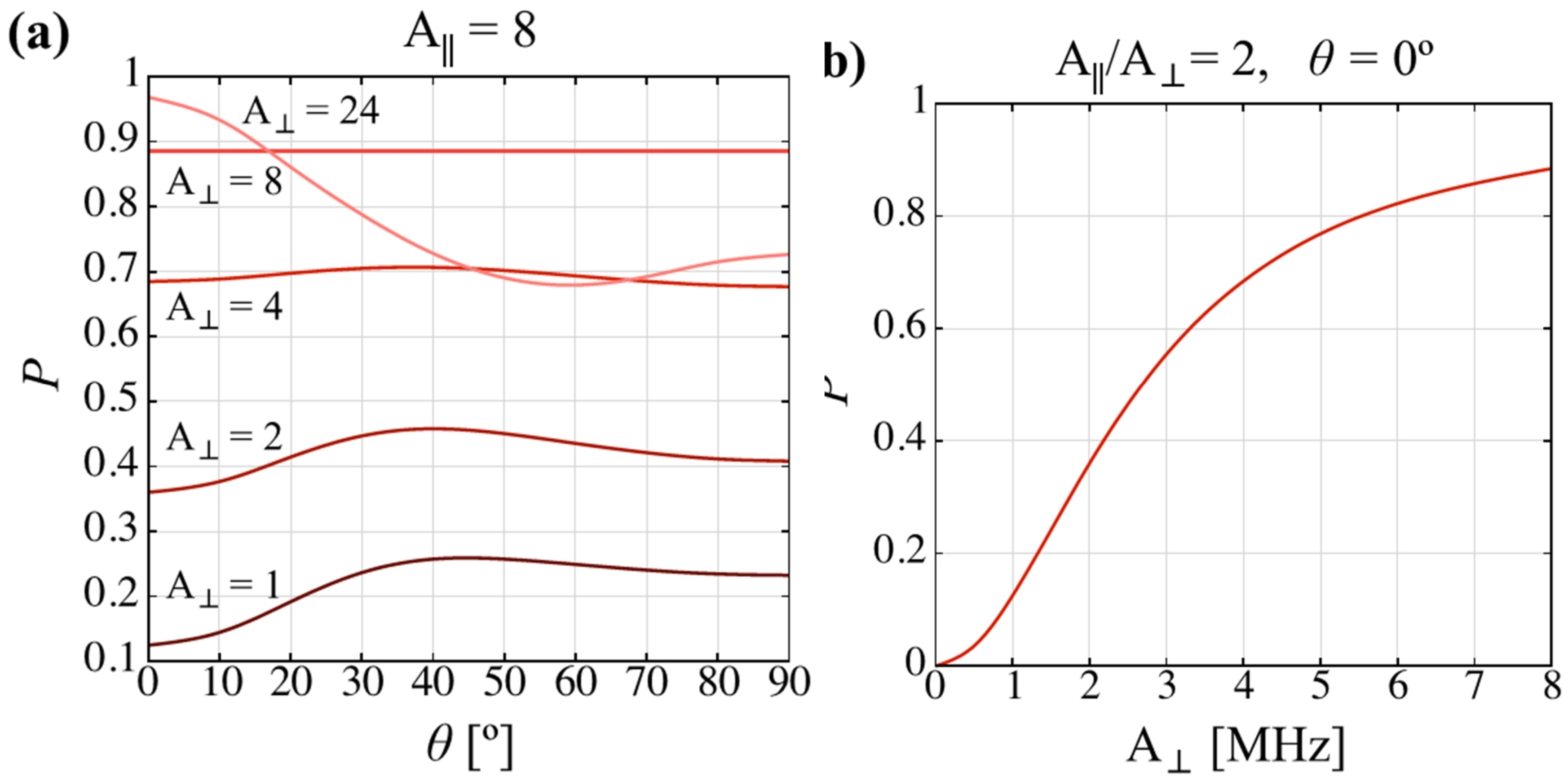

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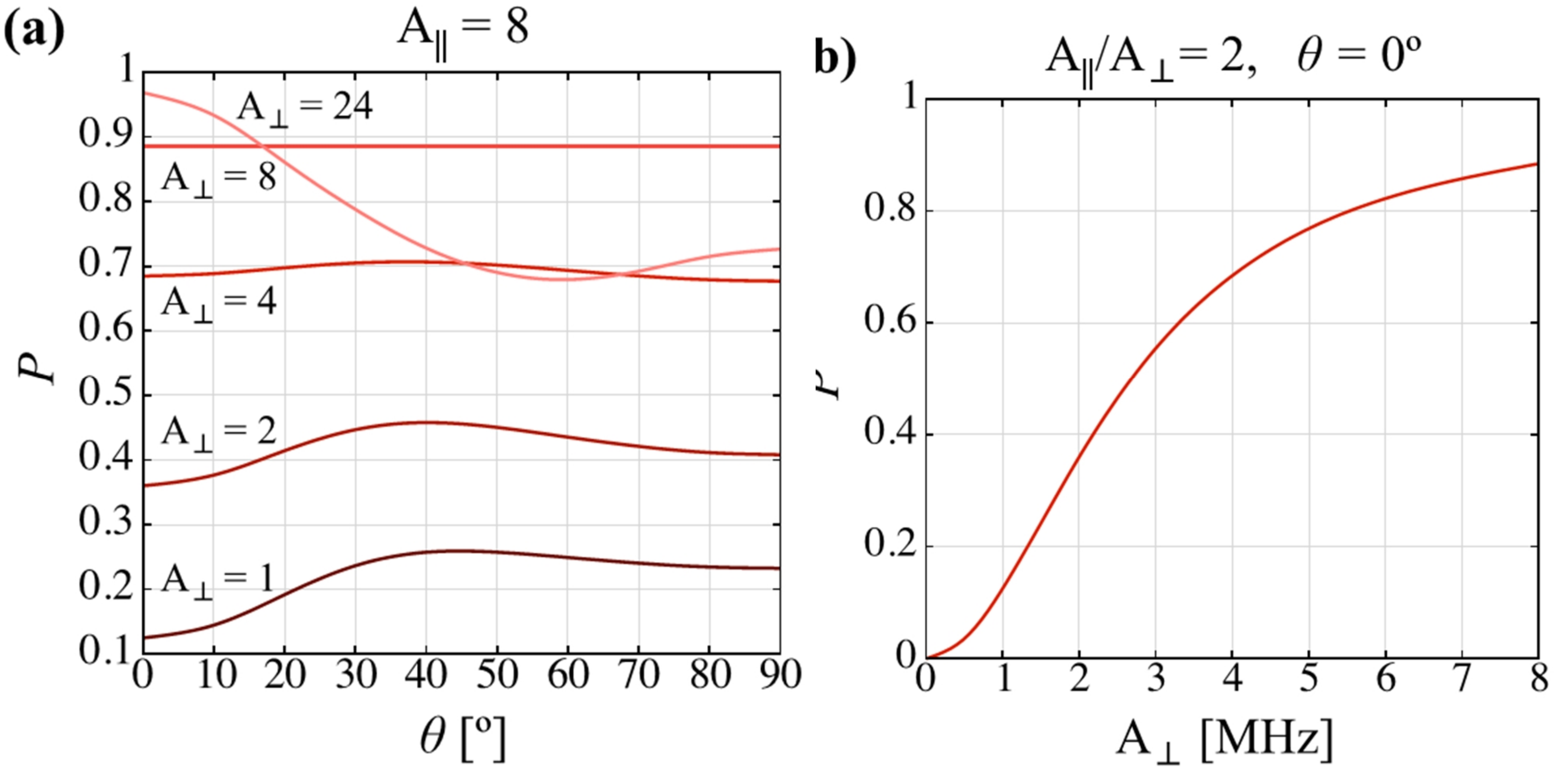
What determines the polarizability?

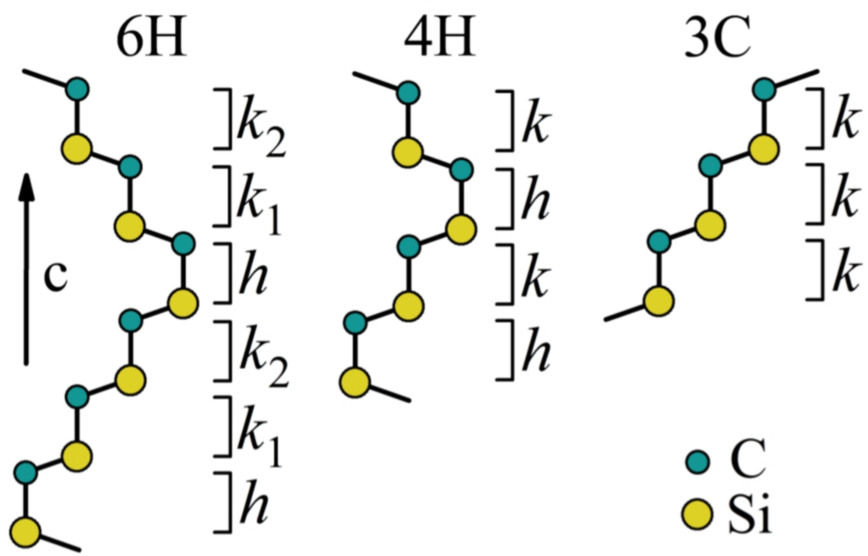
- relatively small angular dependence
- the polarizability depends predominantly on A perpendicular



Excited state, I = ½, quantities are in MHz unit.

Detour:
Silicon carbide polytypes

- SiC is a wide-band-gap semiconductor
- SiC exists in about 250 crystalline form
- Provides good doping possibilities
- Inequivalent defect sites

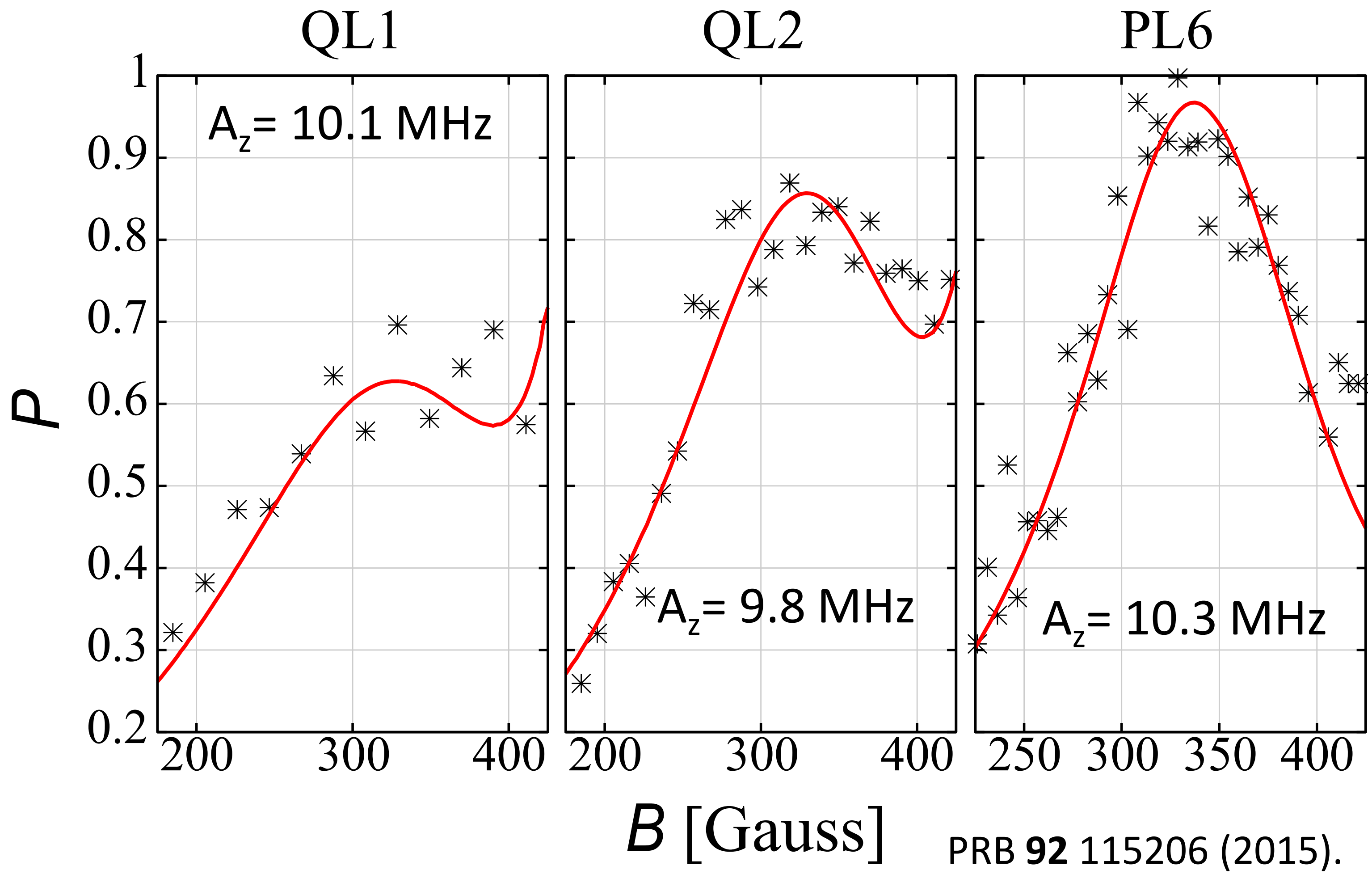


	Most common polytypes		
	3C	4H	6H
Gap [eV]	2.36	3.23	3.0

Configurations of pair defects	
3C-SiC	*kk*
4H-SiC	*hh, kh, kk, hk*
6H-SiC	hh, k_1h, k_1k_1, k_2k_1, k_2k_2, hk_2

What other parameter is important?

Near the ESLAC, ^{29}Si (I = ½) @ Si_{IIb} site close to axial divacancies in SiC.



PRB **92** 115206 (2015).

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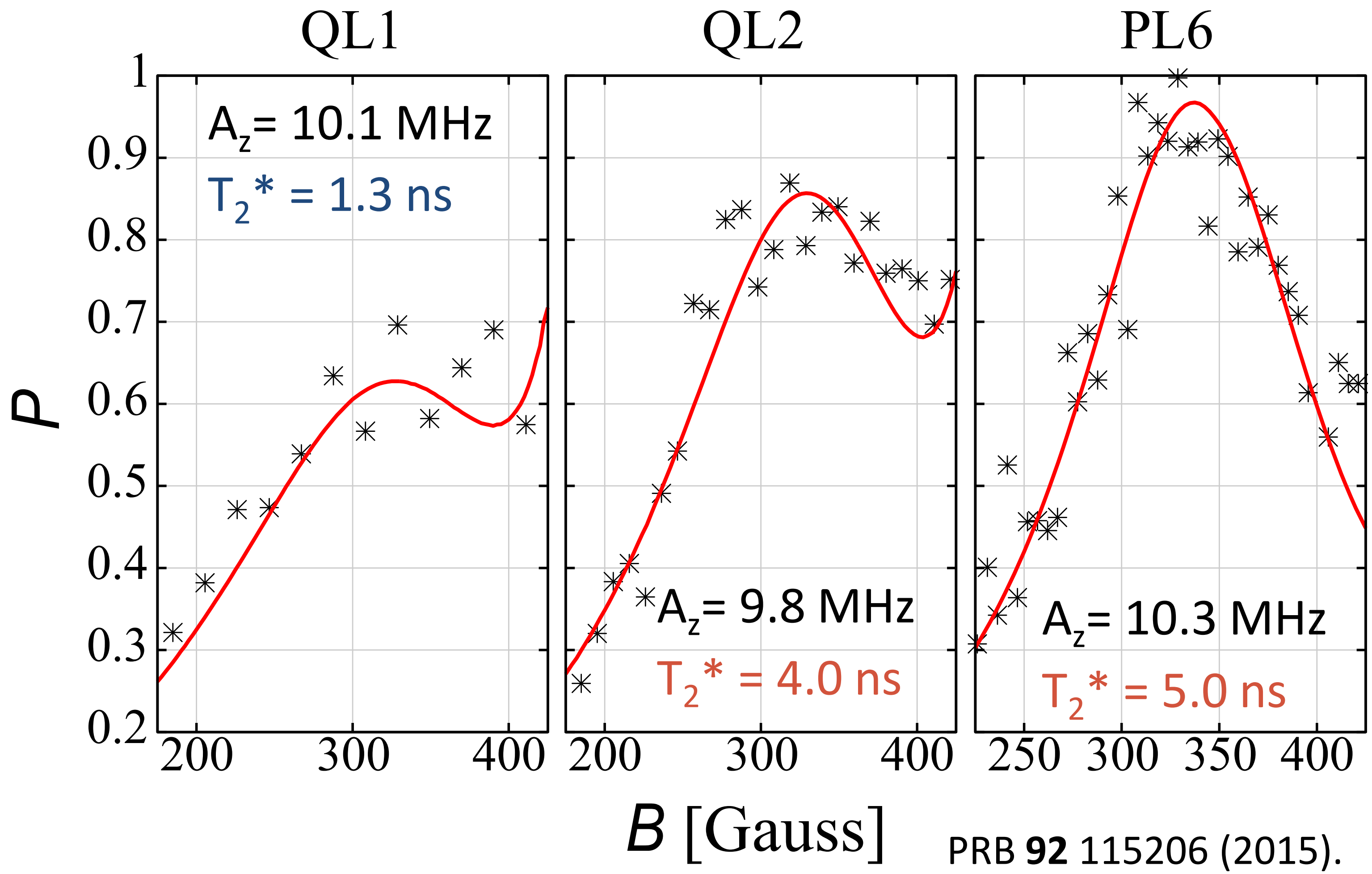
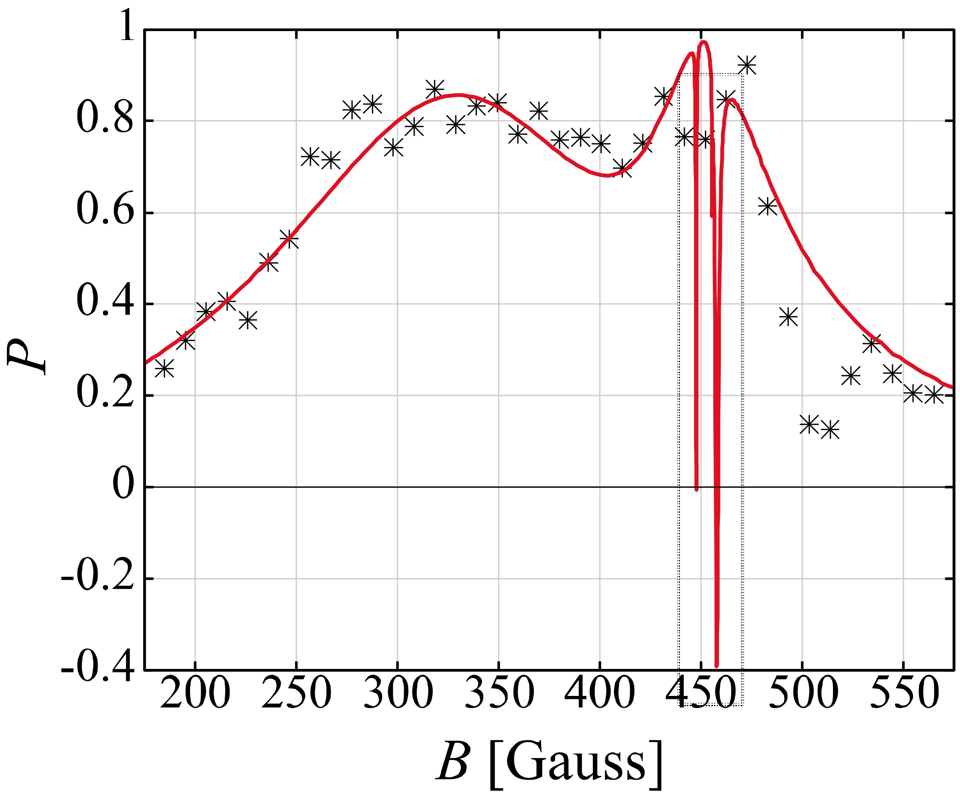
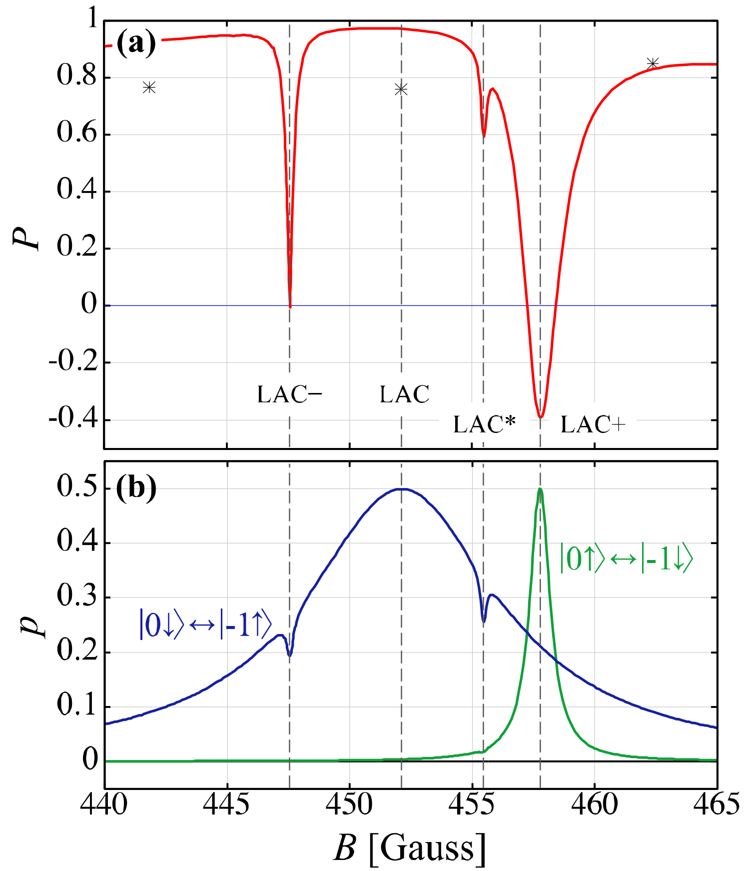


PRB **92** 115206 (2015).

GSLAC fine structure

When the symmetry of the spin Hamiltonian is reduced (divacancy in SiC):





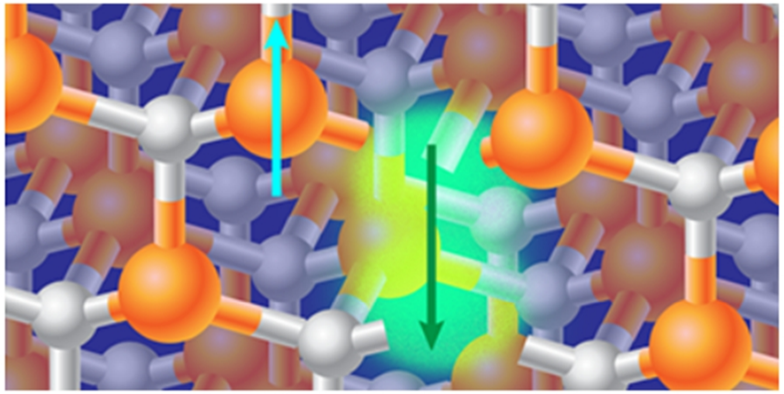
- nuclear spin polarization can flip as a function of *B*

Summary

- Theory of optical nuclear spin polarization in solids is advanced

- A. L. Falk *et al.*, Physical Review Letters **114** 247603 (2015).
- V. Ivády *et al.*, Physical Review B **92** 115206 (2015).

Physics VIEWPOINT



Polarizing Nuclear Spins in Silicon Carbide

Published 17 June 2015

An optical technique polarizes the spin of nuclei in silicon carbide, offering a potential new route to nuclear spin-based quantum memory.

See more in *Physics*