Lecture 10:
Nuclear spin effects in quantum dots 2

Lecture aims to explain:

1. Spin temperature and degree of nuclear spin polarisation
2. Typical nuclear polarisation degrees achievable in QDs
3. Optically detected NMR in QDs
4. ODNMR imaging in QDs: first steps
Spin temperature and degree of nuclear spin polarisation
Spin temperature and population probabilities

Spin temperature is a useful concept for thermodynamic treatment of spin systems.

In most practical cases the high temperature approximation is valid.

Let's consider $N$ spins 1/2 in thermal equilibrium with a reservoir of temperature $\Theta$ in static magnetic field $H_0$.

Energies:

$E_+ = -\frac{1}{2} \gamma \hbar H_0$

$E_+ = +\frac{1}{2} \gamma \hbar H_0$

Fractional probabilities:

$p_+ = \frac{e^{\frac{1}{2} \gamma \hbar H_0 / k_B \Theta}}{Z}$

$p_- = \frac{e^{-\frac{1}{2} \gamma \hbar H_0 / k_B \Theta}}{Z}$

Partition function:

$Z = e^{-\frac{1}{2} \gamma \hbar H_0 / k_B \Theta} + e^{\frac{1}{2} \gamma \hbar H_0 / k_B \Theta}$
Dependence of degree of spin polarisation on spin temperature

Populations of spin levels

\[ N_+ = N \frac{e^{\frac{1}{2} \gamma \hbar H_0 / k_B \Theta}}{Z} \quad N_- = N \frac{e^{-\frac{1}{2} \gamma \hbar H_0 / k_B \Theta}}{Z} \]

Spin polarisation degree:

\[ \langle I \rangle = \frac{N_+ - N_-}{N} = p_+ - p_- \]

High temperature approximation:

\[ \langle I \rangle \approx \gamma \hbar H_0 / k_B \Theta \]

Proton at room temperature, \( H_0 = 1 \text{T} \):

\[ \langle I \rangle \approx 6.8 \times 10^{-6} \]

Proton at \( T = 10 \text{K}, H_0 = 1 \text{T} \):

\[ \langle I \rangle \approx 2.04 \times 10^{-4} \]
Nuclear spin polarisation degree achievable in semiconductor QDs
In many semiconductors circularly polarised optical excitation creates electrons having preferred orientation of the spin.

Optical excitation creates an ultra-cold electron spin reservoir with efficient interaction with nuclear spins: **low spin temperatures of nuclear spins** (down to sub $\mu$K) and high polarisation degree up to 90% becomes achievable.

Efficiency of interaction with electrons can depend on their spin orientation: in that case nuclear spin temperature can be low for a relatively high electron spin temperature.

see Chekhovich PRB (2011)
Complications in measuring nuclear spin polarisation in quantum dots

Hyperfine shifts ($E_{eHF}$) of the electron spin states with contributions from all nuclear subsets:

$$E_{eHS} = \sum \rho_i A_i \langle I_i \rangle$$

Exact material content of most quantum dots (i.e. $\rho_i$) is usually unknown

see Chekhovich arXiv (2011)
Optically detected NMR in semiconductor QDs
Experiments in unstrained QDs

Note, in typical experiments on **optical orientation**, **nuclear spins are aligned** (anti-)parallel the **external magnetic field**. This produces the largest effect in the splitting of the optically detected PL spectrum. Thus optical detection of nuclear spin in PL is completely different from detection of FID, spin-echo etc using detection coils.
ODNMR spectra in unstrained QDs

Note, in strained dots, NMR spectra are complicated due to quadrupole effects, i.e. interaction of nuclei with electric field gradients produced by strain. **Quadrupole effects is one of the reason NMR spectrum may be strongly broadened**
ODNMR imaging in QDs: first steps
Light-induced Knight field in a dot

Knight field experienced by a nuclear spin at \( r_n \)

\[
B_e \propto -sF |\Psi(r_n)|^2
\]

\( s \) - average electron spin on the dot, \( F \) - filling factor, i.e. Time-average electron occupancy on the dot, \( \Psi \) - electron wave-function
Control of NMR lineshape by optical pumping

In case $s$ and $F$ are known, the volume occupied by the wavefunction can be estimated.

At high optical powers ($>1\mu W$) strongly asymmetric broadening due to $B_e \sim |\psi(r_n)|^2$.
NMR mapping of the dot: nano-ODNMR (theory)

Pronounced broadening possible in electron-charged dots in high B-field

NMR map of the dot indicating volumes with differing resonance frequencies: position-selective addressing of nuclear spins possible

Characteristics Knight field gradient of $\sim 0.5 \times 10^6$ T/m is achieved (compare to 10 mT/m usually used in macro-MRI)
Spatial-selectivity with a single ML resolution (theory)

Detection of a ML sheet (0.28 nm) of nuclei displaced from the electron wave-function centre

![Graph showing ODNMR signal vs. radio frequency detuning](graph.png)
Population of spin states in many cases can be described by spin temperature, which is also useful for estimate of the degree of nuclear spin polarisation. Normally, at room temperature very low degrees of polarisation are observed, so very high numbers of nuclei are required to increase the sensitivity of NMR measurement. In semiconductor QDs, very high degrees of nuclear spin polarisation up to 90% can be observed when pumping with optically excited electron spins (optical orientation) is used.

In typical experiments on optical orientation, nuclear spins are aligned (anti-)parallel the external magnetic field. This produces the largest effect in the splitting of the optically detected QD photoluminescence (PL) spectrum. Thus optical detection of nuclear spin in PL is completely different from detection of in-plane magnetisation in FID, spin-echo etc using detection coils.

In strained dots, NMR spectra are complicated due to quadrupole effects, i.e. interaction of nuclei with electric field gradients produced by strain. Quadrupole effects is one of the reason NMR spectrum may be strongly broadened.

Using electron Knight field in the dot, it is possible to perform nano-NMR in a QD. Characteristic Knight field gradient of \( \sim 0.5 \times 10^6 \text{ T/m} \) can achieved (compare to 10 mT/m usually used in macro-MRI).