Determining the Sign of Polarization Relative to the Magnetic Field in the Polarized ^{3}He Target

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Version 1.00

May 2, 2007

Abstract

This document will outline how to obtain (a) the sign of the ^{3}He nuclear polarization, (b) the sign of the alkali polarization, (c) the sign of the laser light polarization, & (d) the magnitude of the laser light polarization. The sign of the ^{3}He polarization relative to the holding field can be determined directly from the sign of the frequency shift extracted from EPR polarimetry. If the ^{3}He and alkali polarizations are at equilibrium, then the signs of their respective polarizations are the same. To determine the sign of the light polarization, one also needs to know the laser beam propagation direction relative to the holding field. Finally, we'll describe a standard technique to measure the degree of circular polarization of the light using a rotatable beam splitting polarizing cube.

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1 Notation & Conventions

Operators, matrices, and unit vectors are denoted by hats $\hat{M}$. Hamiltonians are $\mathcal{H}$, energies are E, frequencies are ν (with units of Hz), and angular frequencies are ω (with units of rad·Hz). All quantities are denoted in SI. Angular momentum operators are unitless:

$$
\begin{aligned}
\hat{\vec{J}}^2 |J, m_J\rangle &= J(J+1)\,|J, m_J\rangle && (1)\\
\hat{J}_z |J, m_J\rangle &= m_J\,|J, m_J\rangle \qquad (m_J = -J..J) && (2)\\
\hat{J}_\pm &= \hat{J}_x \pm i\hat{J}_y && (3)\\
\hat{J}_\pm |J, m_J\rangle &= \sqrt{J(J+1) - m_J(m_J \pm 1)}\,|J, m_J \pm 1\rangle && (4)
\end{aligned}
$$

The longitudinal spin polarization is defined as:

$$P \equiv \frac{\left\langle \hat{\vec{J}} \right\rangle}{J} \tag{5}$$

The statistical weight is denoted by $[J]$ and is defined by $[J] \equiv 2J+1$. The magnetic moment arising from a spin is written as:

$$\vec{\mu} = \left(\frac{\mu}{J}\right)\vec{J} = g\mu_x\vec{J} \tag{6}$$

where g is the unitless g-factor:

$$g = \frac{1}{J}\left(\frac{\mu}{\mu_x}\right) \tag{7}$$

The units are carried in μ_x, which is the Bohr magneton μ_B for the electron and the nuclear magneton μ_N for nuclei. Note that the sign of the magnetic moment is carried implicitly in g or alternatively μ. For example, $g \approx -2$ for the electron, $g \approx 2(+2.79)$ for the proton, and $g \approx 2(-1.91)$ for the neutron. Finally, the various angular momentum are usually labelled as:

- $\vec{S}$ for the alkali electron spin
- $\vec{L}$ for the alkali electron orbital angular momentum
- $\vec{J} = \vec{L} + \vec{S}$ for the total alkali electronic angular momentum
- $\vec{I}$ for the alkali nuclear spin
- $\vec{F} = \vec{I} + \vec{J}$ for the total alkali atomic angular momentum
- $\vec{K}$ for the noble gas nuclear spin

2 Brief Overview of EPR Frequency Shift Polarimetry

EPR frequency shift polarimetry (EPR) [1, 2, 3] is a method used to provide an absolute calibration of the ^{3}He polarization in a target cell. It takes advantage of the Zeeman splitting of the hyperfine levels of an alkali atom in it's ground state:

$$E(m_F, B) = \langle m_F|\,\hat{\mathcal{H}}\,|m_F\rangle = \langle m_F|\,\hat{\mathcal{H}}_0 - \hat{\vec{\mu}}\cdot\vec{B}\,|m_F\rangle = E_0 - g\mu_B m_F B \tag{8}$$

where E_0 is the zero field energy of the alkali atom, $|m_F\rangle$ is the hyperfine state, and B is the magnitude of the magnetic field. The g-factor is given by:

$$g = \frac{g_e}{[I]}\left[1 - \frac{g_I\mu_N}{g_e\mu_B}\right] + \delta g\,(m_F, B) \tag{9}$$

where $g_e\,(g_I)$ is the g-factor associated with the spin of the electron (alkali nucleus) and δg "hides" the dependence of the g-factor on m_F and B. At sufficiently low fields, δg is essentially a small higher order correction. A magnetic field is considered low when the strength of the Zeeman interaction is small relative to the hyperfine interaction. For ^{39}K and ^{85}Rb, the strength of these two interactions are equal at 165 gauss and 1080 gauss respectively. Traditionally, the target cells are located in a magnetic holding field B_0 that is between 10 and 30 gauss. Therefore, for the foregoing discussion, we'll drop the μ_N/μ_B and δg terms since we are only interested in the *sign* of the polarization. (Beware, however, for accurate numerical results for the *magnitude* of the ^{3}He polarization, these two terms must be kept!)

The total field B is given by the vector sum of the holding field B_0 and a small additional effective field due to the presence of polarized ^{3}He gas B_{He}:

$$\vec{B} = \vec{B_0} + \vec{B}_{\mathrm{He}} \tag{10}$$

Under typical operating conditions, roughly 15% of B_{He} is due to the average classical magnetic field produced by the bulk magnetization of the polarized ^{3}He gas in the region where the alkali atoms are probed. The rest of B_{He} comes from an effective field due to the spin-exchange collisions that occur between the alkali and ^{3}He atoms. For a uniformly polarized sphere of ^{3}He, the sum of the classical field and effective spin-exchange field is given by:

$$\vec{B}_{\mathrm{He}} = \frac{2\mu_0}{3}\kappa_0 \vec{M}_{\mathrm{He}} \tag{11}$$

where κ_0 is a unitless temperature dependent (T) quantity. Because the effective field due to spin exchange cannot be calculated accurately from theory, κ_0 must be determined empirically. The part of κ_0 that is due to spin exchange is given by [3, 4]:

$$\begin{aligned}
\kappa_{\mathrm{se}}(T) &= \kappa_0(T) - 1 && (12)\\
\mathrm{Rb} : \kappa_0 &= 6.39 + 0.00924 \cdot (T - 200\ ^\circ\mathrm{C}) && (13)\\
\mathrm{K} : \kappa_0 &= 5.99 + 0.0086 \cdot (T - 200\ ^\circ\mathrm{C}) && (14)\\
\mathrm{Na} : \kappa_0 &= 4.84 + 0.00914 \cdot (T - 200\ ^\circ\mathrm{C}) && (15)
\end{aligned}$$

The magnetization of a uniformly polarized sample of ^{3}He is given by:

$$\vec{M}_{\mathrm{He}} = \rho \langle \vec{\mu} \rangle = \rho g_K \mu_N \left\langle \vec{K} \right\rangle = \rho g_K \mu_N K \vec{P} \tag{16}$$

where ρ, g_K, K, and P are resepectively the number density, g-factor, spin, and polarization of the ^{3}He nuclei. Under typical operating conditions, B_{He} is on the order of 10's of milligauss.

During an EPR measurement, the frequency of transition between the $m_F = s(I + 1/2)$ state and the $m_F = s(I - 1/2)$ state is monitored, where $s = \pm$ is the sign of the alkali polarization. In the low field limit, this frequency is given by a sum of the contributions of the main field and the ^{3}He gas:

$$\nu = \left(\frac{g_e}{[I]}\right)\left(\frac{\mu_B}{h}\right)\left(\vec{B}_0 + \frac{2\mu_0}{3}\kappa_0 \rho g_K \mu_N K \vec{P}\right) \cdot \hat{B} = \nu_0 + \delta\nu_K \left(\vec{P} \cdot \hat{B}\right) \tag{17}$$

where $\hat{B}$ is the unit vector pointing along the direction of the total field. Since the holding field is orders of magnitude larger than the ^{3}He field, the total field $\vec{B}$ and the holding field $\vec{B}_0$ are nearly parallel:

$$\nu = \nu_0 + \delta\nu_K \left(\vec{P} \cdot \hat{B}\right) \approx \nu_0 + \delta\nu_K \left(\vec{P} \cdot \hat{B}_0\right) \tag{18}$$

To isolate the contribution from ^{3}He, the ^{3}He spins are "flipped" adiabatically while keeping the holding field constant. To return the ^{3}He spins to their original state, they are flipped adiabatically once more. At the conclusion of the measurement, three frequencies have been recorded:

1. ν_{bef}, the EPR frequency before flipping the ^{3}He spins
2. ν_{mid}, the EPR frequency after flipping the ^{3}He spins for the first time
3. ν_{aft}, the EPR frequency after flipping the ^{3}He spins for the second time

Two frequency shifts are measured:

$$\Delta\nu_{\mathrm{bm}} \equiv \frac{\nu_{\mathrm{bef}} - \nu_{\mathrm{mid}}}{2} = \delta\nu_K \left[\frac{\vec{P}_{\mathrm{bef}} - \vec{P}_{\mathrm{mid}}}{2}\right] \cdot \hat{B}_0 \tag{19}$$

$$\Delta\nu_{\mathrm{am}} \equiv \frac{\nu_{\mathrm{aft}} - \nu_{\mathrm{mid}}}{2} = \delta\nu_K \left[\frac{\vec{P}_{\mathrm{aft}} - \vec{P}_{\mathrm{mid}}}{2}\right] \cdot \hat{B}_0 \tag{20}$$

Assuming that no ^{3}He polarization is lost in the course of the measurement, then the ^{3}He polarizations are related by:

$$\vec{P}_{\mathrm{bef}} = -\vec{P}_{\mathrm{mid}} = \vec{P}_{\mathrm{aft}} = \vec{P} \tag{21}$$

and the frequency shift is given by:

$$\Delta\nu = \left(\frac{g_e \mu_B}{h[I]}\right) \left(\frac{2\mu_0}{3} \kappa_0 \rho g_K \mu_N K\right) \left(\vec{P} \cdot \hat{B}_0\right) \tag{22}$$

Under typical operating conditions, this frequency shift is ±10's of kHz.

3 The Sign of the ^{3}He Polarization

Before going on further, it is imperative to reemphasize two important points:

1. The frequency shift due to polarized ^{3}He is measured relative to the EPR frequency when the ^{3}He polarization is zero. In other words, the "baseline" EPR frequency is due to the all the fields *not* associated with the ^{3}He.

2. The sign of the ^{3}He polarization is measured relative to the sign of the holding field (which is positive by definition).

The sign of the frequency shift $\Delta\nu$ is determined by the product $g_K \left(\vec{P} \cdot \hat{B}_0\right)$. Since the magnetic moment of ^{3}He is negative ($g_K < 0$), the sign of the polarization is negative to the sign of the frequency shift:

$$\mathrm{sign}\left[\vec{P} \cdot \hat{B}_0\right] = -\mathrm{sign}\left[\Delta\nu\right] \tag{23}$$

The physical interpretation of this result is easy to understand. When the holding field and the field due to ^{3}He are parallel (antiparallel), then the two fields add (subtract). The resulting EPR frequency is consequently greater (smaller) than the zero ^{3}He polarization EPR frequency. Thus the frequency shift is positive (negative). Because the magnetic moment of ^{3}He is negative, the polarization of ^{3}He and the magnetic field due to the ^{3}He are always of opposite sign:

$$\mathrm{sign}\left[\vec{P}\right] = -\mathrm{sign}\left[\vec{B}_{\mathrm{He}}\right] \tag{24}$$

whereas the sign of the expectation value of the spin state is always the same as the sign of the polarization:

$$\mathrm{sign}\left[\vec{P}\right] = \mathrm{sign}\left[\left\langle \vec{K} \right\rangle\right] \tag{25}$$

A graphical depiction of this argument is given at the bottom of Figs. (1) & (2). To summarize:

- well shape $\Rightarrow \Delta\nu > 0 \Rightarrow B_{\mathrm{He}}$ is parallel to B_0 $\left(^3\text{He is in low energy state}\right) \Rightarrow B_{\mathrm{He}} > 0 \Rightarrow P < 0$
- hat shape $\Rightarrow \Delta\nu < 0 \Rightarrow B_{\mathrm{He}}$ is antiparallel to B_0 $\left(^3\text{He is in high energy state}\right) \Rightarrow B_{\mathrm{He}} < 0 \Rightarrow P > 0$

4 The Sign of the Alkali Polarization

If the spin system in the cell is at equilibrium, then one can be certain that the sign of the alkali polarization is the same as the sign of the ^{3}He polarization. The phrase "at equilibrium" means that both the alkali and ^{3}He polarizations have reached their saturation values. The alkali and ^{3}He spins are not at equilibrium after the first spin flip and before the second spin flip during an EPR measurement. During this middle period of an EPR measurement, the signs of the alkali and ^{3}He polarizations are opposite.

This observation can used to define a looser and more useful definition for "at equilibrium:" the spin system is "at equilibrium" when the difference between the number of alkali spin flips and the number of ^{3}He spin flips is even. The alkali spins are usually flipped by rotating the quarter waveplate used to circularly polarize the laser beam by 90 degrees. Suppose that both the alkali and ^{3}He polarizations are zero and the laser has just been turned on. In this case, the polarizations of both the alkali atoms and ^{3}He nuclei are changing with time. However, since neither set of spins have been flipped, the signs of the polarizations should be the same. In summary, as long as nothing "weird" has happened, the sign of the alkali polarization should be the same as the sign of the ^{3}He polarization.

An alternative way to determine the sign of the alkali polarization is from the EPR frequency and the magnitude of the holding field. To lowest order, the frequencies are linear in field and independent of the m_F state. However, the higher order terms (hidden in δg) give an m_F dependence to the EPR frequency. For example, at $B_0 = 25$ gauss, the difference in EPR frequencies between the $m_F = +(I + 1/2) \leftrightarrow +(I - 1/2)$ transition and the $m_F = -(I + 1/2) \leftrightarrow -(I - 1/2)$ transition are -450 kHz and -4000 kHz for ^{85}Rb and ^{39}K respectively. To use this method, one needs to know the magnitude of the holding field to only about 20% for ^{39}K EPR frequencies and to about 4% for ^{85}Rb EPR frequencies. At 25 gauss, this corresponds to only about 5 gauss for ^{39}K EPR frequencies and to about 1 gauss for ^{85}Rb EPR frequencies.

Finally, the signs of the alkali polarizations in a hybrid cell are always essentially the same. This is because the alkali spin exchange is very fast (>MHz). The alkali atoms can always be thought of as being "at equilibrium" with each other.

5 The Sign of the Light Polarization

5.1 Atomic vs. Light Coordinate System

Once the sign of the alkali polarization is known, the sign of the light polarization can be determined from knowledge of the laser beam propagation direction relative to the holding field. This is a very tricky argument for two reasons:

1. There are two different coordinate systems involved in this discussion.

2. There are two different conventions for labelling the circular polarization of light.

First let's start with the two different coordinate systems. From the point of view of the alkali atom, the most natural coordinate system is the one in which the positive z direction points along the direction of the holding field. Let's call this the atomic coordinate system. On the other hand, from the point of view of the photon in the laser beam, the most natural coordinate system is the one in which the positive z direction points along the direction of propagation of the laser beam. Let's call this the light coordinate system.

When the laser beam is traveling parallel to the the holding field ($\Theta = 0$), the atomic system and the light system are one and the same. However, when the laser beam is travelling antiparallel to the holding field ($\Theta = \pi$), the atomic system and the light system point in opposite directions! See App. (A.5) for the general case of $\Theta \neq 0, \pi$.

Suppose we've found that the polarization of ^{85}Rb is negative. This means that the $m_F = -3$ state is being filled and the $m_F = +3$ is being depopulated by the polarized light. If we ignore the nuclear spin, then this corresponds to the $m_J = +1/2$ state being filled while the $m_J = -1/2$ is being depopulated. For this to happen, the Rb atom must be selectively undergoing transitions from the $\left|S_{1/2}, +1/2\right\rangle$ state to the $\left|P_{1/2}, -1/2\right\rangle$ state. This implies that the angular momentum carried by the photon in the atomic system must be -1. Consequently, the electric field vector of the laser light is rotating clockwise around the z-axis of the atomic system. To be clear, "rotating" is really just shorthand for "rotating at a fixed point as a function of time."

If the laser beam is travelling parallel to the holding field, then our work is done. We can conclude that the electric field vector is rotating clockwise around the laser beam propagation direction. Whether we call that "right" or "left" circularly polarized light is a matter of convention that is discussed later. What happens in the scenario where the laser beam is propagating antiparallel to the holding field? Recall that the z direction in the light system points antiparallel to the z direction in the atomic system. This means that the electric field vector is rotating counter-clockwise around the laser beam propagation direction, even though it is rotating clockwise around the holding field. This argument is depicted graphically in the upper and middle portions of Figs. (1) & (2).

5.2 "Helicity" vs. "Optics" Sign Convention

Now we can finally address the question of what "handedness" to label circularly polarized light: "right" or "left." One approach is to define a quantity called helicity, which is the sign of the projection of the angular momentum of the photon about the photon propagation direction $\vec{J}_\gamma$ onto the photon momentum $\vec{k}$:

$$h = \mathrm{sign}\left[\vec{J}_\gamma \cdot \vec{k}\right] \tag{26}$$

The helicity obeys the right hand rule: it is positive when the electric field vector rotates counter-clockwise about the light propagation direction. Once again, to be clear, by "rotate," we mean "rotating as *a function of time* at a fixed point." Let's call this the "helicity" convention. In this convention, it is natural to call light with positive (negative) helicity , "right" ("left") circularly polarized.

The other convention is the "standard optics" convention. In this case, we imagine how the electric field vector rotates as *a function of position* at a fixed time. Suppose we can "freeze" time and "look" at the electric field vector at different positions. If we were to connect that electric field vectors from location to location, we would end up with a "corkscrew" shape. A "right" handed laser beam in the helicity convention looks like a "left" handed corkscrew. The difference between these two conventions is depicted in Fig. (3).

6 The Magnitude of the Light Polarization

In this section, we use the notations and results of Apps. (A) & (B). One can measure the degree of circular polarization of a beam of light by rotating a beam splitting polarizing cube about the beam propagation direction. An input light polarization angle of θ wrt the cube axis is equivalent to having the cube axis be $-\theta$ from the light polarization $\mathcal{P}$ axis. Therefore varying θ is equivalent to rotating the cube. If the incident light is normal to the cube, then the intensity of the light transmitted through the cube is given by:

$$\begin{aligned}
I_t &= \left|\hat{\mathcal{C}}_t\left|E\right\rangle\right|^2 = t_1^2\left\langle E_{\mathcal{P}}|E_{\mathcal{P}}\right\rangle + t_2^2\left\langle E_{\mathcal{S}}|E_{\mathcal{S}}\right\rangle \\
&= \frac{E_0^2 T_t e_t}{1+e_t}\left(\frac{1-P+1+P+2\sqrt{1-P^2}\cos(2\theta)}{4}\right) + \frac{E_0^2 T_t}{1+e_t}\left(\frac{1-P+1+P-2\sqrt{1-P^2}\cos(2\theta)}{4}\right) \\
&= \frac{E_0^2 T_t}{2}\left[1+\left(\frac{e_t-1}{e_t+1}\right)\sqrt{1-P^2}\cos(2\theta)\right]
\end{aligned} \tag{27}$$

The maximum and minimum transmitted intensities are:

$$I_{\max} = \frac{E_0^2 T_t}{2}\left[1+\left(\frac{e_t-1}{e_t+1}\right)\sqrt{1-P^2}\right] \tag{28}$$

$$I_{\min} = \frac{E_0^2 T_t}{2}\left[1-\left(\frac{e_t-1}{e_t+1}\right)\sqrt{1-P^2}\right] \tag{29}$$

Defining the cube efficiency f_c and forming the cube asymmetry A_c:

$$f_c \equiv \frac{e_t-1}{e_t+1} \qquad \& \qquad A_c \equiv \frac{I_{\max}-I_{\min}}{I_{\max}+I_{\min}} = f_c\sqrt{1-P^2} \tag{30}$$

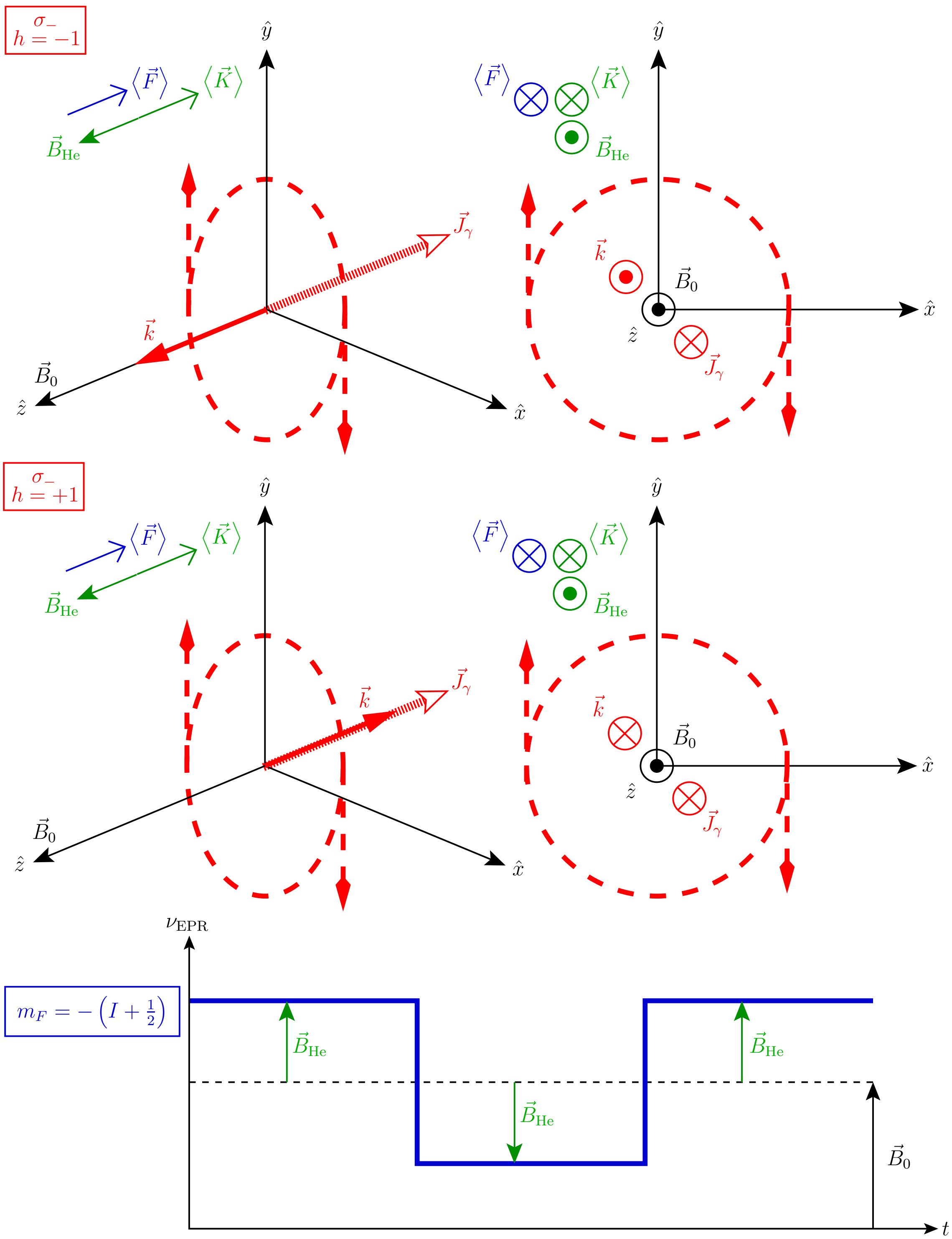

Figure 1: "Well" Spectrum. The field due to ^{3}He points parallel to the holding field. The sign of the ^{3}He and alkali polarizations are negative. The angular momentum of the light is antiparallel to the holding field. The EPR frequency shift measurement probes the $m_F = -(I+1/2) \leftrightarrow -(I-1/2)$ transition. Using the helicity convention, the upper (middle) figure represents "left" ("right") circularly polarized light travelling parallel (antiparallel) to the holding field.

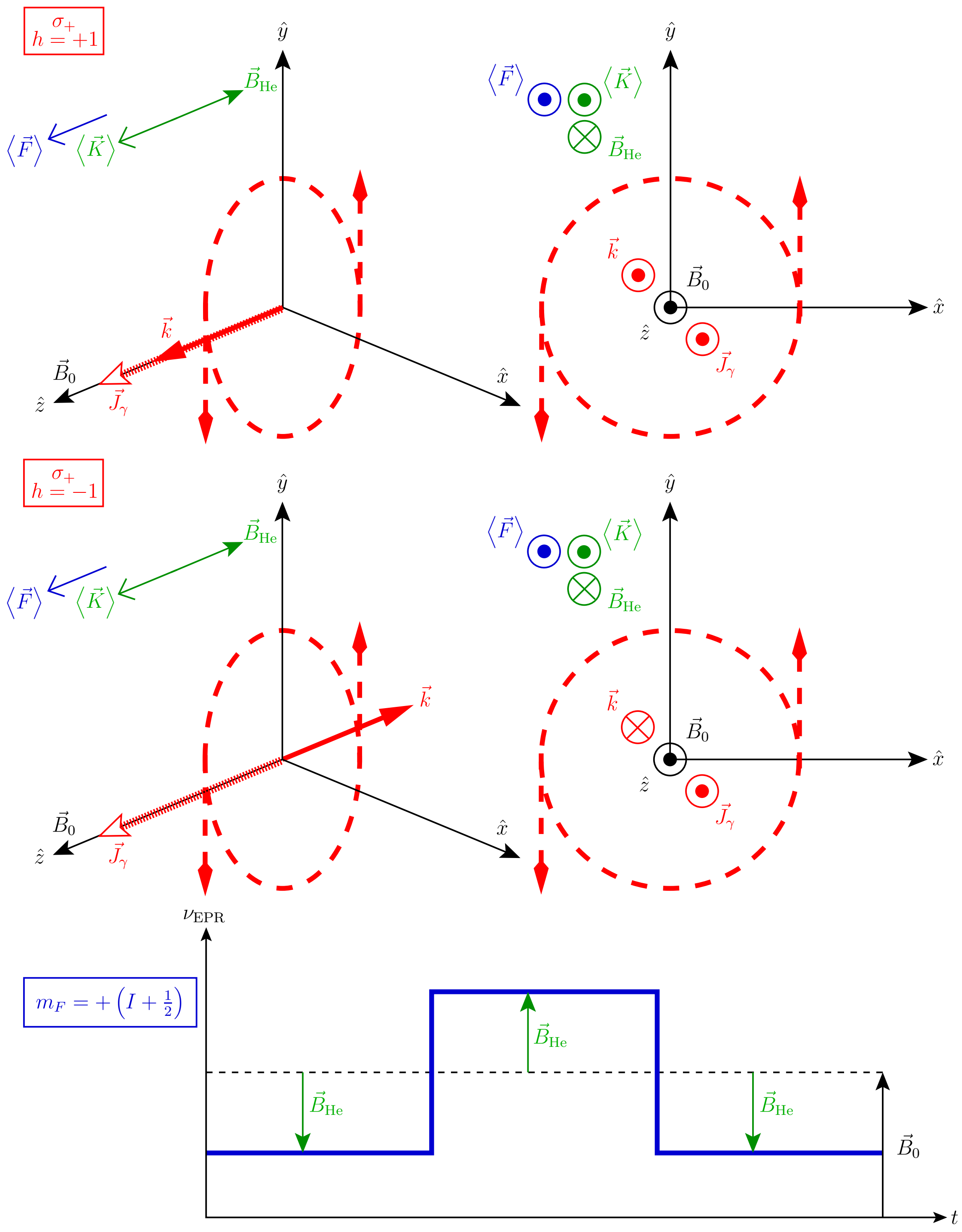

Figure 2: "Hat" Spectrum. The field due to ^{3}He points antiparallel to the holding field. The sign of the ^{3}He and alkali polarizations are positive. The angular momentum of the light is parallel to the holding field. The EPR frequency shift measurement probes the $m_F = +(I + 1/2) \leftrightarrow +(I - 1/2)$ transition. Using the helicity convention, the upper (middle) figure represents "right" ("left") circularly polarized light travelling parallel (antiparallel) to the holding field.

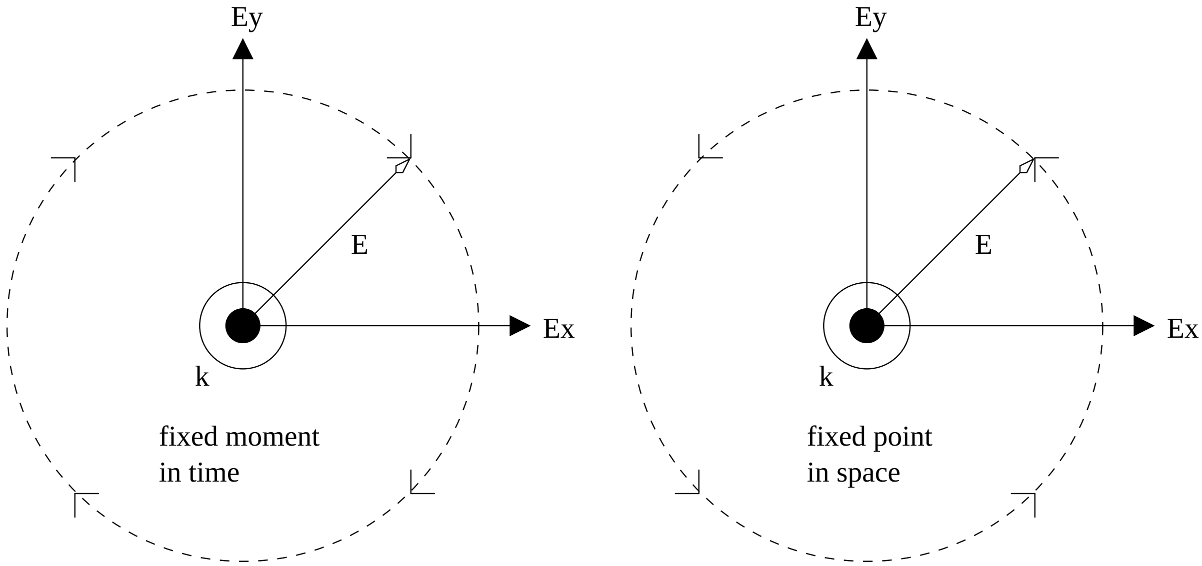

Figure 3: Right (helicity) circularly polarized light. Left: fixed time, forward in space. Right: fixed space, forward in time.

yields a "pythagorean" expression for polarization:

$$P^2 + \left(\frac{A_c}{f_c}\right)^2 = 1 \tag{31}$$

where P is the degree of circular polarization.

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A Describing Polarized Light

A.1 Complex Representation: The Jones Calculus

We will use the Jones convention for defining the polarization state of the light (vectors) and the action of the various optical elements (matrices). This convention uses complex number representation and a linear polarization basis. The electric field component of a monochromatic electromagnetic plane wave with propagation vector $\vec{k} = k\hat{z}$ at time t is:

$$\vec{E}(z,t) = E_x(z,t)\hat{x} + E_y(z,t)\hat{y} = |E\rangle\, e^{ikz-i\omega t} \tag{32}$$

$$E_x(z,t) = E_{0x}\exp(ikz - i\omega t + i\alpha_x) \tag{33}$$

$$E_y(z,t) = E_{0y}\exp(ikz - i\omega t + i\alpha_y) \tag{34}$$

$$|E\rangle \equiv \begin{bmatrix} E_{0x}e^{i\alpha_x} \\ E_{0y}e^{i\alpha_y} \end{bmatrix} \tag{35}$$

where the relative phase shift is $\alpha = \alpha_x - \alpha_y$. *Note that it is assumed that the real part of $\vec{E}$ is taken when the physical field is needed.* At a fixed point is space and over one period ($= \frac{2\pi}{\omega}$) in time, $\vec{E}$ sweeps out an ellipse in the xy-plane given by [5]:

$$\left(\frac{E_x}{E_{0x}}\right)^2 + \left(\frac{E_y}{E_{0y}}\right)^2 - 2\left(\frac{E_x}{E_{0x}}\right)\left(\frac{E_y}{E_{0y}}\right)\cos(\alpha) = \sin^2(\alpha) \tag{36}$$

In this representation, computing the modulus square of the electric field vector gives:

$$\vec{E}^* \cdot \vec{E} = \langle E|E\rangle = E_{0x}^2 + E_{0y}^2 \tag{37}$$

The time averaged modulus squared of electric field vector is therefore:

$$\left|\vec{E}\right|^2_{\text{time}} \equiv \frac{\vec{E}^* \cdot \vec{E}}{2} = \frac{E_{0x}^2 + E_{0y}^2}{2} \tag{38}$$

and finally the intensity is:

$$I = \sqrt{\frac{\epsilon}{\mu}}\left\langle \vec{E}^* \cdot \vec{E}\right\rangle_{\text{time}} = \sqrt{\frac{\epsilon}{\mu}}\frac{\langle E \mid E\rangle}{2} = \frac{\langle B \mid B\rangle}{2\mu\sqrt{\epsilon\mu}} \tag{39}$$

A.2 Linear Polarization Basis

For linear polarization, the relative phase shift is an integer multiple of half a wave,

$$\alpha = \pm n\pi \tag{40}$$

or in other words the two components are in phase. Eqn. (36) becomes degenerate,

$$\left(\frac{E_x}{E_{0x}}\right)^2 + \left(\frac{E_y}{E_{0y}}\right)^2 \mp 2\left(\frac{E_x}{E_{0x}}\right)\left(\frac{E_y}{E_{0y}}\right) = 0 \tag{41}$$

with solutions

$$\frac{E_y}{E_{0y}} = \mp\frac{E_x}{E_{0x}} \tag{42}$$

Two specific solutions are the orthogonal axes of the xy-plane which correspond to horizontal and vertical linearly polarized light. Horizontal linearly polarized light is denoted by

$$|\mathcal{P}\rangle = |x\rangle = \begin{bmatrix} 1 \\ 0 \end{bmatrix} \tag{43}$$

Vertical linearly polarized light is denoted by

$$|\mathcal{S}\rangle = |y\rangle = \begin{bmatrix} 0 \\ 1 \end{bmatrix} \tag{44}$$

Linear polarization at an angle θ counterclockwise from the x-axis is

$$|\theta\rangle = \begin{bmatrix} \cos(\theta) \\ \sin(\theta) \end{bmatrix} \tag{45}$$

A.3 Circular Polarization Basis

When the relative phase shift is a quarter wave,

$$\alpha = \pm(2n+1)\frac{\pi}{2} \tag{46}$$

and the magnitudes of the two components are identical,

$$E_{0x} = E_{0y} \tag{47}$$

then Eqn. (36) reduces to an equation for a circle:

$$E_x{}^2 + E_y{}^2 = 1 \tag{48}$$

The two orthogonal states are labeled by their helicity, namely the sign of the projection of the spin to the propagation vector. Right circularly polarized light,

$$|\mathcal{R}\rangle = |+\rangle = \frac{\sqrt{2}}{2}\begin{bmatrix} 1 \\ +i \end{bmatrix} \tag{49}$$

following the right hand rule such that the spin is parallel to the direction of propagation. Left circularly polarized light,

$$|\mathcal{L}\rangle = |-\rangle = \frac{\sqrt{2}}{2}\begin{bmatrix} 1 \\ -i \end{bmatrix} \tag{50}$$

is antiparallel. Note that the standard optics convention is opposite to the helicity convention. In the helicity convention, for right circularly polarized light, $\vec{E}$ rotates counterclockwise in the xy-plane at a fixed point in space. In the standard optics convention, for right circularly polarized light, $\vec{E}$ rotates counterclockwise in the xy-plane at a fixed moment in time as you move foward in the direction of propagation. See Fig. (3). Unless otherwise noted, *the helicity convention* will be used. See [6] for further discussion regarding handedness convention.

A.4 Stokes Parameters

Since the polarization vector of light has two components with complex coefficients, four real numbers are required to describe it completely. These real numbers are called Stokes parameters. Unfortunately many different conventions exist in the literature. For our purposes, the most useful convention in the circular polarization basis for arbitrarily polarized light is:

$$|E\rangle = E_0 e^{i\phi_p}\left[\sqrt{\frac{1+P}{2}}e^{-i\theta}|\mathcal{R}\rangle + \sqrt{\frac{1-P}{2}}e^{+i\theta}|\mathcal{L}\rangle\right] \tag{51}$$

where ϕ_p is just an overall phase factor that rarely contains any useful information about the light. Equivalently in the linear polarization basis, it is written as:

$$|E\rangle = E_0 e^{i\phi_p}\left[\left(\sqrt{1-P}\frac{e^{+i\theta}}{2} + \sqrt{1+P}\frac{e^{-i\theta}}{2}\right)|\mathcal{P}\rangle + \left(\sqrt{1-P}\frac{e^{+i\theta}}{2i} - \sqrt{1+P}\frac{e^{-i\theta}}{2i}\right)|\mathcal{S}\rangle\right] \tag{52}$$

The magnitude of $\vec{E}$ is:

$$\sqrt{\langle E|E\rangle} = \sqrt{\langle E_{\mathcal{R}}|E_{\mathcal{R}}\rangle + \langle E_{\mathcal{L}}|E_{\mathcal{L}}\rangle} = \sqrt{\left(\frac{1+P}{2}\right)E_0^2 + \left(\frac{1-P}{2}\right)E_0^2} = E_0 \tag{53}$$

The degree of circular polarization of the light is:

$$\frac{\langle E_{\mathcal{R}}|E_{\mathcal{R}}\rangle - \langle E_{\mathcal{L}}|E_{\mathcal{L}}\rangle}{\langle E|E\rangle} = \frac{\left(\frac{1+P}{2}\right)E_0^2 - \left(\frac{1-P}{2}\right)E_0^2}{E_0^2} = P \tag{54}$$

where $P = +(-)1$ for pure right (left) circular polarization and $P = 0$ for pure linear polarization. In the linear basis for pure linear polarization:

$$|E\rangle = E_0 e^{i\phi_p}\left[\left(\frac{e^{+i\theta}}{2} + \frac{e^{-i\theta}}{2}\right)|\mathcal{P}\rangle + \left(\frac{e^{+i\theta}}{2i} - \frac{e^{-i\theta}}{2i}\right)|\mathcal{S}\rangle\right] = E_0 e^{i\phi_p}\left[\cos(\theta)\,|\mathcal{P}\rangle + \sin(\theta)\,|\mathcal{S}\rangle\right] \tag{55}$$

where θ is the angle of the linear polarization vector with respect to the $|\mathcal{P}\rangle$-axis. In general for elliptically polarized light, θ is the angle that the major axis of the polarization ellipse makes with the $|\mathcal{P}\rangle$-axis.

A.5 Projecting onto an Atomic Coordinate System

The rectangular light coordinate system is defined by:

$$1_{\text{axis}} = |\mathcal{P}\rangle \qquad 2_{\text{axis}} = |\mathcal{S}\rangle \qquad 3_{\text{axis}} = |\mathcal{P}\rangle \times |\mathcal{S}\rangle = |\mathcal{Z}\rangle \tag{56}$$

where $|\mathcal{Z}\rangle$ is the light propagation direction. The rectangular atomic coordinate system is defined by:

$$1_{\text{axis}} = \hat{x} \qquad 2_{\text{axis}} = \hat{y} \qquad 3_{\text{axis}} = \hat{z} \tag{57}$$

where the z-axis is traditionally taken to be the quantization axis (direction of the main magnetic "holding" field). One useful way to decompose the light coordinates in the atomic coordinate representation is:

$$\begin{aligned}
|\mathcal{P}\rangle &= \cos(\Phi)\cos(\Theta)\hat{x} + \sin(\Phi)\cos(\Theta)\hat{y} - \sin(\Theta)\hat{z} && (58)\\
|\mathcal{S}\rangle &= -\sin(\Phi)\hat{x} + \cos(\Phi)\hat{y} && (59)\\
|\mathcal{Z}\rangle &= \cos(\Phi)\sin(\Theta)\hat{x} + \sin(\Phi)\sin(\Theta)\hat{y} + \cos(\Theta)\hat{z} && (60)\\
|\mathcal{R}\rangle &= \left[\cos(\Phi)\cos(\Theta) - i\sin(\Phi)\right]\frac{\hat{x}}{\sqrt{2}} + \left[\sin(\Phi)\cos(\Theta) + i\cos(\Phi)\right]\frac{\hat{y}}{\sqrt{2}} - \sin(\Theta)\frac{\hat{z}}{\sqrt{2}} && (61)\\
|\mathcal{L}\rangle &= \left[\cos(\Phi)\cos(\Theta) + i\sin(\Phi)\right]\frac{\hat{x}}{\sqrt{2}} + \left[\sin(\Phi)\cos(\Theta) - i\cos(\Phi)\right]\frac{\hat{y}}{\sqrt{2}} - \sin(\Theta)\frac{\hat{z}}{\sqrt{2}} && (62)
\end{aligned}$$

where Φ and Θ are azimuthal and polar angles of the $|\mathcal{Z}\rangle$ vector with respect to the spherical atomic coordinate system. #check#make a diagram depicting this. The light polarization vector couples to the atom most naturally in the irreducible spherical vector basis:

$$\hat{x} = \frac{\hat{\varepsilon}_- - \hat{\varepsilon}_+}{\sqrt{2}} \qquad \hat{y} = i\left(\frac{\hat{\varepsilon}_- + \hat{\varepsilon}_+}{\sqrt{2}}\right) \qquad \hat{z} = \hat{\varepsilon}_0 \tag{63}$$

Combining the projection and irreducible basis decomposition gives the following for the light coordinates:

$$\begin{aligned}
|\mathcal{P}\rangle &= -\sin(\Theta)\hat{\varepsilon}_0 - \exp(-i\Phi)\cos(\Theta)\frac{\hat{\varepsilon}_+}{\sqrt{2}} + \exp(+i\Phi)\cos(\Theta)\frac{\hat{\varepsilon}_-}{\sqrt{2}} && (64)\\
|\mathcal{S}\rangle &= i\exp(-i\Phi)\frac{\hat{\varepsilon}_+}{\sqrt{2}} + i\exp(+i\Phi)\frac{\hat{\varepsilon}_-}{\sqrt{2}} && (65)\\
|\mathcal{Z}\rangle &= +\cos(\Theta)\hat{\varepsilon}_0 - \exp(-i\Phi)\sin(\Theta)\frac{\hat{\varepsilon}_+}{\sqrt{2}} + \exp(+i\Phi)\sin(\Theta)\frac{\hat{\varepsilon}_-}{\sqrt{2}} && (66)\\
|\mathcal{R}\rangle &= -\sin(\Theta)\frac{\hat{\varepsilon}_0}{\sqrt{2}} - \exp(-i\Phi)\left[\frac{1+\cos(\Theta)}{2}\right]\hat{\varepsilon}_+ - \exp(+i\Phi)\left[\frac{1-\cos(\Theta)}{2}\right]\hat{\varepsilon}_- && (67)\\
|\mathcal{L}\rangle &= -\sin(\Theta)\frac{\hat{\varepsilon}_0}{\sqrt{2}} + \exp(-i\Phi)\left[\frac{1-\cos(\Theta)}{2}\right]\hat{\varepsilon}_+ + \exp(+i\Phi)\left[\frac{1+\cos(\Theta)}{2}\right]\hat{\varepsilon}_- && (68)
\end{aligned}$$

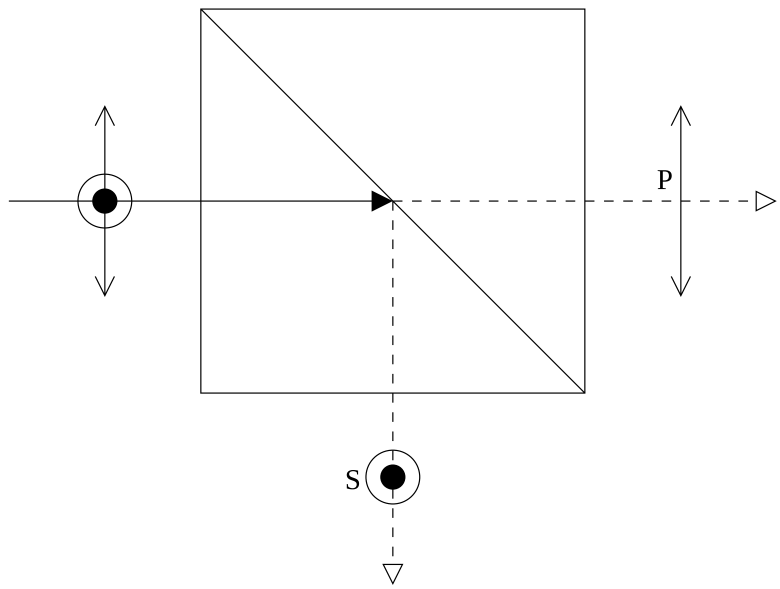

Figure 4: Top view of BSPC

Only a real vector can be decomposed in the spherical basis in a *consistent* way. For example, $|\mathcal{P}\rangle$, $|\mathcal{S}\rangle$, & $|\mathcal{Z}\rangle$ are all real vectors and $|\mathcal{R}\rangle$ & $|\mathcal{L}\rangle$ are complex vectors; therefore their decompostions using the complex conjugates of the irreducible basis are:

$$|\mathcal{P}\rangle = -\sin(\Theta)\hat{\varepsilon}_0^* - \frac{\cos(\Theta)}{\sqrt{2}}\exp(+i\Phi)\,\hat{\varepsilon}_+^* + \frac{\cos(\Theta)}{\sqrt{2}}\exp(-i\Phi)\,\hat{\varepsilon}_-^* \tag{69}$$

$$|\mathcal{S}\rangle = -\frac{i}{\sqrt{2}}\exp(+i\Phi)\,\hat{\varepsilon}_+^* - \frac{i}{\sqrt{2}}\exp(-i\Phi)\,\hat{\varepsilon}_-^* \tag{70}$$

$$|\mathcal{Z}\rangle = +\cos(\Theta)\hat{\varepsilon}_0^* - \frac{\sin(\Theta)}{\sqrt{2}}\exp(+i\Phi)\,\hat{\varepsilon}_+^* + \frac{\sin(\Theta)}{\sqrt{2}}\exp(-i\Phi)\,\hat{\varepsilon}_-^* \tag{71}$$

$$|\mathcal{R}\rangle = -\frac{\sqrt{2}}{2}\sin(\Theta)\hat{\varepsilon}_0^* + \left[\frac{1-\cos(\Theta)}{2}\right]\exp(+i\Phi)\,\hat{\varepsilon}_+^* + \left[\frac{1+\cos(\Theta)}{2}\right]\exp(-i\Phi)\,\hat{\varepsilon}_-^* \tag{72}$$

$$|\mathcal{L}\rangle = -\frac{\sqrt{2}}{2}\sin(\Theta)\hat{\varepsilon}_0^* - \left[\frac{1+\cos(\Theta)}{2}\right]\exp(+i\Phi)\,\hat{\varepsilon}_+^* - \left[\frac{1-\cos(\Theta)}{2}\right]\exp(-i\Phi)\,\hat{\varepsilon}_-^* \tag{73}$$

Note the subtle difference in the two decompositions of $|\mathcal{R}\rangle$ & $|\mathcal{L}\rangle$.

B Polarization Optics

B.1 Beam Splitting Polarizing Cubes

An ideal beam splitting polarizing cube (BSPC) simply splits an incoming beam into it's two linearly polarized components. Once separated, the two beam paths are orthogonal, see Fig. (4). The transmitted beam is selected by

$$\hat{C}_t = \begin{bmatrix} 1 & 0 \\ 0 & 0 \end{bmatrix} \tag{74}$$

and the reflected beam is selected by

$$\hat{C}_r = \begin{bmatrix} 0 & 0 \\ 0 & 1 \end{bmatrix} \tag{75}$$

For the ideal case, the transmitted and reflected beams are pure $\mathcal{P}$ & $\mathcal{S}$ linear polarizations repectively. In practice the splitting and polarizing are imperfect. According to RMI (Dr. Zhiming Lu, zlu@rmico.com,

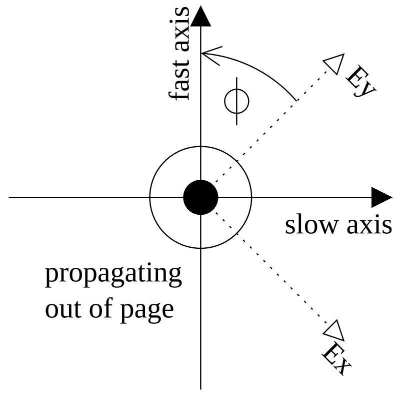

Figure 5: Coordinate System of a Waveplate #check#time or space convention fast axis?

Rocky Mountain Instruments, 106 Laser Drive, Lafayette, CO, 80026, 303-664-5000), our 2" BSPC has an extinction ratio for the transmitted beam of $\geq 1000:1$ whereas for the reflected beam it is $\leq 20:1$. The transmittance is about $\geq 95\%$, whereas the reflectance is about $\geq 99.9\%$. Therefore a more realistic form of $\hat{C}$ can be written. For example, for the transmitted beam:

$$\hat{C}_t = \begin{bmatrix} t_1 & 0 \\ 0 & t_2 \end{bmatrix} \tag{76}$$

$$T_t = \frac{I_{\text{transmitted}}}{I_{\text{input}\mathcal{P}}} = t_1^2 + t_2^2 \tag{77}$$

$$e_t = \frac{I_{\text{transmitted}\mathcal{P}}}{I_{\text{transmitted}\mathcal{S}}} = \frac{t_1^2}{t_2^2} \tag{78}$$

where t is the transmittance and e_t is the extinction ratio for the transmitted beam. Solving for t_1 & t_2 in terms of t & e_t and doing the same for the reflected beam, the more general cube matrices become:

$$\hat{C}_t = \begin{bmatrix} \sqrt{\frac{T_t}{1+e_t^{-1}}} & 0 \\ 0 & \sqrt{\frac{T_t}{1+e_t}} \end{bmatrix} \tag{79}$$

$$\hat{C}_r = \begin{bmatrix} \sqrt{\frac{T_r}{1+e_r}} & 0 \\ 0 & \sqrt{\frac{T_r}{1+e_r^{-1}}} \end{bmatrix} \tag{80}$$

Given the specifications for our cube, the matrices are:

$$\hat{C}_t \approx \begin{bmatrix} 0.974 & 0 \\ 0 & 0.031 \end{bmatrix} \tag{81}$$

$$\hat{C}_r \approx \begin{bmatrix} 0.213 & 0 \\ 0 & 0.951 \end{bmatrix} \tag{82}$$

The fully general cube matrices could be, in principle, complex and have nonzero off diagonal elements.

B.2 Matrix Representation of Waveplates

A waveplate is an optical element that has different indices of refraction along two orthogonal axes, see Fig. (5). This results in a net phase shift between the linear components of the polarization vector. First, the polarization vector has to be expressed in the basis of the waveplate. Therefore, a passive or coordinate system rotation of angle ϕ radians is performed,

$$\hat{R}(\phi) = \begin{bmatrix} \cos(\phi) & \sin(\phi) \\ -\sin(\phi) & \cos(\phi) \end{bmatrix} \tag{83}$$

followed by a relative phase retardation of β radians,

$$\hat{W}(\beta) = \begin{bmatrix} \exp\left(+i\frac{\beta}{2}\right) & 0 \\ 0 & \exp\left(-i\frac{\beta}{2}\right) \end{bmatrix} \tag{84}$$

and finally a rotation back to the orignal basis, $\hat{R}(-\phi)$. The complete waveplate operator is thus:

$$\begin{aligned} \hat{W}(\phi,\beta) &= \hat{R}(-\phi)\hat{W}(\beta)\hat{R}(\phi) &(85)\\ &= \exp\left(-i\frac{\beta}{2}\right)\begin{bmatrix} 1+2i\exp\left(i\frac{\beta}{2}\right)\sin\left(\frac{\beta}{2}\right)\cos^2(\phi) & i\exp\left(i\frac{\beta}{2}\right)\sin\left(\frac{\beta}{2}\right)\sin(2\phi) \\ i\exp\left(i\frac{\beta}{2}\right)\sin\left(\frac{\beta}{2}\right)\sin(2\phi) & 1+2i\exp\left(i\frac{\beta}{2}\right)\sin\left(\frac{\beta}{2}\right)\sin^2(\phi) \end{bmatrix} &(86) \end{aligned}$$

Note that for one complete wave, $\beta = 2\pi$. Typically the fast axis is taken to be vertical.

B.3 Half Waveplate

A half-waveplate has a retardance $\beta = \frac{2\pi}{2} = \pi$. When it is orientated at an angle of ϕ from a set of reference axes, the waveplate matrix becomes:

$$\hat{W}_{\frac{1}{2}}(\phi) = i\begin{bmatrix} \cos(2\phi) & \sin(2\phi) \\ \sin(2\phi) & -\cos(2\phi) \end{bmatrix} \tag{87}$$

This operation implies that each linear polarization component of some arbitrarily polarized light is rotated by twice the angle between the linear polarization axis and the waveplate fast axis. If the the linear polarization is either $\mathcal{S}$ or $\mathcal{P}$, then a half-waveplate at an angle ϕ with respect to the polarization axis rotates the linear polarization by an angle of 2ϕ. A half-waveplate at $\pm 45^o$ simplify flips $\mathcal{P} \leftrightarrow \mathcal{S}$. For pure circularly polarized light, a half-waveplate orientated at *any* angle simply flips $\mathcal{L} \leftrightarrow \mathcal{R}$.

B.4 Quarter Waveplate

For a quarter-waveplate with retardance $\beta = \frac{2\pi}{4} = \frac{\pi}{2}$, orientated at an angle of 45^o, the matrix becomes:

$$\hat{W}_{\frac{1}{4}}\left(\frac{\pi}{4}\right) = \frac{\sqrt{2}}{2}\begin{bmatrix} 1 & i \\ i & 1 \end{bmatrix} \tag{88}$$

To be explicit, a quarter-waveplate with its fast axis rotated counterclockwise by 45^o turns horizontal linearly polarized light into right circularly polarized light,

$$\begin{aligned} \hat{W}_{\frac{1}{4}}\left(\frac{\pi}{4}\right)|\mathcal{P}\rangle &= |\mathcal{R}\rangle &(89)\\ \hat{W}_{\frac{1}{4}}\left(\frac{\pi}{4}\right)|\mathcal{R}\rangle &= i\,|\mathcal{S}\rangle &(90)\\ \hat{W}_{\frac{1}{4}}\left(\frac{\pi}{4}\right)|\mathcal{S}\rangle &= i\,|\mathcal{L}\rangle &(91)\\ \hat{W}_{\frac{1}{4}}\left(\frac{\pi}{4}\right)|\mathcal{L}\rangle &= |\mathcal{P}\rangle &(92) \end{aligned}$$

and so forth following the simple pattern $\mathcal{P} \to \mathcal{R} \to \mathcal{S} \to \mathcal{L} \to \mathcal{P}$. An angle of -45^o simply reverses the direction of the arrows. Note that in the RHS of the two middle equations, there is an overall phase factor (i) which for our purposes is unimportant.

C Physical Constants and Alkali Data

Symbol	Value	Units	Description
g_e	$-2.002\ 319\ 304\ 372$	unitless	electron g-factor
g_K	$-4.254\ 995\ 436$	unitless	^{3}He nuclear g-factor
μ_B	$9.274\ 000\ 95 \times 10^{-24}$	$\mathrm{J \cdot T^{-1}}$	Bohr magneton
μ_N	$5.050\ 783\ 4 \times 10^{-27}$	$\mathrm{J \cdot T^{-1}}$	Nuclear magneton
c	$299\ 792\ 458$	$\mathrm{m \cdot s^{-1}}$	definition of the speed of light
ϵ_0	$8.854\ 187\ 817 \times 10^{-12}$	$\mathrm{C^2 \cdot N^{-1} \cdot m^{-2}}$	permittivity of free space
μ_0	$4\pi \times 10^{-7}$	$\mathrm{N \cdot A^{-2}}$	permeability of free space
h	$6.626\ 069 \times 10^{-34}$	$\mathrm{J \cdot s}$	Planck constant
amu	$1.660\ 538\ 9 \times 10^{-27}$	kg	12·(atomic mass unit) = mass ^{12}C

Table 1: Fundamental Physical Constants. [7]

Isotope	Mass (amu)	Natural Abundance	Nuclear Spin, I	Magnetic Moment (μ_N)	g-factor $g_I(\mu_N)$
^{6}Li	6.015 122 3	0.075 9	1	+0.822 056	+0.822 056
^{7}Li	7.016 004 0	0.924 1	3/2	+3.256 44	+2.170 96
^{23}Na	22.989 769 7	1.0	3/2	+2.217 52	+1.478 35
^{39}K	38.963 706 9	0.932 58	3/2	+0.391 46	+0.260 97
^{40}K	39.963 998 7	0.000 117	4	−1.298	−0.324 5
^{41}K	40.961 826 0	0.067 30	3/2	+0.214 87	+0.143 25
^{85}Rb	84.911 789	0.721 7	5/2	+1.353 02	+0.541 208
^{87}Rb	86.909 184	0.278 3	3/2	+2.751 2	+1.834 1
^{133}Cs	132.905 447	1.0	7/2	+2.579	+0.736 9
Reference	[8]				Eqn. (7)

Table 2: Alkali atom isotopic and nuclear data.