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Analytical Planar Mixing Transfer Functions for Two Coupled Spin-1 Nuclei

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Analytical coherence transfer functions are presented for spin systems consisting of two spins 1 under planar mixing conditions. Compared to isotropic mixing experiments, larger transfer amplitudes are found and differences in the multiplet patterns are discussed. © 2001 Elsevier Science

Key Words: **Hartmann–Hahn transfer; spin 1; planar mixing; isotropic mixing; analytical transfer functions; TOCSY; TACSY.**

1. INTRODUCTION

Coherence and polarization transfer functions are important for the theoretical analysis and practical implementation of mixing periods with maximum transfer efficiency. Homonuclear and heteronuclear Hartmann–Hahn-type experiments can be classified according to the form of the coupling tensors in the effective mixing Hamiltonian (*1, 2*). For example, isotropic coupling tensors are created by heteronuclear isotropic mixing sequences (*1, 3, 4*) and by most homonuclear Hartmann–Hahn sequences if applied to scalar coupled spin systems (*1*, *5*, *6*). Effective planar coupling tensors (*7*) are typical for most heteronuclear Hartmann–Hahn experiments (*1*, *8*, *9*) and for homonuclear Hartmann–Hahn experiments that are based on RF irradiation at multiple frequencies (*10–14*). A large number of analytical coherence and polarization transfer functions are known for Hartmann–Hahn-type mixing experiments in spin-$\frac{1}{2}$ systems (*5*, *8*, *15–29*). However, for spin systems involving spin-1 nuclei, only few analytical transfer functions have been reported. For spin systems consisting of a spin-$\frac{1}{2}$ and a spin-1 nucleus, transfer functions are known for planar (*15*) and isotropic (*30*) mixing conditions. For two coupled spin-1 nuclei, analytical transfer functions are only known for the case of isotropic mixing (*31, 32*). Here, we present analytical coherence and polarization transfer functions for two coupled spin-1 nuclei under planar mixing conditions. These results form the basis for a detailed comparison of isotropic and planar transfer between two spin-1 nuclei.

2. THEORY

The planar mixing Hamiltonian of a spin system consisting of two spins 1 with coupling constant J has the form

$$\mathcal{H}_\mathrm{P} = 2\pi J^\mathrm{eff}\{I_x S_x + I_y S_y\} \qquad [1]$$

with the effective coupling constant $J^\mathrm{eff} \leq J/2$ (*9*). As $\mathcal{H}_\mathrm{P}$ commutes with $F_z = I_z + S_z$, the Hamiltonian $\mathcal{H}_\mathrm{P}$ is represented by a block-diagonal 9×9 matrix in the basis of the product functions $|m_I, m_S\rangle$. The five blocks correspond to the five sets of basis functions $\{|1, 1\rangle\}$, $\{|1, 0\rangle, |0, 1\rangle\}$, $\{|1, -1\rangle, |-1, 1\rangle, |0, 0\rangle\}$, $\{|-1, 0\rangle, |0, -1\rangle\}$, and $\{|-1, -1\rangle\}$ with $m = 2, 1, 0, -1$, and -2, respectively. Hence, coherence or polarization transfer is only possible between states with equal magnetic quantum number m. The individual blocks of $\mathcal{H}_\mathrm{P}$ have the following explicite form. There are two identical 1×1 blocks corresponding to the total magnetic quantum numbers $m = 2$ and $m = -2$,

$$\mathcal{H}_\mathrm{P}^{(2)} = \mathcal{H}_\mathrm{P}^{(-2)} = 0 = \lambda_0, \qquad [2]$$

two identical 2×2 blocks corresponding to $m = 1$ and $m = -1$,

$$\mathcal{H}_\mathrm{P}^{(1)} = \mathcal{H}_\mathrm{P}^{(-1)} = 2\pi\begin{pmatrix} 0 & J^\mathrm{eff} \\ J^\mathrm{eff} & 0 \end{pmatrix} \qquad [3]$$

with eigenvalues

$$\lambda_{1,2} = \mp 2\pi J^\mathrm{eff} \qquad [4]$$

and normalized eigenvectors

$$\kappa_{1,2} = \left(\mp\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}\right), \qquad [5]$$

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and one 3×3 block corresponding to the total magnetic quantum number $m = 0$,

$$\mathcal{H}_{\mathrm{P}}^{(0)} = 2\pi \begin{pmatrix} 0 & J^{\mathrm{eff}} & 0 \\ J^{\mathrm{eff}} & 0 & J^{\mathrm{eff}} \\ 0 & J^{\mathrm{eff}} & 0 \end{pmatrix} \tag{6}$$

with the eigenvalues

$$\begin{aligned} \lambda_3 &= 0 \\ \lambda_{4,5} &= \mp 2\sqrt{2}\pi J^{\mathrm{eff}} \end{aligned} \tag{7}$$

and the eigenvectors

$$\begin{aligned} \kappa_3 &= \left(-\frac{1}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}}\right) \\ \kappa_4 &= \left(\frac{1}{2}, -\frac{1}{\sqrt{2}}, \frac{1}{2}\right) \\ \kappa_5 &= \left(\frac{1}{2}, \frac{1}{\sqrt{2}}, \frac{1}{2}\right). \end{aligned} \tag{8}$$

Based on these eigenvectors, an eigenbasis $|\psi_k\rangle$ can be constructed (cf. Table 1) in which the matrix representation of $\mathcal{H}_{\mathrm{P}}$ is diagonal and the diagonal propagator

$$U_{\mathrm{P}}(\tau) = \exp\{-\mathrm{i}\, \mathcal{H}_{\mathrm{P}} \tau\} \tag{9}$$

can be calculated directly by exponentiating the diagonal elements of $-\mathrm{i}\, \mathcal{H}_{\mathrm{P}} \tau$.

TABLE 1

Eigenbasis of the Planar Mixing Hamiltonian $\mathcal{H}_{\mathrm{P}}$ for a Spin System Consisting of Two Spins 1

$$\begin{aligned}
|\psi_1\rangle &= |1, 1\rangle \\
|\psi_2\rangle &= -\frac{1}{\sqrt{2}}|0, 1\rangle + \frac{1}{\sqrt{2}}|1, 0\rangle \\
|\psi_3\rangle &= -\frac{1}{\sqrt{2}}|1, -1\rangle + \frac{1}{\sqrt{2}}|-1, 1\rangle \\
|\psi_4\rangle &= \frac{1}{\sqrt{2}}|0, 1\rangle + \frac{1}{\sqrt{2}}|1, 0\rangle \\
|\psi_5\rangle &= \frac{1}{2}|1, -1\rangle - \frac{1}{\sqrt{2}}|0, 0\rangle + \frac{1}{2}|-1, 1\rangle \\
|\psi_6\rangle &= -\frac{1}{\sqrt{2}}|-1, 0\rangle + \frac{1}{\sqrt{2}}|0, -1\rangle \\
|\psi_7\rangle &= \frac{1}{2}|1, -1\rangle + \frac{1}{\sqrt{2}}|0, 0\rangle + \frac{1}{2}|-1, 1\rangle \\
|\psi_8\rangle &= \frac{1}{\sqrt{2}}|-1, 0\rangle + \frac{1}{\sqrt{2}}|0, -1\rangle \\
|\psi_9\rangle &= |-1, -1\rangle
\end{aligned}$$

TABLE 2

Transfer Functions $T_{A\rightarrow B}(\tau)$ of a Spin System Consisting of Two Spins 1 under Planar Mixing Conditions with the Initial Operator $A = I_z$

Target operator B	$T_{I_z\rightarrow B}(\tau)$
I_z	$\frac{1}{2} + \frac{1}{3}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) + \frac{1}{6}\cos(4\pi J^{\mathrm{eff}}\tau)$
S_z	$\frac{1}{2} - \frac{1}{3}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) - \frac{1}{6}\cos(4\pi J^{\mathrm{eff}}\tau)$
$I_z S_z^2$	$\frac{1}{2} + \frac{1}{2}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau)$
$I_z^2 S_z$	$\frac{1}{2} - \frac{1}{2}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau)$
$I_z S_x^2$, $I_z S_y^2$	$\frac{1}{2} + \frac{1}{4}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) + \frac{1}{4}\cos(4\pi J^{\mathrm{eff}}\tau)$
$I_x^2 S_z$, $I_y^2 S_z$	$\frac{1}{2} - \frac{1}{4}\cos(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) - \frac{1}{4}\cos(4\pi J^{\mathrm{eff}}\tau)$
$I_x S_y$, $-I_y S_x$	$\frac{1}{2\sqrt{2}}\sin(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) + \frac{1}{4}\sin(4\pi J^{\mathrm{eff}}\tau)$
$[I_x, I_z]_+[S_y, S_z]_+$, $-[I_y, I_z]_+[S_x, S_z]_+$	$-\frac{1}{2\sqrt{2}}\sin(2\sqrt{2}\pi J^{\mathrm{eff}}\tau) + \frac{1}{4}\sin(4\pi J^{\mathrm{eff}}\tau)$

Hence, the coherence and polarization transfer functions [1]

$$T_{A\rightarrow B}(\tau) = \frac{\mathrm{Tr}\{B^{\dagger} U_{\mathrm{P}}(\tau)\; A\; U_{\mathrm{P}}^{\dagger}(\tau)\}}{\mathrm{Tr}\{B^{\dagger}\; B\}} \tag{10}$$

between an initial operator A and a target operator B can be derived if A and B are also expressed in the eigenbasis $|\psi_k\rangle$ of $\mathcal{H}_{\mathrm{P}}$.

For the initial operator $A = I_{1z}$, all nonvanishing coherence and polarization transfer functions of the spin system consisting of two spins 1 under planar mixing conditions are summarized in Table 2. The transfer functions consist of a constant term and of one or two harmonic terms. The frequencies of the harmonic terms are $2J^{\mathrm{eff}}$ and $\sqrt{2}J^{\mathrm{eff}}$, respectively, corresponding to differences of eigenvalues in Eqs. [4] and [7], respectively.

3. RESULTS AND DISCUSSION

It is interesting to compare the results for planar mixing for two coupled spins 1 to the case of isotropic mixing (*31, 32*). The observable transfer functions for isotropic mixing for a spin system consisting of two spins 1 under the normalization condition Eq. [10] are (*32*)

$$T_{I_z\rightarrow S_z}(\tau) = \frac{1}{2} - \frac{2}{9}\cos(2\pi J\tau) - \frac{5}{18}\cos(4\pi J\tau), \tag{11}$$

$$T_{I_z\rightarrow I_x^2 S_z}(\tau) = \frac{1}{2} - \frac{1}{6}\cos(2\pi J\tau) - \frac{1}{3}\cos(4\pi J\tau), \tag{12}$$

$$T_{I_z\rightarrow I_x S_y - I_y S_x}(\tau) = \frac{1}{6}\sin(2\pi J\tau) + \frac{5}{12}\sin(4\pi J\tau). \tag{13}$$

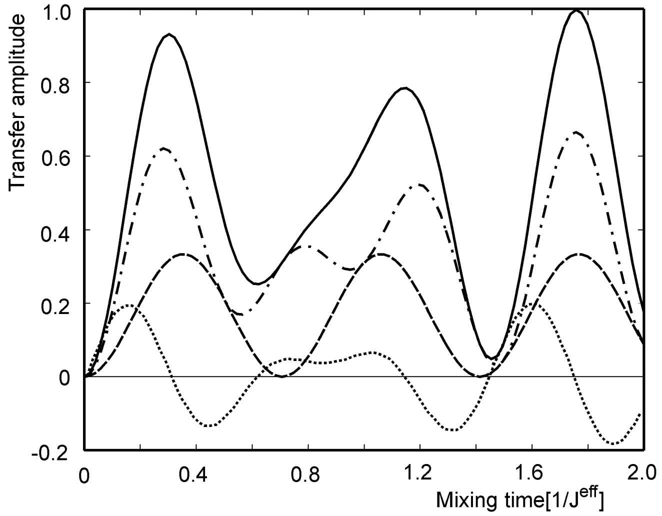

FIG. 1. Transfer functions $T_{I_z \to S_z}$ (τ) (solid line), $\frac{2}{3} T_{I_z \to I_x^2 S_z}$ (τ) (dashed–dotted line) corresponding to the contribution of the sum of the outer triplet components, $T_{I_z \to S_z} - \frac{2}{3} T_{I_z \to I_x^2 S_z}$ (τ) (dashed line) corresponding to the contribution of the inner triplet component, and $\frac{1}{3} T_{I_z \to I_x S_y - I_y S_x}$ (τ) (dotted line) under planar mixing conditions.

The transfer function $T_{I_z \to S_z}$ corresponds to the overall polarization transfer and includes all three components of the triplet, usually denoted as (1,1,1). The transfer function $T_{I_z \to I_x^2 S_z}$ describes the behavior of the outer triplet components (1,0,1) and in order to get their contribution to the overall transfer it must be scaled by the factor $\frac{2}{3}$. The contribution of the inner multiplet component (0,1,0) is the difference $T_{I_z \to S_z} - \frac{2}{3} T_{I_z \to I_x^2 S_z}$. The antiphase term $T_{I_z \to I_x S_y - I_y S_x}$ does not contribute to the overall polarization transfer function $T_{I_z \to S_z}$. To facilitate the comparison, the contributing components are visualized in Figs. 1 and 2 for the planar and isotropic case, respectively.

Whereas planar and isotropic polarization transfer functions are identical for a spin system consisting of two spins $\frac{1}{2}$ (*1*), this is not the case for spin-1 particles. According to Table 2 and Eqs. [11]–[13], the most important differences are the frequencies of the harmonic terms, which determine the characteristic form of the transfer functions. In the case of planar mixing, the ratio of the nonzero frequencies $\sqrt{2}J$ and $2J$ is not a rational number and hence the transfer functions are not periodic. This is in contrast to the case of isotropic mixing, where the nonzero frequencies J and $2J$ are related by a factor of 2 and hence, the transfer functions have a period of J^{-1} and a much simpler and more symmetric form (cf. Fig. 2). The maximum polarization transfer efficiency $T_{I_z \to S_z}$ under isotropic mixing conditions is limited to 80%, whereas under planar mixing conditions complete transfer is possible. Almost complete transfer (>99.7%) is found at a mixing time of about $1.76/J^{\mathrm{eff}}$. However, at a much shorter mixing time of $0.3/J^{\mathrm{eff}}$ a transfer efficiency of 93%,

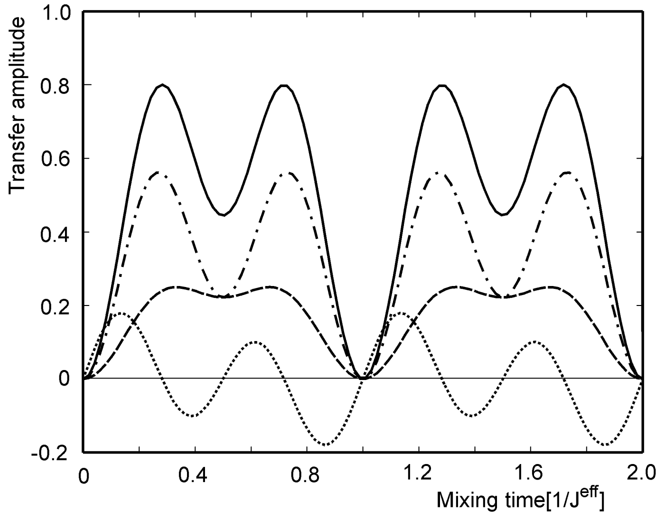

FIG. 2. Transfer functions $T_{I_z \to S_z}$ (τ) (solid line), $\frac{2}{3} T_{I_z \to I_x^2 S_z}$ (τ) (dashed–dotted line) corresponding to the contribution of the sum of the outer triplet components, $T_{I_z \to S_z} - \frac{2}{3} T_{I_z \to I_x^2 S_z}$ (τ) (dashed line) corresponding to the contribution of the inner triplet component, and $\frac{1}{3} T_{I_z \to I_x S_y - I_y S_x}$ (τ) (dotted line) under isotropic mixing conditions (*32*).

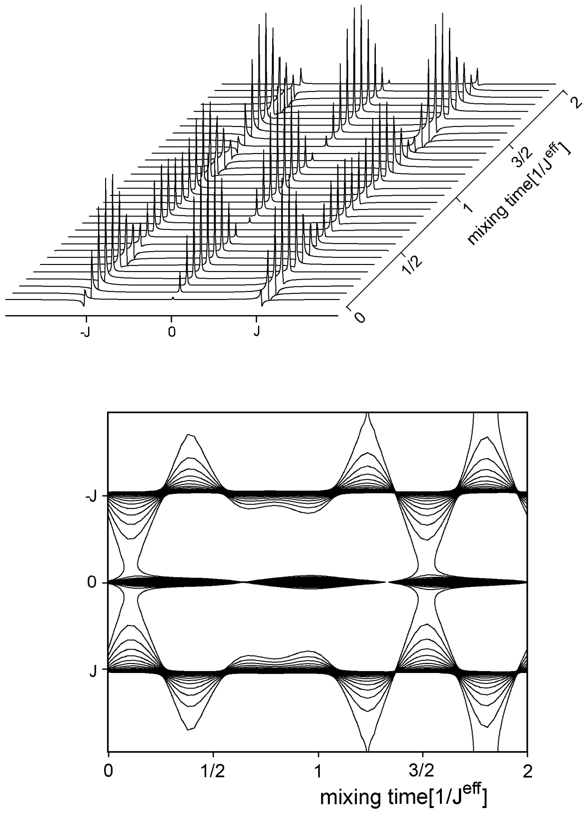

FIG. 3. Contour and stacked plots of a series of simulated 1D spectra with incremented planar mixing times. The integral over the simulated triplets corresponds to the transfer $T_{I_z \to S_z}$, while the transfer functions $\frac{2}{3} T_{I_z \to I_x^2 S_z}$ and $T_{I_z \to S_z} - \frac{2}{3} T_{I_z \to I_x^2 S_z}$ correspond to the outer and central lines, respectively. In the contour plot the effect of the dispersive antiphase magnetization on the lineshape of the multiplets, which results from the transfer $T_{I_z \to I_x S_y - I_y S_x}$ (only positive contour levels are shown), is visualized.

which significantly exceeds the transfer amplitude of isotropic mixing experiments, is found.

The derived analytical transfer functions were verified with the help of independent numerical simulations using an extended version of the program SIMONE (*30, 33*). In Figs. 3 and 4, simulated spectra of spin S are shown as a function of the mixing time τ for planar and isotropic mixing, respectively. In the simulations, planar mixing in the yz plane was applied to the initial operator I_x and the expectation value of the operator S^- was calculated. This case is equivalent to the transfers $T_{I_z\to B}$ in Table 2 and Eqs. [11]–[13], since only the coordinate system is changed from x, y, z to y, z, x. The outer lines of the triplets (corresponding to $\frac{2}{3}\,T_{I_z\to I_x^2 S_z}$ in Table 2 and Eqs. [11]–[13]) show a time dependence different than that of the central line (corresponding to $T_{I_z\to S_z}-\frac{2}{3}\,T_{I_z\to I_x^2 S_z}$). In addition, dispersive antiphase terms corresponding to the operator $I_xS_y-I_yS_x$ result in undesirable phase distortions. The contribution of the antiphase term is best visible in the contour plots of Figs. 3 and 4. While isotropic mixing conditions create pure in-phase signals only at mixing times near integer multiples of $1/(4J^{\text{eff}})$, planar mixing results in approximately pure in-phase signals in a wide range of mixing times $0.6/J^{\text{eff}} \le \tau \le 1.1/J^{\text{eff}}$.

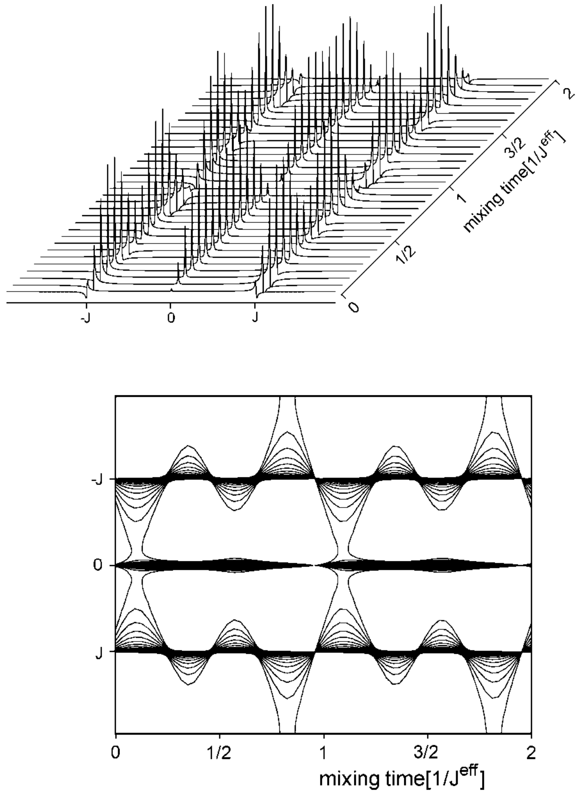

FIG. 4. Contour and stacked plots of a series of simulated 1D spectra with incremented isotropic mixing times. The integral over the simulated triplets corresponds to the transfer $T_{I_z\to S_z}$, while the transfer functions $\frac{2}{3}T_{I_z\to I_x^2 S_z}$ and $T_{I_z\to S_z}-\frac{2}{3}T_{I_z\to I_x^2 S_z}$ correspond to the outer and central lines, respectively. In the contour plot the effect of the dispersive antiphase magnetization on the lineshape of the multiplets, which results from the transfer $T_{I_z\to I_xS_y-I_yS_x}$ (only positive contour levels are shown), is visualized.

In heteronuclear polarization transfer experiments between two spin-1 nuclei (as, for example, in ^{6}Li–^{2}H or ^{14}N–^{2}H spin pairs), planar mixing (P) is preferable to isotropic mixing (I) not only in terms of transfer amplitude but also in terms of transfer time, because $J_{\text{P}}^{\text{eff}} \approx 1.5 J_{\text{I}}^{\text{eff}}$. In homonuclear spin systems consisting of two spin-1 nuclei with short relaxation times due to quadrupolar interactions the transfer is significantly faster under isotropic mixing conditions than under planar mixing conditions ($J_{\text{P}}^{\text{eff}} \approx 0.5 J_{\text{I}}^{\text{eff}}$) and therefore highly preferred. Planar mixing may be of advantage in homonuclear cases where the gain in transfer amplitude is not counterbalanced by relaxation losses or if relatively clean multiplets are desirable for a range of coupling constants. However, in the more practical case of perdeuterated molecules with spin systems consisting of more than two coupled spin-1 nuclei (*31*), either isotropic or planar mixing conditions might be favorable depending on the coupling network of the spins, as was previously shown for the spin-$\frac{1}{2}$ case (e.g., (*26*)).

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