28.04.2010 # 7

Today

- Selection rules allowing for circularly polarized light to create spin polarized carriers
- Dynamic nuclear polarization
- Carrier spins coupled to localized impurity spins

Recall from the effective mass approx (lecture #5)

$$\psi_{cb}(\vec{r}) = \underbrace{\chi_{cb}(\vec{r})}_{\text{envelope function}} \underbrace{u_{cb}(\vec{r})}_{\text{Bloch function of local extrema}}$$

Optical transition rates:

$$\propto |\langle \psi_f | e\vec{r}\cdot\vec{E} | \psi_i \rangle|^2 \delta(E_f - E_i - \hbar\omega)$$

$$\propto |\langle \psi_{cb} | e\vec{r}\cdot\vec{E} | \psi_{vb} \rangle|^2$$

$$\propto \left| \int_{\text{all space}} \chi_{cb}^*(\vec{r})\, u_{cb}^*(\vec{r}) \left(e\vec{r}\cdot\vec{E}\right) \chi_{vb}(\vec{r})\, u_{vb}(\vec{r})\, d\vec{r} \right|^2$$

$\chi(\vec{r})$ varies slowly over unit cell
$u(\vec{r})$ varies over unit cell & is periodic
↳ separable integral

$$\propto \left| \sum_{\text{unit cells}} \int_{\text{unit cell}} \chi_{cb}^*(\vec{r})\, u_{cb}^*(\vec{r}) \left(e\vec{r}\cdot\vec{E}\right) \chi_{vb}(\vec{r})\, u_{vb}(\vec{r})\, d\vec{r} \right|^2$$

$$\propto \left| \int_{\text{all space}} \chi_{cb}^*(\vec{r})\, \chi_{vb}(\vec{r})\, d\vec{r} \cdot \int_{\text{unit cell}} u_{cb}^*(\vec{r}) \left(e\vec{r}\cdot\vec{E}\right) u_{vb}(\vec{r})\, d\vec{r} \right|^2$$

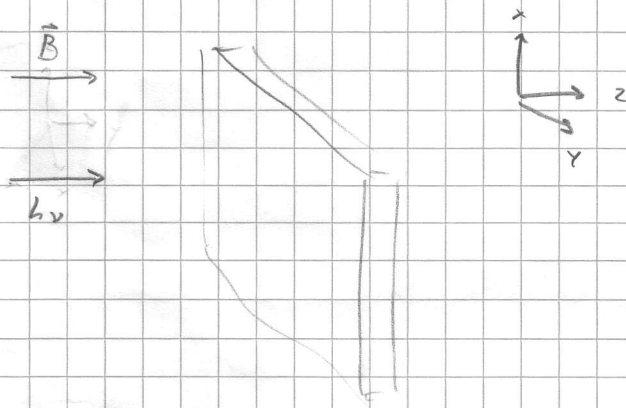
$$\propto \left| \int_{\text{all space}} \phi_{cb}^*(z)\, \phi_{vb}(z)\, e^{i(\vec{k}_{\parallel,vb} - \vec{k}_{\parallel,cb})\cdot\vec{r}}\, d\vec{r}\, \langle s | e\vec{r}\cdot\vec{E} | p \rangle \right|^2$$

$$\propto \left| \int \phi_{cb}^{*}(z)\, \phi_{vb}(z)\, dz\, \langle s | e\hat{r}\cdot\vec{E} | p \rangle \right|^2 \delta\left(\vec{k}_{\perp,vb} - \vec{k}_{\perp,cb}\right)$$

↑ transitions are vertical (no change in momentum $k_\perp$)

Relative transition amplitudes can be solved by symmetry

∴ Calculating optical transitions is all about finding $\left| \langle s | e\hat{r}\cdot\vec{E} | p \rangle \right|^2$ ←



Lin pol light

$\vec{E} = E(t)\,\hat{x}$ or $E(t)\,\hat{y}$

Circ. pol. light (prop along $\hat{z}$)

$\vec{E} = \frac{E(t)}{\sqrt{2}}(\hat{x} + i\hat{y})$ (R) (σ^-)

or

$\vec{E} = \frac{E(t)}{\sqrt{2}}(\hat{x} - i\hat{y})$ (L) (σ^+)

Can write in terms of spherical harmonics:

$e\hat{r}\cdot\vec{E}_{\sigma^+} \propto \langle 1,-1 | \hat{r} \rangle$ ⟸

$e\hat{r}\cdot\vec{E}_{\sigma^-} \propto \langle 1,+1 | \hat{r} \rangle$

This will simplify finding relative transition amplitudes:

E.g. $\left| {}_{CB}\langle \tfrac{1}{2}, +\tfrac{1}{2} | \; e\hat{r}\cdot E_{\sigma^+} \; | \tfrac{3}{2}, -\tfrac{1}{2} \rangle_{LH} \right|^2$

$\left| \langle \uparrow | \langle 0,0 | \langle 1,-1 | \hat{r} \rangle \left(\sqrt{\tfrac{1}{3}} |1,+1\rangle |\downarrow\rangle - \sqrt{\tfrac{2}{3}} |1,0\rangle |\uparrow\rangle \right) \right|^2$

$\left| \langle 0,0 | \langle 1,-1 | 1,0 \rangle \cdot \left(-\sqrt{\tfrac{2}{3}}\right) | \hat{r} \rangle \right|^2$

$\propto 0$

E.g.

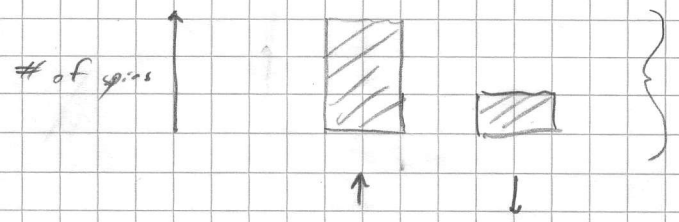
$$\left| {}_{CB}\left\langle \tfrac{1}{2}, -\tfrac{1}{2} \right| e\vec{r}\cdot\vec{E}_{\sigma-} \left| \tfrac{1}{2}, +\tfrac{1}{2} \right\rangle_{SO} \right|^2$$

$$\left| \langle \downarrow | \langle 0,0 | \langle 1,+1 | F \rangle \left(\sqrt{\tfrac{2}{3}} |1,+1\rangle|\downarrow\rangle + \sqrt{\tfrac{1}{3}} |1,0\rangle|\uparrow\rangle \right) \right|^2$$

$$\left| \langle 0,0 | \langle 1,+1 | 1,+1 \rangle \left(\sqrt{\tfrac{2}{3}} \right) |F\rangle \right|^2$$

$$\propto \quad \tfrac{2}{3}\gamma \quad \leftarrow \text{some constant}$$

With above calculations you can show that exciting w/ circ. pol. light you can create a 3:1 spin-up to spin-down (or vice-versa) imbalance in the conduction band. This means you have a CB spin polarization of 50%.



Can use the same principles to measure spin polarizations...

Faraday Rotation

Probe ⟶ linearly pol. light

Probe polarization will be rotated by angle

$$\theta_F \propto \left(E_{\sigma+} - E_{\sigma-}\right) \propto \left(n_\uparrow - n_\downarrow\right) \propto \langle S_z \rangle$$

Spin in a Magnetic Field

$$\vec{M}_s = -\frac{g\mu_B}{\hbar}\vec{S} \longrightarrow \pm\frac{\hbar}{2}$$

Hamiltonian:

$$H = -\vec{M}\cdot\vec{B}$$

$$H_s = \frac{g\mu_B}{\hbar}\vec{B}\cdot\vec{S} = \frac{g\mu_B}{\hbar}BS_z$$

eigen states : $|\uparrow\rangle$, $|\downarrow\rangle$

eigen values : $+\frac{1}{2}g\mu_B B$, $-\frac{1}{2}g\mu_B B$

Time dependence:

$$\frac{d\vec{S}}{dt} = \frac{i}{\hbar}[H_s, \vec{S}] + \frac{\partial\vec{S}}{\partial t} \longrightarrow 0$$

$$\frac{d\vec{S}}{dt} = i g\mu_B B [S_z, \vec{S}] \qquad \rightarrow \quad [S_\alpha, S_\beta] = i\epsilon_{\alpha\beta\gamma}\hbar S_\gamma$$

$$\therefore \quad \frac{d\vec{S}}{dt} = \frac{g\mu_B}{\hbar}\vec{B}\times\vec{S}$$

$$\frac{d\vec{S}}{dt} = \vec{\omega}_L \times \vec{S}$$

$$\boxed{\frac{d}{dt}\langle\vec{S}\rangle = \vec{\omega}_L \times \langle\vec{S}\rangle}$$

Real systems have energy relaxation time (T_1) & decoherence time (T_2).

$$\therefore \quad \frac{d}{dt}\langle\vec{S}\rangle = \vec{\omega}_L\times\langle\vec{S}\rangle - \frac{(\langle\vec{S}\rangle\cdot\vec{\omega}_L - S_{eq})\,\hat{\omega}_L}{T_1} - \frac{\langle\vec{S}\rangle - (\langle\vec{S}\rangle\cdot\hat{\omega}_L)\hat{\omega}_L}{T_2}$$

Conduction Band electrons → Coupling to Nuclear Spins

Recall our general Hamiltonian in a field $\vec{B}$:

$$H = \frac{p^2}{2m} + V(\vec{r}) + \frac{1}{2m^2c^2}(\vec{S}\times\vec{\nabla}V)\cdot\vec{P} + \frac{g\mu_B}{\hbar}\vec{B}\cdot\vec{S}$$

Periodic Potential — Spin-Orbit $\left(\frac{1}{2m^2c^2r}\frac{\partial V}{\partial r}(\vec{L}\cdot\vec{S})\right)$ — Zeeman term

$$\vec{P} = \vec{p} + e\vec{A}$$ ← vector potential

Usually one next proceeded w/ $\vec{k}\cdot\vec{p}$ or Kane Theory ...

Let's now include the effect of lattice nuclear spins:

$$\vec{\mu}_I = \gamma_n \vec{I}$$ ← nuclear spin

$\vec{\mu}_I$: nuclear magnetization; γ_n: gyromagnetic ratio

$$\vec{A} = \frac{\mu_0}{4\pi}\frac{\vec{\mu}_I\times\vec{r}}{r^3} = \frac{-\mu_0}{4\pi}\vec{\nabla}\times\frac{\vec{\mu}_I}{r}$$

$$\vec{A} = \frac{\mu_0}{4\pi}\vec{\nabla}\times\frac{\gamma_n\vec{I}}{r}$$ ←

Now to the Hamiltonian ... only relevant parts for this discussion contain $\vec{B}$ or $\vec{A}$.

$$H = \frac{1}{2m}(\vec{p} - e\vec{A})^2 + \frac{g\mu_B}{\hbar}\vec{B}\cdot\vec{S}$$

To 1st order in $\vec{A}$ →

$$H = \frac{1}{2m}(p^2 + e\vec{p}\cdot\vec{A} + e\vec{A}\cdot\vec{p}) + \frac{g\mu_B}{\hbar}\vec{S}\cdot(\vec{\nabla}\times\vec{A})$$

$$H = \frac{p^2}{2m} + \frac{\mu_0\mu_B}{2\pi}\frac{\vec{L}\cdot\vec{\mu}_I}{r^3} + \frac{g\mu_0\mu_B}{4\pi\hbar}\vec{S}\cdot\left(\vec{\nabla}\times\left(\vec{\nabla}\times\frac{\vec{\mu}_I}{r}\right)\right)$$

Since the angular momentum $\ell = 0$ for the conduction band states (s-like), we can ignore the 2^{nd} term & look only at the last term:

$$\left(\vec{\nabla} \times \left(\vec{\nabla} \times \frac{\vec{\mu}_I}{r}\right)\right) = \frac{1}{r^3}\left[3(\mu_I \cdot \hat{r})\hat{r} - \vec{\mu}_I\right] + \frac{8\pi}{3}\vec{\mu}_I \delta^3(\vec{r})$$

Spin Dependent part of Hamiltonian is then:

$$H_s = \frac{g\mu_0 \mu_B \gamma_N}{4\pi \hbar}\left(\underbrace{\frac{1}{r^3}\left[3(\vec{I}\cdot\hat{r})(\vec{S}\cdot\hat{r}) - \vec{I}\cdot\vec{S}\right]}_{\text{magnetic dipole-dipole coupling of 2 mag. moments}} + \underbrace{\frac{8\pi}{3}(\vec{I}\cdot\vec{S})\delta^3(\vec{r})}_{\text{contact hyperfine coupling}}\right)$$

For s-like CB $\longrightarrow$ dipole-dipole term vanishes; hyperfine term dominates

For p-like VB $\longrightarrow$ dipole-dipole term weak; hyperfine term vanishes

So for CB:

$$H_s = H_{HF} = \frac{2}{3}\frac{g\mu_0\mu_B\gamma_N}{\hbar}(\vec{I}\cdot\vec{S})\delta^3(\vec{r}-\vec{R})$$

$\vec{R}$ → position of nucleus

Integrating over the whole volume to find the expectation value:

$$\langle \psi_{cb}|H_{HF}|\psi_{cb}\rangle = \frac{2}{3}\frac{g\mu_0\mu_B\gamma_N}{\hbar}\frac{1}{V}\int_V |\psi(\vec{r})|^2 d^3r \;\frac{1}{\Omega}\int_\Omega |u_{cb}(\vec{r})|^2 \delta(\vec{r}-\vec{R}) d^3r \;(\vec{I}\cdot\vec{S})$$

$\int_V$ → full volume; $\int_\Omega$ → unit cell

$$= \frac{2}{3}\frac{g\mu_0\mu_B\gamma_N}{\hbar}\frac{1}{V}\int_V |\psi(\vec{r})|^2 d^3r \underbrace{\frac{1}{\Omega}|u_{cb}(\vec{R})|^2}_{\eta}(\vec{I}\cdot\vec{S})$$

$$\langle \psi_{cb} | H_{HF} | \psi_{cb} \rangle = \frac{2}{3} \frac{g \mu_e \mu_B \gamma_N}{\hbar} \eta \, \vec{I} \cdot \vec{S} \quad \Longleftarrow$$

$$\text{Note:} \quad \vec{I} \cdot \vec{S} = \frac{1}{2}\left(I^+ S^- + I^- S^+\right) + I_z S_z$$

↳ Implies that ang. mom. can be transferred from the electron spins to the nuclear spins & vice-versa through "flip-flop" interactions

So, by optically pumping the electron spins (continuously) one can achieve a nuclear polarization. This is b/c the nuclear spin-lattice relaxation time is typically long (seconds to days) while the nuclear dipolar spin-spin time is short ($\sim 100\,\mu s$). So the nuclear spin polarization can be characterized by a spin temperature.

As long as the external applied magnetic field B is much larger than the local fields between nuclear spins B_L ($B \gg B_L$), then nuclear spin-spin interactions can be neglected and the interaction of electron spin & nuclear spin can be treated seperately.

$$T_{ss} = \frac{1}{\gamma_N B_L} \approx 100\,\mu s \rightarrow B_L \approx 1\,mT$$

So, since total spin is conserved in the electron - nuclear system we can write an equation of balance:

$$W_{m,m-1} N_m n_\downarrow = W_{m-1,m} N_{m-1} n_\uparrow$$

w/ N_m : population of level w/ nuclear spin m

$W_{m,n}$: transition probability from m to n

$n_\uparrow$: population of spin up electrons

$$n_\uparrow = n\left(\frac{1}{2} + \frac{1}{\hbar}\langle S\rangle\right)$$
$$n_\downarrow = n\left(\frac{1}{2} - \frac{1}{\hbar}\langle S\rangle\right)$$

w/ n : total population of electron spins

By thermodynamics:

$$W_{m,m-1} = W_{m-1,m}\, e^{\frac{g\mu_B B}{k_B T}}$$

where $g\mu_B B$ is the energy difference between states of the electron & nuclear spin. We are ignoring the Zeeman energy of the nucleus since $g\mu_B >> \hbar\gamma_N$.

$$\therefore \quad \frac{N_m}{N_{m-1}} = \frac{\left(1 + \frac{2}{\hbar}\langle S\rangle\right)}{\left(1 - \frac{2}{\hbar}\langle S\rangle\right)} \frac{\left(1 - \frac{2}{\hbar}S_T\right)}{\left(1 + \frac{2}{\hbar}S_T\right)}$$

w/ $S_T = -\frac{\hbar}{2}\tanh\left(\frac{g\mu_B B}{k_B T}\right)$

↑ equilibrium average electron spin w/o optical pumping

So, since total spin is conserved in the electron - nuclear system we can write an equation of balance:

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w/ $S_T = -\frac{\hbar}{2}\tanh\left(\frac{g\mu_B B}{k_B T}\right)$

↑ equilibrium average electron spin w/o optical pumping

Average nuclear spin : $\langle I \rangle = \sum_{m=-I}^{I} m_m N_m$

Therefore we can express $\langle I \rangle$ as a Brillouin function :

$$\langle I \rangle = B_I \left(\ln \left(\frac{1 + \frac{2}{k}\langle S \rangle}{1 - \frac{2}{k}\langle S \rangle} \; \frac{1 - \frac{2}{k} S_T}{1 + \frac{2}{k} S_T} \right) \right)$$

Typically for optical pumping :

$$\langle S \rangle >> S_T$$

Therefore we can neglect S_T and retain only the first term in the Brillouin expansion :

$$\langle I \rangle = \frac{4}{3} I(I+1) \langle S \rangle \quad \longleftarrow$$

In vector form :

$$\langle \vec{I} \rangle = \frac{4}{3} I(I+1) \frac{(\langle \vec{S} \rangle \cdot \vec{B}) \vec{B}}{B^2}$$

<u>Gist</u> : Dynamic nuclear polarization (DNP) is caused by the component of the pumped electron spin along the applied field.

Nuclear polarization is prop. to electron spin polarization by optical injection.