Nuclear spin relaxation in paramagnetic systems with zero-field splitting and arbitrary electron spin

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Supplementary material

<u>*S* = 1</u>

Coefficients of the relaxation matrix elements:

$$\lambda_{\alpha\beta} = \frac{1}{6}\left\{a^{*}_{\alpha,-1}a_{\beta,-1} - 2a^{*}_{\alpha,0}a_{\beta,0} + a^{*}_{\alpha,1}a_{\beta,1}\right\}^2 + \frac{1}{2}\left\{a^{*}_{\alpha,-1}a_{\beta,0} - a^{*}_{\alpha,0}a_{\beta,1}\right\}^2 + \frac{1}{2}\left\{a^{*}_{\alpha,0}a_{\beta,-1} - a^{*}_{\alpha,1}a_{\beta,0}\right\}^2 + \left\{a^{*}_{\alpha,-1}a_{\beta,1}\right\}^2 + \left\{a^{*}_{\alpha,1}a_{\beta,-1}\right\}$$

$$\varsigma_{\alpha\beta} = -\frac{1}{12}\left\{a^{*}_{\alpha,-1}a_{\alpha,-1} - 2a^{*}_{\alpha,0}a_{\alpha,0} + a^{*}_{\alpha,1}a_{\alpha,1} - a^{*}_{\beta,-1}a_{\beta,-1} + 2a^{*}_{\beta,0}a_{\beta,0} - a^{*}_{\beta,1}a_{\beta,1}\right\}^2 - \frac{1}{2}\left\{a^{*}_{\alpha,-1}a_{\alpha,0} - a^{*}_{\alpha,0}a_{\alpha,1} - a^{*}_{\beta,-1}a_{\beta,0} + a^{*}_{\beta,0}a_{\beta,1}\right\}^2 - \left\{a^{*}_{\alpha,-1}a_{\alpha,1} - a^{*}_{\beta,-1}a_{\beta,1}\right\}^2$$

$$\xi_{\alpha\beta} = -\frac{1}{12}\left\{a^{*}_{\alpha,-1}a_{\beta,-1} - 2a^{*}_{\alpha,0}a_{\beta,0} + a^{*}_{\alpha,1}a_{\beta,1}\right\}^2 - \frac{1}{4}\left\{a^{*}_{\alpha,0}a_{\beta,-1} - a^{*}_{\alpha,1}a_{\beta,0}\right\}^2 - \frac{1}{4}\left\{a^{*}_{\alpha,-1}a_{\beta,0} - a^{*}_{\alpha,0}a_{\beta,1}\right\}^2 - \frac{1}{2}\left\{a^{*}_{\alpha,1}a_{\beta,-1}\right\}^2 - \frac{1}{2}\left\{a^{*}_{\alpha}\right.$$

Tensor operators in the $|I, m'\rangle\langle I, m''|$ representation:

$$S_{-1} = |1, -1\rangle\langle 1, 0| + |1, 0\rangle\langle 1, 1|$$

$$S_{0} = |1, 1\rangle\langle 1, 1| - |1, -1\rangle\langle 1, -1|$$

$$S_{1} = -|1, 0\rangle\langle 1, -1| - |1, 1\rangle\langle 1, 0|$$

Coefficients of the projection vectors:

$$c^{\alpha\beta}_{-1} = a^{*}_{\alpha,1}a_{\beta,0} + a^{*}_{\alpha,0}a_{\beta,-1}$$

$$c_0^{\alpha\beta} = a^*_{\alpha,-1}a_{\beta,-1} - a^*_{\alpha,1}a_{\beta,1}$$

$$c_1^{\alpha\beta} = -a^*_{\alpha,0}a_{\beta,1} - a^*_{\alpha,-1}a_{\beta,0}$$

S = 3/2

Coefficients of the relaxation matrix elements:

$$\begin{aligned}\lambda_{\alpha\beta} = \frac{3}{2}&\left\{a^*_{\alpha,-3/2}a_{\beta,-3/2} - a^*_{\alpha,-1/2}a_{\beta,-1/2} - a^*_{\alpha,1/2}a_{\beta,1/2} + a^*_{\alpha,3/2}a_{\beta,3/2}\right\}^2 + \\ &3\left\{a^*_{\alpha,-3/2}a_{\beta,-1/2} - a^*_{\alpha,1/2}a_{\beta,3/2}\right\}^2 + 3\left\{a^*_{\alpha,-1/2}a_{\beta,-3/2} - a^*_{\alpha,3/2}a_{\beta,1/2}\right\}^2 + \\ &3\left\{a^*_{\alpha,-3/2}a_{\beta,1/2} + a^*_{\alpha,3/2}a_{\beta,-1/2}\right\}^2 + 3\left\{a^*_{\alpha,1/2}a_{\beta,-3/2} + a^*_{\alpha,-1/2}a_{\beta,-3/2}\right\}^2\end{aligned}$$

$$\begin{aligned}\varsigma_{\alpha\beta} = -\frac{3}{4}&\{a^*_{\alpha,-3/2}a_{\alpha,-3/2} - a^*_{\alpha,-1/2}a_{\alpha,-1/2} - a^*_{\alpha,1/2}a_{\alpha,1/2} + a^*_{\alpha,3/2}a_{\alpha,3/2} - \\ &a^*_{\beta,-3/2}a_{\beta,-3/2} + a^*_{\beta,-1/2}a_{\beta,-1/2} + a^*_{\beta,1/2}a_{\beta,1/2} - a^*_{\beta,3/2}a_{\beta,3/2}\}^2 - \\ &3\left\{a^*_{\alpha,-3/2}a_{\alpha,-1/2} - a^*_{\alpha,1/2}a_{\alpha,3/2} - a^*_{\beta,-3/2}a_{\beta,-1/2} + a^*_{\beta,1/2}a_{\beta,3/2}\right\}^2 - \\ &3\left\{a^*_{\alpha,-3/2}a_{\alpha,1/2} - a^*_{\alpha,-1/2}a_{\alpha,3/2} - a^*_{\beta,-3/2}a_{\beta,1/2} + a^*_{\beta,-1/2}a_{\beta,3/2}\right\}^2\end{aligned}$$

$$\begin{aligned}\xi_{\alpha\beta} = -\frac{3}{4}&\left\{a^*_{\alpha,-3/2}a_{\beta,-3/2} - a^*_{\alpha,-1/2}a_{\beta,-1/2} - a^*_{\alpha,1/2}a_{\beta,1/2} + a^*_{\alpha,3/2}a_{\beta,3/2}\right\}^2 - \\ &\frac{3}{2}\left\{a^*_{\alpha,-3/2}a_{\beta,-1/2} - a^*_{\alpha,1/2}a_{\beta,3/2}\right\}^2 - \frac{3}{2}\left\{a^*_{\alpha,-1/2}a_{\beta,-3/2} - a^*_{\alpha,3/2}a_{\beta,1/2}\right\}^2 - \\ &\frac{3}{2}\left\{a^*_{\alpha,-3/2}a_{\beta,1/2} + a^*_{\alpha,3/2}a_{\beta,-1/2}\right\}^2 - \frac{3}{2}\left\{a^*_{\alpha,1/2}a_{\beta,-3/2} + a^*_{\alpha,-1/2}a_{\beta,3/2}\right\}^2\end{aligned}$$

Tensor operators in the $|I,m'\rangle\langle I,m''|$ representation:

$$S_{-1} = \sqrt{2}\left|\frac{3}{2},-\frac{1}{2}\right\rangle\left\langle\frac{3}{2},\frac{1}{2}\right| + \sqrt{\frac{3}{2}}\left|\frac{3}{2},\frac{1}{2}\right\rangle\left\langle\frac{3}{2},\frac{3}{2}\right| + \sqrt{\frac{3}{2}}\left|\frac{3}{2},-\frac{3}{2}\right\rangle\left\langle\frac{3}{2},-\frac{1}{2}\right|$$

$$S_0 = \frac{1}{2}\left|\frac{3}{2},\frac{1}{2}\right\rangle\left\langle\frac{3}{2},\frac{1}{2}\right| - \frac{1}{2}\left|\frac{3}{2},-\frac{1}{2}\right\rangle\left\langle\frac{3}{2},-\frac{1}{2}\right| + \frac{3}{2}\left|\frac{3}{2},\frac{3}{2}\right\rangle\left\langle\frac{3}{2},\frac{3}{2}\right| - \frac{3}{2}\left|\frac{3}{2},-\frac{3}{2}\right\rangle\left\langle\frac{3}{2},-\frac{3}{2}\right|$$

$$S_1 = \sqrt{2}\left|\frac{3}{2},\frac{1}{2}\right\rangle\left\langle\frac{3}{2},-\frac{1}{2}\right| + \sqrt{\frac{3}{2}}\left|\frac{3}{2},-\frac{1}{2}\right\rangle\left\langle\frac{3}{2},-\frac{3}{2}\right| + \sqrt{\frac{3}{2}}\left|\frac{3}{2},\frac{3}{2}\right\rangle\left\langle\frac{3}{2},\frac{1}{2}\right|$$

Coefficients of the projection vectors:

$$c_{-1}^{\alpha\beta} = \sqrt{2}a^{*}_{\alpha,-1/2}a_{\beta,1/2} + \sqrt{\frac{3}{2}}a^{*}_{\alpha,1/2}a_{\beta,3/2} + \sqrt{\frac{3}{2}}a^{*}_{\alpha,-3/2}a_{\beta,-1/2}$$

$$c_{0}^{\alpha\beta} = \frac{3}{2}a^{*}_{\alpha,-3/2}a_{\beta,-3/2} + \frac{1}{2}a^{*}_{\alpha,-1/2}a_{\beta,-1/2} - \frac{1}{2}a^{*}_{\alpha,1/2}a_{\beta,1/2} - \frac{3}{2}a^{*}_{\alpha,3/2}a_{\beta,3/2}$$

$$c_{1}^{\alpha\beta} = \sqrt{2}a^{*}_{\alpha,1/2}a_{\beta,-1/2} + \sqrt{\frac{3}{2}}a^{*}_{\alpha,3/2}a_{\beta,1/2} + \sqrt{\frac{3}{2}}a^{*}_{\alpha,-1/2}a_{\beta,-3/2}$$

***S* = 2**

Coefficients of the relaxation matrix elements:

$$\lambda_{\alpha\beta} = \left\{\sqrt{6}a^{*}_{\alpha,-2}a_{\beta,-2} - \frac{3}{\sqrt{6}}a^{*}_{\alpha,-1}a_{\beta,-1} - \sqrt{6}a^{*}_{\alpha,0}a_{\beta,0} - \frac{3}{\sqrt{6}}a^{*}_{\alpha,1}a_{\beta,1} + \sqrt{6}a^{*}_{\alpha,2}a_{\beta,2}\right\}^{2} +$$
$$\left\{3a^{*}_{\alpha,-2}a_{\beta,-1} + \frac{\sqrt{6}}{2}a^{*}_{\alpha,-1}a_{\beta,0} - \frac{\sqrt{6}}{2}a^{*}_{\alpha,0}a_{\beta,1} - 3a^{*}_{\alpha,1}a_{\beta,2}\right\}^{2} +$$
$$\left\{-3a^{*}_{\alpha,-1}a_{\beta,-2} - \frac{\sqrt{6}}{2}a^{*}_{\alpha,0}a_{\beta,-1} + \frac{\sqrt{6}}{2}a^{*}_{\alpha,1}a_{\beta,0} + 3a^{*}_{\alpha,2}a_{\beta,1}\right\}^{2} +$$
$$\left\{\sqrt{6}a^{*}_{\alpha,-2}a_{\beta,0} - 3a^{*}_{\alpha,-1}a_{\beta,1} + \sqrt{6}a^{*}_{\alpha,0}a_{\beta,1}\right\}^{2} + \left\{\sqrt{6}a^{*}_{\alpha,0}a_{\beta,-2} - 3a^{*}_{\alpha,1}a_{\beta,-1} + \sqrt{6}a^{*}_{\alpha,2}a_{\beta,0}\right\}^{2}$$

$$\varsigma_{\alpha\beta} = -\frac{1}{2}\{\sqrt{6}a^{*}_{\alpha,-2}a_{\alpha,-2} - \frac{3}{\sqrt{6}}a^{*}_{\alpha,-1}a_{\alpha,-1} - \sqrt{6}a^{*}_{\alpha,0}a_{\alpha,0} - \frac{3}{\sqrt{6}}a^{*}_{\alpha,1}a_{\alpha,1} + \sqrt{6}a^{*}_{\alpha,2}a_{\alpha,2} -$$
$$\sqrt{6}a^{*}_{\beta,-2}a_{\beta,-2} + \frac{3}{\sqrt{6}}a^{*}_{\beta,-1}a_{\beta,-1} + \sqrt{6}a^{*}_{\beta,0}a_{\beta,0} + \frac{3}{\sqrt{6}}a^{*}_{\beta,1}a_{\beta,1} - \sqrt{6}a^{*}_{\beta,2}a_{\beta,2}\}^{2} -$$
$$\left\{3a^{*}_{\alpha,-2}a_{\alpha,-1} + \frac{\sqrt{6}}{2}a^{*}_{\alpha,-1}a_{\alpha,0} - \frac{\sqrt{6}}{2}a^{*}_{\alpha,0}a_{\alpha,1} - 3a^{*}_{\alpha,1}a_{\alpha,2} - 3a^{*}_{\beta,-2}a_{\beta,-1} - \frac{\sqrt{6}}{2}a^{*}_{\beta,-1}a_{\beta,0} + \frac{\sqrt{6}}{2}a^{*}_{\beta,0}a_{\beta}\right.$$
$$\left\{\sqrt{6}a^{*}_{\alpha,-2}a_{\alpha,0} + 3a^{*}_{\alpha,-1}a_{\alpha,1} + \sqrt{6}a^{*}_{\alpha,0}a_{\alpha,2} - \sqrt{6}a^{*}_{\beta,-2}a_{\beta,0} - 3a^{*}_{\beta,-1}a_{\beta,1} - \sqrt{6}a^{*}_{\beta,0}a_{\beta,2}\right\}^{2}$$

$$
\begin{aligned}
\xi_{\alpha\beta} = &-\frac{1}{2}\left\{\sqrt{6}a^*_{\alpha,-2}a_{\beta,-2} - \frac{\sqrt{6}}{2}a^*_{\alpha,-1}a_{\beta,-1} - \sqrt{6}a^*_{\alpha,0}a_{\beta,0} - \frac{\sqrt{6}}{2}a^*_{\alpha,1}a_{\beta,1} + \sqrt{6}a^*_{\alpha,2}a_{\beta,2}\right\}^2 - \\
&\frac{1}{2}\left\{3a^*_{\alpha,-2}a_{\beta,-1} + \frac{\sqrt{6}}{2}a^*_{\alpha,-1}a_{\beta,0} - \frac{\sqrt{6}}{2}a^*_{\alpha,0}a_{\beta,1} - 3a^*_{\alpha,1}a_{\beta,2}\right\}^2 - \\
&\frac{1}{2}\left\{-3a^*_{\alpha,-1}a_{\beta,-2} - \frac{\sqrt{6}}{2}a^*_{\alpha,0}a_{\beta,-1} + \frac{\sqrt{6}}{2}a^*_{\alpha,1}a_{\beta,0} + 3a^*_{\alpha,2}a_{\beta,1}\right\}^2 - \\
&\frac{1}{2}\left\{\sqrt{6}a^*_{\alpha,-2}a_{\beta,0} + 3a^*_{\alpha,-1}a_{\beta,1} - \sqrt{6}a^*_{\alpha,0}a_{\beta,2}\right\}^2 - \frac{1}{2}\left\{\sqrt{6}a^*_{\alpha,0}a_{\beta,-2} + 3a^*_{\alpha,1}a_{\beta,-1} - \sqrt{6}a^*_{\alpha,2}a_{\beta,0}\right\}^2
\end{aligned}
$$

Tensor operators in the $|I,m'\rangle\langle I,m''|$ representation:

$$S_{-1} = \sqrt{2}|2,1\rangle\langle 2,2| + \sqrt{3}|2,0\rangle\langle 2,1| + \sqrt{3}|2,-1\rangle\langle 2,0| + \sqrt{2}|2,-2\rangle\langle 2,-1|$$

$$S_0 = -2|2,-2\rangle\langle 2,-2| - |2,-1\rangle\langle 2,-1| + |2,1\rangle\langle 2,1| + 2|2,2\rangle\langle 2,2|$$

$$S_1 = \sqrt{2}|2,-1\rangle\langle 2,-2| + \sqrt{3}|2,0\rangle\langle 2,-1| + \sqrt{3}|2,1\rangle\langle 2,0| + \sqrt{2}|2,2\rangle\langle 2,1|$$

Coefficients of the projection vectors:

$$c^{\alpha\beta}_{-1} = \sqrt{2}a^*_{\alpha,-1}a_{\beta,-2} + \sqrt{3}a^*_{\alpha,0}a_{\beta,-1} + \sqrt{3}a^*_{\alpha,1}a_{\beta,0} + \sqrt{2}a^*_{\alpha,2}a_{\beta,1}$$

$$c^{\alpha\beta}_{0} = 2a^*_{\alpha,-2}a_{\beta,-1} + a^*_{\alpha,-1}a_{\beta,-1} - a^*_{\alpha,1}a_{\beta,1} - 2a^*_{\alpha,2}a_{\beta,2}$$

$$c^{\alpha\beta}_{1} = \sqrt{2}a^*_{\alpha,-2}a_{\beta,-1} + \sqrt{3}a^*_{\alpha,-1}a_{\beta,0} + \sqrt{3}a^*_{\alpha,0}a_{\beta,1} + \sqrt{2}a^*_{\alpha,1}a_{\beta,2}$$

***S* = 5/2**

Coefficients of the relaxation matrix elements:

$$\lambda_{\alpha\beta} = \frac{2}{3}\{5a^*_{\alpha,-5/2}a_{\beta,-5/2} - a^*_{\alpha,-3/2}a_{\beta,-3/2} - 4a^*_{\alpha,-1/2}a_{\beta,-1/2} - 4a^*_{\alpha,1/2}a_{\beta,1/2} - a^*_{\alpha,3/2}a_{\beta,3/2} + 5a^*_{\alpha,5/2}a_{\beta,5/2}\}^2 +$$
$$4\left\{\sqrt{5}a^*_{\alpha,-5/2}a_{\beta,-3/2} + \sqrt{2}a^*_{\alpha,-3/2}a_{\beta,-1/2} - \sqrt{2}a^*_{\alpha,1/2}a_{\beta,3/2} - \sqrt{5}a^*_{\alpha,3/2}a_{\beta,5/2}\right\}^2 +$$
$$4\left\{-\sqrt{5}a^*_{\alpha,-3/2}a_{\beta,-5/2} - \sqrt{2}a^*_{\alpha,-1/2}a_{\beta,-3/2} + \sqrt{2}a^*_{\alpha,3/2}a_{\beta,1/2} + \sqrt{5}a^*_{\alpha,5/2}a_{\beta,3/2}\right\}^2 +$$
$$\left\{\sqrt{10}a^*_{\alpha,-5/2}a_{\beta,-1/2} + 3\sqrt{2}a^*_{\alpha,-3/2}a_{\beta,1/2} + 3\sqrt{2}a^*_{\alpha,-1/2}a_{\beta,3/2} - \sqrt{10}a^*_{\alpha,1/2}a_{\beta,5/2}\right\}^2 +$$
$$\left\{\sqrt{10}a^*_{\alpha,-1/2}a_{\beta,-5/2} + 3\sqrt{2}a^*_{\alpha,1/2}a_{\beta,-3/2} + 3\sqrt{2}a^*_{\alpha,3/2}a_{\beta,-1/2} - \sqrt{10}a^*_{\alpha,5/2}a_{\beta,1/2}\right\}^2$$

$$\varsigma_{\alpha\beta} = -\frac{1}{3}\{5a^*_{\alpha,-5/2}a_{\alpha,-5/2} - a^*_{\alpha,-3/2}a_{\alpha,-3/2} - 4a^*_{\alpha,-1/2}a_{\alpha,-1/2} - 4a^*_{\alpha,1/2}a_{\alpha,1/2} - a^*_{\alpha,3/2}a_{\alpha,3/2} + 5a^*_{\alpha,5/2}a_{\alpha,5/2} - 5a^*_{\beta,-5/2}a_{\beta,-5/2} + a^*_{\beta,-3/2}a_{\beta,-3/2} + 4a^*_{\beta,-1/2}a_{\beta,-1/2} + 4a^*_{\beta,1/2}a_{\beta,1/2} + a^*_{\beta,3/2}a_{\beta,3/2} - 5a^*_{\beta,5/2}a_{\beta,5/2}\}^2 -$$
$$4\{\sqrt{5}a^*_{\alpha,-5/2}a_{\alpha,-3/2} + \sqrt{2}a^*_{\alpha,-3/2}a_{\alpha,-1/2} - \sqrt{2}a^*_{\alpha,1/2}a_{\alpha,3/2} - \sqrt{5}a^*_{\alpha,3/2}a_{\alpha,5/2} - \sqrt{5}a^*_{\beta,-5/2}a_{\beta,-3/2} - \sqrt{2}a^*_{\beta,-3/2}a_{\beta,-1/2} + \sqrt{2}a^*_{\beta,1/2}a_{\beta,3/2} + \sqrt{5}a^*_{\beta,3/2}a_{\beta,5/2}\}^2 -$$
$$\{\sqrt{10}a^*_{\alpha,-5/2}a_{\alpha,-1/2} + 3\sqrt{2}a^*_{\alpha,-3/2}a_{\alpha,1/2} + 3\sqrt{2}a^*_{\alpha,-1/2}a_{\alpha,3/2} + \sqrt{10}a^*_{\alpha,1/2}a_{\alpha,5/2} - \sqrt{10}a^*_{\beta,-5/2}a_{\beta,-1/2} - 3\sqrt{2}a^*_{\beta,-3/2}a_{\beta,1/2} - 3\sqrt{2}a^*_{\beta,-1/2}a_{\beta,3/2} - \sqrt{10}a^*_{\beta,1/2}a_{\beta,5/2}\}^2$$

$$\xi_{\alpha\beta} = -\frac{1}{3}\{5a^*_{\alpha,-5/2}a_{\beta,-5/2} - a^*_{\alpha,-3/2}a_{\beta,-3/2} - 4a^*_{\alpha,-1/2}a_{\beta,-1/2} - 4a^*_{\alpha,1/2}a_{\beta,1/2} - a^*_{\alpha,3/2}a_{\beta,3/2} + 5a^*_{\alpha,5/2}a_{\beta,5/2}\}^2 -$$
$$2\left\{\sqrt{5}a^*_{\alpha,-5/2}a_{\beta,-3/2} + \sqrt{2}a^*_{\alpha,-3/2}a_{\beta,-1/2} - \sqrt{2}a^*_{\alpha,1/2}a_{\beta,3/2} - \sqrt{5}a^*_{\alpha,3/2}a_{\beta,5/2}\right\}^2 +$$
$$2\left\{-\sqrt{5}a^*_{\alpha,-3/2}a_{\beta,-5/2} - \sqrt{2}a^*_{\alpha,-1/2}a_{\beta,-3/2} + \sqrt{2}a^*_{\alpha,3/2}a_{\beta,1/2} + \sqrt{5}a^*_{\alpha,5/2}a_{\beta,3/2}\right\}^2 +$$
$$\frac{1}{2}\left\{\sqrt{10}a^*_{\alpha,-5/2}a_{\beta,-1/2} + 3\sqrt{2}a^*_{\alpha,-3/2}a_{\beta,1/2} + 3\sqrt{2}a^*_{\alpha,-1/2}a_{\beta,3/2} + \sqrt{10}a^*_{\alpha,1/2}a_{\beta,5/2}\right\}^2 +$$
$$\frac{1}{2}\left\{\sqrt{10}a^*_{\alpha,-1/2}a_{\beta,-5/2} + 3\sqrt{2}a^*_{\alpha,1/2}a_{\beta,-3/2} + 3\sqrt{2}a^*_{\alpha,3/2}a_{\beta,-1/2} + \sqrt{10}a^*_{\alpha,5/2}a_{\beta,1/2}\right\}^2$$

Tensor operators in the $|I,m'\rangle\langle I,m''|$ representation:

$$S_{-1} = \frac{3}{\sqrt{2}}\left|\frac{5}{2},-\frac{1}{2}\right\rangle\left\langle\frac{5}{2},\frac{1}{2}\right| + 2\left|\frac{5}{2},\frac{1}{2}\right\rangle\left\langle\frac{5}{2},\frac{3}{2}\right| + 2\left|\frac{5}{2},-\frac{3}{2}\right\rangle\left\langle\frac{5}{2},-\frac{1}{2}\right| + \sqrt{\frac{5}{2}}\left|\frac{5}{2},-\frac{3}{2}\right\rangle\left\langle\frac{5}{2},-\frac{5}{2}\right| + \sqrt{\frac{5}{2}}\left|\frac{5}{2},\frac{5}{2}\right\rangle\left\langle\frac{5}{2},\frac{3}{2}\right|$$

$$S_0 = \frac{1}{2}\left|\frac{5}{2},\frac{1}{2}\right\rangle\left\langle\frac{5}{2},\frac{1}{2}\right| - \frac{1}{2}\left|\frac{5}{2},-\frac{1}{2}\right\rangle\left\langle\frac{5}{2},-\frac{1}{2}\right| + \frac{3}{2}\left|\frac{5}{2},\frac{3}{2}\right\rangle\left\langle\frac{5}{2},\frac{3}{2}\right| - \frac{3}{2}\left|\frac{5}{2},-\frac{3}{2}\right\rangle\left\langle\frac{5}{2},-\frac{3}{2}\right| + \frac{5}{2}\left|\frac{5}{2},\frac{5}{2}\right\rangle\left\langle\frac{5}{2},\frac{5}{2}\right| - \frac{5}{2}\left|\frac{5}{2},-\frac{5}{2}\right\rangle\left\langle\frac{5}{2},-\frac{5}{2}\right|$$

$$S_1 = \frac{3}{\sqrt{2}}\left|\frac{5}{2},\frac{1}{2}\right\rangle\left\langle\frac{5}{2},-\frac{1}{2}\right| + 2\left|\frac{5}{2},\frac{3}{2}\right\rangle\left\langle\frac{5}{2},\frac{1}{2}\right| + 2\left|\frac{5}{2},-\frac{1}{2}\right\rangle\left\langle\frac{5}{2},-\frac{3}{2}\right| + \sqrt{\frac{5}{2}}\left|\frac{5}{2},-\frac{5}{2}\right\rangle\left\langle\frac{5}{2},-\frac{3}{2}\right| + \sqrt{\frac{5}{2}}\left|\frac{5}{2},\frac{3}{2}\right\rangle\left\langle\frac{5}{2},\frac{5}{2}\right|$$

Coefficients of the projection vectors:

$$c_{-1}^{\alpha\beta} = \sqrt{\frac{5}{2}}\,a^*_{\alpha,-5/2}a_{\beta,-3/2} + 2a^*_{\alpha,-3/2}a_{\beta,-1/2} + \frac{3}{\sqrt{2}}\,a^*_{\alpha,-1/2}a_{\beta,1/2} + 2a^*_{\alpha,1/2}a_{\beta,3/2} + \sqrt{\frac{5}{2}}\,a^*_{\alpha,3/2}a_{\beta,5/2}$$

$$c_{0}^{\alpha\beta} = \frac{5}{2}\,a^*_{\alpha,-5/2}a_{\beta,-5/2} + \frac{3}{2}\,a^*_{\alpha,-3/2}a_{\beta,-3/2} + \frac{1}{2}\,a^*_{\alpha,-1/2}a_{\beta,-1/2} - \frac{1}{2}\,a^*_{\alpha,1/2}a_{\beta,1/2} - \frac{3}{2}\,a^*_{\alpha,3/2}a_{\beta,3/2} - \frac{5}{2}\,a^*_{\alpha,5/2}a_{\beta,5/2}$$

$$c_{1}^{\alpha\beta} = \sqrt{\frac{5}{2}}\,a^*_{\alpha,-3/2}a_{\beta,-5/2} + 2a^*_{\alpha,-1/2}a_{\beta,-3/2} + \frac{3}{\sqrt{2}}\,a^*_{\alpha,1/2}a_{\beta,-1/2} + 2a^*_{\alpha,3/2}a_{\beta,1/2} + \sqrt{\frac{5}{2}}\,a^*_{\alpha,5/2}a_{\beta,3/2}$$

S = 3

Coefficients of the relaxation matrix elements:

$$\lambda_{\alpha\beta} = \frac{3}{2}\left\{5a^*_{\alpha,-3}a_{\beta,-3} - 3a^*_{\alpha,-1}a_{\beta,-1} - 4a^*_{\alpha,0}a_{\beta,0} - 3a^*_{\alpha,1}a_{\beta,1} + 5a^*_{\alpha,3}a_{\beta,3}\right\}^2 +$$

$$\frac{1}{4}\{5\sqrt{6}a^*_{\alpha,-3}a_{\beta,-2} + 3\sqrt{10}a^*_{\alpha,-2}a_{\beta,-1} - 2\sqrt{3}a^*_{\alpha,-1}a_{\beta,0} - 2\sqrt{3}a^*_{\alpha,0}a_{\beta,1} - 3\sqrt{10}a^*_{\alpha,1}a_{\beta,2} -$$

$$5\sqrt{6}a^*_{\alpha,2}a_{\beta,3}\}^2 + \frac{1}{4}\{-5\sqrt{6}a^*_{\alpha,-2}a_{\beta,-3} - 3\sqrt{10}a^*_{\alpha,-1}a_{\beta,-2} + 2\sqrt{3}a^*_{\alpha,0}a_{\beta,-1} + 2\sqrt{3}a^*_{\alpha,1}a_{\beta,0} +$$

$$3\sqrt{10}a^*_{\alpha,2}a_{\beta,1} + 5\sqrt{6}a^*_{\alpha,3}a_{\beta,2}\}^2 +$$

$$\left\{\sqrt{15}a^*_{\alpha,-3}a_{\beta,-1} + \sqrt{30}a^*_{\alpha,-2}a_{\beta,0} + 6a^*_{\alpha,-1}a_{\beta,1} + \sqrt{30}a^*_{\alpha,0}a_{\beta,2} + \sqrt{15}a^*_{\alpha,1}a_{\beta,3}\right\}^2 +$$

$$\left\{\sqrt{15}a^*_{\alpha,-1}a_{\beta,-3} + \sqrt{30}a^*_{\alpha,0}a_{\beta,-2} + 6a^*_{\alpha,1}a_{\beta,-1} + \sqrt{30}a^*_{\alpha,2}a_{\beta,0} + \sqrt{15}a^*_{\alpha,3}a_{\beta,1}\right\}^2$$

$$\varsigma_{\alpha\beta} = -\frac{3}{4}\{5a^*_{\alpha,-3}a_{\alpha,-3} - 3a^*_{\alpha,-1}a_{\alpha,-1} - 4a^*_{\alpha,0}a_{\alpha,0} - 3a^*_{\alpha,1}a_{\alpha,1} + 5a^*_{\alpha,3}a_{\alpha,3} -$$

$$5a^*_{\beta,-3}a_{\beta,-3} + 3a^*_{\beta,-1}a_{\beta,-1} + 4a^*_{\beta,0}a_{\beta,0} + 3a^*_{\beta,1}a_{\beta,1} - 5a^*_{\beta,3}a_{\beta,3}\}^2 -$$

$$\frac{1}{4}\{5\sqrt{6}a^*_{\alpha,-3}a_{\alpha,-2} + 3\sqrt{10}a^*_{\alpha,-2}a_{\alpha,-1} + 2\sqrt{3}a^*_{\alpha,-1}a_{\alpha,0} - 2\sqrt{3}a^*_{\alpha,0}a_{\alpha,1} -$$

$$3\sqrt{10}a^*_{\alpha,1}a_{\alpha,2} - 5\sqrt{6}a^*_{\alpha,2}a_{\alpha,3} - 5\sqrt{6}a^*_{\beta,-3}a_{\beta,-2} - 3\sqrt{10}a^*_{\beta,-2}a_{\beta,-1} - 2\sqrt{3}a^*_{\beta,-1}a_{\beta,0} +$$

$$2\sqrt{3}a^*_{\beta,0}a_{\beta,1} + 3\sqrt{10}a^*_{\beta,1}a_{\beta,2} + 5\sqrt{6}a^*_{\beta,2}a_{\beta,3}\}^2 -$$

$$\{\sqrt{15}a^*_{\alpha,-3}a_{\alpha,-1} + \sqrt{30}a^*_{\alpha,-2}a_{\alpha,0} + 6a^*_{\alpha,-1}a_{\alpha,1} + \sqrt{30}a^*_{\alpha,0}a_{\alpha,2} + \sqrt{15}a^*_{\alpha,1}a_{\alpha,3} -$$

$$\sqrt{15}a^*_{\beta,-3}a_{\beta,-1} - \sqrt{30}a^*_{\beta,-2}a_{\beta,0} - 6a^*_{\beta,-1}a_{\beta,1} - \sqrt{30}a^*_{\beta,0}a_{\beta,2} - \sqrt{15}a^*_{\beta,1}a_{\beta,3}\}^2$$

$$\xi_{\alpha\beta} = -\frac{3}{4}\left\{5a^*_{\alpha,-3}a_{\beta,-3} - 3a^*_{\alpha,-1}a_{\beta,-1} - 4a^*_{\alpha,0}a_{\beta,0} - 3a^*_{\alpha,1}a_{\beta,1} + 5a^*_{\alpha,3}a_{\beta,3}\right\}^2 -$$

$$\frac{1}{8}\{5\sqrt{6}a^*_{\alpha,-3}a_{\beta,-2} + 3\sqrt{10}a^*_{\alpha,-2}a_{\beta,-1} - 2\sqrt{3}a^*_{\alpha,-1}a_{\beta,0} - 2\sqrt{3}a^*_{\alpha,0}a_{\beta,1} -$$

$$3\sqrt{10}a^*_{\alpha,1}a_{\beta,2} - 5\sqrt{6}a^*_{\alpha,2}a_{\beta,3}\}^2 - \frac{1}{8}\{-5\sqrt{6}a^*_{\alpha,-2}a_{\beta,-3} - 3\sqrt{10}a^*_{\alpha,-1}a_{\beta,-2} + 2\sqrt{3}a^*_{\alpha,0}a_{\beta,-1} +$$

$$2\sqrt{3}a^*_{\alpha,1}a_{\beta,0} + 3\sqrt{10}a^*_{\alpha,2}a_{\beta,1} + 5\sqrt{6}a^*_{\alpha,3}a_{\beta,2}\}^2 -$$

$$\frac{1}{2}\left\{\sqrt{15}a^*_{\alpha,-3}a_{\beta,-1} + \sqrt{30}a^*_{\alpha,-2}a_{\beta,0} + 6a^*_{\alpha,-1}a_{\beta,1} + \sqrt{30}a^*_{\alpha,0}a_{\beta,2} + \sqrt{15}a^*_{\alpha,1}a_{\beta,3}\right\}^2 -$$

$$\frac{1}{2}\left\{\sqrt{15}a^*_{\alpha,-1}a_{\beta,-3} + \sqrt{30}a^*_{\alpha,0}a_{\beta,-2} + 6a^*_{\alpha,1}a_{\beta,-1} + \sqrt{30}a^*_{\alpha,2}a_{\beta,0} + \sqrt{15}a^*_{\alpha,3}a_{\beta,1}\right\}^2$$

Tensor operators in the $|I,m'\rangle\langle I,m''|$ representation:

$$S_{-1}=\sqrt{3}|3,-3\rangle\langle 3,-2|+\sqrt{5}|3,-2\rangle\langle 3,-1|+\sqrt{6}|3,-1\rangle\langle 3,0|+\sqrt{6}|3,0\rangle\langle 3,1|+\sqrt{5}|3,1\rangle\langle 3,2|+\sqrt{3}|3,2\rangle\langle 3,3|$$

$$S_{0}=-3|3,-3\rangle\langle 3,-3|-2|3,-2\rangle\langle 3,-2|-|3,-1\rangle\langle 3,-1|+|3,1\rangle\langle 3,1|+2|3,2\rangle\langle 3,2|+3|3,3\rangle\langle 3,3|$$

$$S_{1}=\sqrt{3}|3,-2\rangle\langle 3,-3|+\sqrt{5}|3,-1\rangle\langle 3,-2|+\sqrt{6}|3,0\rangle\langle 3,-1|+\sqrt{6}|3,1\rangle\langle 3,0|+\sqrt{5}|3,2\rangle\langle 3,1|+\sqrt{3}|3,3\rangle\langle 3,2|$$

Coefficients of the projection vectors:

$$c_{-1}^{\alpha\beta}=\sqrt{3}a_{\alpha,-2}^{*}a_{\beta,-3}+\sqrt{5}a_{\alpha,-1}^{*}a_{\beta,-2}+\sqrt{6}a_{\alpha,0}^{*}a_{\beta,-1}+\sqrt{6}a_{\alpha,1}^{*}a_{\beta,0}+\sqrt{5}a_{\alpha,2}^{*}a_{\beta,1}+\sqrt{3}a_{\alpha,3}^{*}a_{\beta,2}$$

$$c_{0}^{\alpha\beta}=3a_{\alpha,-3}^{*}a_{\beta,-3}+2a_{\alpha,-2}^{*}a_{\beta,-2}+a_{\alpha,-1}^{*}a_{\beta,-1}-a_{\alpha,1}^{*}a_{\beta,1}-2a_{\alpha,2}^{*}a_{\beta,2}-3a_{\alpha,3}^{*}a_{\beta,3}$$

$$c_{1}^{\alpha\beta}=\sqrt{3}a_{\alpha,-3}^{*}a_{\beta,-2}+\sqrt{5}a_{\alpha,-2}^{*}a_{\beta,-1}+\sqrt{6}a_{\alpha,-1}^{*}a_{\beta,0}+\sqrt{6}a_{\alpha,0}^{*}a_{\beta,1}+\sqrt{5}a_{\alpha,1}^{*}a_{\beta,2}+\sqrt{3}a_{\alpha,2}^{*}a_{\beta,3}$$