Dipole Moment and Line Strengths

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June 2024

1 Basic Definitions

Basic equation for line strength:

$$\mathrm{S}(\tau', \mathrm{J}'; \tau'', \mathrm{J}'') \propto \sum_{M'} \sum_{M''} \left| \langle \tau' J' M' | \vec{E} \cdot \vec{\mu} | \tau'' J'' M'' \rangle \right|^2 \tag{1}$$

$|\tau JM\rangle$ can be expanded in the basis of symmetric rotor wavefunctions:

$$|\tau JM\rangle = \sum_K a_{\tau,K} \, |JKM\rangle \tag{2}$$

2 Dipole moment transformations

We can think of the dipole moment in multiple different coordinate bases, such as cartesian and spherical. The dipole moment can be calculated in a molecule-fixed cartesian basis from electronic structure, which we will call $\vec{\mu_q}$. However, computations of the line strength require the usage of μ as a spherical tensor operator, such that we can transform from the molecular axes to space fixed axes.

In the spherical basis, we define the following two quantities, and their relation:

- $\mu_{\mathbf{p}}^{(\mathbf{1})}$ - the space fixed coordinate system
- $\mu_{\mathbf{q}}^{(\mathbf{1})}$ - the molecular fixed axes
- $\mu_{\mathbf{p}}^{(\mathbf{1})} = \sum_q D_{pq}^{(1)\dagger} \mu_{\mathbf{q}}^{(\mathbf{1})}$

To begin, we will redefine our transformation between $\mu_{\mathbf{p}}^{(\mathbf{1})}$ and $\mu_{\mathbf{q}}^{(\mathbf{1})}$ using matrix notation, to simplify the math.

$$\mu_{\mathbf{p}}^{(\mathbf{1})} = \hat{X} \mu_{\mathbf{q}}^{(\mathbf{1})} \quad \text{with} \quad X_{pq} = D_{pq}^{(1)} \quad p, q \in [-1, 1] \tag{3}$$

$\hat{X}$ can be shown to be unitary, therefore $\hat{X}^{-1} \equiv \hat{X}^\dagger$ and $\langle \psi | \hat{X}^{-1} \hat{X} | \psi \rangle \equiv \langle \psi | \psi \rangle$

Next, we define a coordinate transform operator, which maps from the cartesian basis to the spherical basis (think about spherical harmonics!)

$$\begin{pmatrix} -1 \\ 0 \\ 1 \end{pmatrix} \mu_{\mathbf{q}}^{(\mathbf{1})} = \hat{T} \vec{\mu_q} \quad \text{where} \quad \hat{T} = \begin{bmatrix} \frac{\sqrt{2}}{2} & -i\frac{\sqrt{2}}{2} & 0 \\ 0 & 0 & 1 \\ -\frac{\sqrt{2}}{2} & -i\frac{\sqrt{2}}{2} & 0 \end{bmatrix} \tag{4}$$

Again, T is a unitary operator.

Using these operators, we can find $\vec{\mu_p}$ from $\vec{\mu_q}$ as follows:

$$\vec{\mu_p} = \hat{T}^\dagger \hat{X} \hat{T} \vec{\mu_q} \tag{5}$$

For completeness, we document the following expression for $\hat{T}^\dagger \hat{X} \hat{T}$, noting that it again is a unitary operator:

$$\begin{bmatrix} \frac{1}{2}\left[D^{(1)\dagger}_{-1,-1} - D^{(1)\dagger}_{-1,1} - D^{(1)\dagger}_{1,-1} + D^{(1)\dagger}_{1,1}\right] & -\frac{i}{2}\left[D^{(1)\dagger}_{-1,-1} - D^{(1)\dagger}_{-1,1} - D^{(1)\dagger}_{1,-1} + D^{(1)\dagger}_{1,1}\right] & \frac{\sqrt{2}}{2}\left[D^{(1)\dagger}_{-1,0} - D^{(1)\dagger}_{-1,0}\right] \\ \frac{i}{2}\left[D^{(1)\dagger}_{-1,-1} - D^{(1)\dagger}_{-1,1} + D^{(1)\dagger}_{1,-1} - D^{(1)\dagger}_{1,1}\right] & \frac{1}{2}\left[D^{(1)\dagger}_{-1,-1} + D^{(1)\dagger}_{-1,1} + D^{(1)\dagger}_{1,-1} - D^{(1)\dagger}_{1,1}\right] & i\frac{\sqrt{2}}{2}\left[D^{(1)\dagger}_{-1,0} + D^{(1)\dagger}_{-1,0}\right] \\ \frac{\sqrt{2}}{2}\left[D^{(1)\dagger}_{0,-1} - D^{(1)\dagger}_{0,1}\right] & -i\frac{\sqrt{2}}{2}\left[D^{(1)\dagger}_{0,-1} + D^{(1)\dagger}_{0,1}\right] & D^{(1)\dagger}_{0,0} \end{bmatrix} \tag{6}$$

3 Line Strength Operators

The line strength is given by the expectation value of the following quantity:

$$S \propto \sum_{M'} \sum_{M''} \left| \langle \psi | \vec{E}(t) \cdot \vec{\mu_p} | \psi \rangle \right|^2 \tag{7}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \langle \psi | \vec{E}(t) \cdot \hat{T}^\dagger \hat{X} \hat{T} \vec{\mu_q} | \psi \rangle \right|^2 \tag{8}$$

Choosing $\vec{E}(t)$ to be polarized along the $Z-$axis, we rewrite it as $E_0 \cos{(\Omega t)} \hat{Z}$, and take the time average:

$$S \propto \sum_{M'} \sum_{M''} E_0^2 \left\langle \cos{(\Omega t)}^2 \right\rangle_t \left| \langle \psi | \hat{Z} \cdot \hat{T}^\dagger \hat{X} \hat{T} \vec{\mu_q} | \psi \rangle \right|^2 \tag{9}$$

$$S \propto \sum_{M'} \sum_{M''} \frac{E_0^2}{2} \left| \langle \psi | \hat{Z} \cdot \hat{T}^\dagger \hat{X} \hat{T} \vec{\mu_q} | \psi \rangle \right|^2 \tag{10}$$

Since we are only considering the proportionality, we drop $E_0/2$ in the remainder of the discussion.

$$S \propto \sum_{M'} \sum_{M''} \left| \langle \psi | \left[\frac{\sqrt{2}}{2} \left[D^{(1)\dagger}_{0,-1} - D^{(1)\dagger}_{0,1} \right] \mu_{\mathbf{x}} - i\frac{\sqrt{2}}{2} \left[D^{(1)\dagger}_{0,-1} + D^{(1)\dagger}_{0,1} \right] \mu_{\mathbf{y}} + D^{(1)\dagger}_{0,0} \mu_{\mathbf{z}} \right] | \psi \rangle \right|^2 \tag{11}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \langle \psi | \left[\frac{\sqrt{2}}{2} \left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}} \right] D^{(1)\dagger}_{0,-1} - \frac{\sqrt{2}}{2} \left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}} \right] D^{(1)\dagger}_{0,1} + D^{(1)\dagger}_{0,0} \mu_{\mathbf{z}} \right] | \psi \rangle \right|^2 \tag{12}$$

In addition, we take advantage of the adjoint behavior of wigner D matrices:

$$D^{(j)}_{-m-k} = (-1)^{k-m} D^{(j)\dagger}_{mk} \tag{13}$$

$$D^{(j)\dagger}_{mk} = (-1)^{m-k} D^{(j)}_{-m-k} \tag{14}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \langle \psi | \left[\frac{-\sqrt{2}}{2} \left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}} \right] D^{(1)}_{0,1} + \frac{\sqrt{2}}{2} \left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}} \right] D^{(1)}_{0,-1} + D^{(1)}_{0,0} \mu_{\mathbf{z}} \right] | \psi \rangle \right|^2 \tag{15}$$

For simplicity, the operator expression will be called $\hat{Q}$

4 Evaluating the Line Strengths

We choose the symmetry adapted basis set:

$$|JKMs\rangle = \sqrt{\frac{(-1)^s}{2(1+\delta_{K,0})}} \Big[|JKM\rangle + (-1)^{s+J} |J-KM\rangle \Big] \tag{16}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \langle J''K''M''s''|\hat{Q}|J'K'M's'\rangle \right|^2 \tag{17}$$

For an asymmetric rotor,

$$|J\tau Ms\rangle = \sum_{K=0}^{J} \alpha_{K,\tau}^{(J)} \sqrt{\frac{(-1)^s}{2(1+\delta_{K,0})}} \Big[|JKM\rangle + (-1)^{s+J} |J-KM\rangle \Big] \tag{18}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \sum_{K'} \sum_{K''} \alpha_{K',\tau',s'}^{(J')\dagger} \alpha_{K'',\tau'',s''}^{(J'')\dagger} \langle J'K'M's'|\hat{Q}|J''K''M''s''\rangle \right|^2 \tag{19}$$

5 Derivation of symmetric rotor line strengths

In order to calculate the intensities, we need to calculate eqn. (17), starting by expanding out the states:

For simplicity's sake, we define the following coefficient:

$$\beta_{s,K} = \sqrt{\frac{(-1)^s}{2(1+\delta_{K,0})}} \tag{20}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \beta^{\dagger}_{s',K'} \beta_{s'',K''} \left[\langle J'K'M'| + (-1)^{s'+J'} \langle J' - K'M'| \right] \hat{Q} \left[|J''K''M''\rangle + (-1)^{s''+J''} |J'' - K''M''\rangle \right] \right|^2 \tag{21}$$

Recall foil:

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \sum_{M'} \sum_{M''} \Big| \langle J'K'M'|\hat{Q}|J''K''M''\rangle + (-1)^{s'+J'} \langle J' - K'M'|\hat{Q}|J''K''M''\rangle$$
$$+ (-1)^{s''+J''} \langle J'K'M'|\hat{Q}|J'' - K''M''\rangle + (-1)^{s'+J'+s''+J''} \langle J' - K'M'|\hat{Q}|J'' - K''M''\rangle \Big|^2 \tag{22}$$

We will now derive a generic expression for a matrix element of the kind above, using the wavefunctions as follows:

$$\langle \mathbb{R}_3|\Psi\rangle = |JKM\rangle = \left[\frac{2J+1}{8\pi^2}\right]^{\frac{1}{2}} D^{(J)\dagger}_{MK}(\mathbb{R}_3) \tag{23}$$

$$\langle \mathbb{R}_3|\Psi\rangle = |JKM\rangle = (-1)^{M-K} \left[\frac{2J+1}{8\pi^2}\right]^{\frac{1}{2}} D^{(J)}_{-M-K}(\mathbb{R}_3) \tag{24}$$

$$\langle J'K'M'|\hat{Q}|J''K''M''\rangle =$$
$$(-1)^{M''-K''} \left[\frac{2J'+1}{8\pi^2}\right]^{\frac{1}{2}} \left[\frac{2J''+1}{8\pi^2}\right]^{\frac{1}{2}} \int_{\mathbb{R}} D^{(J')}_{M'K'} \left[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] D^{(1)}_{0,1} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] D^{(1)}_{0,-1} + D^{(1)}_{0,0}\mu_{\mathbf{z}} \right] D^{(J'')}_{-M''-K''} \mathrm{d}\Omega \tag{25}$$

We tabulate the following:

$$\int_{\mathbb{R}} D^{(J')}_{M'K'} D^{(1)}_{0,1} D^{(J'')}_{-M''-K''} \mathrm{d}\Omega = 8\pi^2 \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} \tag{26}$$

$$\int_{\mathbb{R}} D^{(J')}_{M'K'} D^{(1)}_{0,-1} D^{(J'')}_{-M''-K''} \mathrm{d}\Omega = 8\pi^2 \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} \tag{27}$$

$$\int_{\mathbb{R}} D^{(J')}_{M'K'} D^{(1)}_{0,0} D^{(J'')}_{-M''-K''} \mathrm{d}\Omega = 8\pi^2 \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \tag{28}$$

Substituting these in, we find the following:

$$\langle J'K'M'|\hat{Q}|J''K''M''\rangle = (-1)^{M''-K''} [2J'+1]^{\frac{1}{2}} [2J''+1]^{\frac{1}{2}} \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix} \cdot$$
$$\left[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \right] \tag{29}$$

Now putting it all together:

$$
\begin{aligned}
S \propto\ & \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 [2J'+1]\,[2J''+1] \sum_{M'}\sum_{M''} \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix}^2 \Bigg| (-1)^{M''-K''} \Bigg[\\
& \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \right] \\
& + (-1)^{s'+J'} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & -K' \end{pmatrix} \right] \\
& + (-1)^{s''+J''+2K''} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \right] \\
& + (-1)^{s'+J'+s''+J''+2K''} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & -K' \end{pmatrix} \right] \Bigg] \Bigg|^2
\end{aligned} \tag{30}
$$

This is the divergence point for an asymmetric rotor. From here we cancel out (-1) raised to two times an integer, evaluate the M 3j-symbol, and flip 3j coefficients such we can match pairs.

Recall:

$$
\begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} = (-1)^{j_1+j_2+j_3} \begin{pmatrix} j_1 & j_2 & j_3 \\ -m_1 & -m_2 & -m_3 \end{pmatrix} \tag{31}
$$

$$
\begin{aligned}
S \propto\ & \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 \frac{[2J'+1]\,[2J''+1]}{3} \Bigg| \\
& \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \right] \\
& + (-1)^{s'+2J'+J''+1} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \right] \\
& + (-1)^{s''+J''} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \right] \\
& + (-1)^{s'+2J'+s''+2J''+1} \left[\frac{-\sqrt{2}}{2}\,[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \right] \Bigg|^2
\end{aligned} \tag{32}
$$

Getting rid of 2x quantum numbers and going wild with colors...

$$
\begin{aligned}
S \propto & \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 \frac{[2J'+1]\,[2J''+1]}{3} \Bigg| \\
& \left[\frac{-\sqrt{2}}{2}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\right] \\
& - (-1)^{s'+J''}\left[\frac{-\sqrt{2}}{2}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\right] \\
& + (-1)^{s''+J''}\left[\frac{-\sqrt{2}}{2}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\right] \\
& - (-1)^{s'+s''}\left[\frac{-\sqrt{2}}{2}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\right]\Bigg|^2
\end{aligned} \tag{33}
$$

Combining like colors:

$$
\begin{aligned}
S \propto & \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 \frac{[2J'+1]\,[2J''+1]}{3} \Bigg| \\
& - \frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix}\Big[\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right] + (-1)^{s'+s''}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\Big] \\
& + \frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix}\Big[\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right] + (-1)^{s'+s''}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\Big] \\
& + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Big[1 - (-1)^{s'+s''}\Big] \\
& + \frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix}\Big[(-1)^{s'+J''}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right] + (-1)^{s''+J''}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right]\Big] \\
& - \frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix}\Big[(-1)^{s'+J''}\left[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}\right] + (-1)^{s''+J''}\left[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}\right]\Big] \\
& - \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Big[(-1)^{s'+J''} - (-1)^{s''+J''}\Big]\Bigg|^2
\end{aligned} \tag{34}
$$

The requirements for the different 3j-symbols to contribute are:

- $-K'' + 1 + K' = 0 \rightarrow K' - K'' = -1 \rightarrow \Delta K = -1$
- $-K'' - 1 + K' = 0 \rightarrow K' - K'' = 1 \rightarrow \Delta K = +1$
- $-K'' + K' = 0 \rightarrow K' = K'' \rightarrow \Delta K = 0$
- $K'' - 1 + K' = 0 \rightarrow K' = 1 - K'' \rightarrow$ only allows $1 \rightarrow 0$ and $0 \rightarrow 1$
- $K'' + 1 + K' = 0 \rightarrow K' = -1 - K'' \rightarrow$ FORBIDDEN
- $K'' + 0 + K' = 0 \rightarrow K' = -K'' \rightarrow \Delta K = 0, \quad K'' = K' = 0$

5.1 $\Delta K = 0$

5.1.1 Case $K' = K'' \neq 0$:

$$
\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \neq 0 \quad \text{and} \quad \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \equiv 0 \tag{35}
$$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}^2 \left| \left[1 - (-1)^{s'+s''}\right] \right|^2 \tag{36}$$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, [2J'+1]\,[2J''+1] \, \frac{4}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}^2 \, [1 - \delta_{s',s''}] \tag{37}$$

Now, if $s' = s''$, then $S = 0$

5.1.2 Case $K' = K'' = 0$:

$$\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \neq 0 \quad \text{and} \quad \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \neq 0 \tag{38}$$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| \left[1 - (-1)^{s'+s''} - \left[(-1)^{s'} - (-1)^{s''}\right] (-1)^{J''}\right] \right|^2 \tag{39}$$

1. $s' \neq s''$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| \left[1 + 1 - [1+1]\,(-1)^{J''+s'}\right] \right|^2 \tag{40}$$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{4\,[2J'+1]\,[2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| \left[1 - (-1)^{J''+s'}\right] \right|^2 \tag{41}$$

2. $s' = s''$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| \left[1 - 1 - [0]\,(-1)^{J''}\right] \right|^2 \tag{42}$$

$$S \propto 0 \tag{43}$$

Again, if $s' = s''$, then $S = 0$

5.2 $\Delta K = -1$

5.2.1 Case $K'' \neq 1$:

$$\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} \neq 0 \quad \text{and} \quad \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & (K''-1) \end{pmatrix} \equiv 0 \tag{44}$$

$$S \propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \left| -\frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] \right|^2 \tag{45}$$

$$\propto |\beta_{s',K'}|^2 \, |\beta_{s'',K''}|^2 \, \frac{[2J'+1]\,[2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \frac{1}{2} \left| \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''}\,[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] \right|^2 \tag{46}$$

We now examine the cases that $s'' = s'$ and $s'' \neq s'$
1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \frac{1}{2} \left| \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{2s'} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right] \right|^2 \tag{47}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \frac{1}{2} \left| [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}} + \mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right|^2 \tag{48}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 {\mu_{\mathbf{x}}}^2 \tag{49}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \frac{1}{2} \left| [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^1 [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] \right|^2 \tag{50}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \frac{1}{2} \left| [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}} - \mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \right|^2 \tag{51}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \tag{52}$$

5.2.2 Case $K'' = 1$:

$$\begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & (1-1) \end{pmatrix} \neq 0 \quad \text{and} \quad \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & (1-1) \end{pmatrix} \neq 0 \tag{53}$$

$$\begin{aligned} S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \Bigg| \\ -\frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix} \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right] \\ + \frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0 \end{pmatrix} \left[(-1)^{s'+J''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''+J''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right] \Bigg|^2 \end{aligned} \tag{54}$$

Recall eqn. (31):

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \Bigg| - \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix} \Big[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] - (-1)^{J''+J'} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix} \Big[(-1)^{s'+J''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''+J''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] \Bigg|^2 \tag{55}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Bigg| \Big[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] + (-1)^{J''+J'} \Big[(-1)^{s'+J''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''+J''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] \Bigg|^2 \tag{56}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Bigg| \Big[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] + (-1)^{J'} \Big[(-1)^{s'} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] \Bigg|^2 \tag{57}$$

We now examine the cases that $s'' = s'$ and $s'' \neq s'$

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Bigg| \Big[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{2s'} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] + (-1)^{J'} \Big[(-1)^{s'} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \Big] \Bigg|^2 \tag{58}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Bigg| [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] + (-1)^{J'+s'} [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] \Bigg|^2 \tag{59}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Big| [2\mu_{\mathbf{x}}] + (-1)^{J'+s'} [2\mu_{\mathbf{x}}] \Big|^2 \tag{60}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 {\mu_{\mathbf{x}}}^2 \Big| 1 + (-1)^{J'+s'} \Big|^2 \tag{61}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Big| \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^1 [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] + (-1)^{J'+s'} \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''-s'} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] \Big|^2 \tag{62}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Big| [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] - [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] + (-1)^{J'+s'} [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] - [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] \Big|^2 \tag{63}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \Big| -2i\mu_{\mathbf{y}} + (-1)^{J'+s'} [-2i\mu_{\mathbf{y}}] \Big|^2 \tag{64}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \Big| 1 + (-1)^{J'+s'} \Big|^2 \tag{65}$$

5.3 $\Delta K = +1$

5.3.1 Case $K'' \neq 0$:

$$\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} \neq 0 \quad \text{and} \quad \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} \equiv 0 \tag{66}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \Big| \left[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\right] \Big|^2 \tag{67}$$

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \Big| \left[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{2s'} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\right] \Big|^2 \tag{68}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \Big| [[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]] \Big|^2 \tag{69}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 |2\mu_{\mathbf{x}}|^2 \tag{70}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 {\mu_{\mathbf{x}}}^2 \tag{71}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \left| [[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^1 [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]] \right|^2 \tag{72}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \left| [[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] - [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]] \right|^2 \tag{73}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 |2i\mu_{\mathbf{y}}|^2 \tag{74}$$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \tag{75}$$

5.3.2 Case $K'' = 0$:

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \Bigg| \left[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\right] + (-1)^{J''} \left[(-1)^{s'} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] \Bigg|^2 \tag{76}$$

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \Bigg| \left[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{2s'} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\right] + (-1)^{J''+s'} [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] \Bigg|^2 \tag{77}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \left| [2\mu_{\mathbf{x}}] + (-1)^{J''+s'} [2\mu_{\mathbf{x}}] \right|^2 \tag{78}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 {\mu_{\mathbf{x}}}^2 \left| 1 + (-1)^{J''+s'} \right|^2 \tag{79}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \Bigg| [[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^1 [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]] + (-1)^{J''+s'} \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''-s'} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\right] \Bigg|^2 \tag{80}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \Bigg| [[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] - [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]] + (-1)^{J''+s'} [[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] - [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]] \Bigg|^2 \tag{81}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{[2J'+1][2J''+1]}{3} \frac{1}{2} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 \left| [-2i\mu_{\mathbf{y}}] + (-1)^{J''+s'} [-2i\mu_{\mathbf{y}}] \right|^2 \tag{82}$$

$$\propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \left| 1 + (-1)^{J''+s'} \right|^2 \tag{83}$$

6 Table of symmetric rotor line strengths

6.1 $\Delta K = 0$

6.1.1 Case $K' = K'' \neq 0$:

1. $s'' = s'$

$$S \propto 0 \tag{84}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{4[2J'+1][2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}^2 \tag{85}$$

6.1.2 Case $K' = K'' = 0$:

1. $s'' = s'$

$$S \propto 0 \tag{86}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{4[2J'+1][2J''+1]}{3} \mu_{\mathbf{z}}{}^2 \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix}^2 \left| \left[1 - (-1)^{J''+s'}\right] \right|^2 \tag{87}$$

6.2 $\Delta K = -1$

6.2.1 Case $K'' \neq 1$:

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \mu_{\mathbf{x}}{}^2 \tag{88}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix}^2 \mu_{\mathbf{y}}{}^2 \tag{89}$$

6.2.2 Case $K'' = 1$:

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \mu_{\mathbf{x}}{}^2 \left| 1 + (-1)^{J'+s'} \right|^2 \tag{90}$$

2. $s'' \neq s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s'',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -1 & 1 & 0 \end{pmatrix}^2 \mu_{\mathbf{y}}{}^2 \left| 1 + (-1)^{J'+s'} \right|^2 \tag{91}$$

6.3 $\Delta K = +1$

6.3.1 Case $K'' \neq 0$:

1. $s'' = s'$

$$S \propto |\beta_{s',K'}|^2 |\beta_{s',K''}|^2 \frac{2[2J'+1][2J''+1]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 \mu_{\mathbf{x}}{}^2 \tag{92}$$

2. $s'' \neq s'$

$$S \propto \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \tag{93}$$

6.3.2 Case $K'' = 0$:

1. $s'' = s'$

$$S \propto \left|\beta_{s',K'}\right|^2 \left|\beta_{s',K''}\right|^2 \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 {\mu_{\mathbf{x}}}^2 \left|1+(-1)^{J''+s'}\right|^2 \tag{94}$$

2. $s'' \neq s'$

$$S \propto \left|\beta_{s',K'}\right|^2 \left|\beta_{s'',K''}\right|^2 \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3} \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix}^2 {\mu_{\mathbf{y}}}^2 \left|1+(-1)^{J''+s'}\right|^2 \tag{95}$$

7 Derivation of asymmetric rotor line strengths

Recall:

$$|J\tau M s\rangle = \sum_{K=0}^{J} \alpha_{K,\tau}^{(J)} \beta_{s,K} \Big[|JKM\rangle + (-1)^{s+J} |J-KM\rangle \Big] \tag{96}$$

$$S \propto \sum_{M'} \sum_{M''} \left| \sum_{K'} \sum_{K''} \alpha_{K',\tau',s'}^{(J')\dagger} \alpha_{K'',\tau'',s''}^{(J'')\dagger} \langle J'K'M's' | \hat{Q} | J''K''M''s'' \rangle \right|^2 \tag{97}$$

$$\begin{aligned} S \propto \sum_{M'} \sum_{M''} \Bigg| \sum_{K'} \sum_{K''} & \alpha_{K',\tau',s'}^{(J')\dagger} \alpha_{K'',\tau'',s''}^{(J'')\dagger} \beta_{s',K'}^{\dagger} \beta_{s'',K''} \cdot \\ & \Big[\langle J'K'M'| + (-1)^{s'+J'} \langle J'-K'M'| \Big] \hat{Q} \Big[|J''K''M''\rangle + (-1)^{s''+J''} |J''-K''M''\rangle \Big] \Bigg|^2 \end{aligned} \tag{98}$$

$$\begin{aligned} S \propto \sum_{M'} \sum_{M''} \Bigg| \sum_{K'} \sum_{K''} & \alpha_{K',\tau',s'}^{(J')\dagger} \alpha_{K'',\tau'',s''}^{(J'')\dagger} \beta_{s',K'}^{\dagger} \beta_{s'',K''} \Big[\langle J'K'M' | \hat{Q} | J''K''M''\rangle + (-1)^{s'+J'} \langle J'-K'M' | \hat{Q} | J''K''M''\rangle \\ & + (-1)^{s''+J''} \langle J'K'M' | \hat{Q} | J''-K''M''\rangle + (-1)^{s'+J'+s''+J''} \langle J'-K'M' | \hat{Q} | J''-K''M''\rangle \Big] \Bigg|^2 \end{aligned} \tag{99}$$

We now jump to the evaluation of these matrix elements:

$$\begin{aligned} S \propto [2J'+1][2J''+1] \sum_{M'} \sum_{M''} & \begin{pmatrix} J'' & 1 & J' \\ -M'' & 0 & M' \end{pmatrix}^2 \Bigg| \sum_{K'} \sum_{K''} \alpha_{K',\tau',s'}^{(J')\dagger} \alpha_{K'',\tau'',s''}^{(J'')\dagger} \beta_{s',K'}^{\dagger} \beta_{s'',K''} (-1)^{M''-K''} \Bigg[\\ & \Bigg[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \Bigg] \\ & + (-1)^{s'+J'} \Bigg[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & -K' \end{pmatrix} \Bigg] \\ & + (-1)^{s''+J''+2K''} \Bigg[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \Bigg] \\ & + (-1)^{s'+J'+s''+J''+2K''} \Bigg[\frac{-\sqrt{2}}{2} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & -K' \end{pmatrix} \Bigg] \Bigg] \Bigg|^2 \end{aligned} \tag{100}$$

Getting rid of two times a quantum number and simplifying the M 3j-symbols:

$$
\begin{aligned}
S \propto \frac{[2J'+1]\,[2J''+1]}{3}\Bigg| \sum_{K'}\sum_{K''} \alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}(-1)^{-K''}\Bigg[\\
\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s'+J'}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & -K' \end{pmatrix}\Bigg] \\
+(-1)^{s''+J''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s'+J'+s''+J''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & -K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & -K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & -K' \end{pmatrix}\Bigg]\Bigg]\Bigg|^2
\end{aligned}
\tag{101}
$$

Using eqn. (31)

$$
\begin{aligned}
S \propto \frac{[2J'+1]\,[2J''+1]}{3}\Bigg| \sum_{K'}\sum_{K''} \alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}(-1)^{-K''}\Bigg[\\
\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s'+2J'+J''+1}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s''+J''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s'+2J'+s''+2J''+1}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Bigg]\Bigg]\Bigg|^2
\end{aligned}
\tag{102}
$$

Again, cancelling doubled quantum numbers and using colors...

$$
\begin{aligned}
S \propto \frac{[2J'+1]\,[2J''+1]}{3}\Bigg| \sum_{K'}\sum_{K''} \alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}(-1)^{-K''}\Bigg[\\
\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Bigg] \\
-(-1)^{s'+J''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Bigg] \\
+(-1)^{s''+J''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Bigg] \\
-(-1)^{s'+s''}\Bigg[\frac{-\sqrt{2}}{2}[\mu_{\mathbf{x}}-i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \frac{\sqrt{2}}{2}[\mu_{\mathbf{x}}+i\mu_{\mathbf{y}}]\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Bigg]\Bigg]\Bigg|^2
\end{aligned}
\tag{103}
$$

$$
\begin{aligned}
S \propto \frac{[2J'+1]\,[2J''+1]}{3} \Bigg| \sum_{K''}\sum_{K'} & \alpha^{(J')\dagger}_{K',\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',K'} \beta_{s'',K''} (-1)^{-K''} \Bigg[\\
& - \frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} \left[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right] \\
& + \frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} \left[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s'+s''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \right] \\
& + \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix} \left[1 - (-1)^{s'+s''} \right] \\
& + \frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} \left[(-1)^{s'+J''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''+J''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] \right] \\
& - \frac{\sqrt{2}}{2} \begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix} \left[(-1)^{s'+J''} [\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s''+J''} [\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] \right] \\
& - \mu_{\mathbf{z}} \begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix} \left[(-1)^{s'+J''} - (-1)^{s''+J''} \right] \Bigg] \Bigg|^2
\end{aligned} \tag{104}
$$

Again, the requirements for the different 3j-symbols to contribute are:

- $-K'' + 1 + K' = 0 \rightarrow K' - K'' = -1 \rightarrow \Delta K = -1$
- $-K'' - 1 + K' = 0 \rightarrow K' - K'' = 1 \rightarrow \Delta K = +1$
- $-K'' + K' = 0 \rightarrow K' = K'' \rightarrow \Delta K = 0$
- $K'' - 1 + K' = 0 \rightarrow K' = 1 - K'' \rightarrow \quad \text{only allows} \quad 1 \rightarrow 0 \quad \text{and} \quad 0 \rightarrow 1$
- $K'' + 1 + K' = 0 \rightarrow K' = -1 - K'' \rightarrow \text{FORBIDDEN}$
- $K'' + 0 + K' = 0 \rightarrow K' = -K'' \rightarrow \Delta K = 0, \quad K'' = K' = 0$

From here, it is important to note that $\langle J'\tau'M's'|$ and $|J''\tau''M''s''\rangle$ will only contain states with even or odd K, which we denote as $|J\tau Ms\rangle_e$ and $|J\tau Ms\rangle_o$ respectively. (make other section to describe this $\rightarrow$ good group meeting topic?!?!?!?). This, alongside previously explored methods of comparing $s' = s'', s' \neq s''$ can be used to simplify the line strength. We also can simplify the $(-1)^{K''}$ term, due to all K'' being the same evenness.

8 Simplifying the asymmetric rotor

8.1 Steps for simplification

The steps for simplification are as follows:

1. compare the phase of the transition ($s' \quad \text{vs} \quad s''$)
2. compare the evenness or oddness of the K's involved in each $|J\tau Ms\rangle$ state, denoted $|J\tau Ms\rangle_n$
3. simplify the summation based of of selection rules for K, making sure to take into account dimensionality problems posed by the different J levels.

$$
\begin{aligned}
S \propto \frac{[2J'+1]\,[2J''+1]}{3}\Bigg| & \sum_{K''}\sum_{K'} \alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}(-1)^{-K''}\Bigg[\\
& -\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix}\Big[[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s'+s''}[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\Big] \\
& +\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix}\Big[[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s'+s''}[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\Big] \\
& +\mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K' \end{pmatrix}\Big[1 - (-1)^{s'+s''}\Big] \\
& +\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix}\Big[(-1)^{s'+J''}[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}] + (-1)^{s''+J''}[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}]\Big] \\
& -\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ K'' & 1 & K' \end{pmatrix}\Big[(-1)^{s'+J''}[\mu_{\mathbf{x}} + i\mu_{\mathbf{y}}] + (-1)^{s''+J''}[\mu_{\mathbf{x}} - i\mu_{\mathbf{y}}]\Big] \\
& -\mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K' \end{pmatrix}\Big[(-1)^{s'+J''} - (-1)^{s''+J''}\Big]\Bigg]\Bigg|^2
\end{aligned} \tag{105}
$$

8.2 $s' = s''$

$$
\begin{aligned}
S \propto \frac{2\,[2J'+1]\,[2J''+1]}{3}{\mu_{\mathbf{x}}}^2\Bigg| & \sum_{K''}\sum_{K'} \alpha^{(J')\dagger}_{K',\tau',s}\alpha^{(J'')\dagger}_{K'',\tau'',s}\beta^{\dagger}_{s,K'}\beta_{s,K''}\Bigg[\\
& -\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix}(-1)^{s+J''}\Bigg]\Bigg|^2
\end{aligned} \tag{106}
$$

Next, we consider the different combinations of e/o states.

8.2.1 Case $\langle J'\tau'M's|_n\,, |J''\tau''M''s\rangle_n$:

This term will always be rigorously zero, as $\min[\Delta K] \quad \forall \quad K', K'' = 2$, neglecting $K' = K''$

8.2.2 Case $\langle J'\tau'M's|_n\,, |J''\tau''M''s\rangle_{\bar{n}}$:

We next consider the cases that $J' > J''$, $J' < J''$ and $J' = J''$

Since ΔK can equal $\pm$ 1, we can remove the 3j-symbols which are zero, and expand the summation over K'

$$
\begin{aligned}
S \propto \frac{2\,[2J'+1]\,[2J''+1]}{3}{\mu_{\mathbf{x}}}^2\Bigg| & \sum_{K''}\Bigg[\\
& \alpha^{(J')\dagger}_{(K''-1),\tau',s}\alpha^{(J'')\dagger}_{K'',\tau'',s}\beta^{\dagger}_{s,(K''-1)}\beta_{s,K''}\Bigg[\\
& -\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & (K''-1) \end{pmatrix}(-1)^{s+J''}\Bigg] \\
& +\alpha^{(J')\dagger}_{(K''+1),\tau',s}\alpha^{(J'')\dagger}_{K'',\tau'',s}\beta^{\dagger}_{s,(K''+1)}\beta_{s,K''}\Bigg[\\
& -\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & (K''+1) \end{pmatrix}(-1)^{s+J''}\Bigg]\Bigg]\Bigg|^2
\end{aligned} \tag{107}
$$

$$S \propto \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{x}}}^2\Bigg|\sum_{K''}\Bigg[$$
$$\alpha^{(J')\dagger}_{(K''-1),\tau',s}\alpha^{(J'')\dagger}_{K'',\tau'',s}\beta^{\dagger}_{s,(K''-1)}\beta_{s,K''}\left[-\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1)\end{pmatrix}+\begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0\end{pmatrix}(-1)^{s+J''}\delta_{K'',1}\right]$$
$$+\alpha^{(J')\dagger}_{(K''+1),\tau',s}\alpha^{(J'')\dagger}_{K'',\tau'',s}\beta^{\dagger}_{s,(K''+1)}\beta_{s,K''}\left[\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1)\end{pmatrix}+\begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1\end{pmatrix}(-1)^{s+J''}\delta_{K'',0}\right]\Bigg]\Bigg|^2 \quad (108)$$

8.3 $s' \neq s''$

$$S \propto \frac{\left[2J'+1\right]\left[2J''+1\right]}{3}\Bigg|\sum_{K''}\sum_{K'}\alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}\Bigg[$$
$$-\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K'\end{pmatrix}\left[-2i\mu_{\mathbf{y}}\right]$$
$$+\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K'\end{pmatrix}\left[-2i\mu_{\mathbf{y}}\right]$$
$$+\mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K'\end{pmatrix}\left[2\right]$$
$$+\frac{\sqrt{2}}{2}\begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K'\end{pmatrix}(-1)^{s'+J''}\left[-2i\mu_{\mathbf{y}}\right]$$
$$-\mu_{\mathbf{z}}\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K'\end{pmatrix}(-1)^{s'+J''}\left[2\right]\Bigg]\Bigg|^2 \quad (109)$$

8.3.1 Case $\left\langle J'\tau'M's\right|_n, \left|J''\tau''M''s\right\rangle_n$:

$\Delta K \equiv 2n, \quad n\in\mathbb{Z} \quad \forall \quad K''$

$$S \propto \frac{4\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{z}}}^2\Bigg|\sum_{K''}\sum_{K'}\alpha^{(J')\dagger}_{K',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K'}\beta_{s'',K''}\Bigg[$$
$$\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K'\end{pmatrix}-\begin{pmatrix} J'' & 1 & J' \\ K'' & 0 & K'\end{pmatrix}(-1)^{s'+J''}\Bigg]\Bigg|^2 \quad (110)$$

Now, K'' must equal K', so we reduce the summation:

$$S \propto \frac{4\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{z}}}^2\Bigg|\sum_{K''}\alpha^{(J')\dagger}_{K'',\tau',s'}\alpha^{(J'')\dagger}_{K'',\tau'',s''}\beta^{\dagger}_{s',K''}\beta_{s'',K''}\Bigg[$$
$$\begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K''\end{pmatrix}-\begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0\end{pmatrix}(-1)^{s'+J''}\delta_{K'',0}\Bigg]\Bigg|^2 \quad (111)$$

8.3.2 Case $\left\langle J'\tau'M's\right|_n, \left|J''\tau''M''s\right\rangle_{\bar{n}}$:

$\Delta K \equiv 2n+1, \quad n\in\mathbb{Z} \quad \forall \quad K''$

$$S \propto \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{y}}}^2 \left| \sum_{K''}\sum_{K'} \alpha^{(J')\dagger}_{K',\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',K'} \beta_{s'',K''} \left[- \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & K' \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & K' \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ K'' & -1 & K' \end{pmatrix} (-1)^{s'+J''} \right] \right|^2 \tag{112}$$

Expanding out sum over K' into allowed terms:

$$\begin{aligned} S \propto \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{y}}}^2 \Bigg| \sum_{K''} \Bigg[& \\ & \alpha^{(J')\dagger}_{(K''-1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''-1)} \beta_{s'',K''} \Bigg[\\ & - \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',1} \Bigg] \\ & + \alpha^{(J')\dagger}_{(K''+1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''+1)} \beta_{s'',K''} \Bigg[\\ & - \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',0} \Bigg] \Bigg] \Bigg|^2 \end{aligned} \tag{113}$$

$$\begin{aligned} S \propto \frac{2\left[2J'+1\right]\left[2J''+1\right]}{3}{\mu_{\mathbf{y}}}^2 \Bigg| \sum_{K''} \Bigg[& \\ & \alpha^{(J')\dagger}_{(K''-1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''-1)} \beta_{s'',K''} \left[- \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',1} \right] \\ & + \alpha^{(J')\dagger}_{(K''+1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''+1)} \beta_{s'',K''} \left[\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',0} \right] \Bigg] \Bigg|^2 \end{aligned} \tag{114}$$

9 Table of Asymmetric Rotor Line Strengths

9.1 $s' = s''$

9.1.1 Case $\langle J'\tau'M's|_n, |J''\tau''M''s\rangle_n$:

$$S = 0 \tag{115}$$

9.1.2 Case $\langle J'\tau'M's|_n, |J''\tau''M''s\rangle_{\bar{n}}$:

$$\begin{aligned} S \propto \frac{2\,[2J'+1]\,[2J''+1]}{3} {\mu_{\mathbf{x}}}^2 \Bigg| \sum_{K''} \Bigg[\\ \alpha^{(J')\dagger}_{(K''-1),\tau',s} \alpha^{(J'')\dagger}_{K'',\tau'',s} \beta^{\dagger}_{s,(K''-1)} \beta_{s,K''} \left[- \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0 \end{pmatrix} (-1)^{s+J''} \delta_{K'',1} \right] \\ + \alpha^{(J')\dagger}_{(K''+1),\tau',s} \alpha^{(J'')\dagger}_{K'',\tau'',s} \beta^{\dagger}_{s,(K''+1)} \beta_{s,K''} \left[\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix} (-1)^{s+J''} \delta_{K'',0} \right] \Bigg] \Bigg|^2 \end{aligned} \tag{116}$$

9.2 $s' \neq s''$

9.2.1 Case $\langle J'\tau'M's|_n, |J''\tau''M''s\rangle_n$:

$$\begin{aligned} S \propto \frac{4\,[2J'+1]\,[2J''+1]}{3} {\mu_{\mathbf{z}}}^2 \Bigg| \sum_{K''} \alpha^{(J')\dagger}_{K'',\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',K''} \beta_{s'',K''} \Bigg[\\ \begin{pmatrix} J'' & 1 & J' \\ -K'' & 0 & K'' \end{pmatrix} - \begin{pmatrix} J'' & 1 & J' \\ 0 & 0 & 0 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',0} \Bigg] \Bigg|^2 \end{aligned} \tag{117}$$

9.2.2 Case $\langle J'\tau'M's|_n, |J''\tau''M''s\rangle_{\bar{n}}$:

$$\begin{aligned} S \propto \frac{2\,[2J'+1]\,[2J''+1]}{3} {\mu_{\mathbf{y}}}^2 \Bigg| \sum_{K''} \Bigg[\\ \alpha^{(J')\dagger}_{(K''-1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''-1)} \beta_{s'',K''} \left[- \begin{pmatrix} J'' & 1 & J' \\ -K'' & 1 & (K''-1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 1 & -1 & 0 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',1} \right] \\ + \alpha^{(J')\dagger}_{(K''+1),\tau',s'} \alpha^{(J'')\dagger}_{K'',\tau'',s''} \beta^{\dagger}_{s',(K''+1)} \beta_{s'',K''} \left[\begin{pmatrix} J'' & 1 & J' \\ -K'' & -1 & (K''+1) \end{pmatrix} + \begin{pmatrix} J'' & 1 & J' \\ 0 & -1 & 1 \end{pmatrix} (-1)^{s'+J''} \delta_{K'',0} \right] \Bigg] \Bigg|^2 \end{aligned} \tag{118}$$