Spin Dynamics and Experimental Review

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1 Spin-Orbit Coupling

Relatively simple but very much worth noting to understand at full. This is the effect of a carrier's spin with respect to the orbital motion around its atomic nucleus, or the greater crystalline lattice, in an E-field. In solid-state, it couples an electron's intrinsic magnetic moment with the H field it experiences due to moving in an E-field. In atoms and crystalline lattices the Hamiltonian H_{SOC} is proportional to L (orbital angular momentum) x S (spin angular momentum).

The process of why and how it happens essentially goes:

- ➢ E-field generated from the positively charged atomic nucleus (protons)
- ➢ Electron moves, orbits in this field
- ➢ From the electron's frame of reference, the nucleus appears to be moving
- ➢ Moving charge produces magnetic field (Ampère)
- ➢ Looks like nucleus is generating a magnetic field
- ➢ Electron's spin moment now interacts with this seen magnetic field
- ➢ Now you have a coupling energy from this interaction

Quantum mechanically this Hamiltonian looks like:

$$H_{\text{SOC}} \propto \frac{1}{r}\frac{dV}{dr}\,\mathbf{L}\cdot\mathbf{S}$$

- Dependent on E-field
- Stronger nuclear potential means stronger coupling
- Same goes for atomic mass (heavy atoms)

2 Spin Current

This is a bit more involved but still fairly comprehensible off bat. This is the "flow" of spin angular momentum ($\pm\frac{\hbar}{2}$ for an electron). Essentially, this is analogous to conventional charge current in a few different ways. I.e. a rank two tensor, J_i^α, combining the spatial direction of particle motion, denoted by i, with the direction of spin polarization, α. This will allow for magnetic state manipulation without any conventional charge transport.

There is essentially two key cases on how this can exist in the first place.

Case 1: Spin-polarized normal charge current

- We have a conventional current, but the spin per carrier is imbalanced. For example 70% of carriers may be spin-up, while 30% may be spin-down. This is the case in ferromagnets (unbalanced spin moments).
- Carries charge and spin angular momentum (key for spin torque in spintronic devices).
- Some pretty important context here is that in a normal conductor at thermodynamic equilibrium, the spin of carriers is of course equally balanced and distributed in all directions, resulting in net-zero magnetization.

Case 2: Pure spin current with no charge current

- Conceptually a bit weird, as carriers can move far from their origin point, or stay localized (magnons). Spin $\uparrow$ electrons travel in an arbitrary direction, while spin $\downarrow$ travel equal and opposite in the other direction.
- Thus, we get $\mathbf{J}_\uparrow = -\mathbf{J}_\downarrow$, meaning the $\mathbf{J}_c = 0$.
- In typical heavy metals (Cu, Pt, etc), electrons move with a randomized quantum Fermi velocity ($10^5 - 10^6$ m/s). I.e. they are essentially non-localized.
- To give more context as to HOW far they move here, in heavy metals like Pt, it might be 1-10 nm, or 10-100 nm in something like Cu.
- Very interesting edge case here: magnons. In materials like YIG, there are no mobile charge carriers. Instead, localized electrons stay spinning on their lattice sites. They are in-turn exchange coupled to their nearby neighbors. So a precession phase will actually propagate amongst the lattice. Think of "the wave" when you're watching a football game or similar.

So all of this of course begs the question: *Where does it come from or how can we even get spin current in the first place?* Somewhat conventionally (most recent experimental developments), there are three main ways:

1. Ferromagnetic Injection
This is the most relatively simple and straightforward method. You are simply dumping carriers from a ferromagnetic material into a normal metal. Since the ferromagnet is already polarized and has a net spin moment (unequal DOS), you've basically now injected spin-polarized carriers into the metal.

2. Spin-Hall Effect
In materials with very strong spin-orbit coupling (Pt, Ta, W), injected moving carriers enter and pass through, pushing spin $\uparrow$ electrons to one side and spin $\downarrow$ to the other. Thus creating a pure transverse spin current, with no ferromagnets needed.

3. Spin Pumping
The most complex and involved method. In short, a precessing magnetization (ferromagnetic resonance,

FMR) literally "pumps" spin current into an adjacent material. Ferromagnetic resonance couples an external microwave field to the precessing magnetic moments of electrons in the ferromagnet. This resonance occurs because the microwave magnetic field applies torque in phase with the natural Larmor precession, allowing energy to accumulate coherently and amplify the precession amplitude.

Physically, this resonant driving torque appears in the Landau-Lifshitz Equation as:

$$\frac{d\mathbf{M}}{dt} = -\gamma\, \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\lambda}{M_s}\mathbf{M} \times (\mathbf{M} \times \mathbf{H}_{\text{eff}})$$

and the part we care about that drives the microwave excitation as:

$$\frac{d\mathbf{M}}{dt} = -\gamma\, \mathbf{M} \times \mathbf{B}$$

But let's start to dive into the actual process here, once the microwave field has been applied and provides the needed resonant driving torque. In a ferromagnet, there is a clear collective magnetization vector:

$$\mathbf{M}$$

If we apply a static magnetic field:

$$\mathbf{H}_0$$

The key here is this magnetization here feels a *torque*. Which literally looks like:

$$\tau = \mathbf{M} \times \mathbf{H}_0$$

Torque is perpendicularly applied, which starts to literally rotate $\mathbf{M}$ about $\mathbf{H}_0$. This is the precession being described. I think the best way to conceptually grasp this is to consider a physical and experimental methodology.

Let us consider some sort of bilayer. The two layers being a ferromagnet, and a non-magnetic metal. A common combination encountered in much theoretical and experimental literature is Yttrium-Iron-Garnett (YIG) as the FM (a ferrimagnetic insulator technically, not a strict ferromagnet), and Platinum (PT), as the nonmagnetic metal. The process looks something like:

- Apply microwave excitation at FMR.
- $\mathbf{M}$ begins to rotate around the effective microwave $\mathbf{H}_0$.
- Rotates with a time dependence. The bilayer interface begins to "feel" this exchange field.
- Conduction band electrons in the NM want to follow this motion, as the localized spins in the FM interact with the conduction electron spins around the boundary.
- But the key here is that since there is a time dependency, phase lag, and finite response time, you get a "torque" potential aspect on the NM electron spin moments.
- This torque is what transfers spin angular momentum from the ferromagnet into the NM. But we don't quite have a spin current just yet. Just a nonzero angular momentum density at the boundary.
- The current comes into play with diffusion. Diffusion is crucial here, because spin angular momentum is comprised in a gradient at the boundary of the NM.
- Free moving charges in the NM on one localized side have different spin angular momentum, and this imbalance, with time, propagates as a diffusion spin current (due to the random motion of electrons at thermal equilibirum).

This is essentially the entire process of spin pumping, from environment architecture, to field excitation, to spin current diffusion across the NM. To engineer this, there are steady-state decay equations for diffusion, boundary conditions that set "how good" the initial precession is at transferring torque to the NM, and the actual physical thickness of the NM (it is finite, thus absorption will saturate across the width).

3 Inverse Spin-Hall Effect

Obviously we can't just stick an ammeter next to a conductor or material to measure a spin current, as it carries angular momentum, not charge. So we have to reverse engineer this process a bit. This is where the inverse spin hall effect (often denoted as ISHE) comes into play. We can detect spin by essentially converting it back to charge. But, if you recall from the spin-hall effect earlier, since a net charge current J cancels out to zero, we don't actually have a voltage/current we can measure either. The methodology physicists and engineers typically use works like this:

- Lets say we have a spin current that's pointed in the +x direction. And our spin polarization moment is along the +z direction.
- Send this current into a heavy metal (Pt, W)
- Spin up and spin down electrons are moving in opposite directions, and both experience a spin-dependent transverse deflection due to spin–orbit coupling. Both deflections produce charge accumulation on the same transverse side.
- Essentially, spin different charges move away from each other, but transversally the same direction of deflection. So they're all being "pushed" from one side to another.
- This is a weird part - the time this process takes to hit steady state is quite fast. So once the first carriers start to reach one side, an electric field starts to form which cancels further transverse flow (imbalance).
- Now we have a charge imbalance which means an actual measurable nonzero transverse voltage across the width (open-circuit voltage across the width of the surface).

The efficiency of converting spin current into charge current is governed by the spin Hall angle (θSH), which quantifies how strongly spin–orbit coupling generates transverse charge flow.

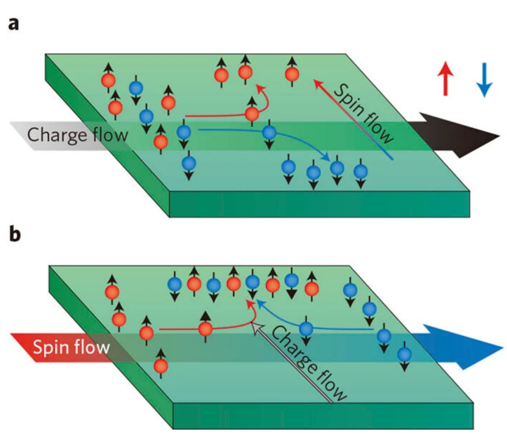

Figure 1: Inverse Spin Hall Effect schematic on metal side. Source [1]

A spin current $\mathbf{J}_s$ with spin polarization σ is converted to a transverse charge current $\mathbf{J}_c$ via spin-orbit coupling. The conversion efficiency is governed by the spin-Hall angle θSH, and the resulting charge current is perpendicular to both the spin current direction and it's polarization:

$$\mathbf{J}_c \propto \theta_{SH} \, \mathbf{J}_s \times \boldsymbol{\sigma}$$

4 Magnetic Resonance Dynamics

The idea of magnetic resonance begins with a simple fact: an electron's spin carries an intrinsic magnetic moment that is proportional to its angular momentum. When placed in an external magnetic field, this magnetic moment experiences a torque, causing it to precess at a characteristic frequency set by the field's strength. Much more broadly, it is also important to note that spin is simply an intrinsic quantum number that can take integer or half-integer values (0, 1/2, 1, 3/2, 2, ...), depending on the particle or system. The value 1/2 spin is just for electrons and other fundamental fermions like protons and neutrons.

The vector quantity $\vec{\mu}$ is known as the nuclear magnetic dipole moment or more generally, just the magnetic moment. A fundamental relationship in particle physics is that the spin angular momentum and magnetic moment vectors are related to each other by:

$$\vec{\mu} = \gamma \vec{S}$$

Where γ is a physical constant known as the *gyromagnetic ratio*. This simply defines how strong of a magnetic moment is generated by a given spin. And $\vec{S}$ is of course the spin angular momentum vector, denoted as quantum number $\pm\frac{1}{2}$ for electrons. For an electron spin system, this relationship further reduces to [1]. The negative sign arises because the electron carries negative charge, a subtle difference meaning its magnetic moment is oriented opposite to its spin angular momentum. For electrons, $\gamma_e \approx 28$ GHz/T.

$$\vec{\mu_e} = -\gamma_e \vec{S} \tag{1}$$

This is also typically expressed in literature in units of the *Bohr magneton*, or the fundamental constant that sets the natural scale for electron magnetic moments. The electron magnetic moment happens to be equal to about one Bohr magneton.

$$\mu_B = \frac{e\hbar}{2m_e} \approx 9.274 \times 10^{-24} \,\text{J/T}$$

A key concept to consider is the idea of Larmor precession, and the Larmor frequency. When a magnetic moment is placed in a static magnetic field, it experiences a torque rather than a direct net force, as from earlier. This torque does not align the moment directly with the field; instead, it causes the moment to start to precess around the field's direction. This specific motion is called Larmor precession. The angular frequency of this nuclear precession is defined as:

$$\omega_0 = \gamma B_0$$

Another point of emphasis here is that every nucleus has its own resonance frequency (basically each isotope has a different γ, i.e. $^1H, ^{13}C, ^{31}P$ all differ slightly), and this Larmor frequency is completely independent of any spin magnitude. It's also important to note that, when describing the collective behavior of any spin system, a quasi-macroscopic magnetization vector $\vec{M}$ is introduced, which is simply just the vector sum of all microscopic magnetic moments of a system. In ideal electron spin resonance (ESR), all electrons of a uniform material would resonate at the same frequency, i.e. $\omega_1 = \omega_2 = \omega_3 ....$ But in real materials, each electron actually experiences a local effective field. This local effective field is comprised of the dominant external field B_0, an exchange field between neighbors $B_{exchange}$ (which is very strong in ferromagnets), general dipole fields from neighbors (tiny localized perturbations), and hyperfine fields (interaction with nearby nuclear spins). Because of this, it is likely to get slightly different resonant frequencies between electrons in one lattice. In most literature, this is often referred to as *line broadening.*

On the concept of energy; spins in different orientations have different energy interactions with the external magnetic field B_0. Specifically, this quantum Hamiltonian is denoted by:

$$E = -\vec{\mu} \cdot \vec{B_0}$$

Substituting in our relationship from earlier, we can get:

$$E = -\gamma \vec{S} \cdot \vec{B_0}$$

Assuming $\vec{B_0} = B_0 \hat{z}$, then:

$$E = -\gamma B_0 S_z$$

Since we're dealing with a spin $\pm\frac{1}{2}$ system,

$$S_z = \pm\frac{\hbar}{2}$$

Then,

$$E = \mp\frac{\gamma \hbar B_0}{2}$$

With an energy difference of,

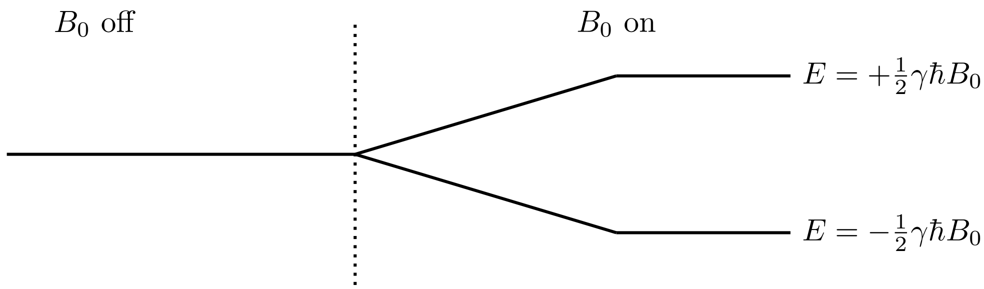

Figure 2: Zeeman splitting for a spin-$\frac{1}{2}$ system.

$$\Delta E = E\downarrow - E\uparrow = \gamma\hbar B_0 = \hbar\omega_0$$

The nonzero difference in energy levels between these two spin states is known as the *Zeeman splitting phenomenon*, illustrated above in Fig. 2.

5 Optically Detected Magnetic Resonance (ODMR)

Optically detected magnetic resonance (ODMR) draws upon and extends much of the principles of traditional magnetic resonance, by replacing inductive detection with optical readout. In this resonance technique, microwave magnetic fields drive spin transitions at their resonance condition as we know, but fluorescence intensity allows for a direct measure of the spin state. This approach is particularly powerful in long-coherence time, highly localized systems, such as nitrogen-vacancy (NV) centers in diamond (we will dive into this architecture later), where single-spin readouts actually become possible.

The relevant Hamiltonian for an NV center state looks like:

$$E = DS_z^2 + \gamma_e B_0 S_z$$

You'll notice this looks very similar to before (general magnetic resonance), but adds in a few terms. D here represents the zero-magnetic field splitting parameter. A crucial fact of NV center spin dynamics is; the ground-state is not spin-$\frac{1}{2}$ like an electron in ESR, but rather spin-1. So, in the equation above, S must be equal to either 1, 0, or -1. Essentially, even if B_0 equals zero (Zeeman term from earlier vanishes), this DS_z^2 term tells us that the total energy is still not zero for the ± 1 spin states in the NV center. This a strong fundamental architecture difference from previous definitions of spin state and transport manipulation energies.

Since we now know that the NV center has a nonzero energy splitting at zero magnetic field, it possesses a unique well-defined microwave resonance even without applying B_0. This zero-field splitting provides a sort of built-in resonance baseline, and then magnetic fields are measured by how well they perturb that frequency. This is what the D in our energy equation represents - for NV centers it is ≈ 2.87 GHz. 2.87 GHz corresponds to the energy difference between the 0, and ± 1 spin states under zero magnetic field. I.e., in the absence of an applied B_0, the ± 1 spin states lie about 2.87 GHz higher in energy than the 0 state. Meaning when we apply a microwave field at 2.87 GHz, we are able to "flip the spin" in the NV center.

This is incredibly powerful because it is what leads to the entire idea of ”optical detection”. The NV center is ”pumped” into its 0 state (a naturally bright state) by a laser ($\approx$ 532nm), and when we apply 2.87 GHz microwave fields, we drive it into its ± 1 states, which happen to be much, much darker. Thus, fluorescence decreases, and 2.87 GHz is the bridge that connects microwave control, to spin state perturbation, to physical optical brightness.

Of course experimentally, 2.87 GHz also happens to be in an extremely friendly microwave regime. It is relatively easy to generate with standard electronics, not too high or too low of a frequency (a pretty solid spectral separation), and maybe most importantly, large enough of a frequency to have thermally stable splitting at room temp. At room temp:

$$kT/h \approx 6\,\mathrm{THz}$$

So, 2.87 GHz is relatively tiny in comparison to thermal energy, and the population differences are very small. To give some context on scale and magnitude for experimentation, a single NV center under optical excitation typically produces photon count rates on the order of $10^4 - 10^6$ counts per second, enabling ODMR detection with avalanche photodiodes. In ensemble architectures (larger scale localization), fluorescence intensities are much, much higher, and can actually be detected using conventional photodiodes or cameras. Experimentally, ODMR spectra are obtained by sweeping the applied microwave frequency while actively monitoring fluorescence; resonant transitions then appear as noticeable dips in the detected photon count rates.

References

[1] Liang Cheng, Ziqi Li, Daming Zhao, and Elbert EM Chia. Studying spin–charge conversion using terahertz pulses. *Apl Materials*, 9(7), 2021.