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Mechanical and Electrical Control of Spin Qubits in Li-doped Silicon

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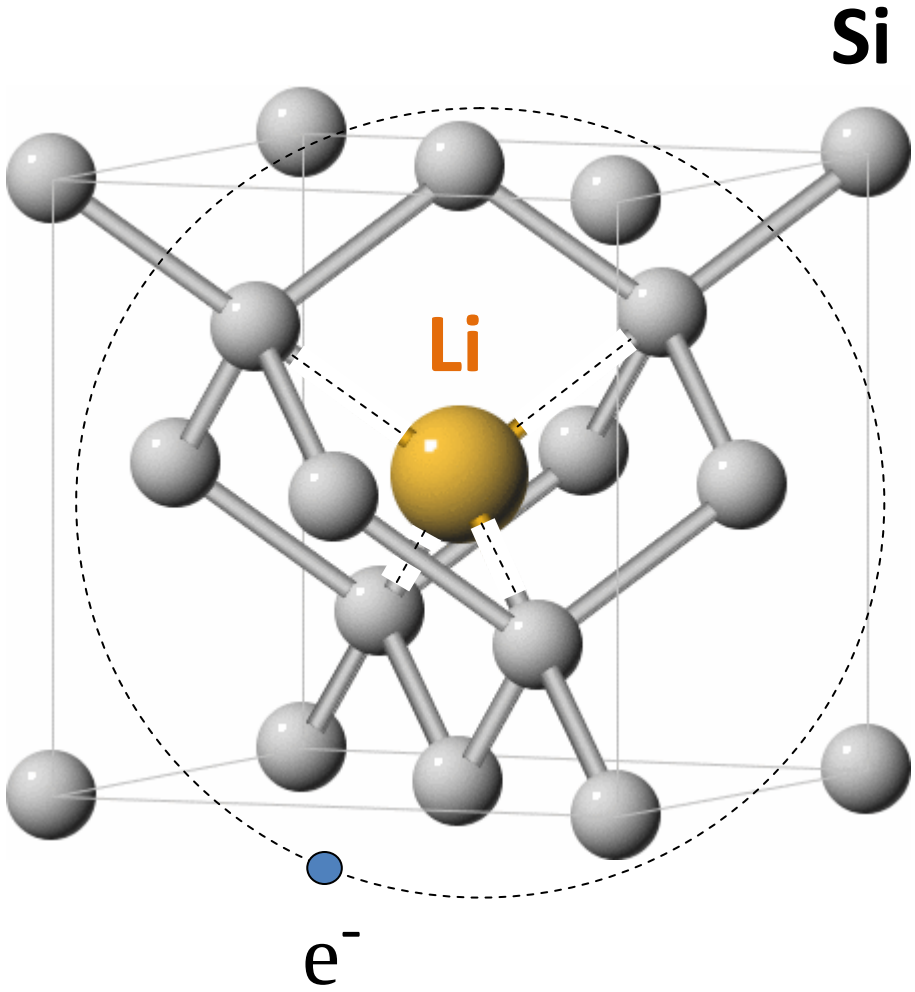
Why Li in Si ?

1) ground state is 5-fold valley-orbit degenerate
2) 3 orders of magnitude enhancement of SO interaction compared to phosphorus donor
3) Strong spin-to-charge conversion
4) Stress, magnetic and electric field controls
5) T_1 time of spin-orbital excitation is several seconds at 0.34 K
6) T_2 time is in the millisecond range at 0.34 K
7) Effective mass theory provides accurate description of EPR and IR experiments. Valley-orbit splitting of ground state energy level 1s(T+E) is 7.5 μeV and is about 100 times smaller then in Si:P
8) 1 Angstrom induced electric dipole moment in 3KV/cm electric field
9) Qubits are formed by “spin-orbital” excitations of Li donor electron
10) Strong inter-qubit elastic-dipole coupling mediated by virtual acoustic phonons. Can be controlled by stress or electric field.

Interstitial donor Li in Si

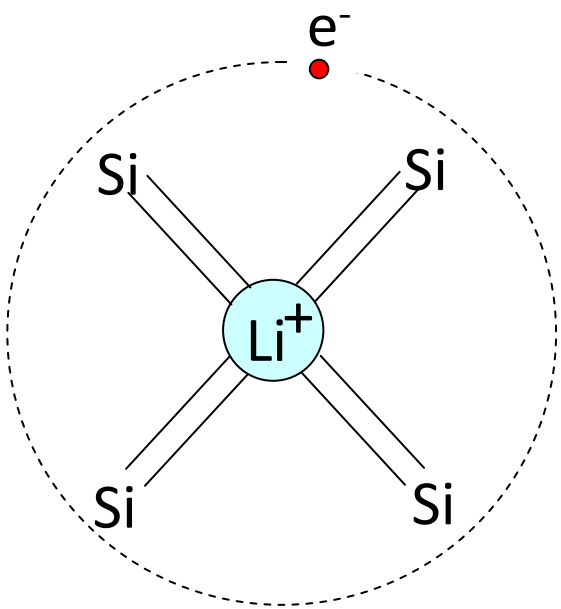
$1s^2 2s$

- **interstitial impurity**
- **local symmetry the same as in substitutional donors**
- **weak, repulsive central cell potential**



Shallow donors in Si

Effective mass theory (EMT), Kohn & Luttinger, Phys. Rev. (1955)



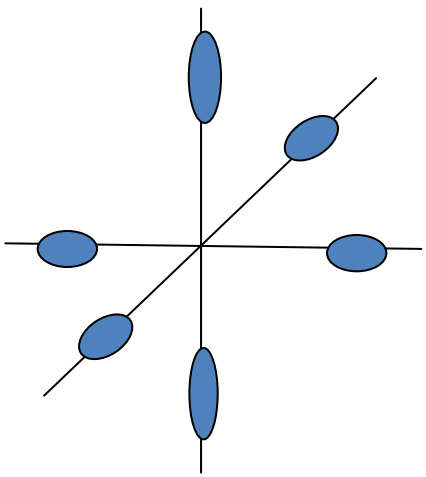
Length and energy scales:

$$\gamma = \frac{m_\perp}{m_\parallel} \approx 0.19, \quad Ry^* = \frac{m_\perp e^4}{2\hbar^2 \varepsilon^2} \approx 31\,\text{meV}$$

$$a_\perp = 2.34\,\text{nm} \qquad a_\parallel = 1.35\,\text{nm}$$

Dimensionless Hamiltonian:

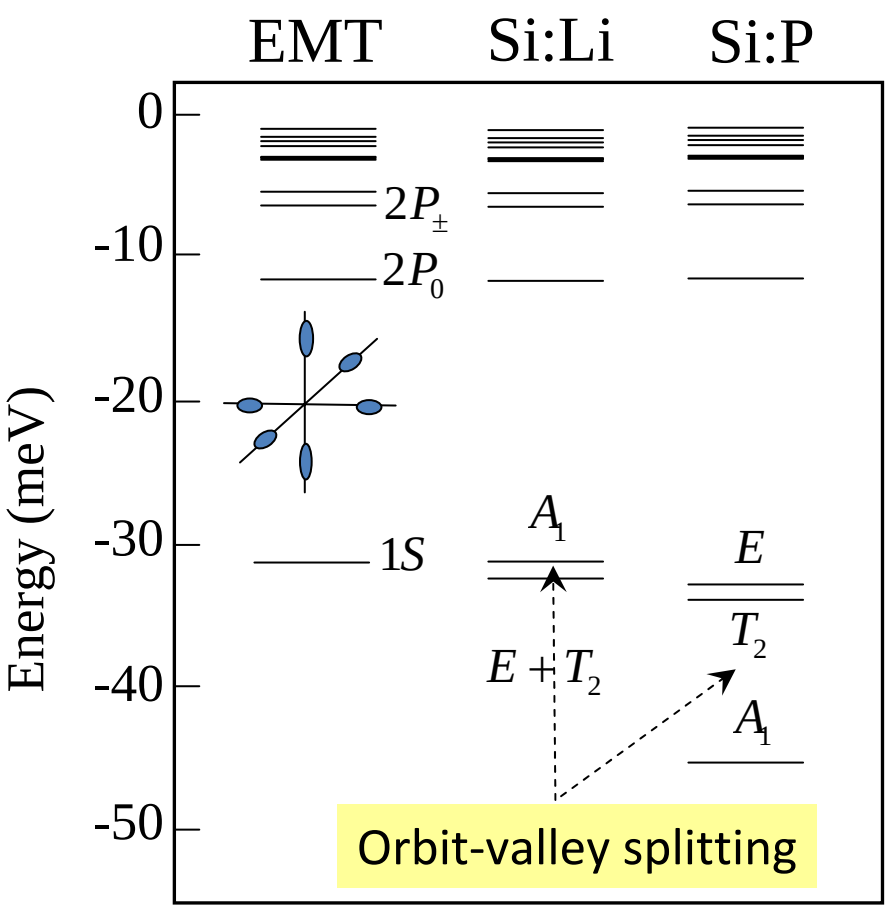
$$H_0 = -\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \gamma \frac{\partial^2}{\partial z^2} \right) - \frac{2}{r}$$



1s envelope function:

$$F_{1s}^z(\mathbf{r}) = \left(\frac{1}{\pi a_\perp^2 a_\parallel} \right)^{1/2} \exp\left[-\sqrt{\left(x^2 + y^2\right)/a_\perp^2 + z^2/a_\parallel^2} \right]$$

Li donor in silicon: background



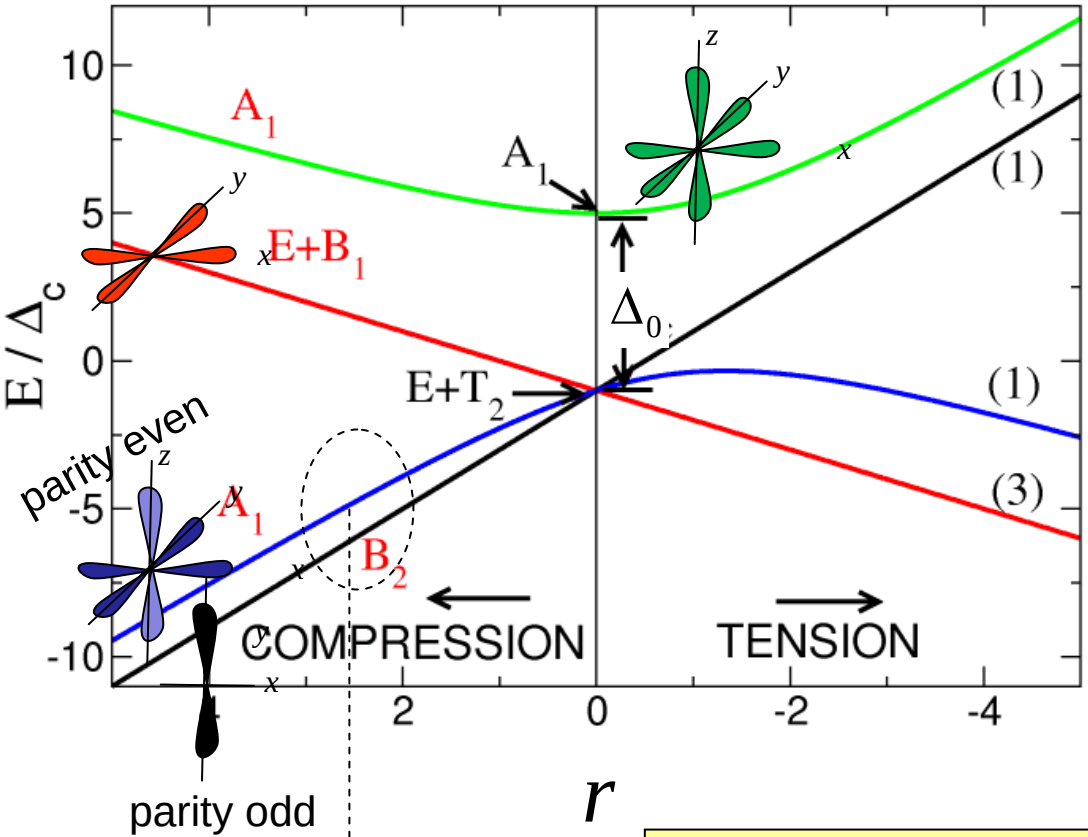
$$\Psi^{\mu}(\vec{r}) = \sum_{j=1}^{6} \alpha_{\mu,j} F_{\mu}^{j}(\vec{r}) \psi_{\vec{k}_j}(\vec{r})$$

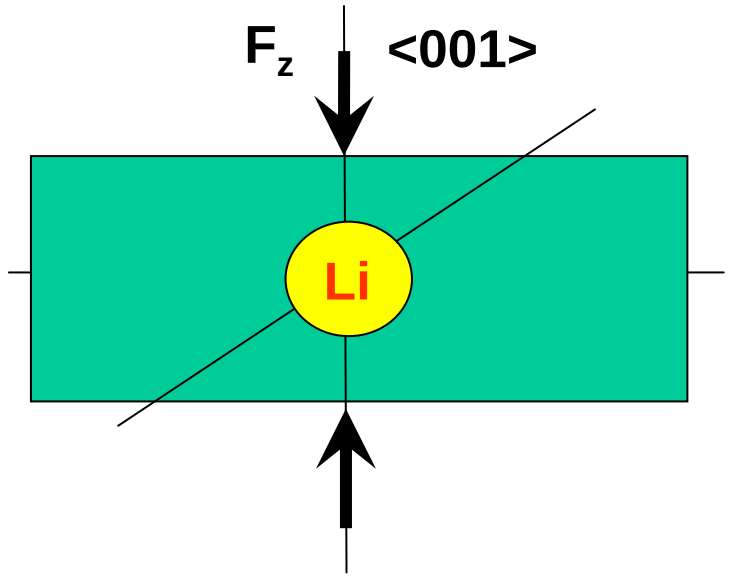
Level	P (meV)	Li (meV)
1s(A_1)	45.59	31.24
1s(E)	32.58	33.02
1s(T_2)	33.89	33.02

- Inverse order of 1s states
- Ground state 1s(T+E) is degenerate
- Splitting of T_2 and E 7.5 μeV

Li – 5-fold valley-degenerate ground state!

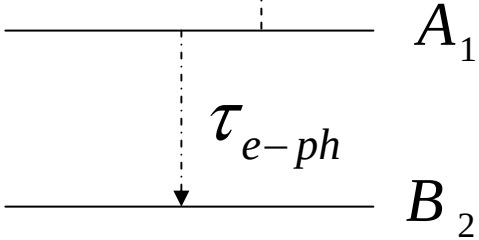
Si:Li spectrum under uniaxial stress





$\Delta_0 = 1.76 \quad \text{meV}$

$$r = (3\Delta_0)^{-1}\Xi_u(s_{11} - s_{12})F_z$$



$B_2 \quad \alpha = (0,0,0,0,1,-1)/\sqrt{2} \Rightarrow |0\rangle$ parity odd

$A_1 \quad \alpha = (a,a,a,a,b,b) \Rightarrow |1\rangle$ parity even

$\tau_{e-ph} > 1s$ Smelyanskiy,Petukhov,Osipov PRB (2005),

Electron-phonon interaction

$$\left\langle \Psi_\mu \middle| H_{el-ph} \middle| \Psi_{\mu'} \right\rangle = \sum_{\mathbf{q}\nu} V_{\mu\mu'}(\mathbf{q}\nu)\left(a_{\mathbf{q}\nu} + a^\dagger_{-\mathbf{q}\nu}\right)$$

where

$$V_{\mu\mu'}(\mathbf{q}\nu) = \sum_{|i|,|j|=1}^{3} \alpha_i^\mu \alpha_j^{\mu'} \sum_{\mathbf{k},\mathbf{G}} A_i(\mathbf{k}) A_j(\mathbf{k}+\mathbf{q}+\mathbf{G}) M_{\mathbf{k}}(\mathbf{q}\nu)$$

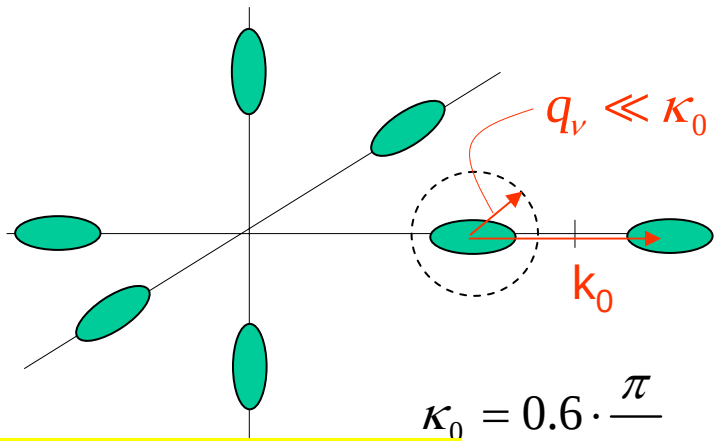
Envelope Functions

Long-wave acoustic phonons:

$$M_{\mathbf{k}}(\mathbf{q}\nu) \simeq \left(\hbar / 2\rho V \Omega_{\mathbf{q}\nu}\right)^{1/2} \sum_{\alpha\beta} D_{\alpha\beta}(\mathbf{k}) q_\alpha e_\beta(\mathbf{q}\nu)$$

$$D_{\alpha\beta}(\mathbf{k}) = \Xi_u \hat{k}_{i\alpha} \hat{k}_{j\beta} + \Xi_d \delta_{\alpha\beta}$$

Deformation potential constants



$$\kappa_0 = 0.6 \cdot \frac{\pi}{a_0}$$

$$\alpha_i^{A_1} = \alpha_{-i}^{A_1} \qquad \alpha_i^{B_2} = -\alpha_{-i}^{B_2} \qquad \mathbf{D}(\hat{k}_i) = \mathbf{D}(-\hat{k}_i)$$

Different parity intra-valley transitions are forbidden in all orders!

Smelyanskiy,Petukhov,Osipov PRB (2005),
Hasegawa, Phys. Rev. (1961); Castner, PRL (1963)

Probabilities of one-phonon decay

Weak transition: inter-valley Umklapp process:

$$W_w^{\mu\nu} \propto \left(\frac{\lambda}{\Delta_0}\right)^2 \frac{\Xi_u^2 \left(a_\parallel \kappa_0\right)^2}{\pi\hbar\rho u_t^2 a_\parallel^3 \left[1+\left(a_\parallel \kappa_0 / 2\right)^2\right]^6} \left(\frac{\omega_{\mu\nu} a_\parallel}{u_t}\right)^5$$

$\rightarrow$ $O(10^{-5})$

Smelyanskiy,Petukhov,Osipov PRB (2005)

Strong transition: intravalley process:

$$W_s^{\mu\nu} \propto \left(\frac{\lambda}{\Delta_0}\right)^2 \frac{\Xi_u^2}{\pi\hbar\rho u_t^2 a_\parallel^3} \left(\frac{\omega_{\mu\nu} a_\parallel}{u_t}\right)^3$$

Weak transitions involve umklapp processes
Strong transitions do not!

Hamiltonian of Li Donor in Si: Stress+Magnetic Field

$$H^{(0)} = H_0 + H_{ST} + H_Z + H_{SO}$$

Strain Zeeman Spin-Orbit

$$\langle \psi_\mu | H_0 | \psi_{\mu'} \rangle = \Delta_0 \delta_{\mu, A_1} \delta_{\mu', A_1} \hat{I}, \quad |\psi_\mu\rangle = \sum_{|i|=1}^{3} \alpha_i^\mu |i\rangle$$

Valley-orbit states

$$\langle \psi_\mu | H_{ST} | \psi_{\mu'} \rangle = \Xi_u \hat{I} \sum_{i=1}^{3} A_{\mu\mu'}^i e_{ii}, \qquad A_{\mu\mu'}^i = \alpha_i^\mu \alpha_i^{\mu'} + \alpha_{-i}^\mu \alpha_{-i}^{\mu'},$$

e_{ii} → Strain tensor

$$\langle \psi_\mu | H_Z | \psi_{\mu'} \rangle = g_\perp \mu_B H \sum_{i=1}^{3} n_i \hat{S}_i \left(\delta_{\mu\mu'} + \varepsilon A_{\mu\mu'}^i \right), \qquad \varepsilon = (g_\perp - g_\parallel) / g_\perp$$

Spin-Orbit Hamiltonian

Valley-orbit states: $|A_1\rangle, |E_\theta\rangle, |E_\varepsilon\rangle -$ even $\quad |T_x\rangle, |T_y\rangle, |T_z\rangle -$ odd

$$H_{SO} = i\hat{S}_z\left(\lambda'|E_\varepsilon\rangle\langle T_z| + \lambda|T_x\rangle\langle T_y|\right) + i\hat{S}_x\left(\lambda'|E_+\rangle\langle T_x| + \lambda|T_y\rangle\langle T_z|\right) + i\hat{S}_y\left(\lambda'|E_-\rangle\langle T_y| + \lambda|T_z\rangle\langle T_x|\right) + h.c.$$

$$|E_\pm\rangle = \frac{1}{2}\left(|E_\varepsilon\rangle \pm \sqrt{3}|E_\theta\rangle\right).$$

Spin-orbit interaction couples the states of different parity!

$$|\lambda'| = 0.056\ \mathrm{cm}^{-1},$$
$$|\lambda| = 0.021\ \mathrm{cm}^{-1}.$$

Origin of the spin-orbit coupling Si host!

Watkins and Ham, Phys. Rev. (1970)

Chiral Symmetry

The Hamiltonian $H^{(0)}$ of shallow donors in Si which includes:

- Valley-orbit splitting
- Arbitrary biaxial strain
- Zeeman term with B parallel to one of the crystal axes
- Spin-orbit interaction

possess chiral symmetry:

Chirality operator $\hat{Z} = \hat{R}_z(\pi)\hat{\sigma}_z, \quad \hat{R}_z(\pi)$ - Rotation through angle π around z-axis

$$[H^{(0)}, \hat{Z}] = 0$$

Eigenstates of $H^{(0)}$:

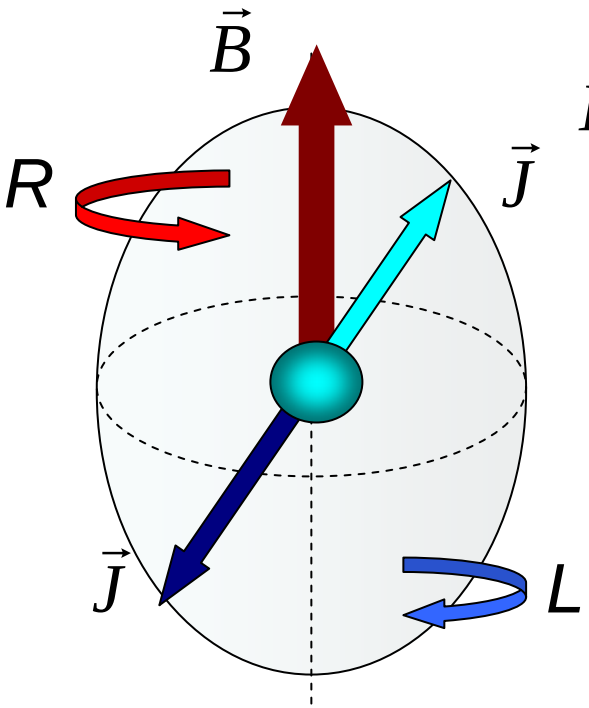
$$\left|\Phi_\sigma^{(0)}\right\rangle = \left|\chi_\sigma\right\rangle\left(c_z\left|T_{2z}\right\rangle + c_\theta\left|E_\theta\right\rangle + c_\varepsilon\left|E_\varepsilon\right\rangle + c_{A_1}\left|A_1\right\rangle\right) + \left|\chi_{-\sigma}\right\rangle\left(c_x\left|T_{2x}\right\rangle + c_y\left|T_{2y}\right\rangle\right)$$

are also eigenstates of $\hat{Z}$:

$$\hat{Z}\left|\Phi_\pm^{(0)}\right\rangle = \pm\left|\Phi_\pm^{(0)}\right\rangle\left|\chi_\sigma\right\rangle, \ \pm \equiv R, L$$

Chiral Symmetry

Example: Particle in an ellipsoidal potential



$$H = -\frac{1}{2}\Delta + \frac{1}{2}\left(\omega_x^2 x^2 + \omega_y^2 y^2 + \omega_z^2 z^2\right) + \lambda \vec{L}\vec{S} + g\mu_B B\sigma_z$$

$\vec{J} = \vec{L} + \vec{S}$ is not conserved

J_z is not conserved

$Sign(J_z)$ is a good quantum number!

$Sign(J_z) = \pm 1 -$ Chirality

$\mathbf{B} \;||\; \mathbf{F} \;||\; \hat{z}$

Spin-flip transitions: Compressive stress

even

odd

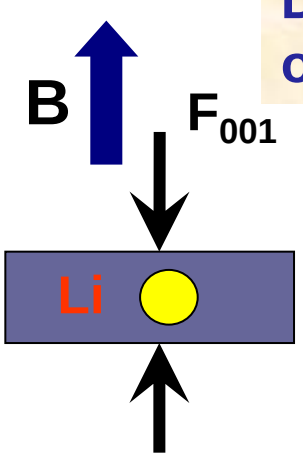
$$|R\rangle = |\chi_\downarrow\rangle\left(c^d_{A_1}|A_1\rangle + c^d_\theta|E_\theta\rangle + c^d_\varepsilon|E_\varepsilon\rangle + c^d_z|T_z\rangle\right) + |\chi_\uparrow\rangle\left(c^d_x|T_x\rangle + c^d_y|T_y\rangle\right)$$

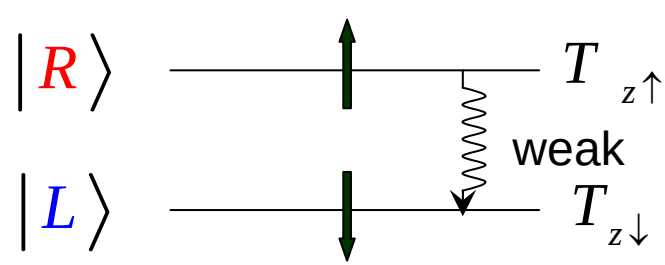
$$|L\rangle = |\chi_\downarrow\rangle\left(c^u_x|T_x\rangle + c^u_y|T_y\rangle\right) + |\chi_\uparrow\rangle\left(c^u_{A_1}|A_1\rangle + c^u_\theta|E_\theta\rangle + c^u_\varepsilon|E_\varepsilon\rangle + c^u_z|T_z\rangle\right)$$

odd

even

Due to chirality (parity) selection rule spin can decay only via weak inter-valley umklapp process!

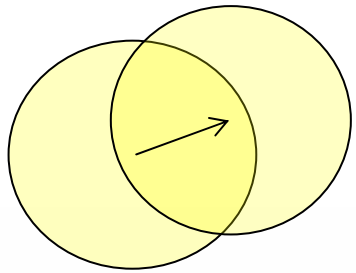




Zeeman doublet

T_1 **long!**

Li atom displacement off the Td site



$\delta \vec{r}$

$$U = \frac{U_0}{8\pi a^3} e^{-\frac{r}{a}}$$

$$\delta U = \frac{U_0}{8\pi a^3}\left(e^{-\frac{|\vec{r}|}{a}} - e^{-\frac{|\vec{r}-\delta\vec{r}|}{a}}\right) = U(r)\,\frac{\delta\vec{r}\cdot\vec{r}}{a\,r}$$

$$\mathcal{V}_j = \frac{1}{\pi a_{\parallel} a_{\perp}^2}\int U(r)\cos(\kappa_0 x_j)\sin(\kappa_0 x_j)\,\frac{x_j\,\delta x_j}{a\,r}\,d^3r$$

$$1.76\ \text{meV} = \Delta_0 = \frac{1}{\pi a_{\parallel} a_{\perp}^2}\int U(r)\,(2\cos^2(\kappa_0 z))\,d^3r$$

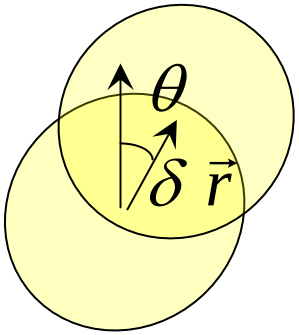
In the limit : $a \ll \frac{\pi}{\kappa_0} \ll a_{\parallel}, a_{\perp}$

$$\mathcal{V}_j = \Delta_0\,\kappa_0\,\delta x_j$$

Li-displacement Dipole Potential

$$\Psi^{\mu}(\vec{r}) = \sum_{j=1}^{6} \alpha_j^{\mu} F_{\mu}^{j}(\vec{r}) e^{i\vec{k}_j \vec{r}} u_{\vec{k}_j}(\vec{r})$$

Very weak short-range dipole potential due to the small off-site displacement of Li donor



$$x\sin(k_0 x)\cos(k_0 x)$$
$$y\sin(k_0 y)\cos(k_0 y)$$
$$z\sin(k_0 z)\cos(k_0 z)$$

Total Hamiltonian $H = H^{(0)} + H_d$

$$\langle \psi_{\mu} | H_d | \psi_{\mu'} \rangle = 2iV \sum_{j=1}^{3} \left(\alpha_j^{\mu} \alpha_{-j}^{\mu'} - \alpha_{-j}^{\mu} \alpha_j^{\mu'} \right) \delta r_j / \delta r$$

Dipole term H_d violates chiral symmetry!

$$\langle L | H_d | R \rangle \propto V \sin\theta$$

Transition rates and kinetics

Spin lifetime

$$\tau_{T_{2z\uparrow}}^{-1} \simeq \tau_{A_1}^{-1}\left(\frac{\lambda\lambda' V_d \sin\theta_d}{\Delta_0^3}\right)^2 \frac{41 v^3}{x^8}\left(\left(\frac{x}{3}\right)^3 + v^2\right)$$

Dipole dominates at moderate stress

$$\tau_{T_{2z\uparrow}}^{-1} \simeq \tau_{A_1}^{-1}\xi_u\left(\frac{\lambda\lambda'\Delta_0 a_{\parallel}}{\hbar u_{\perp}}\right)^2 \frac{7 v^5}{3\left(x^2 - v^2\right)^2}$$

Umklapp dominates at large stress

$$\tau_{A_1}^{-1} \sim 10^{-12}\,\mathrm{sec}, \quad v = g_{\perp}\mu_B B/\Delta_0, \quad x = \Xi_u\left(S_{12} - S_{11}\right)F_{001}/6\Delta_0,$$

$\xi_u \simeq 5\cdot 10^{-5}$ – intervalley overlap factor, $u_{\perp}$ – transverse speed of sound

Multi-level kinetics

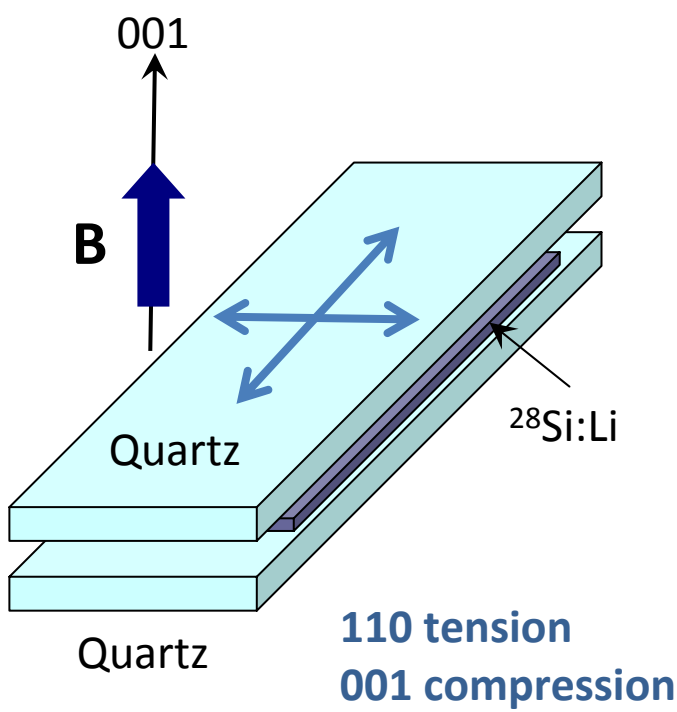
Master equations

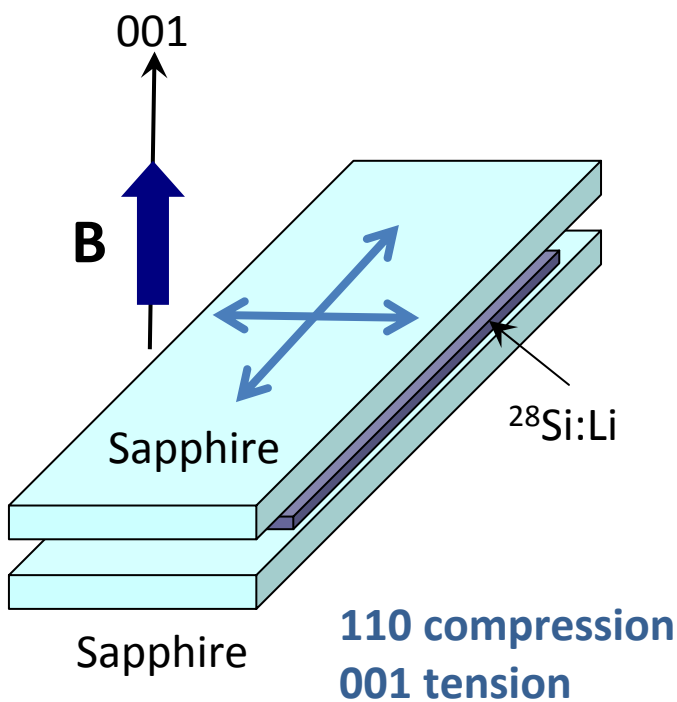
$$\dot{\rho}_n = \sum_{m=1}^{12} L_{nm}\rho_m, \quad \rho_n = \left\langle \Phi_n \middle| \hat{\rho} \middle| \Phi_n \right\rangle$$ - State population

$$L_{nm} = W_{m\to n} - \delta_{nm}\sum_{k=1}^{12} W_{n\to k}$$ - Relaxation operator

Geometry of the sandwiches used in ESR experiments (Steve Lyon , and Alexei Tyryshkin) using thermal compression and tension in a silicon sample

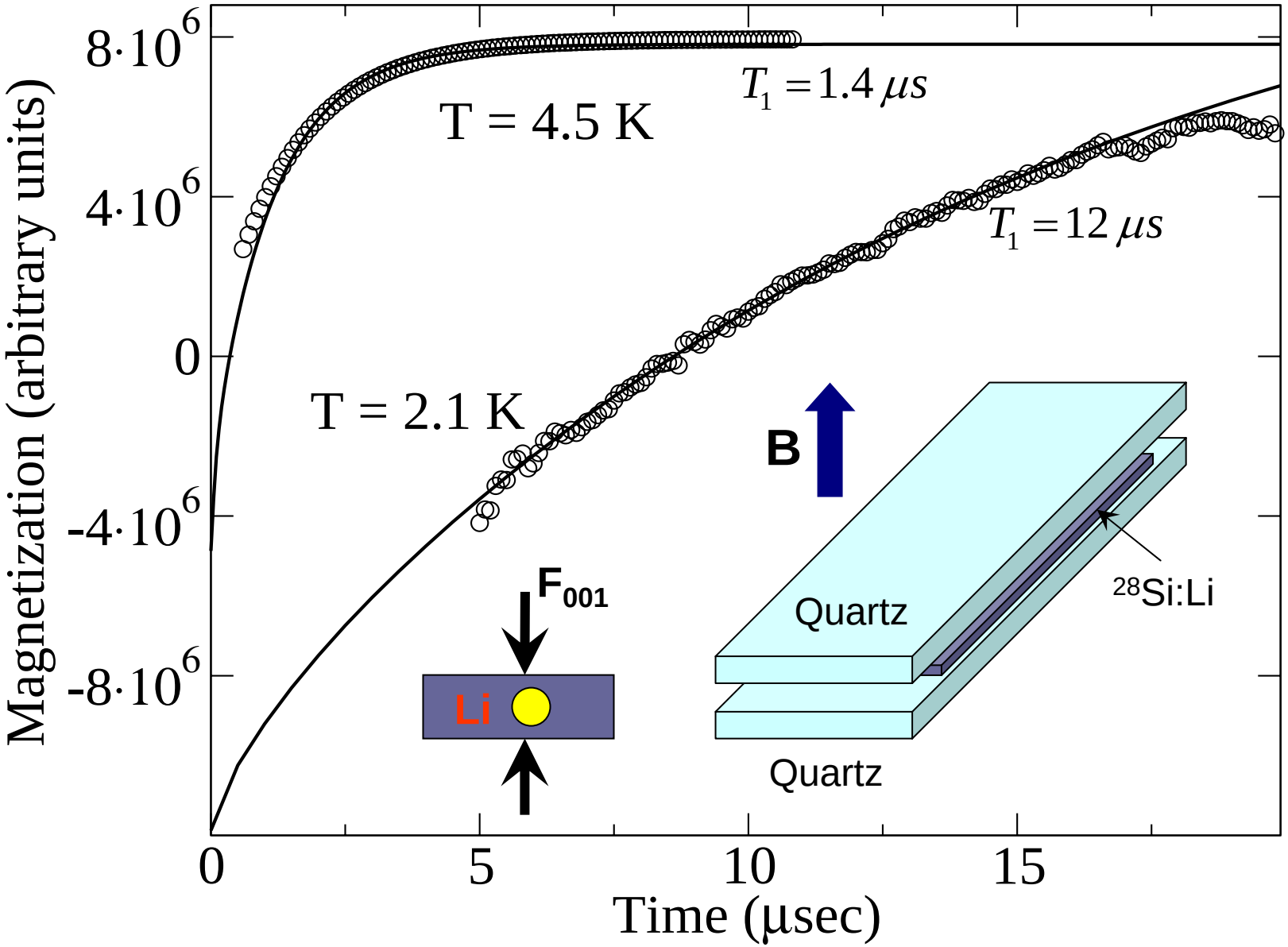
- Quartz sandwich us used to form compressive 001 stress 9 MPa
- Sapphire sandwich is used to create a 001 tension of 30 MPa



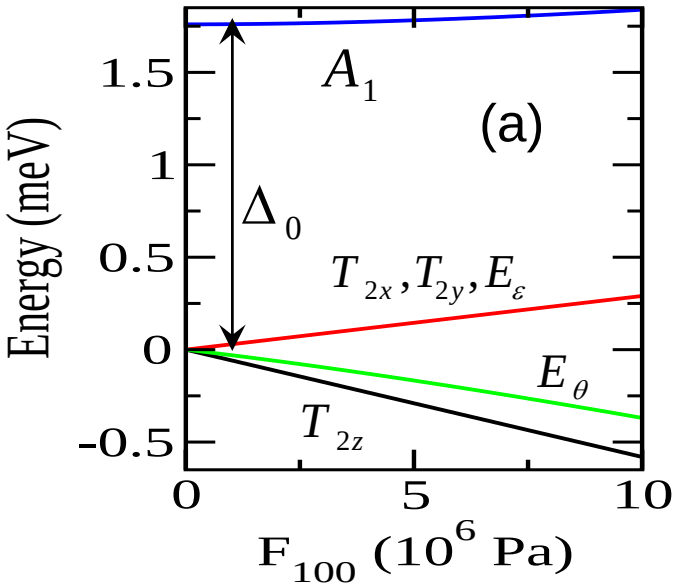


Inversion recovery: Comparison with Experiment

V.N. Smelyanskiy *et al*, arXiv:0807.3928v1

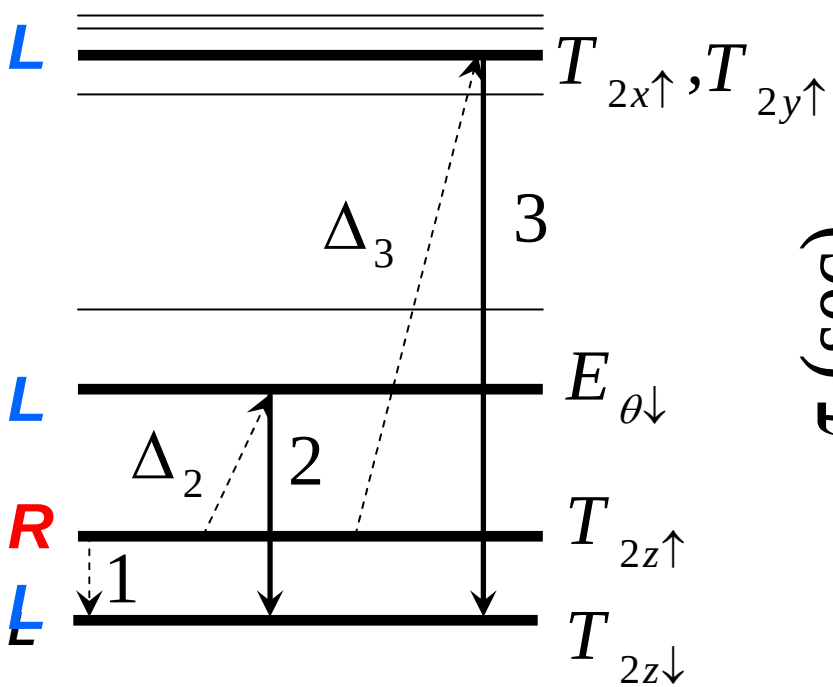


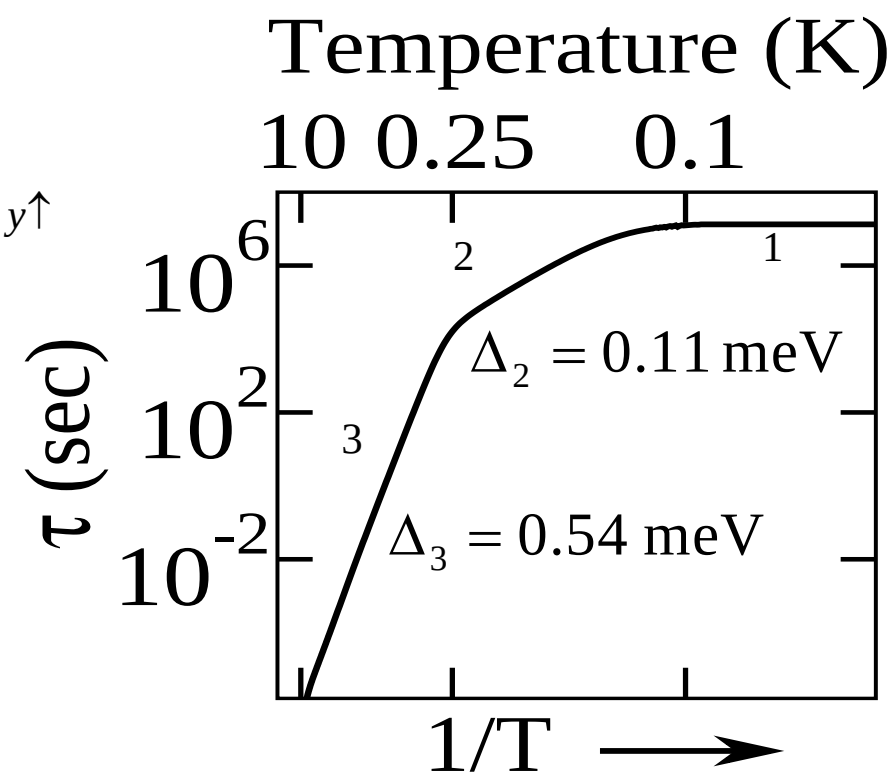
Activation energies



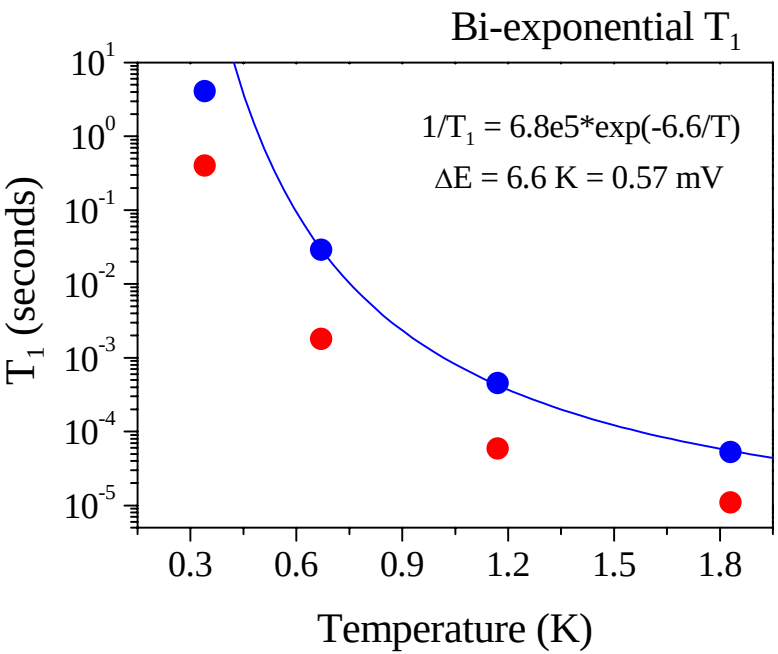
Under compressive stress the Li spin decays mainly via activation transitions that are frozen below 0.3 K

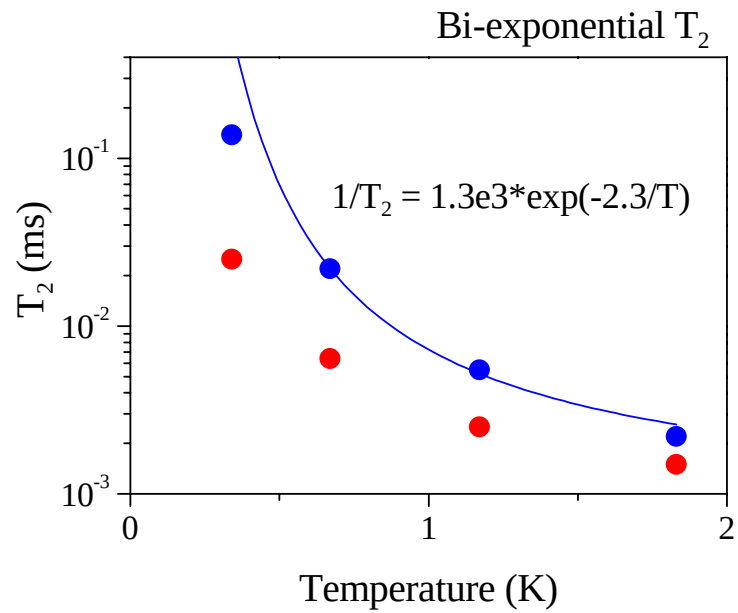
V.N. Smelyanskiy *et al*, arXiv:0807.3928v1





Quartz Sandwich

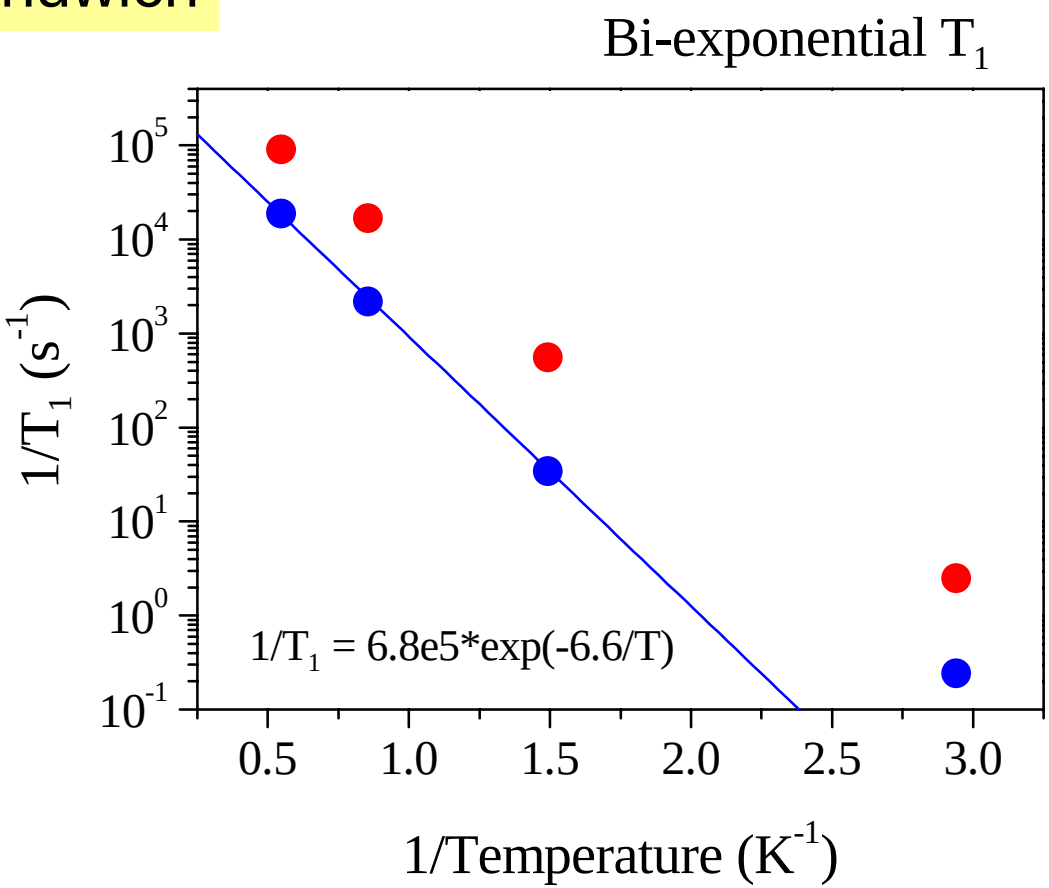




- Temperature dependence of T_1 and T_2 in ^{28}Si:Li under compressive stress and magnetic field applied along [001].

- Two T_1 (or T_2) times shown for each temperature

Quartz Sandwich



The same T_1 temperature dependence from above is plotted now in Arrhenius coordinates. It is clearly seen that the three high-temperature points form a good line, but the last point (0.34K) deviates substantially.

Electrical Manipulation of Li spin Qubits

What to expect for Li?

- Effective mass impurity
- Quasi-degenerate ground state
- Larger induced dipole moment
- Possibility of controlling levels of ground-state manifold with electric field
- Enhanced role of the spin-orbit interaction
- Coupling of the electro-dipole and spin-flip transitions

Previous work: Stark Effect for P and Sb in Si

Experiment: Bradbury et al., PRL (2006)

Theory:
Rahman *et al.*, PRL (2007)
Landsbergen *et al*., NP (2008)
Friesen, PRL (2005)
Smit *et al.* , PRB (2004)
Kettle *et al.* PRB (2003)
Martins *et al.*, PRB (2004)
Debernardi *et al.,* PRB (2006)
Calderon *et al.*, PRL (2006)
...
Classic Quadratic Stark Effect:
Privman, PRB (1980); Ivanov, PRB (1997), and many more ...

Rayleigh-Ritz method

The orthonormal basis relies on normalized hydrogenic wavefunctions and two variational parameters: α, β

$$\varphi_{nlm}(x,y,z) = \left(\frac{\beta}{\gamma}\right)^{\frac{1}{4}} \psi_{nlm}\left(x, y, \left(\frac{\beta}{\gamma}\right)^{\frac{1}{2}} z\right)$$

$$\psi_{nlm}(x,y,z) = R_{nl}(r,\alpha) Y_{lm}(\theta,\varphi)$$

Using the dimensionless Hamiltonian from Kohn-Luttinger and the Stark shift term V, we construct a finite-dimensional Hamiltonian from orthonormal functions ϕ_j:

$$H_{n'l'm'nlm} = \int \varphi^*_{n'l'm'}(x)\ H\ \varphi_{nlm}(x)\ d^3x$$

$$H = H_0 + V$$

Higher Donor Excited States, Faulkner, 1969

Selection rules for mixing states

Matrix elements in H_0 are nonzero only if certain selection rules are obeyed:

$$m_1 = m$$

$$l_1 = l + 2i, \qquad i = 0,1,2,3,\ldots$$

Likewise, matrix elements in V are nonzero only for selection rules, depending on the field direction.

For the z-direction of a valley

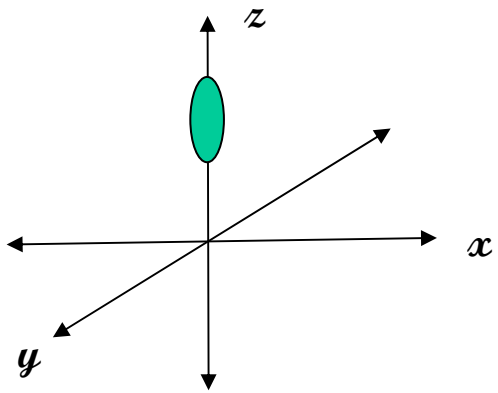
$$m' = m$$

$$l' = l + 1$$

For the x-direction of a valley:

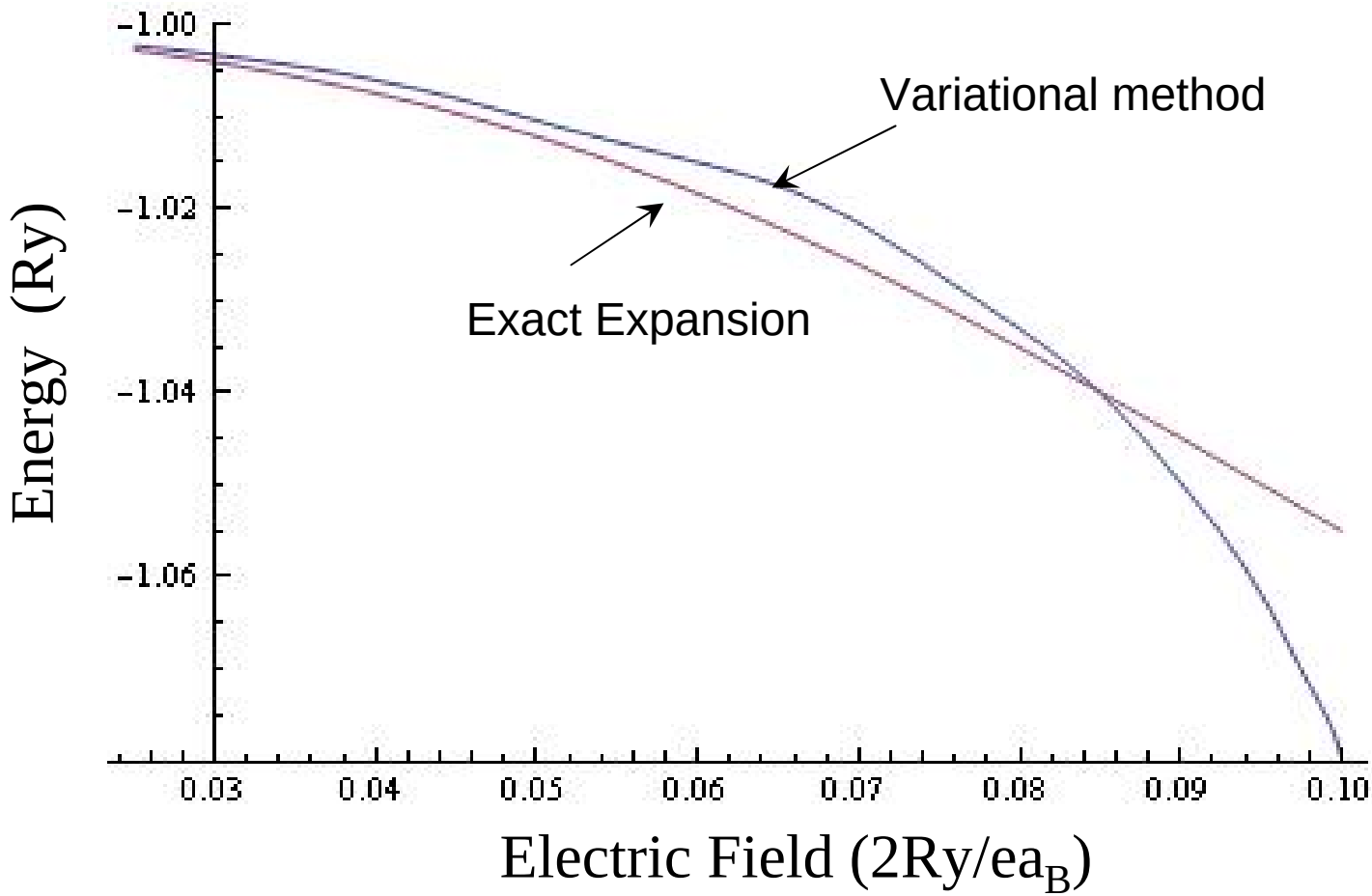
$$m' = m \pm 1$$

$$l' = l + 1$$



$$n = l + 1, l + 2, \ldots, l + l_{\max} + 1$$

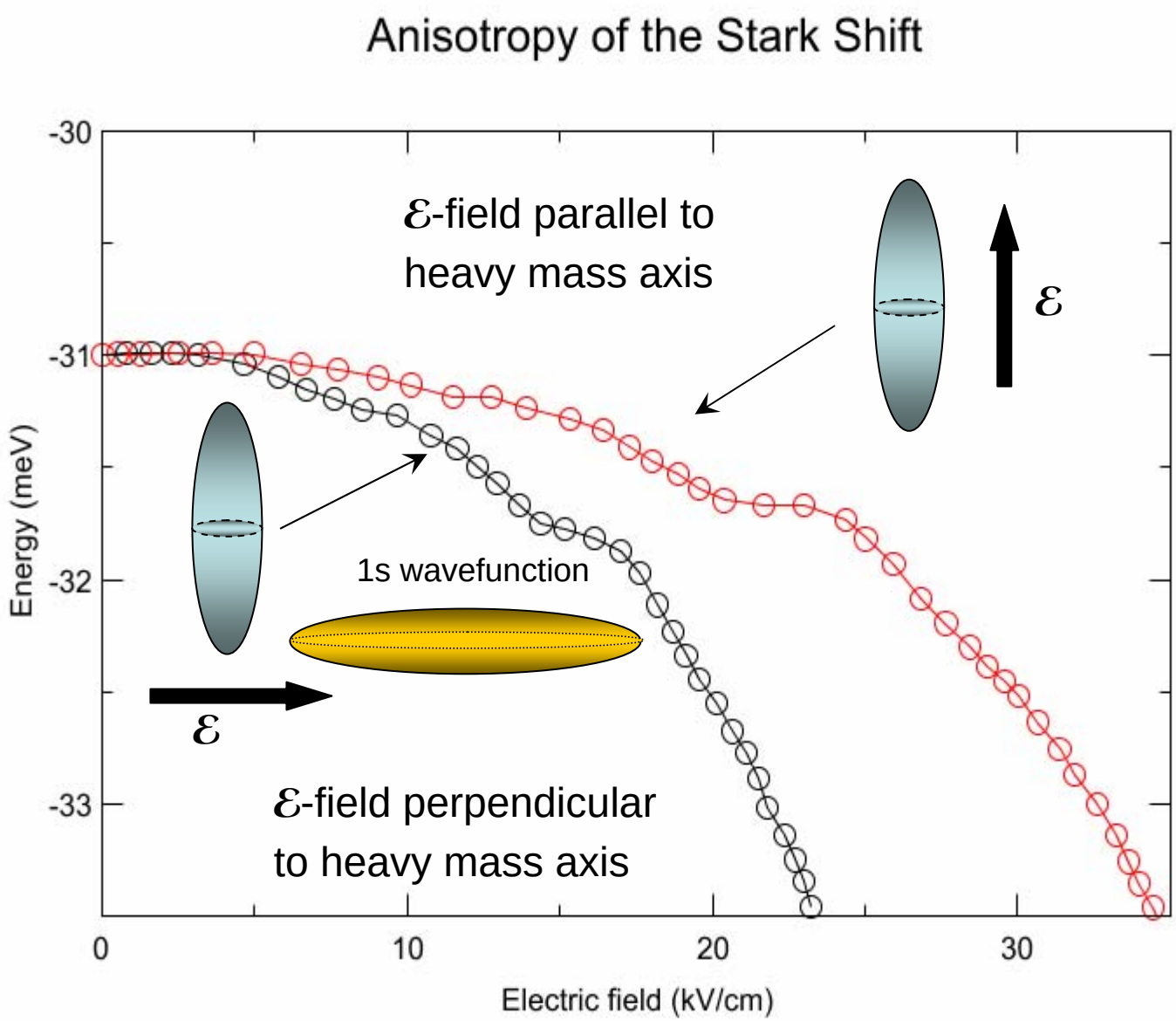
Quadratic Stark Effect for a Hydrogen Atom



Exact expansion
Privman, PRB (1980)

$$E = -Ry \sum_n e_n \left(\frac{\mathcal{E}}{8Ry / ea_B} \right)^n, \quad e_1 = 1, \quad e_2 = 72, \quad e_3 = 28430, \ldots$$

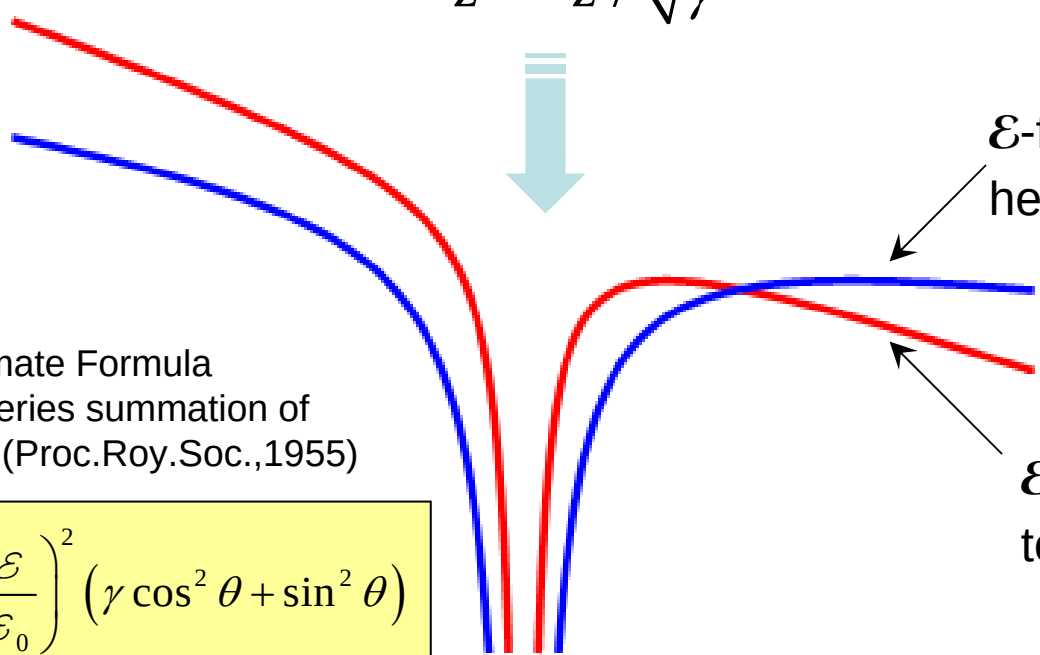
Quadratic Stark Effect for EMA Donors in Si



Anisotropy of Quadratic Stark Effect for EMA Donors in Si

$$H = -\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \gamma \frac{\partial^2}{\partial z^2}\right) - \frac{2}{r} - e\mathcal{E}\mathbf{r}$$

$$z' = z / \sqrt{\gamma}$$



$\mathcal{E}$-field parallel to heavy mass axis

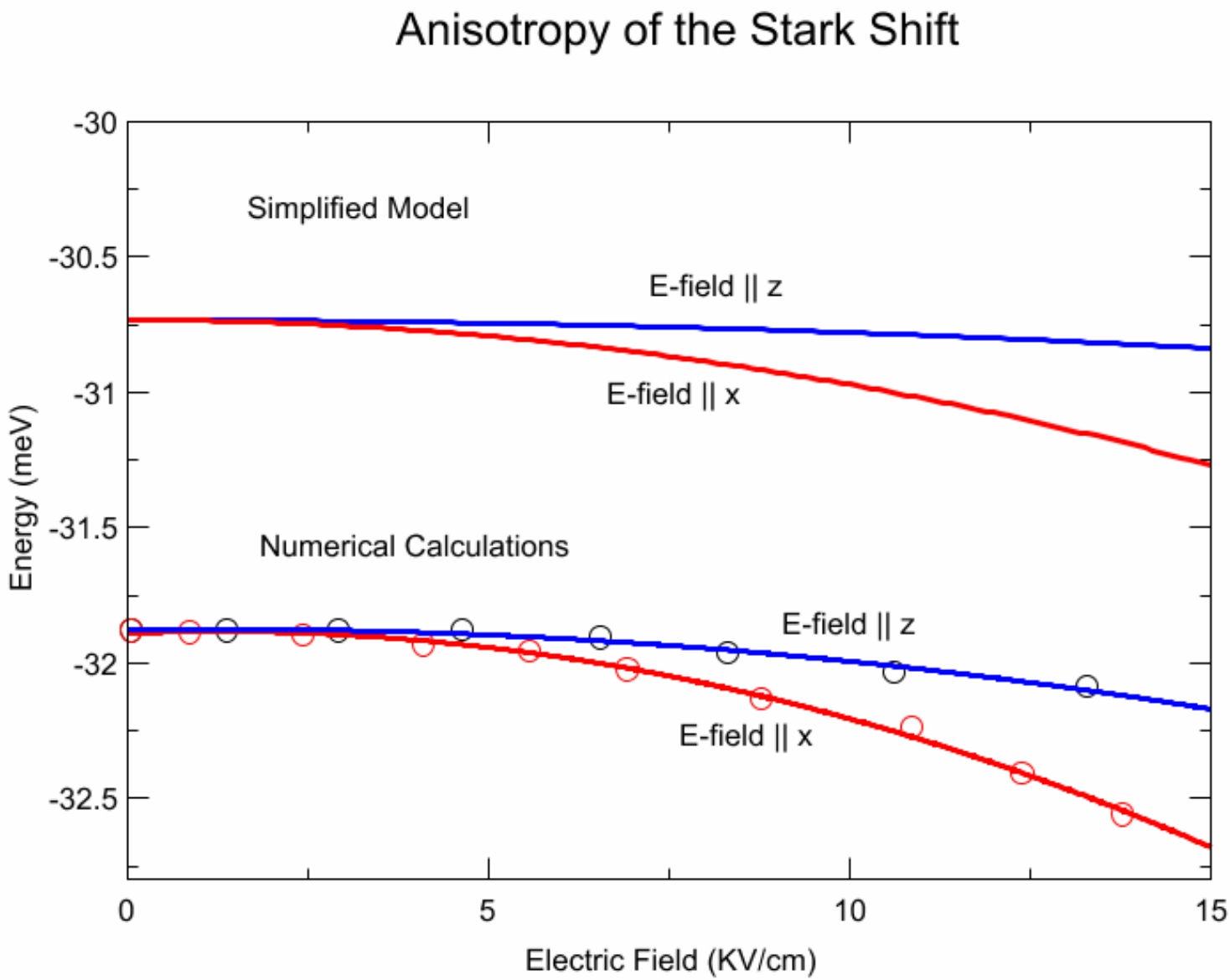
$\mathcal{E}$-field perpendicular to heavy mass axis

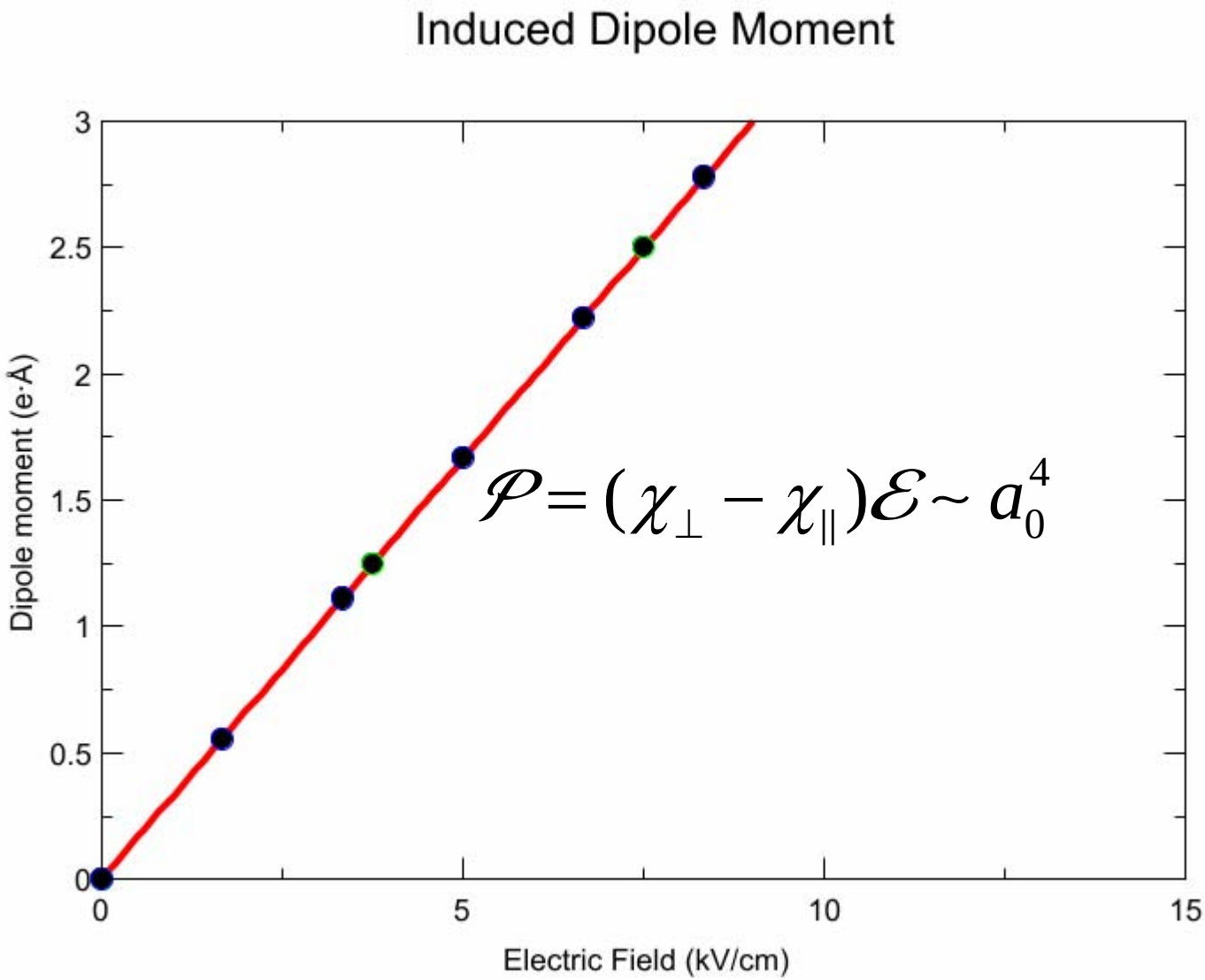
Approximate Formula
using infinite series summation of
Dalgarno & Lewis (Proc.Roy.Soc.,1955)

$$\Delta E \approx -\frac{9}{2} Ry^* \left(\frac{\mathcal{E}}{\mathcal{E}_0}\right)^2 \left(\gamma \cos^2\theta + \sin^2\theta\right)$$

$$\mathcal{E}_0 = 2Ry^* / ea_0^*, \quad \gamma = m_\perp / m_\parallel \approx 0.2$$

Stark shift anisotropy leads to a coupling of Li spin-flip transition and electric dipole moment!


Anisotropy of the Stark Shift
Simplified Model
E-field || z
E-field || x
Numerical Calculations
E-field || z
E-field || x
-30
-30.5
-31
-31.5
-32
-32.5
Energy (meV)
0
5
10
15
Electric Field (KV/cm)


Induced Dipole Moment
Dipole moment (e·Å)
Electric Field (kV/cm)
0
0.5
1
1.5
2
2.5
3
0
5
10
15
$\mathcal{P} = (\chi_\perp - \chi_\parallel)\mathcal{E} \sim a_0^4$

Valley-Orbit Effects

$$E_z = E_z^{(0)} - (1/2)(\chi_\parallel \mathcal{E}_\parallel^2 + \chi_\perp \mathcal{E}_\perp^2)$$

$$\chi_\parallel = 2\sum_{n\geq 2} \frac{|\langle 1,s|e\cdot z|n,p_z\rangle|^2}{E_{n,p_0} - E_{1,s}}$$

$$\chi_\perp = 2\sum_{n\geq 2} \frac{|\langle 1,s|e\cdot x|n,p_x\rangle|^2}{E_{n,p_\pm} - E_{1,s}}$$

Valley-Orbit Hamiltonian Matrix:

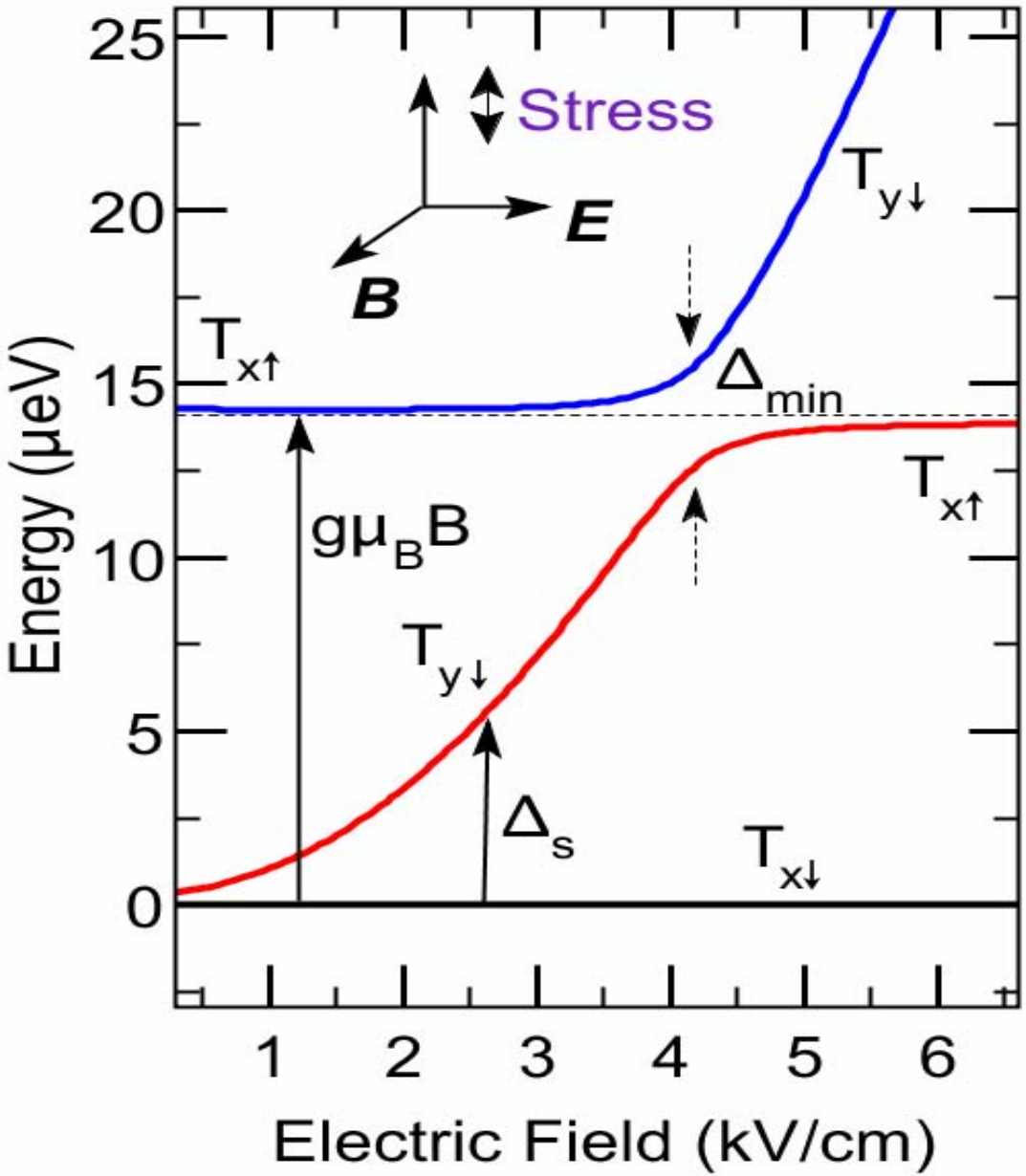
$$H_{ij}^{(0)} = E_i(\mathcal{E})\delta_{ij} + (1/6)\Delta_c(1-\delta_{ij})$$

Dipole-Moment Matrix:

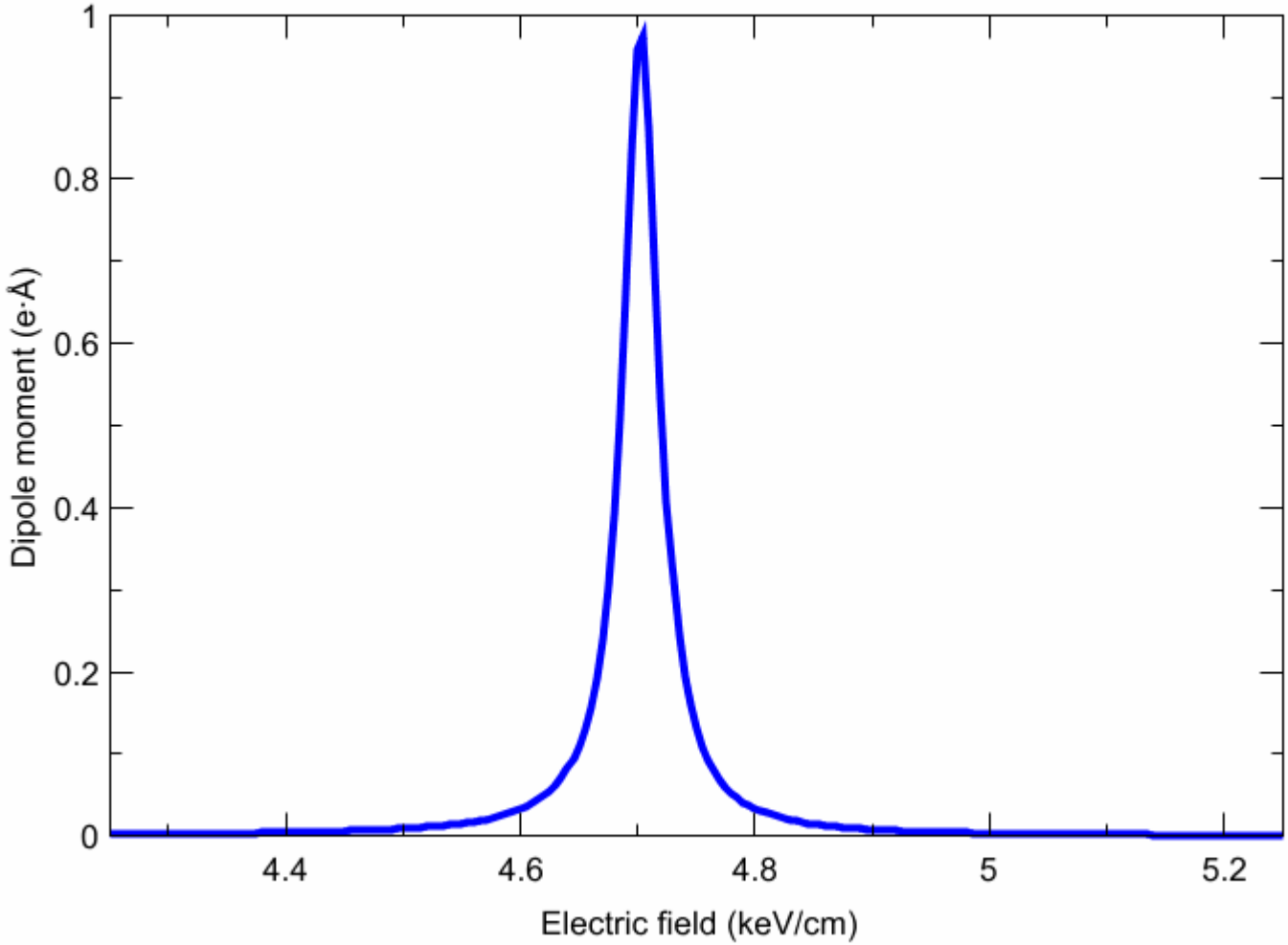
$$\vec{\mathcal{P}}_{ij} = \chi_\parallel \vec{\mathcal{E}}\delta_{ij} + (\chi_\perp - \chi_\parallel)\left[\vec{\mathcal{E}}\delta_{ij} - \vec{n}_i(\vec{n}_j\cdot\vec{\mathcal{E}})\right]$$

$$\boxed{H_{tot} = H^{(0)} + H_{so} + H_{Zeeman}}$$

Three-level System



Transitional Dipole Moment



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Stark Tuning of Donor Electron Spins in Silicon

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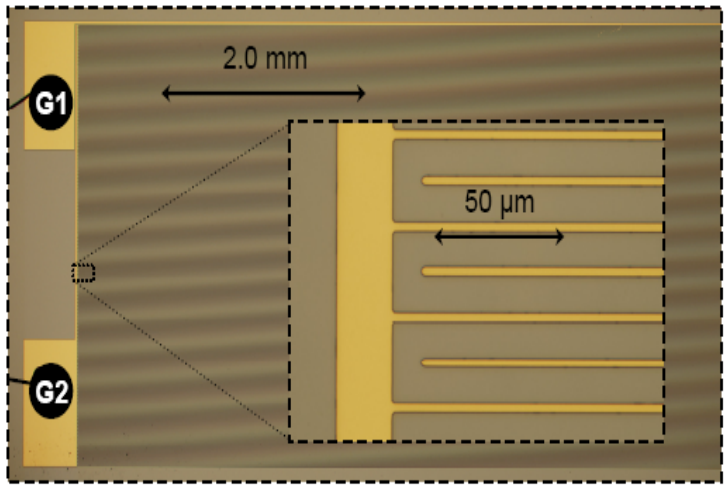
We report Stark shift measurements for ^{121}Sb donor electron spins in silicon using pulsed electron spin resonance. Interdigitated metal gates on a Sb-implanted ^{28}Si epilayer are used to apply the electric fields. Two quadratic Stark effects are resolved: a decrease of the hyperfine coupling between electron and nuclear spins of the donor and a decrease in electron Zeeman g factor. The hyperfine term prevails at magnetic fields of 0.35 T, while the g factor term is expected to dominate at higher magnetic fields. We discuss the results in the context of the Kane model quantum computer.

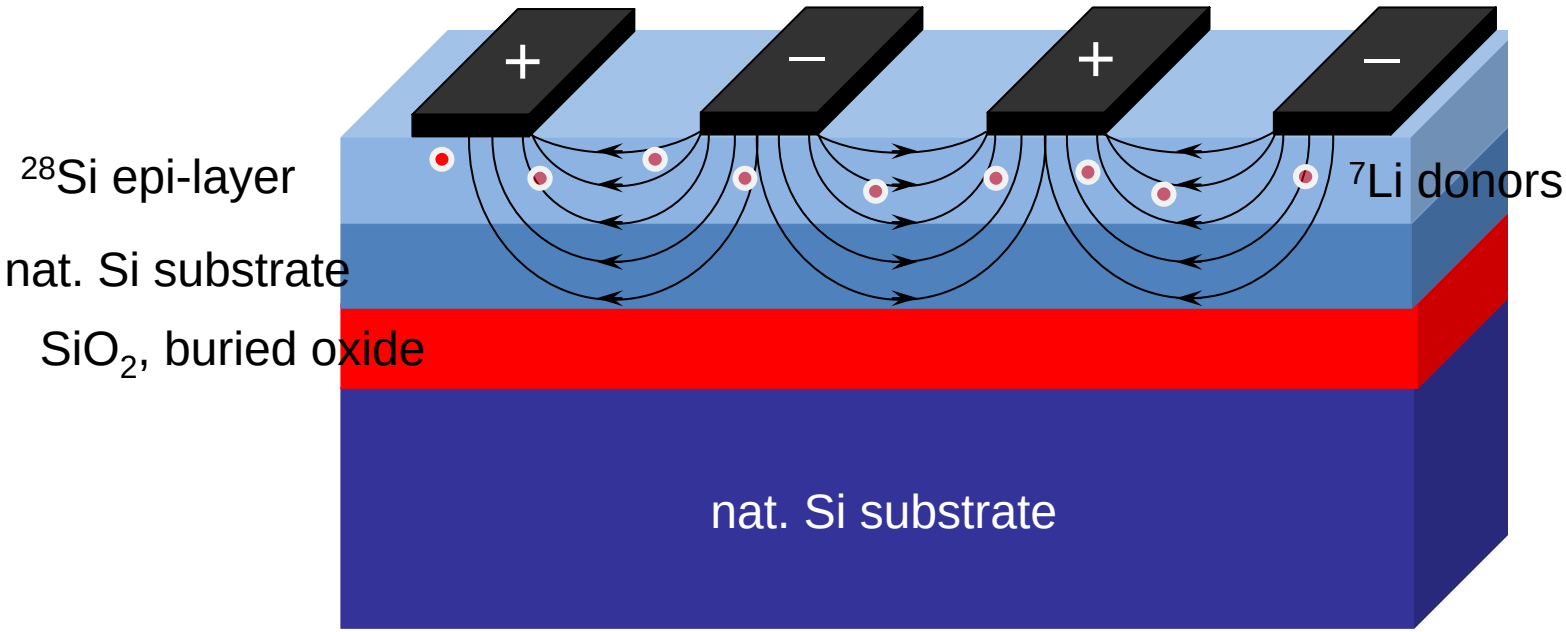
$$\Delta a(E) = \eta_a \cdot a \cdot E^2, \quad \eta_a = -3.7\cdot 10^{-3} \text{ (in } \mu m^2/V^2)$$

$$\Delta g(E) = \eta_g \cdot g \cdot E^2, \quad \eta_g = -1\cdot 10^{-5} \text{ (in } \mu m^2/V^2)$$



- substitutional donors (P, As, Sb) vs. interstitial donor (Li)

Lithium doped ^{28}Silicon – on – Insulator (^{28}SOI) for Stark shift experiments



•Layout of the device for Stark shift experiment. Interdigitated gate structure on surface of ^{28}Si epi-wafer produces a dipolar electrical field pattern across the implanted ^{7}Li donors.

• Electrical fields of ~1000 V/cm to 4000 V/cm have been achieved in earlier experiments on Stark shift in Sb donors and are predicted to yield relatively large electrical dipole moments of order 0.1 nm

Summary

Si:Li Predictions:

- Large T_1 ~ 1s and T_2 spin times ~ 1ms for T<0.3 K (confirmed)
- Possibility to manipulate spin qubits electrically due to unique electronic structure
- Enhanced role of the spin-orbit interaction
- Large induced dipole moment associated with spin-flip transition
- Large induced g-factor shift

What to explore?

- E-field induced g-factor shift (expected to be 3 orders of magnitude larger than in other donors)
- Possibility of Li spin coupling with a resonator cavity