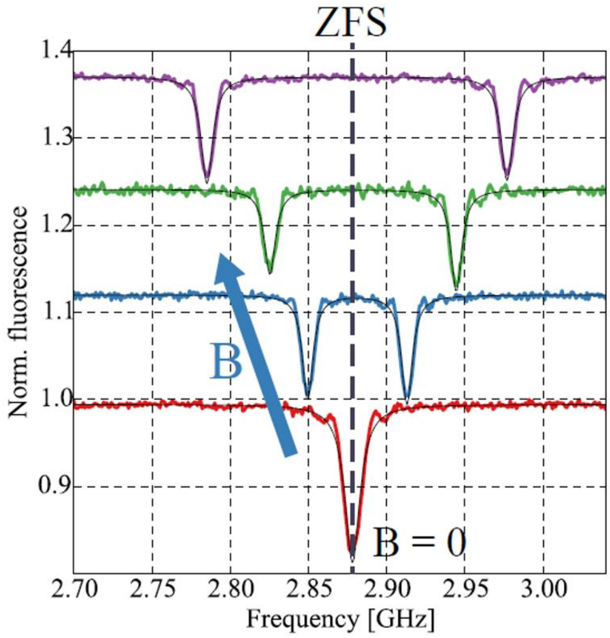
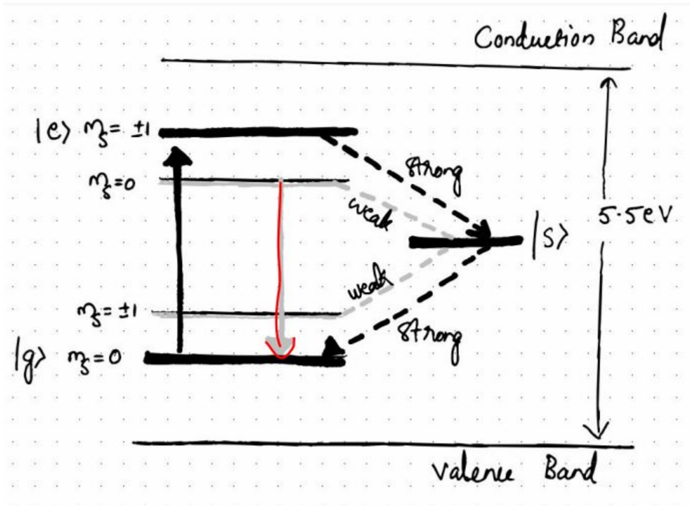
Problem 1

Negatively charged nitrogen-vacancy color centers in diamond has a ground-state triplet (^3A), and an excited-state triplet (^3E) along with SO coupled two intermediate-singlet states (^1A and ^1E).

1. Use the energy level diagram to discuss its spin state dependent fluorescence and optical polarization for quantum state preparation. What are the important consequences?
2. Typical ODMR spectra are recorded by sweeping the frequency of the microwave to drive the system from $m_S = 0 \leftrightarrow m_S = \pm 1$ transition and record the corresponding response by observing the change in the fluorescence of the color centers, as shown below. Discuss the approach in detail and derive an expression to find the magnitude of unknown magnetic field B.



The negatively charged NV-color centers has a ground state triplet (^3A) and an excited state triplet ^3E along with SO coupled two intermediate singlet states (^1A and ^1E).

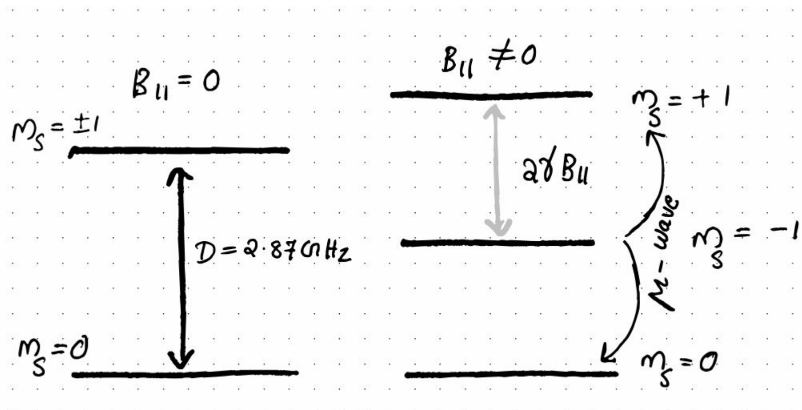


Optical transitions preserve the total spin and occur only between levels of the same total spin. Transitions from $m_s = 0, |g\rangle$ state to an excited $m_s = 0, |e\rangle$ state is accompanied by high fluorescence as indicated by red arrow.

Spin dependent intersystem crossing (due to SO coupling) leads to excited state spin triplet to the singlet state level transitions. This transition mainly affects $m_s = \pm 1, |e\rangle$ state, leads to less fluorescence. Continous optical pumping will polarize the system to the $|0\rangle$ sublevel. This is called spin polarization. Hence high fluorescence indicates the $|0\rangle$ sub-state and low fluorescence indicates the $|1\rangle$ sub-state.

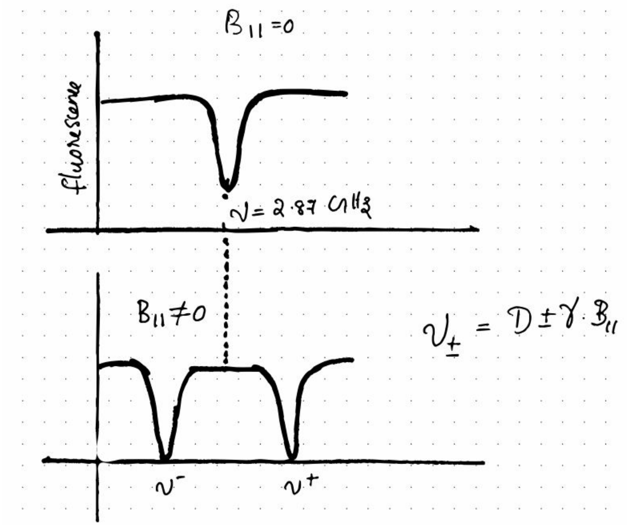
In the presence of an external magnetic field $\vec{B_0}$ there will be a Zeeman shift of the NV frequency associated with the spin state given by:

$$\boxed{\Delta\nu = \frac{m_s.g.\mu_B.B_{11}}{h}}$$



Sweeping the frequency of the microwave will eventually be in resonance with one of the $m_s = 0 \leftrightarrow m_s = \pm 1$ transitions and flip the NV to the $m_s = \pm 1$ states. This results in a decrease of fluorescence due to non-radiative decay via the singlet states.

At zero magnetic field, there is a single dip corresponding to the double degenerate ± 1 level transitions.

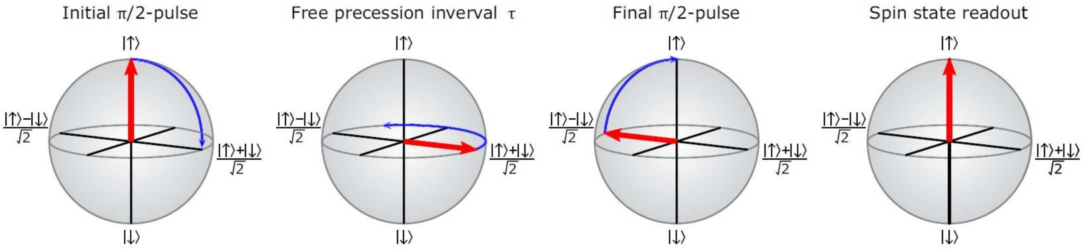


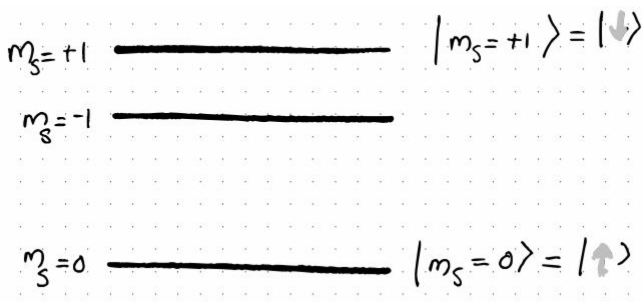
Problem 2

You want to measure a weak magnetic field $B_{unknown}$ of unknown quantum system (it can be a spin) using quantum coherence that involves Ramsey sequences, Ramsey-type pulsed magnetometry, as depicted in figure below. A bias magnetic field $B_0\hat{z}$ along the NV-symmetry axis Zeeman-splits $m_s = \pm 1$ ground state allowing a two-level subspace that can be used as a basis to describe the Hamiltonian to study the dynamics.

1. What is the Hamiltonian of the system before the application $\frac{\pi}{2}$-pulses.
2. The application of oscillating magnetic field $\vec{B_1}(t) = B_1 \cos(\omega\ t)\hat{y}$ perpendicular to the NV-symmetry axis perturb the system. Determine the Hamiltonian driven by this oscillating field? Use the interaction picture to find the state vector as a function of time.
3. Suppose the oscillating field is turned off abruptly after a duration of $\tau\frac{\pi}{2} = \frac{\pi}{2\Omega} = \frac{\pi}{\gamma_e\ B_1}$. What will be the (interaction) Hamiltonian and the evolution of the state vector, remind that the weak magnetic field of the unknown source perturbs the system.
4. Another oscillating field $B_2(t)$ is chosen to be along the xy plane at an angle θ with respect to the polarization direction of the first $\frac{\pi}{2}$-pulse $B_1(t)$. Find the transformed Hamiltonian and the corresponding time evolution of the state vector.
5. Use the information from (1) to (4) to determine the magnitude of the unknown magnetic field.
6. What is the effect of the integration time in the signal-to-noise ratio of the measured magnetic field? I suppose the contrast degrades with increasing integration time due to dephasing, decoherence, and spin lattice interaction. Is there any technique to recover the coherence and rephase?

The bias magnetic field $B_0\hat{z}$ along the NV symmetry axis Zeeman splits $m_s = \pm 1$ ground states allowing a two level subspace.





The Hamiltonian can be expressed in the basis $|\downarrow\rangle$ and $|\uparrow\rangle$.

$$\boxed{\begin{aligned}\hat{H} &= (2\pi D + \gamma_e B)S_z \\ &= \begin{pmatrix} \langle\uparrow|\hat{H}|\uparrow\rangle & \langle\uparrow|\hat{H}|\downarrow\rangle \\ \langle\downarrow|\hat{H}|\uparrow\rangle & \langle\downarrow|\hat{H}|\downarrow\rangle \end{pmatrix} \\ &= \frac{\hbar}{2}\begin{pmatrix} 2\pi D + \gamma_e B & 0 \\ 0 & -2\pi D - \gamma_e B \end{pmatrix}\end{aligned}}$$

Where $B = B_0 + B_{unknown}$. In the bias field B_0 the spin resonance frequency is :

$$\boxed{\omega_0 = 2\pi D + \gamma_e B_0}$$

Since spin operators can be expressed in terms of the Pauli matrices,

$$\boxed{\vec{S} = \frac{\hbar}{2}\vec{\sigma}}$$

One can write:

$$\boxed{\hat{H} = \frac{\hbar\omega_0}{2}\,\sigma_z + \frac{\hbar}{2}\gamma_e\, B_{unknown}\,\sigma_z}$$

Thanks to spin polarization due to optical transitions, the initial state of the spin will be $|\psi(0)\rangle = |\uparrow\rangle$ before the application of an applied AC magnetic field.

$$\boxed{\vec{B_1}(t) = B_1 \cos(\omega\ t)\hat{y}, \qquad \text{with} \quad \omega \approx \omega_0}$$

For $B_1 >>> B_{unknown}$, (typical) second term in H can be droped, then the Hamiltonian for the system driven by this oscillating field, denoted H_{driv} becomes:

$$\boxed{H_{driv} = \frac{\hbar\omega_0}{2}\sigma_z + \frac{\hbar}{2}\gamma_e B_1 \cos(\omega t)\sigma_y}$$

From the interaction picture,

$$H_0 = \frac{\hbar\omega_0}{2}\sigma_z$$

and

$$H_i = \frac{\hbar}{2}\gamma_e B_1 \cos(\omega t)\sigma_y$$

The ineraction picture state vector:

$$\left|\psi'(t)\right\rangle = U_0^+(t)\left|\psi(t)\right\rangle \quad \text{with} \quad U_0(t) = e^{-\frac{iH_0 t}{\hbar}}$$

the state evolves according to,

$$\left|\psi'(t)\right\rangle = U_1'\left|\psi'(0)\right\rangle \quad \text{with} \quad U_1'(t) = e^{-iH_i' t}$$

and

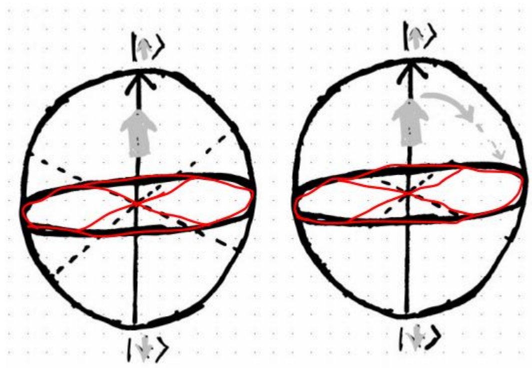
$$\boxed{\begin{aligned} H_i' &= U_0^+(t) H_1 U_0(t) \\ &= \frac{\hbar}{4}\gamma_e B_1 \begin{pmatrix} 0 & -i\left(e^{i(\omega_0+\omega)t} + e^{i(\omega_0-\omega)t}\right) \\ i\left(e^{-i(\omega_0-\omega)t} + e^{-i(\omega_0+\omega)t}\right) & 0 \end{pmatrix} \end{aligned}}$$

Assuming resonant driving of the spin with $\omega = \omega_0$ and by making the rotating wave approximation, dropping off-resonant terms rotating at $2\omega_0$ yields,

$$\boxed{H_i' \approx \frac{\hbar}{4}\gamma_e B_1 \sigma_y}$$

This Hamiltonian cause the spin system to undergo Rabi oscillations at angular frequency :

$$\boxed{\Omega = \frac{\gamma_e B_1}{2}}$$



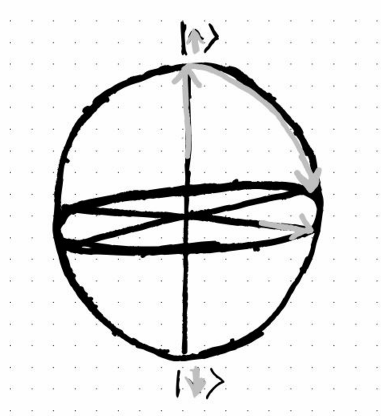
When the oscillating field $B_1(t)$ is turned off abruptly after a duration of $\tau_{\pi/2} = \frac{\pi}{2\Omega} = \frac{\pi}{\gamma_e B_1}$, the state vector becomes:

$$\boxed{\begin{aligned} \left|\psi'(\tau_{\pi/2})\right\rangle &= \exp\left(-i\frac{\gamma_e B_1 \sigma_y \tau_{\pi/2}}{4}\right)\left|\psi'(0)\right\rangle \\ &= \exp\left(-i\frac{\pi}{4}\sigma_y\right)\left|\uparrow\right\rangle \end{aligned}}$$

Using $e^{-i\theta\hat{n}.\vec{\sigma}} = \cos(\theta)\hat{I} - i\sin\theta(\hat{n}.\vec{\sigma})$

$$\boxed{\begin{aligned} \left|\psi'(\tau)\right\rangle &= \exp\left(-i\frac{\pi}{4}\sigma_y\right)\left|\uparrow\right\rangle \\ &= \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & -1 \\ 1 & 1 \end{pmatrix}\begin{pmatrix} 0 \\ 1 \end{pmatrix} \\ &= \frac{1}{\sqrt{2}}(-\left|\downarrow\right\rangle) + (\left|\uparrow\right\rangle) \end{aligned}}$$

Hence,



In the absence of $B_1(t)$ for a sensing time τ, the system Hamiltonian returns to H:

$$\boxed{\begin{aligned} H &= (2\pi D + \gamma_e B)S_z \\ &= \frac{\hbar}{2}\begin{pmatrix} 2\pi D + \gamma_e B & 0 \\ 0 & -2\pi D - \gamma_e B \end{pmatrix} \end{aligned}}$$

where $B = B_0 + B_{unknown}$.

Using again the interaction picture, $H_0 = \frac{\hbar\omega_0}{2}\sigma_z$, with new interaction Hamiltonian H_i determined by $\vec{B}_{unknown} = B_{unknown}\hat{z}$ as:

$$\boxed{H_i' = \frac{\hbar}{2}\gamma_e\ B_{unknown}\ \sigma_z}$$

Taking into consideration that H_i' commutes with H_0, the transformed interaction Hamiltonian takes the form:

$$\widetilde{H_i'} = U_0^+(t)\ H_i'\ U_0(t) = H_i'$$

The interaction picture state vector $|\psi'(t)\rangle$ that evolves under H_i' as:

$$\boxed{\begin{aligned} \big|\psi'(\tau_{\pi/2+\tau})\big\rangle &= e^{-iH_i\ \frac{\tau}{\hbar}}\ \big|\psi'(\tau_{\pi/2})\big\rangle \\ &= \frac{1}{\sqrt{2}}\big(- e^{-i\frac{\phi}{2}}\ |{\downarrow}\rangle + e^{i\frac{\phi}{2}}\ |{\uparrow}\rangle\big) \end{aligned}}$$

where, $\boxed{\phi = \gamma_e\ B_{unknown}\ \tau}$ is the phase accumulated due to $B_{unknown}$ in the interaction picture.

To complete the sequence, a second oscillating field $\vec{B_2(t)} = \vec{B_2}\cos(\omega t)$ with $\omega = \omega_0$ is applied along the xy-plane at an angle θ with respect to the y. Hence:

$$\boxed{H_i'' \approx \frac{\hbar}{4}\gamma_e B_2\Big(\cos(\theta)\sigma_y - \sin(\theta)\sigma_x\Big)}$$

$$\boxed{\begin{aligned} \big|\psi'(\tau_{\pi/2} + \tau + \tau_{\pi/2})\big\rangle &= e^{-i\hat{H}_i''\ \frac{\tau_{\pi/2}}{\hbar}}\ \big|\psi'(\tau_{\pi/2} + \tau)\big\rangle \\ &= \frac{1}{\sqrt{2}}\begin{pmatrix} 1 & -e^{i\theta} \\ e^{i\theta} & 1 \end{pmatrix} \cdot \frac{1}{\sqrt{2}}\begin{pmatrix} -e^{i\frac{\phi}{2}} \\ e^{i\frac{\phi}{2}} \end{pmatrix} \end{aligned}}$$

which is equal to:

$$\boxed{|\psi(t)\rangle = \cos\left(\frac{\phi-\theta}{2}\right)|{\downarrow}\rangle + i\ e^{i\phi}\sin\left(\frac{\phi-\theta}{2}\right)|{\uparrow}\rangle}$$

The phase accumulated during τ is thus mapped on to a population difference between $|{\downarrow}\rangle$ and $|{\uparrow}\rangle$ states:

$$\boxed{|\psi\rangle = \cos\left(\frac{\phi-\theta}{2}\right)|{\downarrow}\rangle + i\ e^{i\theta}\sin\left(\frac{\phi-\theta}{2}\right)|{\uparrow}\rangle}$$

The population difference is detected by measuring the rotating frame observable $\vec{S_z}$.

The value of $B_{unknown}$ is determined by:

$$\boxed{\begin{aligned} < S_z > &= \frac{\hbar}{2}\left\langle\hat{\psi}\right|\sigma_z\left|\hat{\psi}\right\rangle \\ &= \frac{\hbar}{2}\left(\cos^2\left(\frac{\phi-\theta}{2}\right) - \sin^2\left(\frac{\phi-\theta}{2}\right)\right) \\ &= \frac{\hbar}{2}\cos(\phi-\theta) \\ &= \frac{\hbar}{2}\cos(\gamma_e\ B_{unknown}\ \tau - \theta) \end{aligned}}$$

where $\phi = \gamma_e B_{unknown}\tau$.

For small $B_{unknown}$ such that $\phi << 2\pi$, the solution can be linearized about $\phi = 0$ for any value of $\theta = 0$. The value of $B_{unknown}$ and ϕ can then be related to a small change in the observable:

$$\delta < S_z > = < S_z > |_\phi - < S_z > |_0$$

$$\boxed{\begin{aligned} B_{unknown} = \frac{\phi}{\gamma_e \tau} &\approx \frac{1}{\gamma_e \tau} \times \frac{\delta < S_z >}{\frac{d}{d\phi} < S_z > |_0} \\ &\approx \frac{1}{\gamma_e \tau} \times \frac{\frac{2}{\hbar} \times < S_z > |_\phi - \cos\theta}{\sin\theta} \end{aligned}}$$

For $\theta = \frac{\pi}{2}$, the slope of the Ramsey fringe is maximized leading to:

$$\boxed{B_{unknown} \approx \frac{2}{\hbar \gamma_e \tau} < S_z >}$$

Pulsed magnetomety benefit from long sensing time τ, as the accumulated magnetic field dependent phase ϕ increases with τ according to:

$$\boxed{\phi = \gamma_e B \tau \qquad \text{with} \qquad \gamma_e = \frac{g_e \mu_B}{\hbar}}$$

Maximal sensitivity of the observable ϕ to change is when $\boxed{\frac{d\phi}{dB} = \gamma_e \tau}$ is maximized.

But contrast degrades with increasing τ due to dephasing, decoherence and spin-lattice interaction.

If the dephasing is associated with the static or slowly varying inhomogeneties in a spin system, e.g, dipolar fields from other spin impurities, it can be reversed by application of a π-pulse halway through the free precession interval, the so called Hahn protocol. In this protocol, the phase accumulated due to static fields during the second half of the sequence conceals the phase from the first half.