Supporting Information

Synthesis and Characterization of Dimethyl(di(2-pyridyl))borate Nickel(II) Complexes: A Unimolecular Square Planar to Square Planar Rotation Around Nickel(II)

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I. Graphical Spectra

\[
\text{[(py)$_2$BMe$_2$]Ni(PPh$_3$)$_2$Cl (4)}
\]

\[\text{N} \quad \text{N} \quad \text{B} \quad \text{C} \quad \text{Cl} \]

**Figure S-I.1.** $^1$H NMR Spectrum of 4 at 25 °C in CD$_2$Cl$_2$.

**Figure S-I.2.** $^1$H NMR Spectrum of 4 at –40 °C in CD$_2$Cl$_2$. 

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Figure S-I.3. $^{13}$C NMR Spectrum of 4 at 25 °C in CD$_2$Cl$_2$.

Figure S-I.4. $^{11}$B NMR Spectrum of 4 at 25 °C in CD$_2$Cl$_2$. 
Figure S-I.5. $^{31}$P NMR Spectrum of 4 at -40 °C in CD$_2$Cl$_2$.

Figure S-I.6. 1D NOESY Spectrum of 4 at -50 °C in CD$_2$Cl$_2$ (the upfield B(Me) is exo).
**Figure S-I.7.** 1D NOESY Spectrum of 4 at −50 °C in CD$_2$Cl$_2$ (the downfield B(Me) is endo).

**Figure S-I.8.** IR Spectrum of 4.
[(py)$_2$BMe$_2$]Ni(acac) (6)

Figure S-1.9. $^1$H NMR Spectrum of 6 at 25 °C in CD$_2$Cl$_2$.

Figure S-1.10. $^{13}$C NMR Spectrum of 6 at 25 °C in CD$_2$Cl$_2$. 
Figure S-I.11. $^{11}$B NMR Spectrum of 6 at 25 °C in CD$_2$Cl$_2$.

Figure S-I.12. IR Spectrum of 6.
II. Studies on the Effect of PPh₃ Addition on the Rate of Rotation

Inversion recovery data were acquired on a VNMRS 500 or a VNMRS 600 according to previously published procedures. Inversion recovery data were then fitted into CIFIT 2.0 to obtain a rate constant (with a corresponding error) for each set of inversion recovery data.

Inversion recovery data were acquired using a screw cap NMR tube beginning with a 12.0 mM CD₂Cl₂ solution (1 mL) of nickel 4 and 0 equivalent of PPh₃. Inversion recovery data were then obtained for solutions of 4 with 0.1, 0.2, 0.3, 0.4, 0.5, 0.75, 1, 1.25, 1.5, 2.0, 5, and 10 equivalents of PPh₃ at 41.1 °C. The PPh₃ was added via syringe as a concentrated solution (1.0 M PPh₃; 0.3 μL solution is added to obtain 0.1 equivalent). The temperature was calibrated (tempcal) using a methanol standard. Table S-II.1 shows the rate constants obtained when each set of inversion recovery data were fitted into CIFIT 2.0. A plot of ln[PPh₃] vs ln kₜₐₜ (Chart S-II.1) shows that the rate of rotation is independent of [PPh₃]. The error on the slope of the trend line and the y-intercept were calculated using a least squares optimization: 0.005(10) and 3.28(5), respectively. See Section VIII of the SI for inversion recovery data.

**Table S-II.1. Rotation Rate Constants Obtained from Inversion Recovery Experiments.**

| Run | [Ni] (M) | [PPh₃] (M) | PPh₃ Equiv. | kₜₐₜ (s⁻¹) | er(kₜₐₜ) (s⁻¹) | ln[PPh₃] | ln kₜₐₜ | er (ln kₜₐₜ) |
|-----|----------|-------------|-------------|-------------|----------------|-----------|---------|--------------|
| 1   | 0.0120   | 0.0000      | 0           | 24.6496     | 0.3624         | 3.2048    | 0.0147  |
| 2   | 0.0120   | 0.0012      | 0.1         | 25.1335     | 0.1980         | -6.7254   | 3.2242  | 0.007877    |
| 3   | 0.0119   | 0.0024      | 0.2         | 24.7189     | 0.4233         | -6.0323   | 3.2076  | 0.017126    |
| 4   | 0.0119   | 0.0036      | 0.3         | 25.0113     | 0.2475         | -5.6268   | 3.2193  | 0.009897    |
| 5   | 0.0119   | 0.0048      | 0.4         | 25.6950     | 0.2316         | -5.3391   | 3.2463  | 0.009012    |
| 6   | 0.0118   | 0.0059      | 0.5         | 27.8497     | 0.5792         | -5.1309   | 3.3268  | 0.020796    |
| 7   | 0.0118   | 0.0089      | 0.75        | 27.4584     | 0.4796         | -4.7217   | 3.3127  | 0.017467    |
| 8   | 0.0118   | 0.0118      | 1           | 26.2667     | 0.3792         | -4.4436   | 3.2683  | 0.014436    |
| 9   | 0.0117   | 0.0120      | 1.25        | 27.1235     | 0.3685         | -4.4228   | 3.3022  | 0.01356     |
| 10  | 0.0117   | 0.0175      | 1.5         | 25.7745     | 0.2856         | -4.0440   | 3.2494  | 0.011081    |
| 11  | 0.0116   | 0.0232      | 2           | 24.9912     | 0.3304         | -3.7622   | 3.2185  | 0.013222    |
| 12  | 0.0112   | 0.0561      | 5           | 26.1666     | 0.3257         | -2.8801   | 3.2645  | 0.012446    |
| 14  | 0.0106   | 0.1063      | 10          | 25.5716     | 0.3028         | -2.2416   | 3.2415  | 0.011841    |
Chart S-II.1. Plot of ln $[\text{PPh}_3]$ vs ln $k_{\text{obs}}$. 

\[ y = m_1 + m_2 \cdot M_0 \]

Error

| Value   | m_1    | m_2    |
|---------|--------|--------|
|         | 0.0095771 | 0.0051408 |

Chisq

| Value   | 0.016556 |

$R^2$

| Value   | 0.0028007 |
III. $^{31}$P NMR Inversion Recovery Experiment for PPh$_3$ Exchange

To a 1 mL 12 mM CD$_2$Cl$_2$ solution of 4, 0.5 equiv PPh$_3$ (6 μmol; 1.5 μL of a 1 M PPh$_3$ stock solution) was added via syringe. $^{31}$P NMR inversion recovery was performed using a Varian VNMRS 500 at $-42.4$ °C. The temperature was calibrated (tempcal) using a methanol standard. When coordinated PPh$_3$ (Figure S–III.1; 12.60 ppm) is pulsed, no magnetization transfer from coordinated PPh$_3$ to free PPh$_3$ (−7.23 ppm) is observed. The inversion recovery stacked NMR spectra using different mixing times (d2) is shown in Figure S–III.2. A plot of Integration vs d2 is shown in Chart S–III.1. The integration for the free PPh$_3$ peak does not change, indicating that no magnetization transfer is occurring. See Section IX of the SI for inversion recovery data.

Figure S–III.1. $^{31}$P NMR Spectrum of 4 and Free PPh$_3$ at $-42.4$ °C.
**Figure S–III.2.** Stacked Spectra of $^{31}$P NMR Inversion Recovery Data.

**Chart S–III.1.** $^{31}$P NMR Inversion Recovery Data (Plot of Integration vs d2).

| Parameter | Value 1 | Value 2 |
|-----------|---------|---------|
| m1        | 0.0073478 | 0.00040232 |
| m2        | 1.767 | 0.00041336 |
| m3        | NA | 0.010868 |
| Chisq     | NA | 0.020278 |
| R²        | -0.76701 | 0.52845 |

Equation: $y = m1 + m2 \cdot (1 - \exp(-x/m3))$
IV. Studies on the Effect of \((n\text{-Bu})_4\text{NCl}\) and LiCl Addition on the Rate of Rotation

Addition of \((n\text{-Bu})_4\text{NCl}\) to a solution of complex 4 results in a reaction leading to formation of an unidentified paramagnetic side product (see Figure S-IV.1 below). Nevertheless, a small amount of complex 4 remains in solution and measurement of the rate of rotation using inversion recovery experiments is possible.

![Figure S-IV.1. \(^1\)H NMR Spectrum of 4 in the presence of 1 equiv. \((n\text{-Bu})_4\text{NCl}\) at -43.0 °C.](image)

Inversion recovery data were acquired using a J-Young NMR tube beginning with a 12.0 mM solution (1 mL) of nickel 4 and 0.4 equiv. \((n\text{-Bu})_4\text{NCl}\). Inversion recovery data were then obtained for solutions of 4 with 0.7, 1.1, 1.5, and 2.0 equivalents of \((\text{Bu})_4\text{NCl}\) at -43.0 °C. \((n\text{-Bu})_4\text{NCl}\) were added in one portion into the J-Young tube containing nickel complex 4 in the glove box. The temperature was calibrated using a methanol standard (tempcal). Table S-IV.1 shows the rate constants obtained when each set of inversion recovery data were fitted into CIFIT 2.0. A plot of \(\ln[(n\text{-Bu})_4\text{NCl}]\) added vs \(\ln k_{\text{obs}}\) (Chart S-IV.1) shows that the rate of rotation is independent of added \((n\text{-Bu})_4\text{NCl}\). The error on the slope of the trend line and the y-intercept were calculated using a least squares optimization: -0.01(3) and 3.0(1), respectively.
Table S-IV.1. Rotation Rate Constants With Added (n-Bu)$_4$NCl.

| Run | Equiv. NBu4Cl | [NBu4Cl] (M) | $k$ (s$^{-1}$) | er$k$ (s$^{-1}$) |
|-----|---------------|--------------|---------------|-----------------|
| 1   | 0.4           | 0.0048       | 21.509204     | 0.386532        |
| 2   | 0.7           | 0.0084       | 21.374766     | 0.656295        |
| 3   | 1.1           | 0.0132       | 22.238574     | 1.172882        |
| 4   | 1.5           | 0.018        | 22.2577       | 0.514013        |
| 5   | 2             | 0.024        | 20.394057     | 0.832915        |

Chart S-IV.1. Plot of Equivalents of (n-Bu)$_4$NCl Added vs $k_{obs}$.

The rate of rotation was also measured in the presence of excess LiCl, which is insoluble in CD$_2$Cl$_2$. Unlike addition (n-Bu)$_4$NCl, no reaction occurs upon addition of LiCl. Inversion recovery data were acquired using a J–Young NMR tube beginning with a 12.0 mM solution (1 mL) of nickel 4 and 0 equivalent of LiCl. Inversion recovery data were then obtained for solutions of 4 with 6, 12, 24, and 48 equivalents of LiCl at −42.0 °C. LiCl was added in one portion into the J–Young tube containing nickel complex 4 in the glove box. The temperature was calibrated (tempcal) using a methanol standard. Table S-IV.2 shows the rate constants obtained when each set of inversion recovery data were fitted into CIFIT 2.0. A plot of equivalents of LiCl added vs $k_{obs}$ (Chart S-IV.2) shows that the rate of rotation is independent of added LiCl. The error on the slope of the trend line and the y-intercept were calculated using a least squares optimization: −0.02(3) and 22.7(7), respectively. See Section IX of the SI for inversion recovery data.
### Table S-IV.2. Rotation Rate Constants with Added LiCl.

| Run | Equiv. LiCl | $k_{\text{obs}}$ (s$^{-1}$) | $\text{er}(k_{\text{obs}})$ (s$^{-1}$) |
|-----|-------------|-----------------|-----------------|
| 1   | 0           | 21.321882       | 0.3571153       |
| 2   | 6           | 23.315415       | 0.250699        |
| 3   | 12          | 23.126912       | 0.221502        |
| 4   | 24          | 22.650877       | 0.371693        |
| 5   | 48          | 21.093599       | 0.255429        |

### Chart S-IV.2. Plot of Equivalents of LiCl Added vs $k_{\text{obs}}$.

![Chart of LiCl Added vs $k_{\text{obs}}$]
V. Tl(OTf) Experiments

To a solution of 4 (5.0 mg, 9 μmol) in CD₂Cl₂, Tl(OTf) (6.5 mg, 18.4 μmol) was added. The ¹H NMR spectrum of 4 remained largely the same even after standing for 4 days at room temperature. ¹H NMR inversion recovery experiments were performed and rate constants ca. 1 h and 4 d after addition were obtained at −42.0 °C. The rate constants of rotation for both experiments were the same as the rate constant in the absence of thallium. See Section X of the SI for inversion recovery data.

Figure S–V.1. ¹H NMR Spectrum of 4 at 25 °C 1 h after addition of 2 equiv. Tl(OTf).
Figure S–V.2. $^1$H NMR Spectrum of 4 at 25 °C 4 d after addition of 2 equiv. Tl(OTf).

Table S–V.1. Rotation Rate Constants with Added Tl(OTf).

| Run | Time  | Tl(OTf) Equiv. | $k_{obs}$ (s$^{-1}$) | er($k_{obs}$) (s$^{-1}$) |
|-----|-------|----------------|----------------------|--------------------------|
| 1   | 1 h   | 2              | 23.10981             | 0.361239                 |
| 2   | 4d    | 2              | 23.52746             | 0.617042                 |
VI. Eyring Plot for Rotation

Inversion recovery data were acquired on a Varian VNMRS 500 and VNMRS 600 (using a 12 mM CD$_2$Cl$_2$ solution of nickel 4) according to previously published procedure. Inversion recovery data were then fitted into CIFIT 2.0 to obtain a rate constant of each set of inversion recovery data. Table S–VI.1 shows the rate constants of rotation at different temperatures obtained from inversion recovery data. Standard error values for the activation parameters, σ(ΔH$^\ddagger$) and σ(ΔS$^\ddagger$), were calculated based on the equations derived by Girolami et al. Temperatures were calibrated using a methanol standard. From the Eyring plot (Chart S–VI.1), ΔH$^\ddagger$ = 12.2(1) kcal mol$^{-1}$ and ΔS$^\ddagger$ = 0.8(5) eu. See Section XI of the SI for inversion recovery data.

Table S–VI.1. Data for Eyring Analysis of Rotation.

| T (°C) | T (K) | $k_{obs}$ (s$^{-1}$) | $1000/T$ (K$^{-1}$) | $R\ln(k/T) - R\ln(k_B/h)$ | σ(ΔH$^\ddagger$) (kcal/mol) | σ(ΔS$^\ddagger$) (eu) |
|--------|-------|----------------------|---------------------|-----------------------------|-----------------------------|------------------------|
| -61.6  | 211.5 | 1.10                 | 4.73                | -57.63                      | 0.10                        | 0.43                   |
| -61.4  | 211.8 | 1.92                 | 4.72                | -56.53                      | 0.11                        | 0.45                   |
| -53.7  | 219.5 | 6.00                 | 4.56                | -54.33                      | 0.10                        | 0.41                   |
| -46.8  | 226.3 | 12.78                | 4.42                | -52.89                      | 0.09                        | 0.40                   |
| -42.0  | 231.2 | 23.23                | 4.33                | -51.74                      | 0.10                        | 0.16                   |
| -32.6  | 240.5 | 60.20                | 4.16                | -49.93                      | 0.09                        | 0.19                   |
| -31.8  | 241.3 | 81.68                | 4.14                | -49.33                      | 0.22                        | 0.87                   |
| -18.9  | 254.2 | 204.53               | 3.93                | -47.61                      | 0.23                        | 0.94                   |

<σ(ΔH$^\ddagger$)> 0.13  <σ(ΔS$^\ddagger$)> 0.48

Chart S–VII.1. Eyring Plot for Rotation.
VII. Eyring Plots for Ring Flip

Inversion recovery data were acquired on a Varian VNMRS 600 (using a 12 mM CD$_2$Cl$_2$ solution of nickel 4) according to previously published procedure. Inversion recovery data were then fitted into CIFIT 2.0 to obtain a rate constant of each set of inversion recovery data. Table S-VII.1 shows the rate constants of ring flip in CD$_2$Cl$_2$ at different temperatures obtained from inversion recovery data. Standard error values for the activation parameters, $\sigma(\Delta H^\ddagger)$ and $\sigma(\Delta S^\ddagger)$, were calculated based on the equations derived by Girolami et al. Temperatures were calibrated using a methanol standard. From the Eyring plot (Chart S-VII.1), $\Delta H^\ddagger = 15.0(2)$ kcal mol$^{-1}$ and $\Delta S^\ddagger = -4.2(7)$ eu. See Section XII of the SI for inversion recovery data.

**Table S-VII.1. Data for Eyring Analysis of Ring Flip in CD$_2$Cl$_2$.**

| T (°C) | T (K) | $k_{obs}$ (s$^{-1}$) | 1000/T (K$^{-1}$) | Rln(k/T) - Rln(k_B/h) | $\sigma(\Delta H^\ddagger)$ (kcal/mol) | $\sigma(\Delta S^\ddagger)$ (eu) |
|-------|-------|----------------------|-------------------|------------------------|------------------------------------------|-------------------------------|
| 286.8 | 13.7  | 2.69 (4)             | 3.49              | -56.45                 | 0.25                                     | 0.82                          |
| 292.0 | 18.8  | 4.35 (2)             | 3.43              | -55.54                 | 0.17                                     | 0.63                          |
| 301.3 | 28.1  | 10.02 (4)            | 3.32              | -53.94                 | 0.17                                     | 0.61                          |
| 311.4 | 38.2  | 23.16 (17)           | 3.21              | -52.34                 | 0.18                                     | 0.63                          |

<\sigma(\Delta H^\ddagger)> 0.19  
<\sigma(\Delta S^\ddagger)> 0.67

**Chart S-VII.1. Eyring Plot for Ring Flip in CD$_2$Cl$_2$.**

Ring flip rate constants in CD$_2$Cl$_2$ were collected over a temperature range of only 24.5 °C because the solvent boils at 40 °C. Because it is recommended that data collected
over a temperature range of 40 °C be obtained for accurate values of $\Delta S^\ddagger$, we attempted to obtain an Eyring plot for the ring flip in C$_6$D$_6$. Table S–VII.2 shows the rate constants of ring flip (using a 12 mM C$_6$D$_6$ solution of 4) at different temperatures. Data were collected over a temperature range of 37 °C because attempts to obtain a rate constant at 47 °C were unsuccessful (data would not converge when fitted into CIFIT). From the Eyring plot (Chart S–VII.2), $\Delta H^\ddagger = 15.5(3)$ kcal mol$^{-1}$ and $\Delta S^\ddagger = -3.3(11)$ eu.

**Table S–VII.2.** Data for Eyring Analysis of Ring Flip in C$_6$D$_6$.

| T (°C) | T (K) | $k_{obs}$ (s$^{-1}$) | 1000/T (K$^{-1}$) | Rln(k/T) - Rln(k$_B$ / h) | σ($\Delta H^\ddagger$) (kcal/mol) | σ($\Delta S^\ddagger$) (eu) |
|--------|-------|---------------------|-------------------|--------------------------|----------------------------------|-----------------------------|
| 7.0    | 280.2 | 1.13                | 3.57              | -58.14                   | 0.68                            | 2.28                        |
| 13.8   | 287.0 | 1.61                | 3.48              | -57.47                   | 0.15                            | 0.50                        |
| 24.0   | 297.1 | 3.88                | 3.37              | -55.79                   | 0.18                            | 0.61                        |
| 34.3   | 307.4 | 11.71               | 3.25              | -53.67                   | 0.17                            | 0.57                        |
| 44.2   | 317.3 | 31.59               | 3.15              | -51.76                   | 0.42                            | 1.41                        |

$<\sigma(\Delta H^\ddagger)>$ 0.32  $<\sigma(\Delta S^\ddagger)>$ 1.07

**Chart S–VII.2.** Eyring Plot for Ring Flip in C$_6$D$_6$. 

![Eyring Plot](image)
VIII. Inversion Recovery Data for Section III (PPh₃ Addition).

1) Data for 0 Equivalent PPh₃:

![Stacked Spectra of Inversion Recovery Data at 0 Equivalent PPh₃](image)

**Figure S–VIII.1. Stacked Spectra of Inversion Recovery Data at 0 Equivalent PPh₃.**

**Table S–VIII.1. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|------|------|
| 0.001 | -0.3416     | 0.8998      | -0.3289| 0.9086 | -0.0126 | -0.0088 |
| 0.002 | -0.3179     | 0.8771      | -0.3102| 0.8845 | -0.0077 | -0.0074 |
| 0.003 | -0.2954     | 0.8555      | -0.2959| 0.8578 | 0.0005  | -0.0022 |
| 0.004 | -0.2740     | 0.8350      | -0.2733| 0.8348 | -0.0008 | 0.0002  |
| 0.005 | -0.2536     | 0.8155      | -0.2483| 0.8219 | -0.0053 | -0.0064 |
| 0.007 | -0.2157     | 0.7793      | -0.2122| 0.7837 | -0.0035 | -0.0045 |
| 0.010 | -0.1654     | 0.7314      | -0.1559| 0.7403 | -0.0095 | -0.0088 |
| 0.015 | -0.0963     | 0.6662      | -0.0950| 0.6685 | -0.0014 | -0.0023 |
| 0.020 | -0.0422     | 0.6157      | -0.0391| 0.6242 | -0.0031 | -0.0085 |
| 0.025 | 0.0003      | 0.5766      | 0.0019 | 0.5778 | -0.0016 | -0.0012 |
| 0.030 | 0.0338      | 0.5465      | 0.0358 | 0.5509 | -0.0020 | -0.0045 |
| 0.040 | 0.0811      | 0.5055      | 0.0712 | 0.4939 | 0.0099  | 0.0116  |
| 0.050 | 0.1109      | 0.4817      | 0.1079 | 0.4756 | 0.0030  | 0.0061  |
| 0.060 | 0.1301      | 0.4684      | 0.1254 | 0.4575 | 0.0047  | 0.0109  |
| 0.070 | 0.1428      | 0.4613      | 0.1440 | 0.4577 | -0.0011 | 0.0036  |
| 0.080 | 0.1516      | 0.4581      | 0.1545 | 0.4594 | -0.0028 | -0.0013 |
| d2  | Inversion Recovery Data for 0 Equiv. PPh₃ |
|-----|------------------------------------------|
| 0.090 | 0.1581 0.4573 0.1597 0.4549 -0.0017 0.0024 |
| 0.100 | 0.1630 0.4579 0.1621 0.4484 0.0009 0.0094 |
| 0.150 | 0.1796 0.4683 0.1783 0.4601 0.0014 0.0082 |
| 0.200 | 0.1930 0.4814 0.1985 0.4844 -0.0055 -0.0031 |
| 0.250 | 0.2058 0.4943 0.2053 0.4803 0.0005 0.0140 |
| 0.300 | 0.2182 0.5069 0.2148 0.4980 0.0035 0.0089 |
| 0.350 | 0.2304 0.5193 0.2280 0.5096 0.0024 0.0097 |
| 0.400 | 0.2422 0.5313 0.2470 0.5335 -0.0048 -0.0022 |
| 0.500 | 0.2650 0.5544 0.2567 0.5338 0.0083 0.0206 |
| 0.600 | 0.2867 0.5764 0.2870 0.5722 -0.0003 0.0042 |
| 0.700 | 0.3073 0.5974 0.3089 0.5972 -0.0016 0.0001 |
| 0.800 | 0.3269 0.6173 0.3273 0.6187 -0.0004 -0.0014 |
| 0.900 | 0.3456 0.6362 0.3421 0.6297 0.0035 0.0065 |
| 1.000 | 0.3633 0.6542 0.3636 0.6572 -0.0002 -0.0030 |
| 1.500 | 0.4398 0.7319 0.4391 0.7336 0.0008 -0.0018 |
| 2.000 | 0.4994 0.7923 0.4992 0.8017 0.0002 -0.0094 |
| 2.500 | 0.5457 0.8393 0.5471 0.8496 -0.0014 -0.0103 |
| 3.000 | 0.5817 0.8758 0.5819 0.8871 -0.0002 -0.0113 |
| 5.000 | 0.6617 0.9571 0.6589 0.9615 0.0028 -0.0044 |
| 6.000 | 0.6800 0.9756 0.6768 0.9799 0.0031 -0.0043 |
| 8.000 | 0.6977 0.9936 0.6958 0.9961 0.0020 -0.0025 |
| 10.000 | 0.7042 1.0002 0.6982 1.0091 0.0060 -0.0088 |
| 15.000 | 0.7077 1.0037 0.6997 1.0007 0.0080 0.0030 |
| 20.000 | 0.7080 1.0040 0.7020 1.0049 0.0060 -0.0009 |

**Chart S–VIII.1.** Plot of Inversion Recovery Data at 0 Equivalent of PPh₃₂.
CIFIT Guesses and Fit Parameters

Initial values of parameters:
1/T1's
No. 0 =  0.8000 No. 1 =  0.2300
M(\infty)'s
No. 2 =  0.9000 No. 3 =  1.0000
M(0)-M(\infty)'s
No. 4 = -1.4000 No. 5 =  0.5000
and M(0)'s for reference
No. 4 = -0.5000 No. 5 =  1.5000
Rates
No. 6 =  20.0000

Final Values of Fitted Parameters and Uncertainties:
#  0 =     0.870554 +/- 0.172420
#  1 =     0.138770 +/- 0.170388
#  2 =     0.708001 +/- 0.002499
#  3 =     1.004030 +/- 0.002663
#  4 =    -1.074394 +/- 0.003546
#  5 =    -0.080304 +/- 0.004447
#  6 =    24.649642 +/- 0.362351

2) Data for 0.1 Equivalent PPh3-

![Figure S-VIII.2. Stacked Spectra of Inversion Recovery Data at 0.1 Equivalent PPh3-](image)

**Table S-VIII.2. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|-------|-------|-----|-----|
| 0.001  | -0.3273     | 0.8696      | -0.3280 | 0.8705 | 0.0007 | -0.0009 |
| 0.002  | -0.3010     | 0.8442      | -0.3030 | 0.8511 | 0.0020 | -0.0069 |
| 0.003  | -0.2759     | 0.8202      | -0.2701 | 0.8215 | -0.0058 | -0.0014 |
| 0.004  | -0.2521     | 0.7973      | -0.2476 | 0.8003 | -0.0045 | -0.0030 |
| 0.005  | -0.2294     | 0.7756      | -0.2208 | 0.7826 | -0.0086 | -0.0070 |
| 0.007  | -0.1873     | 0.7354      | -0.1773 | 0.7362 | -0.0100 | -0.0008 |
| 0.010  | -0.1316     | 0.6823      | -0.1340 | 0.6826 | 0.0024 | -0.0003 |
|       |       |       |       |       |       |
|-------|-------|-------|-------|-------|-------|
| 0.015 | -0.0554 | 0.6104 | -0.0483 | 0.6160 | -0.0071 | -0.0056 |
| 0.020 | 0.0041 | 0.5549 | 0.0001 | 0.5551 | 0.0040 | -0.0002 |
| 0.025 | 0.0506 | 0.5122 | 0.0500 | 0.5123 | 0.0006 | -0.0001 |
| 0.030 | 0.0870 | 0.4795 | 0.0794 | 0.4762 | 0.0076 | 0.0033 |
| 0.040 | 0.1382 | 0.4354 | 0.1331 | 0.4314 | 0.0050 | 0.0040 |
| 0.050 | 0.1702 | 0.4101 | 0.1675 | 0.4079 | 0.0027 | 0.0022 |
| 0.060 | 0.1907 | 0.3961 | 0.1910 | 0.3940 | -0.0002 | 0.0022 |
| 0.070 | 0.2043 | 0.3890 | 0.2035 | 0.3858 | 0.0008 | 0.0032 |
| 0.080 | 0.2137 | 0.3860 | 0.2144 | 0.3823 | -0.0007 | 0.0036 |
| 0.090 | 0.2206 | 0.3854 | 0.2178 | 0.3766 | 0.0028 | 0.0087 |
| 0.100 | 0.2260 | 0.3863 | 0.2324 | 0.3871 | -0.0064 | -0.0008 |
| 0.150 | 0.2444 | 0.3986 | 0.2433 | 0.3924 | 0.0011 | 0.0062 |
| 0.200 | 0.2595 | 0.4134 | 0.2596 | 0.4100 | -0.0002 | 0.0033 |
| 0.250 | 0.2740 | 0.4280 | 0.2773 | 0.4281 | -0.0033 | -0.0001 |
| 0.300 | 0.2881 | 0.4423 | 0.2871 | 0.4385 | 0.0009 | 0.0038 |
| 0.350 | 0.3018 | 0.4562 | 0.3004 | 0.4516 | 0.0014 | 0.0046 |
| 0.400 | 0.3152 | 0.4698 | 0.3143 | 0.4658 | 0.0009 | 0.0039 |
| 0.500 | 0.3409 | 0.4959 | 0.3432 | 0.4959 | -0.0022 | 0.0000 |
| 0.600 | 0.3654 | 0.5207 | 0.3644 | 0.5195 | 0.0010 | 0.0011 |
| 0.700 | 0.3887 | 0.5442 | 0.3906 | 0.5427 | -0.0020 | 0.0015 |
| 0.800 | 0.4107 | 0.5666 | 0.4127 | 0.5675 | -0.0019 | -0.0009 |
| 0.900 | 0.4317 | 0.5878 | 0.4313 | 0.5873 | 0.0004 | 0.0005 |
| 1.000 | 0.4516 | 0.6080 | 0.4520 | 0.6121 | -0.0003 | -0.0041 |
| 1.500 | 0.5372 | 0.6947 | 0.5358 | 0.6954 | 0.0014 | -0.0006 |
| 2.000 | 0.6033 | 0.7618 | 0.6011 | 0.7625 | 0.0023 | -0.0007 |
| 2.500 | 0.6545 | 0.8136 | 0.6523 | 0.8139 | 0.0022 | -0.0003 |
| 3.000 | 0.6940 | 0.8537 | 0.6940 | 0.8589 | 0.0000 | -0.0052 |
| 5.000 | 0.7806 | 0.9414 | 0.7807 | 0.9459 | -0.0001 | -0.0044 |
| 6.000 | 0.8000 | 0.9611 | 0.7987 | 0.9668 | 0.0012 | -0.0057 |
| 8.000 | 0.8185 | 0.9786 | 0.8174 | 0.9806 | 0.0011 | -0.0008 |
| 10.000 | 0.8251 | 0.9865 | 0.8219 | 0.9868 | 0.0032 | -0.0003 |
| 15.000 | 0.8285 | 0.9899 | 0.8274 | 0.9915 | 0.0010 | -0.0016 |
| 20.000 | 0.8287 | 0.9902 | 0.8223 | 0.9907 | 0.0065 | -0.0005 |
Chart S-VIII.2. Plot of Inversion Recovery Data at 0.1 Equivalent of PPh₃.

CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 = 0.8000  No. 1 = 0.2300

M(∞)'s
No. 2 = 0.9000  No. 3 = 1.0000

M(0)-M(∞)'s
No. 4 = -1.4000  No. 5 = 0.5000

and M(0)'s for reference
No. 4 = -0.5000  No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 0.852153 +/- 0.092977
# 1 = 0.181609 +/- 0.092132
# 2 = 0.828748 +/- 0.001450
# 3 = 0.990222 +/- 0.001542
# 4 = -1.183752 +/- 0.002075
# 5 = -0.093950 +/- 0.002597
# 6 = 25.133540 +/- 0.197980
3) **Data for 0.2 Equivalent PPh$_3$:**

![Figure S-VIII.3. Stacked Spectra of Inversion Recovery Data at 0.2 Equivalent PPh$_3$.](image)

**Table S-VIII.3. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.0016     | 0.6848      | -0.0105| 0.6782 | 0.0089| 0.0065|
| 0.002  | 0.0115      | 0.6710      | 0.0004 | 0.6636 | 0.0112| 0.0074|
| 0.003  | 0.0241      | 0.6580      | 0.0176 | 0.6535 | 0.0064| 0.0045|
| 0.004  | 0.0360      | 0.6456      | 0.0321 | 0.6391 | 0.0039| 0.0065|
| 0.005  | 0.0473      | 0.6338      | 0.0409 | 0.6272 | 0.0065| 0.0066|
| 0.007  | 0.0685      | 0.6120      | 0.0652 | 0.6073 | 0.0032| 0.0047|
| 0.010  | 0.0965      | 0.5832      | 0.0949 | 0.5807 | 0.0016| 0.0025|
| 0.015  | 0.1350      | 0.5440      | 0.1339 | 0.5407 | 0.0011| 0.0032|
| 0.020  | 0.1652      | 0.5137      | 0.1662 | 0.5143 | -0.0010| -0.0006|
| 0.025  | 0.1890      | 0.4904      | 0.1911 | 0.4904 | -0.0021| 0.0000|
| 0.030  | 0.2078      | 0.4725      | 0.2089 | 0.4744 | -0.0012| -0.0019|
| 0.040  | 0.2345      | 0.4483      | 0.2370 | 0.4520 | -0.0025| -0.0037|
| 0.050  | 0.2515      | 0.4345      | 0.2545 | 0.4390 | -0.0029| -0.0045|
| 0.060  | 0.2628      | 0.4270      | 0.2668 | 0.4340 | -0.0040| -0.0070|
| 0.070  | 0.2705      | 0.4232      | 0.2738 | 0.4280 | -0.0034| -0.0048|
| 0.080  | 0.2760      | 0.4218      | 0.2770 | 0.4273 | -0.0010| -0.0055|
| 0.090  | 0.2802      | 0.4217      | 0.2839 | 0.4287 | -0.0037| -0.0070|
| 0.100  | 0.2837      | 0.4225      | 0.2856 | 0.4289 | -0.0019| -0.0064|
| 0.150  | 0.2964      | 0.4308      | 0.2992 | 0.4379 | -0.0028| -0.0070|
Chart S–VIII.3. Plot of Inversion Recovery Data at 0.2 Equivalent of PPh₃₂.
CIFIT Guesses and Fit Parameters

Initial values of parameters:

\[ \frac{1}{T_1} \text{'s} \]
No. 0 = 0.8000 No. 1 = 0.2300

\[ M(\text{inf}) \text{'s} \]
No. 2 = 0.9000 No. 3 = 1.0000

\[ M(0)-M(\text{inf}) \text{'s} \]
No. 4 = -1.4000 No. 5 = 0.5000

and \( M(0) \)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | \( \frac{1}{T_1} \) | \( M(\text{inf}) \) | \( M(0) \) |
|----|---------------------|---------------------|-------------|
| 0  | -1.281365 +/- 0.167198 | 0.714064 +/- 0.001721 | 0.817754 +/- 0.001764 |
| 1  | 2.463663 +/- 0.195686 | 0.729518 +/- 0.002366 | -0.118545 +/- 0.002963 |
| 2  | 0.714064 +/- 0.001721 | 0.729518 +/- 0.002366 | 0.817754 +/- 0.001764 |
| 3  | 0.817754 +/- 0.001764 | 0.729518 +/- 0.002366 | -0.118545 +/- 0.002963 |
| 4  | -0.729518 +/- 0.002366 | 0.817754 +/- 0.001764 | -0.118545 +/- 0.002963 |
| 5  | -0.118545 +/- 0.002963 | 0.817754 +/- 0.001764 | -0.118545 +/- 0.002963 |
| 6  | 24.718904 +/- 0.423347 | 0.817754 +/- 0.001764 | -0.118545 +/- 0.002963 |

Data for 0.3 Equivalent PPh\(_3\): 

![Figure S-VIII.4. Stacked Spectra of Inversion Recovery Data at 0.3 Equivalent PPh\(_3\)](image)

Table S-VIII.4. CIFIT Plot File.

| \( d_2 \) (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | \( \sigma M1 \) | \( \sigma M2 \) |
|----------------|-------------|-------------|-------|-------|--------------|--------------|
| 0.001          | -0.4625     | 0.7588      | -0.4548 | 0.7628 | -0.0077      | -0.0040      |
| 0.002          | -0.4346     | 0.7323      | -0.4489 | 0.7320 | 0.0144       | 0.0003       |
| 0.003          | -0.4080     | 0.7071      | -0.4117 | 0.7029 | 0.0037       | 0.0042       |
| 0.004          | -0.3827     | 0.6832      | -0.3758 | 0.6836 | -0.0069      | -0.0003      |
| 0.005          | -0.3587     | 0.6605      | -0.3538 | 0.6607 | -0.0049      | -0.0002      |
| 0.007          | -0.3140     | 0.6184      | -0.3157 | 0.6147 | 0.0017       | 0.0037       |
| 0.010          | -0.2548     | 0.5630      | -0.2515 | 0.5642 | -0.0033      | -0.0012      |
| 0.015 | -0.1736 | 0.4878 | -0.1760 | 0.4890 | 0.0024 | -0.0012 |
| 0.020 | -0.1100 | 0.4299 | -0.1138 | 0.4289 | 0.0038 | 0.0010  |
| 0.025 | -0.0602 | 0.3854 | -0.0591 | 0.3874 | -0.0011 | -0.0020 |
| 0.030 | -0.0210 | 0.3513 | -0.0238 | 0.3476 | 0.0028 | 0.0037  |
| 0.040 | 0.0344  | 0.3057 | 0.0323  | 0.3057 | 0.0022 | 0.0000  |
| 0.050 | 0.0696  | 0.2800 | 0.0654  | 0.2778 | 0.0042 | 0.0022  |
| 0.060 | 0.0925  | 0.2662 | 0.0985  | 0.2685 | -0.0060 | -0.0023 |
| 0.070 | 0.1080  | 0.2597 | 0.1099  | 0.2563 | -0.0019 | 0.0033  |
| 0.080 | 0.1191  | 0.2574 | 0.1212  | 0.2578 | -0.0021 | -0.0003 |
| 0.090 | 0.1275  | 0.2578 | 0.1271  | 0.2556 | 0.0003  | 0.0022  |
| 0.100 | 0.1342  | 0.2597 | 0.1298  | 0.2565 | 0.0044  | 0.0033  |
| 0.150 | 0.1586  | 0.2778 | 0.1657  | 0.2913 | -0.0070 | -0.0135 |
| 0.200 | 0.1794  | 0.2984 | 0.1808  | 0.3031 | -0.0015 | -0.0047 |
| 0.250 | 0.1993  | 0.3186 | 0.2006  | 0.3213 | -0.0012 | -0.0026 |
| 0.300 | 0.2187  | 0.3384 | 0.2224  | 0.3437 | -0.0037 | -0.0053 |
| 0.350 | 0.2376  | 0.3577 | 0.2409  | 0.3622 | -0.0033 | -0.0045 |
| 0.400 | 0.2561  | 0.3764 | 0.2554  | 0.3739 | 0.0007  | 0.0025  |
| 0.500 | 0.2914  | 0.4125 | 0.2903  | 0.4151 | 0.0011  | -0.0026 |
| 0.600 | 0.3250  | 0.4467 | 0.3196  | 0.4416 | 0.0054  | 0.0051  |
| 0.700 | 0.3568  | 0.4791 | 0.3547  | 0.4746 | 0.0021  | 0.0045  |
| 0.800 | 0.3870  | 0.5098 | 0.3858  | 0.5037 | 0.0012  | 0.0062  |
| 0.900 | 0.4156  | 0.5390 | 0.4174  | 0.5383 | -0.0018 | 0.0007  |
| 1.000 | 0.4427  | 0.5666 | 0.4362  | 0.5615 | 0.0066  | 0.0051  |
| 1.500 | 0.5588  | 0.6849 | 0.5564  | 0.6830 | 0.0024  | 0.0019  |
| 2.000 | 0.6478  | 0.7756 | 0.6452  | 0.7740 | 0.0026  | 0.0016  |
| 2.500 | 0.7161  | 0.8452 | 0.7154  | 0.8424 | 0.0007  | 0.0028  |
| 3.000 | 0.7685  | 0.8985 | 0.7751  | 0.9047 | -0.0066 | -0.0062 |
| 5.000 | 0.8813  | 1.0135 | 0.8911  | 1.0206 | -0.0098 | -0.0072 |
| 6.000 | 0.9059  | 1.0385 | 0.9122  | 1.0463 | -0.0063 | -0.0078 |
| 8.000 | 0.9289  | 1.0619 | 0.9323  | 1.0691 | -0.0035 | -0.0072 |
| 10.000| 0.9368  | 1.0700 | 0.9318  | 1.0662 | 0.0050  | 0.0038  |
| 15.000| 0.9407  | 1.0740 | 0.9334  | 1.0666 | 0.0074  | 0.0075  |
| 20.000| 0.9410  | 1.0743 | 0.9374  | 1.0665 | 0.0037  | 0.0078  |
**Chart S-VIII.4.** Plot of Inversion Recovery Data at 0.3 Equivalent of PPh$_3$.

![Inversion Recovery Data for 0.3 Equiv. PPh$_3$](image)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/T1's
- No. 0 = 0.8000
- No. 1 = 0.2300

M(inf)'s
- No. 2 = 0.9000
- No. 3 = 1.0000

M(0)–M(inf)'s
- No. 4 = -1.4000
- No. 5 = 0.5000

and M(0)'s for reference
- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

---

Final Values of Fitted Parameters and Uncertainties:

- # 0 = 1.001771 +/- 0.097441
- # 1 = 0.067543 +/- 0.095210
- # 2 = 0.941040 +/- 0.001922
- # 3 = 1.074323 +/- 0.002032
- # 4 = -1.432870 +/- 0.002740
- # 5 = -0.287627 +/- 0.003356
- # 6 = 25.011297 +/- 0.247532
5) Data for 0.4 Equivalent PPh$_3$:

![Figure S-VIII.5. Stacked Spectra of Inversion Recovery Data at 0.4 Equivalents PPh$_3$.](image)

**Table S-VIII.5. CIFIT Plot File.**

| $d_2$ (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-----------|-------------|-------------|--------|--------|------|------|
| 0.001     | -0.0693     | 0.7084      | -0.0642| 0.7098 | -0.0051| -0.0014|
| 0.002     | -0.0536     | 0.6927      | -0.0642| 0.6928 | 0.0106 | -0.0001|
| 0.003     | -0.0386     | 0.6778      | -0.0367| 0.6779 | -0.0019| -0.0001|
| 0.004     | -0.0244     | 0.6636      | -0.0218| 0.6646 | -0.0026| -0.0009|
| 0.005     | -0.0108     | 0.6502      | -0.0080| 0.6503 | -0.0028| -0.0001|
| 0.007     | 0.0142      | 0.6254      | 0.0181 | 0.6261 | -0.0038| -0.0006|
| 0.010     | 0.0473      | 0.5928      | 0.0490 | 0.5949 | -0.0017| -0.0021|
| 0.015     | 0.0924      | 0.5488      | 0.0934 | 0.5495 | -0.0010| -0.0008|
| 0.020     | 0.1275      | 0.5150      | 0.1281 | 0.5164 | -0.0006| -0.0014|
| 0.025     | 0.1548      | 0.4893      | 0.1540 | 0.4899 | 0.0008 | -0.0006|
| 0.030     | 0.1761      | 0.4696      | 0.1768 | 0.4708 | -0.0007| -0.0012|
| 0.040     | 0.2059      | 0.4435      | 0.2060 | 0.4434 | -0.0001| 0.0001|
| 0.050     | 0.2246      | 0.4288      | 0.2244 | 0.4302 | 0.0002 | -0.0013|
| 0.060     | 0.2366      | 0.4210      | 0.2365 | 0.4213 | 0.0001 | -0.0003|
| 0.070     | 0.2446      | 0.4172      | 0.2450 | 0.4163 | -0.0004| 0.0009|
| 0.080     | 0.2503      | 0.4158      | 0.2513 | 0.4146 | -0.0010| 0.0012|
| 0.090     | 0.2546      | 0.4158      | 0.2549 | 0.4158 | -0.0003| 0.0000|
| 0.100     | 0.2580      | 0.4167      | 0.2583 | 0.4158 | -0.0004| 0.0009|
| T (100) | b (0.1) | b (0.2) | b (0.4) | b (0.3) | b (0.4) | b (0.0015) | b (0.0009) |
|--------|---------|---------|---------|---------|---------|------------|------------|
| 0.150  | 0.2704  | 0.4255  | 0.2719  | 0.4246  | -0.0015 | 0.0009     |
| 0.200  | 0.2810  | 0.4356  | 0.2786  | 0.4330  | 0.0024  | 0.0026     |
| 0.250  | 0.2913  | 0.4455  | 0.2915  | 0.4439  | -0.0002 | 0.0017     |
| 0.300  | 0.3013  | 0.4553  | 0.3020  | 0.4543  | -0.0007 | 0.0009     |
| 0.350  | 0.3110  | 0.4647  | 0.3098  | 0.4633  | 0.0012  | 0.0015     |
| 0.400  | 0.3205  | 0.4740  | 0.3216  | 0.4727  | -0.0010 | 0.0013     |
| 0.500  | 0.3388  | 0.4918  | 0.3376  | 0.4894  | 0.0012  | 0.0024     |
| 0.600  | 0.3562  | 0.5087  | 0.3561  | 0.5077  | 0.0001  | 0.0010     |
| 0.700  | 0.3727  | 0.5248  | 0.3716  | 0.5241  | 0.0011  | 0.0007     |
| 0.800  | 0.3884  | 0.5401  | 0.3849  | 0.5383  | 0.0035  | 0.0018     |
| 0.900  | 0.4034  | 0.5547  | 0.4003  | 0.5529  | 0.0030  | 0.0018     |
| 1.000  | 0.4176  | 0.5685  | 0.4156  | 0.5672  | 0.0020  | 0.0013     |
| 1.500  | 0.4787  | 0.6281  | 0.4770  | 0.6265  | 0.0018  | 0.0016     |
| 2.000  | 0.5262  | 0.6743  | 0.5253  | 0.6757  | 0.0010  | -0.0014    |
| 2.500  | 0.5631  | 0.7102  | 0.5659  | 0.7131  | -0.0028 | -0.0029    |
| 3.000  | 0.5917  | 0.7381  | 0.5955  | 0.7414  | -0.0038 | -0.0033    |
| 5.000  | 0.6551  | 0.7998  | 0.6580  | 0.8044  | -0.0029 | -0.0046    |
| 6.000  | 0.6695  | 0.8138  | 0.6727  | 0.8186  | -0.0032 | -0.0048    |
| 8.000  | 0.6834  | 0.8273  | 0.6836  | 0.8297  | -0.0003 | -0.0024    |
| 10.000 | 0.6884  | 0.8323  | 0.6885  | 0.8307  | -0.0001 | 0.0015     |
| 15.000 | 0.6911  | 0.8349  | 0.6885  | 0.8335  | 0.0026  | 0.0013     |
| 20.000 | 0.6913  | 0.8351  | 0.6840  | 0.8299  | 0.0073  | 0.0051     |

**Chart S-VII.5.** Plot of Inversion Recovery Data at 0.4 Equivalent of PPh₃.  

![Inversion Recovery Data for 0.4 Equiv. PPh₃](image-url)
CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 =  0.8000  No. 1 =  0.2300

M(inf)'s
No. 2 =  0.9000  No. 3 =  1.0000

M(0) - M(inf)'s
No. 4 = -1.4000  No. 5 =  0.5000

and M(0)'s for reference
No. 4 = -0.5000  No. 5 =  1.5000

Rates
No. 6 =  20.0000

Final Values of Fitted Parameters and Uncertainties:

#  0 =  -0.166572 +/-  0.095998
#  1 =  1.196100 +/-  0.102556
#  2 =  0.691299 +/-  0.001012
#  3 =  0.835076 +/-  0.001057
#  4 =  -0.777191 +/-  0.001420
#  5 =  -0.110073 +/-  0.001770
#  6 =  25.695039 +/-  0.231560

6) Data for 0.6 Equivalent PPh$_3$:

![Graph showing stacked spectra of inversion recovery data at 0.5 Equivalent PPh$_3$.]

Figure S–VIII.6. Stacked Spectra of Inversion Recovery Data at 0.5 Equivalent PPh$_3$.

Table S–VIII.6. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|------|------|
| 0.001 | -0.5488     | 0.8086      | -0.5234| 0.8250 | -0.0254| -0.0164|
| 0.002 | -0.5145     | 0.7767      | -0.5247| 0.7927 | 0.0102 | -0.0160|
| 0.003 | -0.4821     | 0.7466      | -0.4613| 0.7591 | -0.0208| -0.0125|
| 0.004 | -0.4515     | 0.7182      | -0.4315| 0.7357 | -0.0200| -0.0175|
| 0.005 | -0.4225     | 0.6914      | -0.4121| 0.6981 | -0.0103| -0.0067|
| 0.007 | -0.3691 | 0.6421 | -0.3489 | 0.6580 | -0.0202 | -0.0159 |
| 0.010 | -0.2993 | 0.5780 | -0.2908 | 0.5817 | -0.0086 | -0.0038 |
| 0.015 | -0.2059 | 0.4928 | -0.2017 | 0.5055 | -0.0042 | -0.0126 |
| 0.020 | -0.1349 | 0.4291 | -0.1400 | 0.4299 | 0.0051  | -0.0008 |
| 0.025 | -0.0809 | 0.3815 | -0.0828 | 0.3877 | 0.0020  | -0.0062 |
| 0.030 | -0.0396 | 0.3461 | -0.0453 | 0.3394 | 0.0057  | 0.0067  |
| 0.040 | 0.0163  | 0.3005 | 0.0138  | 0.2976 | 0.0025  | 0.0029  |
| 0.050 | 0.0498  | 0.2762 | 0.0448  | 0.2645 | 0.0050  | 0.0117  |
| 0.060 | 0.0706  | 0.2641 | 0.0618  | 0.2465 | 0.0088  | 0.0177  |
| 0.070 | 0.0840  | 0.2590 | 0.0831  | 0.2506 | 0.0009  | 0.0084  |
| 0.080 | 0.0932  | 0.2577 | 0.0915  | 0.2425 | 0.0018  | 0.0152  |
| 0.090 | 0.1001  | 0.2587 | 0.1010  | 0.2457 | -0.0009 | 0.0130  |
| 0.100 | 0.1056  | 0.2609 | 0.1001  | 0.2435 | 0.0055  | 0.0174  |
| 0.150 | 0.1262  | 0.2781 | 0.1304  | 0.2697 | -0.0042 | 0.0084  |
| 0.200 | 0.1443  | 0.2968 | 0.1430  | 0.2800 | 0.0013  | 0.0168  |
| 0.250 | 0.1618  | 0.3151 | 0.1606  | 0.3043 | 0.0012  | 0.0109  |
| 0.300 | 0.1788  | 0.3330 | 0.1781  | 0.3185 | 0.0008  | 0.0145  |
| 0.350 | 0.1954  | 0.3504 | 0.1935  | 0.3381 | 0.0019  | 0.0123  |
| 0.400 | 0.2116  | 0.3674 | 0.2077  | 0.3517 | 0.0039  | 0.0157  |
| 0.500 | 0.2427  | 0.4001 | 0.2393  | 0.3895 | 0.0035  | 0.0106  |
| 0.600 | 0.2723  | 0.4311 | 0.2766  | 0.4252 | -0.0043 | 0.0059  |
| 0.700 | 0.3004  | 0.4606 | 0.3029  | 0.4573 | -0.0025 | 0.0034  |
| 0.800 | 0.3271  | 0.4887 | 0.3251  | 0.4782 | 0.0020  | 0.0104  |
| 0.900 | 0.3525  | 0.5153 | 0.3494  | 0.5156 | 0.0031  | -0.0003 |
| 1.000 | 0.3766  | 0.5406 | 0.3788  | 0.5393 | -0.0022 | 0.0013  |
| 1.500 | 0.4801  | 0.6492 | 0.4782  | 0.6496 | 0.0019  | -0.0003 |
| 2.000 | 0.5602  | 0.7333 | 0.5652  | 0.7446 | -0.0050 | -0.0113 |
| 2.500 | 0.6222  | 0.7983 | 0.6246  | 0.8082 | -0.0024 | -0.0099 |
| 3.000 | 0.6701  | 0.8486 | 0.6724  | 0.8663 | -0.0023 | -0.0177 |
| 5.000 | 0.7752  | 0.9590 | 0.7773  | 0.9745 | -0.0021 | -0.0155 |
| 6.000 | 0.7988  | 0.9837 | 0.7938  | 1.0001 | 0.0050  | -0.0164 |
| 8.000 | 0.8213  | 1.0073 | 0.8158  | 1.0186 | 0.0055  | -0.0113 |
| 10.000| 0.8294  | 1.0158 | 0.8181  | 1.0230 | 0.0113  | -0.0071 |
| 15.000| 0.8335  | 1.0202 | 0.8133  | 1.0230 | 0.0203  | -0.0028 |
| 20.000| 0.8338  | 1.0205 | 0.8074  | 1.0227 | 0.0265  | -0.0022 |
**Chart S-VIII.6. Plot of Inversion Recovery Data at 0.5 Equivalent of PPh$_3$.**

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/\(T_1\)'s
- No. 0 = 0.8000
- No. 1 = 0.2300

\(M(\text{inf})\)'s
- No. 2 = 0.9000
- No. 3 = 1.0000

\(M(0) - M(\text{inf})\)'s
- No. 4 = -1.4000
- No. 5 = 0.5000

and \(M(0)\)'s for reference
- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value              | Uncertainty   |
|----|--------------------|---------------|
| 0  | 1.894732           | 0.257126      |
| 1  | -0.802632          | 0.236547      |
| 2  | 0.833876           | 0.004341      |
| 3  | 1.020544           | 0.004659      |
| 4  | -1.418897          | 0.006409      |
| 5  | -0.178219          | 0.007887      |
| 6  | 27.849716          | 0.579175      |
7) Data for 0.75 Equivalent PPh₃⁻:

![Stacked Spectra of Inversion Recovery Data at 0.75 Equivalents PPh₃⁻](image)

**Figure S-VIII.7.** Stacked Spectra of Inversion Recovery Data at 0.75 Equivalents PPh₃⁻.

**Table S-VIII.7.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|------|------|
| 0.001 | -0.3683     | 0.5956      | -0.3578 | 0.6042 | -0.0104 | -0.0086 |
| 0.002 | -0.3437     | 0.5727      | -0.3556 | 0.5802 | 0.0119  | -0.0075 |
| 0.003 | -0.3205     | 0.5510      | -0.3120 | 0.5572 | -0.0084 | -0.0063 |
| 0.004 | -0.2985     | 0.5305      | -0.2845 | 0.5375 | -0.0140 | -0.0071 |
| 0.005 | -0.2777     | 0.5111      | -0.2641 | 0.5161 | -0.0136 | -0.0049 |
| 0.007 | -0.2393     | 0.4755      | -0.2254 | 0.4830 | -0.0138 | -0.0075 |
| 0.010 | -0.1889     | 0.4291      | -0.1877 | 0.4348 | -0.0013 | -0.0057 |
| 0.015 | -0.1213     | 0.3674      | -0.1076 | 0.3772 | -0.0137 | -0.0098 |
| 0.020 | -0.0696     | 0.3211      | -0.0731 | 0.3227 | 0.0034  | -0.0016 |
| 0.025 | -0.0300     | 0.2864      | -0.0341 | 0.2872 | 0.0041  | -0.0007 |
| 0.030 | 0.0004      | 0.2606      | -0.0065 | 0.2563 | 0.0069  | 0.0043  |
| 0.040 | 0.0420      | 0.2274      | 0.0356  | 0.2218 | 0.0064  | 0.0055  |
| 0.050 | 0.0674      | 0.2098      | 0.0619  | 0.2074 | 0.0054  | 0.0024  |
| 0.060 | 0.0834      | 0.2012      | 0.0820  | 0.1960 | 0.0014  | 0.0052  |
| 0.070 | 0.0940      | 0.1977      | 0.0929  | 0.1901 | 0.0012  | 0.0076  |
| 0.080 | 0.1015      | 0.1972      | 0.0989  | 0.1886 | 0.0026  | 0.0086  |
| 0.090 | 0.1073      | 0.1984      | 0.1064  | 0.1930 | 0.0009  | 0.0054  |
| 0.100 | 0.1120      | 0.2005      | 0.1096  | 0.1908 | 0.0024  | 0.0098  |
| d2 (s) | Integration |
|-------|-------------|
| 0.150 | 0.1301      |
| 0.200 | 0.1462      |
| 0.250 | 0.1618      |
| 0.300 | 0.1770      |
| 0.350 | 0.1918      |
| 0.400 | 0.2063      |
| 0.500 | 0.2340      |
| 0.600 | 0.2604      |
| 0.700 | 0.2854      |
| 0.800 | 0.3091      |
| 0.900 | 0.3316      |
| 1.000 | 0.3530      |
| 1.500 | 0.4448      |
| 2.000 | 0.5156      |
| 2.500 | 0.5702      |
| 3.000 | 0.6123      |
| 5.000 | 0.7041      |
| 6.000 | 0.7244      |
| 8.000 | 0.7437      |
| 10.000| 0.7505      |
| 15.000| 0.7540      |
| 20.000| 0.7543      |

**Chart S–VIII.7. Plot of Inversion Recovery Data at 0.75 Equivalent of PPh₃**

Inversion Recovery Data for 0.75 Equiv. PPh₃

![Inversion Recovery Data](chart.png)
CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 = 0.8000 No. 1 = 0.2300

M(inf)’s
No. 2 = 0.9000 No. 3 = 1.0000

M(0) - M(inf)’s
No. 4 = -1.4000 No. 5 = 0.5000

and M(0)’s for reference
No. 4 = -0.5000 No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 1.559682 +/- 0.184452
# 1 = -0.482491 +/- 0.173228
# 2 = 0.754272 +/- 0.002681
# 3 = 0.863477 +/- 0.002847
# 4 = -1.148466 +/- 0.003911
# 5 = -0.243599 +/- 0.004746
# 6 = 27.458241 +/- 0.479607

8) Data for 1 Equivalent PPh₃⁻:

![Figure S–VIII.8. Stacked Spectra of Inversion Recovery Data at 1.0 Equivalent PPh₃⁻.](image)

Table S–VIII.8. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.4305     | 0.7275      | -0.4404| 0.7240 | 0.0099| 0.0035|
| 0.002  | -0.4026     | 0.7017      | -0.4282| 0.7024 | 0.0256| -0.0007|
| 0.003  | -0.3762     | 0.6773      | -0.3675| 0.6782 | -0.0087| -0.0008|
| 0.004  | -0.3511     | 0.6542      | -0.3386| 0.6557 | -0.0125| -0.0015|
| 0.005  | -0.3273     | 0.6323      | -0.3107| 0.6362 | -0.0165| -0.0039|
|       |       |       |       |       |       |       |       |
|-------|-------|-------|-------|-------|-------|-------|-------|
| 0.007 | -0.2832 | 0.5920 | -0.2784 | 0.5920 | -0.0048 | 0.0000 |
| 0.010 | -0.2251 | 0.5390 | -0.2135 | 0.5427 | -0.0116 | -0.0037 |
| 0.015 | -0.1463 | 0.4680 | -0.1316 | 0.4803 | -0.0147 | -0.0124 |
| 0.020 | -0.0855 | 0.4139 | -0.0887 | 0.4159 | 0.0032  | -0.0020 |
| 0.025 | -0.0384 | 0.3730 | -0.0382 | 0.3769 | -0.0002 | -0.0039 |
| 0.030 | -0.0018 | 0.3421 | -0.0102 | 0.3414 | 0.0084  | 0.0006  |
| 0.040 | 0.0489  | 0.3015 | 0.0470  | 0.3044 | 0.0019  | -0.0030 |
| 0.050 | 0.0803  | 0.2792 | 0.0798  | 0.2825 | 0.0005  | -0.0033 |
| 0.060 | 0.1003  | 0.2678 | 0.1015  | 0.2683 | -0.0012 | -0.0005 |
| 0.070 | 0.1136  | 0.2627 | 0.1108  | 0.2633 | 0.0028  | -0.0007 |
| 0.080 | 0.1230  | 0.2613 | 0.1211  | 0.2582 | 0.0019  | 0.0030  |
| 0.090 | 0.1300  | 0.2621 | 0.1322  | 0.2623 | -0.0022 | -0.0003 |
| 0.100 | 0.1357  | 0.2641 | 0.1376  | 0.2650 | -0.0019 | -0.0008 |
| 0.150 | 0.1565  | 0.2809 | 0.1526  | 0.2715 | 0.0039  | 0.0094  |
| 0.200 | 0.1745  | 0.2995 | 0.1678  | 0.2925 | 0.0067  | 0.0070  |
| 0.250 | 0.1919  | 0.3177 | 0.1940  | 0.3155 | -0.0022 | 0.0022  |
| 0.300 | 0.2087  | 0.3354 | 0.2012  | 0.3225 | 0.0076  | 0.0129  |
| 0.350 | 0.2252  | 0.3527 | 0.2217  | 0.3478 | 0.0035  | 0.0049  |
| 0.400 | 0.2412  | 0.3696 | 0.2446  | 0.3770 | -0.0034 | -0.0074 |
| 0.500 | 0.2720  | 0.4020 | 0.2733  | 0.3966 | -0.0013 | 0.0053  |
| 0.600 | 0.3013  | 0.4327 | 0.3014  | 0.4296 | -0.0001 | 0.0031  |
| 0.700 | 0.3290  | 0.4618 | 0.3268  | 0.4559 | 0.0022  | 0.0060  |
| 0.800 | 0.3554  | 0.4895 | 0.3573  | 0.4864 | -0.0019 | 0.0031  |
| 0.900 | 0.3803  | 0.5158 | 0.3786  | 0.5103 | 0.0017  | 0.0055  |
| 1.000 | 0.4040  | 0.5407 | 0.4048  | 0.5433 | -0.0008 | -0.0027 |
| 1.500 | 0.5055  | 0.6473 | 0.5059  | 0.6477 | -0.0004 | -0.0004 |
| 2.000 | 0.5836  | 0.7294 | 0.5860  | 0.7266 | -0.0024 | 0.0027  |
| 2.500 | 0.6436  | 0.7924 | 0.6460  | 0.7980 | -0.0024 | -0.0056 |
| 3.000 | 0.6897  | 0.8409 | 0.6913  | 0.8437 | -0.0016 | -0.0028 |
| 5.000 | 0.7895  | 0.9458 | 0.7961  | 0.9528 | -0.0066 | -0.0069 |
| 6.000 | 0.8115  | 0.9689 | 0.8184  | 0.9753 | -0.0070 | -0.0064 |
| 8.000 | 0.8321  | 0.9906 | 0.8354  | 0.9965 | -0.0033 | -0.0059 |
| 10.000| 0.8393  | 0.9982 | 0.8349  | 0.9991 | 0.0045  | -0.0009 |
| 15.000| 0.8429  | 1.0019 | 0.8322  | 1.0003 | 0.0107  | 0.0016  |
| 20.000| 0.8432  | 1.0022 | 0.8305  | 0.9968 | 0.0127  | 0.0054  |
**Chart S–VIII.8. Plot of Inversion Recovery Data at 1.0 Equivalent of PPh₃₂**

![Plot of Inversion Recovery Data for 1.0 Equiv. PPh₃](image)

**CIFIT Guesses and Fit Parameters**

**Initial values of parameters:**
- 1/T1's
  - No. 0 = 0.8000
  - No. 1 = 0.2300
- M(inf)'s
  - No. 2 = 0.9000
  - No. 3 = 1.0000
- M(0)–M(inf)'s
  - No. 4 = -1.4000
  - No. 5 = 0.5000
  - For reference
- M(0)'s
  - No. 4 = -0.5000
  - No. 5 = 1.5000
- Rates
  - No. 6 = 20.0000

**Final Values of Fitted Parameters and Uncertainties:**
- # 0 = 1.865732 +/− 0.154685
- # 1 = -0.749558 +/− 0.141844
- # 2 = 0.843196 +/− 0.002595
- # 3 = 1.002243 +/− 0.002775
- # 4 = -1.303100 +/− 0.003779
- # 5 = -0.247599 +/− 0.004607
- # 6 = 26.266672 +/− 0.379183
9) **Data for 1.25 Equivalents PPh$_3$:**

![Figure S-VIII.9](image)

**Figure S-VIII.9.** Stacked Spectra of Inversion Recovery Data at 1.25 Equivalents PPh$_3$.

**Table S-VIII.9.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.4649     | 0.7619      | -0.4557| 0.7689 | -0.0092| -0.0070|
| 0.002  | -0.4356     | 0.7346      | -0.4528| 0.7394 | 0.0172 | -0.0049|
| 0.003  | -0.4079     | 0.7087      | -0.3961| 0.7146 | -0.0117| -0.0059|
| 0.004  | -0.3816     | 0.6843      | -0.3720| 0.6883 | -0.0097| -0.0040|
| 0.005  | -0.3568     | 0.6611      | -0.3446| 0.6699 | -0.0122| -0.0087|
| 0.007  | -0.3109     | 0.6186      | -0.2952| 0.6255 | -0.0157| -0.0069|
| 0.010  | -0.2507     | 0.5630      | -0.2427| 0.5663 | -0.0080| -0.0033|
| 0.015  | -0.1697     | 0.4889      | -0.1675| 0.4948 | -0.0022| -0.0059|
| 0.020  | -0.1078     | 0.4331      | -0.1078| 0.4358 | 0.0001 | -0.0027|
| 0.025  | -0.0603     | 0.3910      | -0.0646| 0.3923 | 0.0043 | -0.0012|
| 0.030  | -0.0238     | 0.3596      | -0.0212| 0.3665 | -0.0025| -0.0070|
| 0.040  | 0.0262      | 0.3186      | 0.0213 | 0.3169 | 0.0049 | 0.0017 |
| 0.050  | 0.0565      | 0.2965      | 0.0508 | 0.2891 | 0.0057 | 0.0074 |
| 0.060  | 0.0755      | 0.2852      | 0.0681 | 0.2773 | 0.0074 | 0.0080 |
| 0.070  | 0.0878      | 0.2802      | 0.0868 | 0.2730 | 0.0010 | 0.0072 |
| 0.080  | 0.0964      | 0.2788      | 0.0953 | 0.2683 | 0.0011 | 0.0105 |
| 0.090  | 0.1028      | 0.2795      | 0.1050 | 0.2743 | -0.0022| 0.0051 |
| 0.100  | 0.1079      | 0.2814      | 0.1045 | 0.2722 | 0.0033 | 0.0092 |
| 0.150  | 0.1266      | 0.2965      | 0.1265 | 0.2869 | 0.0001 | 0.0097 |
| T (ppm) | Inversion Recovery Data at 1.25 Equivalent of PPh₃ |
|---------|--------------------------------------------------|
| 0.200   | 0.1429 0.3133 0.1434 0.3061 -0.0005 0.0072       |
| 0.250   | 0.1587 0.3297 0.1628 0.3266 -0.0041 0.0030       |
| 0.300   | 0.1740 0.3457 0.1707 0.3359 0.0033 0.0098        |
| 0.350   | 0.1889 0.3612 0.1865 0.3511 0.0024 0.0101        |
| 0.400   | 0.2035 0.3764 0.2053 0.3715 -0.0018 0.0049       |
| 0.500   | 0.2314 0.4056 0.2326 0.4054 -0.0011 0.0002       |
| 0.600   | 0.2580 0.4333 0.2546 0.4241 0.0034 0.0091        |
| 0.700   | 0.2832 0.4596 0.2851 0.4611 -0.0019 -0.0015      |
| 0.800   | 0.3071 0.4845 0.3084 0.4861 -0.0013 -0.0016      |
| 0.900   | 0.3298 0.5082 0.3300 0.5097 -0.0002 -0.0015      |
| 1.000   | 0.3513 0.5306 0.3498 0.5293 0.0015 0.0014        |
| 1.500   | 0.4435 0.6269 0.4404 0.6256 0.0031 0.0013        |
| 2.000   | 0.5145 0.7010 0.5160 0.7056 -0.0015 -0.0046      |
| 2.500   | 0.5692 0.7580 0.5723 0.7657 -0.0031 -0.0077      |
| 3.000   | 0.6112 0.8019 0.6133 0.8094 -0.0021 -0.0075      |
| 5.000   | 0.7024 0.8970 0.7026 0.9030 -0.0001 -0.0059      |
| 6.000   | 0.7225 0.9180 0.7176 0.9237 0.0050 -0.0057      |
| 8.000   | 0.7415 0.9378 0.7369 0.9414 0.0046 -0.0035      |
| 10.000  | 0.7482 0.9448 0.7419 0.9508 0.0063 -0.0061      |
| 15.000  | 0.7515 0.9482 0.7453 0.9507 0.0062 -0.0025      |
| 20.000  | 0.7517 0.9485 0.7414 0.9487 0.0103 -0.0002      |

**Chart S-VIII.9.** Plot of Inversion Recovery Data at 1.25 Equivalent of PPh₃.
CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 = 0.8000 No. 1 = 0.2300

M(inf)'s
No. 2 = 0.9000 No. 3 = 1.0000

M(0) - M(inf)'s
No. 4 = -1.4000 No. 5 = 0.5000
and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000
Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 1.703998 +/- 0.163166
# 1 = -0.607661 +/- 0.152018
# 2 = 0.751755 +/- 0.002477
# 3 = 0.948506 +/- 0.002652
# 4 = -1.247507 +/- 0.003641
# 5 = -0.157704 +/- 0.004488
# 6 = 27.172459 +/- 0.368455

10) Data for 1.5 Equivalents PPh₃₂:

Figure S-VIII.10. Stacked Spectra of Inversion Recovery Data at 1.5 Equivalents PPh₃₂.

Table S-VIII.10. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.53490    | 0.73780     | -0.53450 | 0.73350 | -0.00040 | 0.00430 |
| 0.002  | -0.50380    | 0.70780     | -0.53510 | 0.70510 | 0.03120 | 0.00270 |
| 0.003  | -0.47440    | 0.67940     | -0.48520 | 0.67410 | 0.01080 | 0.00530 |
| 0.004  | -0.44630    | 0.65240     | -0.44760 | 0.64440 | 0.00130 | 0.00800 |
| 0.005  | -0.41970    | 0.62690     | -0.42150 | 0.62070 | 0.00180 | 0.00610 |
|     |      |      |      |      |      |      |      |      |      |      |      |      |      |
|-----|------|------|------|------|------|------|------|------|------|------|------|------|------|
| 0.007 | -0.37050 | 0.57960 | -0.35900 | 0.57170 | -0.01150 | 0.00790 |
| 0.010 | -0.30540 | 0.51740 | -0.29670 | 0.51220 | -0.00870 | 0.00530 |
| 0.015 | -0.21680 | 0.43350 | -0.21990 | 0.43040 | 0.00300 | 0.00320 |
| 0.020 | -0.14820 | 0.36930 | -0.15100 | 0.37120 | 0.00280 | -0.00190 |
| 0.025 | -0.09480 | 0.32020 | -0.08920 | 0.32820 | -0.00560 | -0.00790 |
| 0.030 | -0.05330 | 0.28290 | -0.05180 | 0.28500 | -0.00140 | -0.00210 |
| 0.040 | 0.00460 | 0.23300 | 0.00080 | 0.23740 | 0.00380 | -0.00440 |
| 0.050 | 0.04050 | 0.20490 | 0.03940 | 0.20810 | 0.00110 | -0.00320 |
| 0.060 | 0.06330 | 0.18970 | 0.05870 | 0.19300 | 0.00460 | -0.00320 |
| 0.070 | 0.07830 | 0.18220 | 0.07520 | 0.18550 | 0.00310 | -0.00330 |
| 0.080 | 0.08870 | 0.17920 | 0.09160 | 0.18880 | -0.00280 | -0.00960 |
| 0.090 | 0.09640 | 0.17900 | 0.09770 | 0.18490 | -0.00140 | -0.00590 |
| 0.100 | 0.10240 | 0.18030 | 0.10520 | 0.18490 | -0.00290 | -0.00470 |
| 0.150 | 0.12340 | 0.19510 | 0.12620 | 0.20090 | -0.00280 | -0.00590 |
| 0.200 | 0.14100 | 0.21230 | 0.14590 | 0.22110 | -0.00490 | -0.00880 |
| 0.250 | 0.15790 | 0.22940 | 0.15620 | 0.23240 | 0.00170 | -0.00300 |
| 0.300 | 0.17430 | 0.24600 | 0.17730 | 0.25200 | -0.00290 | -0.00610 |
| 0.350 | 0.19030 | 0.26210 | 0.19130 | 0.26370 | -0.00090 | -0.00160 |
| 0.400 | 0.20590 | 0.27790 | 0.20530 | 0.28070 | 0.00070 | -0.00280 |
| 0.500 | 0.23590 | 0.30810 | 0.23820 | 0.31060 | -0.00230 | -0.00250 |
| 0.600 | 0.26420 | 0.33680 | 0.26210 | 0.33880 | 0.00210 | -0.00210 |
| 0.700 | 0.29110 | 0.36390 | 0.29080 | 0.36750 | 0.00040 | -0.00360 |
| 0.800 | 0.31660 | 0.38970 | 0.31560 | 0.38690 | 0.00100 | 0.00270 |
| 0.900 | 0.34080 | 0.41400 | 0.33850 | 0.41220 | 0.00220 | 0.00180 |
| 1.000 | 0.36370 | 0.43720 | 0.36170 | 0.43420 | 0.00190 | 0.00290 |
| 1.500 | 0.46140 | 0.53590 | 0.46100 | 0.53370 | 0.00040 | 0.00210 |
| 2.000 | 0.53610 | 0.61140 | 0.53180 | 0.60750 | 0.00430 | 0.00380 |
| 2.500 | 0.59330 | 0.66910 | 0.59410 | 0.66840 | -0.00080 | 0.00070 |
| 3.000 | 0.63700 | 0.71320 | 0.63530 | 0.70920 | 0.00170 | 0.00400 |
| 5.000 | 0.73050 | 0.80770 | 0.73650 | 0.80540 | -0.00600 | 0.00230 |
| 6.000 | 0.75070 | 0.82800 | 0.75990 | 0.82580 | -0.00920 | 0.00220 |
| 8.000 | 0.76940 | 0.84690 | 0.77580 | 0.84770 | -0.00640 | -0.00080 |
| 10.000 | 0.77580 | 0.85340 | 0.78370 | 0.85230 | -0.00790 | 0.00110 |
| 15.000 | 0.77890 | 0.85650 | 0.78050 | 0.85050 | -0.00160 | 0.00600 |
| 20.000 | 0.77910 | 0.85670 | 0.77850 | 0.84610 | 0.00060 | 0.01070 |
**Chart S-VIII.10. Plot of Inversion Recovery Data at 1.5 Equivalent of PPh₃.**

**CIFIT Guesses and Fit Parameters**

**Initial values of parameters:**

1/T₁'s  
No. 0 = 0.8000  No. 1 = 0.2300  
M(∞)'s  
No. 2 = 0.9000  No. 3 = 1.0000  
M(0)–M(∞)'s  
No. 4 = -1.4000  No. 5 = 0.5000  
and M(0)'s for reference  
No. 4 = -0.5000  No. 5 = 1.5000  
Rates  
No. 6 = 20.0000

**Final Values of Fitted Parameters and Uncertainties:**

| #  | Value          | Uncertainty |  Value          | Uncertainty |
|----|----------------|-------------|----------------|-------------|
| 0  | 0.792707       | +/- 0.135379| 0.135379       |             |
| 1  | 0.282554       | +/- 0.135140| 0.135140       |             |
| 2  | 0.779104       | +/- 0.002309| 0.002309       |             |
| 3  | 0.856755       | +/- 0.002450| 0.002450       |             |
| 4  | -1.346653      | +/- 0.003349| 0.003349       |             |
| 5  | -0.087352      | +/- 0.004194| 0.004194       |             |
| 6  | 25.774528      | +/- 0.285602| 0.285602       |             |
Figure S–VIII.11. Stacked Spectra of Inversion Recovery Data at 2.0 Equivalents PPh₃₂.

Table S–VIII.11. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.6112     | 0.8798      | -0.6257 | 0.8737 | 0.0146 | 0.0061 |
| 0.002  | -0.5772     | 0.8481      | -0.6200 | 0.8352 | 0.0429 | 0.0129 |
| 0.003  | -0.5448     | 0.8180      | -0.5503 | 0.8114 | 0.0055 | 0.0065 |
| 0.004  | -0.5141     | 0.7893      | -0.5146 | 0.7830 | 0.0005 | 0.0063 |
| 0.005  | -0.4848     | 0.7622      | -0.4725 | 0.7539 | -0.0123 | 0.0082 |
| 0.007  | -0.4305     | 0.7118      | -0.4166 | 0.7102 | -0.0139 | 0.0016 |
| 0.010  | -0.3585     | 0.6455      | -0.3443 | 0.6434 | -0.0142 | 0.0021 |
| 0.015  | -0.2599     | 0.5554      | -0.2638 | 0.5530 | 0.0039 | 0.0024 |
| 0.020  | -0.1829     | 0.4860      | -0.1804 | 0.4895 | -0.0025 | -0.0036 |
| 0.025  | -0.1226     | 0.4326      | -0.1242 | 0.4365 | 0.0016 | -0.0039 |
| 0.030  | -0.0753     | 0.3917      | -0.0737 | 0.3948 | -0.0016 | -0.0031 |
| 0.040  | -0.0086     | 0.3367      | -0.0164 | 0.3389 | 0.0078 | -0.0022 |
| 0.050  | 0.0333      | 0.3054      | 0.0313  | 0.3112 | 0.0020 | -0.0058 |
| 0.060  | 0.0603      | 0.2883      | 0.0570  | 0.2921 | 0.0033 | -0.0038 |
| 0.070  | 0.0783      | 0.2799      | 0.0773  | 0.2861 | 0.0010 | -0.0062 |
| 0.080  | 0.0909      | 0.2766      | 0.0874  | 0.2822 | 0.0036 | -0.0057 |
| 0.090  | 0.1002      | 0.2764      | 0.0984  | 0.2874 | 0.0019 | -0.0111 |
| 0.100  | 0.1075      | 0.2780      | 0.1053  | 0.2804 | 0.0023 | -0.0024 |
| d2 (s) | Integration |
|-------|-------------|
| 5     | 0.150       |
| 10    | 0.200       |
| 15    | 0.250       |
| 20    | 0.300       |
| 25    | 0.350       |

**Chart S-VIII.11. Plot of Inversion Recovery Data at 2.0 Equivalent of PPh$_3$**

Inversion Recovery Data for 2.0 Equiv. PPh$_3$
CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 = 0.8000 No. 1 = 0.2300

M(inf)'s
No. 2 = 0.9000 No. 3 = 1.0000

M(0) - M(inf)'s
No. 4 = -1.4000 No. 5 = 0.5000

and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 1.639768 +/− 0.159698
# 1 = -0.395038 +/− 0.150298
# 2 = 0.830426 +/− 0.002911
# 3 = 1.021867 +/− 0.003118
# 4 = -1.477308 +/− 0.004337
# 5 = -0.108696 +/− 0.005390
# 6 = 24.991213 +/− 0.330442

12) Data for 5.0 Equivalents PPh₃:

Figure S–VIII.12. Stacked Spectra of Inversion Recovery Data at 5.0 Equivalents PPh₃.

Table S–VIII.12. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.6112     | 0.8798      | -0.6257| 0.8737 | 0.0146| 0.0061|
| 0.002  | -0.5772     | 0.8481      | -0.6200| 0.8352 | 0.0429| 0.0129|
| 0.003  | -0.5448     | 0.8180      | -0.5503| 0.8114 | 0.0055| 0.0065|
| 0.004  | -0.5141     | 0.7893      | -0.5146| 0.7830 | 0.0005| 0.0063|
| 0.005  | -0.4848     | 0.7622      | -0.4725| 0.7539 | -0.0123| 0.0082|

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|     |      |      |      |      |      |      |
|----|-----|-----|-----|-----|-----|-----|
| 0.007 | -0.4305 | 0.7118 | -0.4166 | 0.7102 | -0.0139 | 0.0016 |
| 0.010 | -0.3585 | 0.6455 | -0.3443 | 0.6434 | -0.0142 | 0.0021 |
| 0.015 | -0.2599 | 0.5554 | -0.2638 | 0.5530 | 0.0039 | 0.0024 |
| 0.020 | -0.1829 | 0.4860 | -0.1804 | 0.4895 | -0.0025 | -0.0036 |
| 0.025 | -0.1226 | 0.4326 | -0.1242 | 0.4365 | 0.0016 | -0.0039 |
| 0.030 | -0.0753 | 0.3917 | -0.0737 | 0.3948 | -0.0016 | -0.0031 |
| 0.040 | -0.0086 | 0.3367 | -0.0164 | 0.3389 | 0.0078 | -0.0022 |
| 0.050 | 0.0333 | 0.3054 | 0.0313 | 0.3112 | 0.0020 | -0.0058 |
| 0.060 | 0.0603 | 0.2883 | 0.0570 | 0.2921 | 0.0033 | -0.0038 |
| 0.070 | 0.0783 | 0.2799 | 0.0773 | 0.2861 | 0.0010 | -0.0062 |
| 0.080 | 0.0909 | 0.2766 | 0.0874 | 0.2822 | 0.0036 | -0.0057 |
| 0.090 | 0.1002 | 0.2764 | 0.0984 | 0.2874 | 0.0019 | -0.0111 |
| 0.100 | 0.1075 | 0.2780 | 0.1053 | 0.2804 | 0.0023 | -0.0024 |
| 0.150 | 0.1330 | 0.2962 | 0.1328 | 0.2973 | 0.0002 | -0.0011 |
| 0.200 | 0.1540 | 0.3174 | 0.1628 | 0.3281 | -0.0088 | -0.0107 |
| 0.250 | 0.1741 | 0.3382 | 0.1775 | 0.3451 | -0.0034 | -0.0068 |
| 0.300 | 0.1935 | 0.3585 | 0.1928 | 0.3635 | 0.0007 | -0.0050 |
| 0.350 | 0.2124 | 0.3782 | 0.2160 | 0.3879 | -0.0037 | -0.0098 |
| 0.400 | 0.2307 | 0.3972 | 0.2339 | 0.4041 | -0.0032 | -0.0069 |
| 0.500 | 0.2657 | 0.4337 | 0.2668 | 0.4342 | -0.0011 | -0.0005 |
| 0.600 | 0.2987 | 0.4680 | 0.3030 | 0.4737 | -0.0043 | -0.0056 |
| 0.700 | 0.3297 | 0.5004 | 0.3285 | 0.4983 | 0.0012 | 0.0021 |
| 0.800 | 0.3590 | 0.5308 | 0.3577 | 0.5327 | 0.0013 | -0.0019 |
| 0.900 | 0.3865 | 0.5595 | 0.3868 | 0.5607 | -0.0003 | -0.0012 |
| 1.000 | 0.4124 | 0.5865 | 0.4126 | 0.5829 | -0.0002 | 0.0036 |
| 1.500 | 0.5210 | 0.6996 | 0.5171 | 0.6932 | 0.0040 | 0.0065 |
| 2.000 | 0.6014 | 0.7833 | 0.5961 | 0.7733 | 0.0053 | 0.0100 |
| 2.500 | 0.6609 | 0.8453 | 0.6614 | 0.8427 | -0.0005 | 0.0026 |
| 3.000 | 0.7049 | 0.8912 | 0.7017 | 0.8831 | 0.0032 | 0.0081 |
| 5.000 | 0.7928 | 0.9826 | 0.8033 | 0.9826 | -0.0105 | 0.0001 |
| 6.000 | 0.8098 | 1.0004 | 0.8206 | 0.9997 | -0.0108 | 0.0006 |
| 8.000 | 0.8242 | 1.0154 | 0.8297 | 1.0137 | -0.0055 | 0.0017 |
| 10.000 | 0.8286 | 1.0199 | 0.8323 | 1.0179 | -0.0038 | 0.0020 |
| 15.000 | 0.8303 | 1.0218 | 0.8335 | 1.0162 | -0.0031 | 0.0056 |
| 20.000 | 0.8304 | 1.0219 | 0.8333 | 1.0136 | -0.0028 | 0.0082 |
**Chart S-VIII.12. Plot of Inversion Recovery Data at 5.0 Equivalent of PPh$_3$.**

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/T1's
No. 0 = 0.8000 No. 1 = 0.2300
M(inf)'s
No. 2 = 0.9000 No. 3 = 1.0000
M(0)-M(inf)'s
No. 4 = -1.4000 No. 5 = 0.5000
and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000
Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 1.153897 +/− 0.151221
# 1 = 0.051444 +/− 0.147752
# 2 = 1.103346 +/− 0.002131
# 3 = 1.088249 +/− 0.002263
# 4 = -1.167800 +/− 0.003175
# 5 = -0.095688 +/− 0.003948
# 6 = 26.166560 +/− 0.325674
13) Data for 10 Equivalents PPh$_3$:

**Figure S-VIII.13.** Stacked Spectra of Inversion Recovery Data at 10 Equivalents PPh$_3$.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | 0.1751      | 0.9324      | 0.1785 | 0.9367 | -0.0033 | -0.0043 |
| 0.002  | 0.1853      | 0.9225      | 0.1762 | 0.9222 | 0.009    | 0.0004 |
| 0.003  | 0.1949      | 0.9132      | 0.1982 | 0.9132 | -0.0033 | 0      |
| 0.004  | 0.2041      | 0.9043      | 0.208  | 0.906  | -0.0039 | -0.0017 |
| 0.005  | 0.2128      | 0.8959      | 0.2171 | 0.8973 | -0.0042 | -0.0015 |
| 0.007  | 0.229       | 0.8803      | 0.2323 | 0.879  | -0.0033 | 0.0013 |
| 0.01   | 0.2503      | 0.8598      | 0.2528 | 0.8624 | -0.0025 | -0.0026 |
| 0.015  | 0.2794      | 0.8321      | 0.2802 | 0.8318 | -0.0008 | 0.0003 |
| 0.02   | 0.3021      | 0.8108      | 0.3018 | 0.811  | 0.0003  | -0.0002 |
| 0.025  | 0.3197      | 0.7946      | 0.3176 | 0.7944 | 0.0022  | 0.0002 |
| 0.03   | 0.3335      | 0.7822      | 0.3306 | 0.7843 | 0.0029  | -0.0021 |
| 0.04   | 0.3528      | 0.7657      | 0.3506 | 0.7627 | 0.0022  | 0.003  |
| 0.05   | 0.3648      | 0.7564      | 0.3634 | 0.7539 | 0.0015  | 0.0025 |
| 0.06   | 0.3726      | 0.7514      | 0.3704 | 0.7496 | 0.0022  | 0.0018 |
| 0.07   | 0.3777      | 0.749       | 0.3771 | 0.7481 | 0.0006  | 0.0008 |
| 0.08   | 0.3813      | 0.748       | 0.3807 | 0.7467 | 0.0007  | 0.0013 |
| 0.09   | 0.384       | 0.748       | 0.3836 | 0.7468 | 0.0004  | 0.0013 |
| 0.1    | 0.3862      | 0.7486      | 0.3857 | 0.7471 | 0.0004  | 0.0014 |
| 0.15   | 0.3938      | 0.754       | 0.3935 | 0.7536 | 0.0003  | 0.0004 |
| d2 (s) | Integration | Inversion Recovery Data for 10 Equiv. PPh₃ |
|-------|-------------|------------------------------------------|
| 0.2   | 0.4003      | 0.7603 0.4018 0.7598 -0.0016 0.0005      |
| 0.25  | 0.4064      | 0.7665 0.4071 0.7659 -0.0007 0.0006      |
| 0.3   | 0.4125      | 0.7725 0.413 0.7706 -0.0006 0.0019      |
| 0.35  | 0.4183      | 0.7784 0.4204 0.7791 -0.002 -0.0008     |
| 0.4   | 0.424       | 0.7841 0.4259 0.783 -0.0019 0.001       |
| 0.5   | 0.4349      | 0.795 0.4359 0.7937 -0.001 0.0013      |
| 0.6   | 0.4452      | 0.8053 0.4465 0.8053 -0.0013 -0.0001   |
| 0.7   | 0.4549      | 0.815 0.4556 0.8157 -0.0007 -0.0007   |
| 0.8   | 0.4641      | 0.8242 0.4628 0.8239 0.0013 0.0004     |
| 0.9   | 0.4728      | 0.8329 0.4725 0.8349 0.0003 -0.002     |
| 1     | 0.481       | 0.8411 0.4805 0.8395 0.0006 0.0016     |
| 1.5   | 0.5158      | 0.8759 0.5158 0.8749 0 0.001       |
| 2     | 0.542       | 0.9021 0.5435 0.9007 -0.0015 0.0014   |
| 2.5   | 0.5618      | 0.9219 0.5597 0.9212 0.002 0.0007  |
| 3     | 0.5766      | 0.9368 0.5763 0.9386 0.0003 -0.0019   |
| 5     | 0.6074      | 0.9676 0.6099 0.9719 -0.0025 -0.0043 |
| 6     | 0.6138      | 0.9739 0.6134 0.9739 0.0003 0       |
| 8     | 0.6194      | 0.9796 0.6179 0.9802 0.0015 -0.0007 |
| 10    | 0.6212      | 0.9814 0.6199 0.9826 0.0014 -0.0012 |
| 15    | 0.6222      | 0.9822 0.6202 0.9839 0.0019 -0.0017 |
| 20    | 0.6221      | 0.9822 0.6193 0.9818 0.0028 0.0005   |

**Chart S-VIII.13.** Plot of Inversion Recovery Data at 10 Equivalents PPh₃.
CIFIT Guesses and Fit Parameters

Initial values of parameters:

$1/T_1$'s
No. 0 = 0.8000  No. 1 = 0.2300

$M(\infty)$'s
No. 2 = 0.9000  No. 3 = 1.0000

$M(0)$-$M(\infty)$'s
No. 4 = -1.4000  No. 5 = 0.5000

and $M(0)$'s for reference
No. 4 = -0.5000  No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values and Uncertainties:

# 0 = 0.578493 +/- 0.138359
# 1 = 0.553659 +/- 0.140746
# 2 = 0.622091 +/- 0.000805
# 3 = 0.982237 +/- 0.000850
# 4 = -0.457643 +/- 0.001171
# 5 = -0.039427 +/- 0.001464
# 6 = 25.571599 +/- 0.302792
IX. Inversion Recovery Data for Section V (\((n-\text{Bu})_4\text{NCl}\) and LiCl Addition)

1) Data for 0.4 Equivalent \((n-\text{Bu})_4\text{NCl}\):

![Stacked Spectra of Inversion Recovery Data at 0.4 Equivalents \((n-\text{Bu})_4\text{NCl}\).]

*Figure S-IX.1. Stacked Spectra of Inversion Recovery Data at 0.4 Equivalents \((n-\text{Bu})_4\text{NCl}\).*

*Table S-IX.1. CIFIT Plot File.*

| \(d_2\) (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | \(\sigma\)M1 | \(\sigma\)M2 |
|--------------|-------------|-------------|--------|--------|-------------|-------------|
| 0.001        | -0.0441     | 0.8503      | -0.0604| 0.8397 | 0.0162      | 0.0106      |
| 0.002        | -0.0247     | 0.8333      | -0.0386| 0.8245 | 0.0139      | 0.0088      |
| 0.003        | -0.0062     | 0.8171      | -0.0167| 0.8124 | 0.0106      | 0.0047      |
| 0.004        | 0.0116      | 0.8017      | 0.0055 | 0.7982 | 0.0061      | 0.0035      |
| 0.005        | 0.0287      | 0.787       | 0.0278 | 0.7783 | 0.0009      | 0.0087      |
| 0.006        | 0.0451      | 0.773       | 0.0396 | 0.7684 | 0.0055      | 0.0045      |
| 0.007        | 0.0608      | 0.7596      | 0.0558 | 0.759  | 0.005       | 0.0006      |
| 0.008        | 0.0758      | 0.7468      | 0.0771 | 0.7406 | -0.0013     | 0.0062      |
| 0.009        | 0.0902      | 0.7347      | 0.0842 | 0.7286 | 0.006       | 0.0061      |
| 0.01         | 0.104       | 0.7231      | 0.1084 | 0.7168 | -0.0043     | 0.0064      |
| 0.015        | 0.1651      | 0.6731      | 0.168  | 0.6675 | -0.0029     | 0.0056      |
| 0.02         | 0.2149      | 0.634       | 0.2211 | 0.6334 | -0.0062     | 0.0006      |
| 0.025        | 0.2556      | 0.6038      | 0.2585 | 0.603  | -0.003      | 0.0008      |
| 0.03         | 0.289       | 0.5805      | 0.2914 | 0.5854 | -0.0024     | -0.0049     |
| 0.035        | 0.3166      | 0.5628      | 0.3217 | 0.5663 | -0.0051     | -0.0035     |
|   | 0.04 | 0.3395 | 0.5496 | 0.341 | 0.5585 | -0.0015 | -0.0089 |
|---|------|--------|--------|-------|--------|---------|---------|
| 0.045 | 0.3587 | 0.54 | 0.3623 | 0.5477 | -0.0036 | -0.0077 |
| 0.05 | 0.3749 | 0.5332 | 0.376 | 0.5408 | -0.0011 | -0.0076 |
| 0.06 | 0.4007 | 0.5258 | 0.4013 | 0.5374 | -0.0007 | 0.0115 |
| 0.07 | 0.4201 | 0.5243 | 0.426 | 0.5387 | -0.0059 | -0.0144 |
| 0.08 | 0.4356 | 0.5264 | 0.4405 | 0.5408 | -0.0049 | -0.0145 |
| 0.09 | 0.4484 | 0.5307 | 0.4517 | 0.5399 | -0.0033 | -0.0092 |
| 0.1 | 0.4594 | 0.5364 | 0.4586 | 0.5458 | 0.0008 | -0.0094 |
| 0.15 | 0.503 | 0.5722 | 0.503 | 0.582 | 0 | -0.0098 |
| 0.2 | 0.5391 | 0.6083 | 0.5378 | 0.6178 | 0.0014 | -0.0095 |
| 0.25 | 0.5717 | 0.6416 | 0.5684 | 0.6494 | 0.0033 | -0.0078 |
| 0.3 | 0.6014 | 0.672 | 0.5958 | 0.6762 | 0.0056 | -0.0043 |
| 0.35 | 0.6285 | 0.6997 | 0.6306 | 0.7036 | -0.0021 | -0.0038 |
| 0.4 | 0.6533 | 0.7251 | 0.6532 | 0.7277 | 0.0001 | -0.0026 |
| 0.45 | 0.6758 | 0.7482 | 0.6727 | 0.7528 | 0.0031 | -0.0046 |
| 0.5 | 0.6964 | 0.7693 | 0.6934 | 0.7699 | 0.003 | -0.0006 |
| 0.6 | 0.7323 | 0.806 | 0.7239 | 0.8062 | 0.0084 | -0.0003 |
| 0.7 | 0.7622 | 0.8367 | 0.7584 | 0.8271 | 0.0038 | 0.0095 |
| 0.8 | 0.7871 | 0.8621 | 0.7854 | 0.8559 | 0.0017 | 0.0063 |
| 0.9 | 0.8078 | 0.8834 | 0.8007 | 0.8762 | 0.0071 | 0.0072 |
| 1 | 0.825 | 0.901 | 0.8197 | 0.8942 | 0.0053 | 0.0068 |
| 1.5 | 0.8765 | 0.9537 | 0.8778 | 0.9443 | -0.0013 | 0.0094 |
| 2 | 0.897 | 0.9747 | 0.9019 | 0.9713 | -0.0049 | 0.0035 |
| 2.5 | 0.9052 | 0.9831 | 0.9123 | 0.9821 | -0.0071 | 0.001 |
| 3 | 0.9085 | 0.9865 | 0.9173 | 0.984 | -0.0088 | 0.0025 |
| 4 | 0.9103 | 0.9884 | 0.9181 | 0.9884 | -0.0078 | -0.0001 |
| 6 | 0.9107 | 0.9887 | 0.919 | 0.9887 | -0.0083 | 0 |
| 8 | 0.9107 | 0.9887 | 0.922 | 0.9866 | -0.0113 | 0.0022 |
| 10 | 0.9107 | 0.9887 | 0.9143 | 0.9863 | -0.0036 | 0.0024 |
| 12 | 0.9107 | 0.9887 | 0.9167 | 0.984 | -0.006 | 0.0047 |
| 16 | 0.9107 | 0.9887 | 0.9131 | 0.9812 | -0.0024 | 0.0075 |
| 20 | 0.9107 | 0.9887 | 0.9086 | 0.984 | 0.002 | 0.0047 |
Chart S-IX.1. Plot of Inversion Recovery Data at 0.4 Equivalents \((n-\text{Bu})_4\text{NCl}\).

CIFIT Guesses and Fit Parameters

Initial values of parameters:

\(1/T1's\):
- No. 0 = 0.8000  
- No. 1 = 0.2300

\(M(\text{inf})'s\):
- No. 2 = 0.9000  
- No. 3 = 1.0000

\(M(0)-M(\text{inf})'s\):
- No. 4 = -1.4000  
- No. 5 = 0.5000

and \(M(0)'s\) for reference:
- No. 4 = -0.5000  
- No. 5 = 1.5000

Rates:
- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

\# 0 = 2.355768 +/− 0.185780  
\# 1 = 1.325387 +/− 0.189420  
\# 2 = 0.910675 +/− 0.001832  
\# 3 = 0.988722 +/− 0.001927  
\# 4 = -0.975053 +/− 0.003073  
\# 5 = -0.120648 +/− 0.003802  
\# 6 = 21.509204 +/− 0.386532
2) **Data for 0.7 Equivalent \((n{-}\text{Bu})_2\text{NCl}\):**

![Stacked Spectra of Inversion Recovery Data at 0.7 Equivalents \((n{-}\text{Bu})_2\text{NCl}\).]

**Figure S-IX.2.** Stacked Spectra of Inversion Recovery Data at 0.7 Equivalents \((n{-}\text{Bu})_2\text{NCl}\).

**Table S-IX.2.** CIFIT Plot File.

| \(d2\) (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | \(\sigma\)M1 | \(\sigma\)M2 |
|-----------|------------|------------|--------|--------|-------------|-------------|
| 0.001     | 0.1638     | 0.7203     | 0.1478 | 0.7072 | 0.016       | 0.0131      |
| 0.002     | 0.1756     | 0.7106     | 0.1611 | 0.6978 | 0.0145      | 0.0128      |
| 0.003     | 0.1869     | 0.7015     | 0.1823 | 0.696  | 0.0046      | 0.0055      |
| 0.004     | 0.1978     | 0.6927     | 0.1918 | 0.6893 | 0.006       | 0.0035      |
| 0.005     | 0.2082     | 0.6844     | 0.2019 | 0.6832 | 0.0064      | 0.0013      |
| 0.006     | 0.2183     | 0.6765     | 0.2162 | 0.6659 | 0.0021      | 0.0107      |
| 0.007     | 0.2279     | 0.669      | 0.2227 | 0.6665 | 0.0052      | 0.0025      |
| 0.008     | 0.2371     | 0.6619     | 0.2397 | 0.6547 | -0.0025     | 0.0072      |
| 0.009     | 0.246      | 0.6551     | 0.2435 | 0.655  | 0.0025      | 0.0001      |
| 0.01      | 0.2546     | 0.6487     | 0.2496 | 0.6476 | 0.005       | 0.0011      |
| 0.015     | 0.2926     | 0.6211     | 0.2913 | 0.6172 | 0.0013      | 0.0039      |
| 0.02      | 0.3239     | 0.6001     | 0.3262 | 0.5989 | -0.0023     | 0.0011      |
| 0.025     | 0.3499     | 0.5842     | 0.3535 | 0.5859 | -0.0036     | -0.0017     |
| 0.03      | 0.3716     | 0.5724     | 0.3778 | 0.574  | -0.0061     | -0.0015     |
| 0.035     | 0.3899     | 0.564      | 0.3909 | 0.5671 | -0.001      | -0.0031     |
| 0.04      | 0.4055     | 0.5582     | 0.415  | 0.5677 | -0.0096     | -0.0095     |
| 0.045     | 0.4188     | 0.5545     | 0.4257 | 0.5626 | -0.0069     | -0.0081     |
| 0.05 | 0.4304 | 0.5524 | 0.4361 | 0.558 | -0.0057 | -0.0056 |
| 0.06 | 0.4495 | 0.552 | 0.4538 | 0.5633 | -0.0043 | -0.0113 |
| 0.07 | 0.4649 | 0.5548 | 0.4722 | 0.5628 | -0.0073 | -0.0079 |
| 0.08 | 0.4778 | 0.5598 | 0.4815 | 0.5773 | -0.0037 | -0.0175 |
| 0.09 | 0.489 | 0.566 | 0.4909 | 0.5802 | -0.0019 | -0.0142 |
| 0.1 | 0.4992 | 0.573 | 0.5004 | 0.5783 | -0.0011 | -0.0053 |
| 0.15 | 0.5419 | 0.6108 | 0.5398 | 0.618 | 0.0021 | -0.0072 |
| 0.2 | 0.5782 | 0.6469 | 0.5822 | 0.6593 | -0.0039 | -0.0124 |
| 0.25 | 0.6108 | 0.6797 | 0.6111 | 0.6856 | -0.0003 | -0.0058 |
| 0.3 | 0.6401 | 0.7093 | 0.6377 | 0.7106 | 0.0024 | -0.0013 |
| 0.35 | 0.6666 | 0.7359 | 0.664 | 0.7296 | 0.0026 | 0.0064 |
| 0.4 | 0.6904 | 0.76 | 0.6853 | 0.7591 | 0.0051 | 0.0008 |
| 0.45 | 0.7119 | 0.7816 | 0.7071 | 0.7811 | 0.0048 | 0.0005 |
| 0.5 | 0.7312 | 0.8011 | 0.7217 | 0.7995 | 0.0095 | 0.0017 |
| 0.6 | 0.7644 | 0.8346 | 0.7647 | 0.8326 | -0.0003 | 0.002 |
| 0.7 | 0.7914 | 0.8618 | 0.7862 | 0.8588 | 0.0052 | 0.0029 |
| 0.8 | 0.8133 | 0.8839 | 0.8139 | 0.8803 | -0.0006 | 0.0036 |
| 0.9 | 0.8311 | 0.9018 | 0.8263 | 0.8972 | 0.0048 | 0.0046 |
| 1 | 0.8455 | 0.9164 | 0.8442 | 0.91 | 0.0013 | 0.0064 |
| 1.5 | 0.886 | 0.9572 | 0.8872 | 0.9513 | -0.0012 | 0.0058 |
| 2 | 0.9003 | 0.9716 | 0.903 | 0.966 | -0.0027 | 0.0055 |
| 2.5 | 0.9054 | 0.9767 | 0.908 | 0.972 | -0.0026 | 0.0047 |
| 3 | 0.9072 | 0.9785 | 0.9154 | 0.9783 | -0.0083 | 0.0003 |
| 4 | 0.908 | 0.9794 | 0.9147 | 0.9762 | -0.0067 | 0.0031 |
| 6 | 0.9081 | 0.9795 | 0.9092 | 0.9747 | -0.0011 | 0.0048 |
| 8 | 0.9081 | 0.9795 | 0.9091 | 0.977 | -0.0009 | 0.0025 |
| 10 | 0.9081 | 0.9795 | 0.9117 | 0.9791 | -0.0036 | 0.0004 |
| 12 | 0.9081 | 0.9795 | 0.9098 | 0.983 | -0.0017 | -0.0035 |
| 16 | 0.9081 | 0.9795 | 0.9106 | 0.9822 | -0.0025 | -0.0027 |
| 20 | 0.9081 | 0.9795 | 0.9169 | 0.9795 | -0.0088 | 0.0 |
**Chart S-IX.2. Plot of Inversion Recovery Data at 0.7 Equivalents (n-Bu)$_4$NCl.**

![Inversion Recovery Data Plot](image)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

| Parameter Type | Initial Value | Uncertainty |
|----------------|---------------|-------------|
| 1/T1's         |               |             |
| No. 0          | 0.8000        |             |
| No. 1          | 0.2300        |             |
| M(inf)'s       |               |             |
| No. 2          | 0.9000        |             |
| No. 3          | 1.0000        |             |
| M(0)-M(inf)'s  |               |             |
| No. 4          | -1.4000       |             |
| No. 5          | 0.5000        |             |
| M(0)'s         |               |             |
| No. 4          | -0.5000       |             |
| No. 5          | 1.5000        |             |
| Rates          |               |             |
| No. 6          | 20.0000       |             |

Final Values of Fitted Parameters and Uncertainties:

| Parameter | Value       | Uncertainty |
|-----------|-------------|-------------|
| # 0       | 2.253219    | 0.219951    |
| # 1       | 1.902524    | 0.227138    |
| # 2       | 0.908146    | 0.001750    |
| # 3       | 0.979502    | 0.001816    |
| # 4       | -0.756649   | 0.002971    |
| # 5       | -0.249115   | 0.003533    |
| # 6       | 21.374766   | 0.656295    |
3) **Data for 1.1 Equivalents \((n\text{-Bu})_2\text{NCl}\):**

![Figure S-IX.3. Stacked Spectra of Inversion Recovery Data at 1.1 Equivalents \((n\text{-Bu})_2\text{NCl}\).](image)

**Table S-IX.3. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | 0.1353      | 0.7829      | 0.1064 | 0.7566 | 0.0289 | 0.0263 |
| 0.002  | 0.151       | 0.7711      | 0.1524 | 0.7745 | -0.0013 | -0.0034 |
| 0.003  | 0.1661      | 0.7599      | 0.1645 | 0.76   | 0.0017 | -0.0001 |
| 0.004  | 0.1806      | 0.7494      | 0.1852 | 0.747  | -0.0046 | 0.0024 |
| 0.005  | 0.1945      | 0.7394      | 0.191  | 0.7363 | 0.0035 | 0.003  |
| 0.006  | 0.2078      | 0.7299      | 0.2101 | 0.733  | -0.0023 | -0.0031 |
| 0.007  | 0.2206      | 0.721       | 0.2287 | 0.7257 | -0.0081 | -0.0047 |
| 0.008  | 0.2328      | 0.7126      | 0.236  | 0.7184 | -0.0032 | -0.0058 |
| 0.009  | 0.2446      | 0.7047      | 0.2502 | 0.711  | -0.0056 | -0.0063 |
| 0.01   | 0.2559      | 0.6972      | 0.2599 | 0.7029 | -0.004  | -0.0057 |
| 0.015  | 0.306       | 0.666       | 0.3028 | 0.6578 | 0.0032 | 0.0081 |
| 0.02   | 0.3472      | 0.6433      | 0.3062 | 0.6048 | 0.041  | 0.0385 |
| 0.025  | 0.3814      | 0.6272      | 0.3844 | 0.6292 | -0.003 | -0.002 |
| 0.03   | 0.4101      | 0.6164      | 0.4209 | 0.6216 | -0.0108 | -0.0052 |
| 0.035  | 0.4344      | 0.6096      | 0.4309 | 0.6082 | 0.0035 | 0.0014 |
| 0.04   | 0.4552      | 0.606       | 0.46   | 0.6148 | -0.0048 | -0.0087 |
| 0.045  | 0.4732      | 0.6049      | 0.4771 | 0.6097 | -0.0038 | -0.0048 |
| 0.05 | 0.4891 | 0.6056 | 0.4985 | 0.6159 | -0.0095 | -0.0103 |
| 0.06 | 0.5158 | 0.6112 | 0.5292 | 0.6205 | -0.0134 | -0.0094 |
| 0.07 | 0.5379 | 0.6202 | 0.546  | 0.6252 | -0.0081 | -0.005  |
| 0.08 | 0.5569 | 0.6311 | 0.5632 | 0.6467 | -0.0063 | -0.0156 |
| 0.09 | 0.5738 | 0.6431 | 0.5695 | 0.6467 | 0.0043  | -0.0036 |
| 0.1  | 0.5893 | 0.6555 | 0.5851 | 0.6625 | 0.0041  | -0.007  |
| 0.15 | 0.6539 | 0.7156 | 0.6564 | 0.727  | -0.0025 | -0.0114 |
| 0.2  | 0.7061 | 0.7673 | 0.7074 | 0.7704 | -0.0013 | -0.0031 |
| 0.25 | 0.7493 | 0.8105 | 0.7508 | 0.8165 | -0.0015 | -0.0059 |
| 0.3  | 0.7852 | 0.8465 | 0.7722 | 0.8373 | 0.013   | 0.0092  |
| 0.35 | 0.8151 | 0.8764 | 0.8149 | 0.8763 | 0.0002  | 0.0001  |
| 0.4  | 0.84   | 0.9013 | 0.8293 | 0.8924 | 0.0107  | 0.0089  |
| 0.45 | 0.8607 | 0.922  | 0.8535 | 0.9213 | 0.0073  | 0.0008  |
| 0.5  | 0.878  | 0.9392 | 0.8682 | 0.9327 | 0.0097  | 0.0066  |
| 0.6  | 0.9042 | 0.9655 | 0.9154 | 0.9675 | -0.0112 | -0.002  |
| 0.7  | 0.9224 | 0.9837 | 0.9215 | 0.9843 | 0.0009  | -0.0006 |
| 0.8  | 0.935  | 0.9963 | 0.9282 | 0.9914 | 0.0068  | 0.0049  |
| 0.9  | 0.9437 | 1.005  | 0.9485 | 1.0069 | -0.0048 | -0.0019 |
| 1    | 0.9497 | 1.011  | 0.9283 | 0.9932 | 0.0214  | 0.0179  |
| 1.5  | 0.9611 | 1.0225 | 0.9733 | 1.0286 | -0.0122 | -0.0061 |
| 2    | 0.9629 | 1.0243 | 0.9697 | 1.027  | -0.0068 | -0.0027 |
| 2.5  | 0.9632 | 1.0246 | 0.9749 | 1.0363 | -0.0116 | -0.0117 |
| 3    | 0.9633 | 1.0246 | 0.9704 | 1.0264 | -0.0072 | -0.0018 |
| 4    | 0.9633 | 1.0246 | 0.9689 | 1.0291 | -0.0056 | -0.0045 |
| 6    | 0.9633 | 1.0246 | 0.9663 | 1.0146 | -0.003  | 0.01    |
| 8    | 0.9633 | 1.0246 | 0.9731 | 1.0295 | -0.0098 | -0.0049 |
| 10   | 0.9633 | 1.0246 | 0.9444 | 1.0012 | 0.0189  | 0.0234  |
| 12   | 0.9633 | 1.0246 | 0.9777 | 1.0333 | -0.0144 | -0.0087 |
| 16   | 0.9633 | 1.0246 | 0.9807 | 1.0403 | -0.0174 | -0.0157 |
| 20   | 0.9633 | 1.0246 | 0.944  | 1.0045 | 0.0193  | 0.0202  |
**Chart S-IX.3. Plot of Inversion Recovery Data at 1.1 Equivalents (n-Bu)₄NCl.**

![Inversion Recovery Data for 1.1 Equiv. N(Bu)₄Cl](chart.png)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:
- $1/T_1$'s:
  - No. 0 = 0.8000
  - No. 1 = 0.2300
- $M(\infty)$'s:
  - No. 2 = 0.9000
  - No. 3 = 1.0000
- M(0)–M(\infty)'s:
  - No. 4 = -1.4000
  - No. 5 = 0.5000
- and M(0)'s for reference:
  - No. 4 = -0.5000
  - No. 5 = 1.5000
- Rates:
  - No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
- # 0 = 3.685563 +/− 0.443326
- # 1 = 3.669400 +/− 0.488563
- # 2 = 0.963288 +/− 0.002882
- # 3 = 1.024618 +/− 0.003020
- # 4 = -0.844435 +/− 0.005449
- # 5 = -0.229181 +/− 0.006601
- # 6 = 22.238473 +/− 1.172882
4) **Data for 1.5 Equivalents \((n-\text{Bu})_4\text{NCl}\):**

![Stacked Spectra of Inversion Recovery Data at 1.5 Equivalents \((n-\text{Bu})_4\text{NCl}\).](image)

**Figure S-IX.4.** Stacked Spectra of Inversion Recovery Data at 1.5 Equivalents \((n-\text{Bu})_4\text{NCl}\).

**Table S-IX.4.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1  | Fit M2  | σM1  | σM2  |
|--------|-------------|-------------|---------|---------|------|------|
| 0.001  | 0.1392      | 0.8458      | 0.1378  | 0.8469  | 0.0015 | -0.0012 |
| 0.002  | 0.1577      | 0.8335      | 0.1451  | 0.8266  | 0.0127 | 0.0069 |
| 0.003  | 0.1754      | 0.8219      | 0.1702  | 0.8189  | 0.0052 | 0.0031 |
| 0.004  | 0.1924      | 0.8111      | 0.1967  | 0.8071  | -0.0043 | 0.004 |
| 0.005  | 0.2086      | 0.8009      | 0.2061  | 0.8008  | 0.0026 | 0    |
| 0.006  | 0.2242      | 0.7913      | 0.2165  | 0.7913  | 0.0077 | 0    |
| 0.007  | 0.2392      | 0.7823      | 0.2443  | 0.7789  | -0.0051 | 0.0034 |
| 0.008  | 0.2535      | 0.7739      | 0.2548  | 0.7693  | -0.0013 | 0.0046 |
| 0.009  | 0.2672      | 0.7661      | 0.2741  | 0.7707  | -0.0069 | -0.0047 |
| 0.01   | 0.2804      | 0.7587      | 0.2819  | 0.7589  | -0.0015 | -0.0002 |
| 0.015  | 0.3389      | 0.7288      | 0.3432  | 0.7301  | -0.0043 | -0.0013 |
| 0.02   | 0.3871      | 0.7083      | 0.3912  | 0.7119  | -0.0041 | -0.0036 |
| 0.025  | 0.4271      | 0.695       | 0.4267  | 0.7011  | 0.0004  | -0.0061 |
| 0.03   | 0.4608      | 0.6873      | 0.4569  | 0.6904  | 0.0039  | -0.0031 |
| 0.035  | 0.4893      | 0.6838      | 0.488   | 0.683   | 0.0014  | 0.0008 |
| 0.04   | 0.5139      | 0.6835      | 0.5182  | 0.6913  | -0.0043 | -0.0078 |
| 0.045  | 0.5354      | 0.6856      | 0.5381  | 0.6838  | -0.0027 | 0.0018 |
| 0.05 | 0.5543 | 0.6896 | 0.5605 | 0.686 | -0.0062 | 0.0036 |
|------|--------|--------|--------|-------|---------|--------|
| 0.06 | 0.5864 | 0.7011 | 0.5864 | 0.7018 | 0.0008  | -0.0048|
| 0.07 | 0.6131 | 0.7154 | 0.6153 | 0.7202 | -0.0022 | -0.0048|
| 0.08 | 0.6361 | 0.7311 | 0.6276 | 0.7318 | 0.0084  | -0.0008|
| 0.09 | 0.6564 | 0.7476 | 0.6564 | 0.7538 | 0.0069  | -0.0068|
| 0.1  | 0.6748 | 0.7628 | 0.6708 | 0.7636 | 0.0039  | -0.0007|
| 0.15 | 0.7479 | 0.8329 | 0.75   | 0.8365 | -0.0021 | -0.0036|
| 0.2  | 0.8007 | 0.8861 | 0.7983 | 0.8835 | 0.0024  | 0.0025 |
| 0.25 | 0.8397 | 0.9255 | 0.8365 | 0.9227 | 0.0032  | 0.0027 |
| 0.3  | 0.8685 | 0.9546 | 0.8607 | 0.953  | 0.0078  | 0.0117 |
| 0.35 | 0.8898 | 0.9762 | 0.8813 | 0.9721 | 0.0085  | 0.0042 |
| 0.4  | 0.9056 | 0.9922 | 0.9144 | 0.9919 | -0.0088 | 0.0003 |
| 0.45 | 0.9173 | 1.004  | 0.9186 | 0.9982 | -0.0013 | 0.0058 |
| 0.5  | 0.9259 | 1.0128 | 0.9304 | 1.0103 | -0.0044 | 0.0025 |
| 0.6  | 0.9371 | 1.024  | 0.9381 | 1.0217 | -0.0011 | 0.0024 |
| 0.7  | 0.9432 | 1.0302 | 0.9487 | 1.0361 | -0.0056 | -0.0059|
| 0.8  | 0.9465 | 1.0336 | 0.9501 | 1.0343 | -0.0036 | -0.0007|
| 0.9  | 0.9483 | 1.0354 | 0.9522 | 1.0387 | -0.0039 | -0.0033|
| 1    | 0.9493 | 1.0364 | 0.9528 | 1.0353 | -0.0035 | 0.0011 |
| 1.5  | 0.9505 | 1.0376 | 0.9548 | 1.0463 | -0.0043 | -0.0087|
| 2    | 0.9505 | 1.0377 | 0.9474 | 1.0236 | 0.0032  | 0.0141 |
| 2.5  | 0.9505 | 1.0377 | 0.9509 | 1.0331 | -0.0003 | 0.0045 |
| 3    | 0.9505 | 1.0377 | 0.9532 | 1.0405 | -0.0027 | -0.0028|
| 4    | 0.9505 | 1.0377 | 0.9472 | 1.0374 | 0.0034  | 0.0003 |
| 6    | 0.9505 | 1.0377 | 0.9584 | 1.042  | -0.0079 | -0.0043|
| 8    | 0.9505 | 1.0377 | 0.9467 | 1.0419 | 0.0039  | -0.0043|
| 10   | 0.9505 | 1.0377 | 0.9498 | 1.0334 | 0.0008  | 0.0042 |
| 12   | 0.9505 | 1.0377 | 0.9457 | 1.0351 | 0.0049  | 0.0026 |
| 16   | 0.9505 | 1.0377 | 0.9469 | 1.0341 | 0.0037  | 0.0036 |
| 20   | 0.9505 | 1.0377 | 0.9474 | 1.0429 | 0.0031  | -0.0052|
Chart S-IX.4. Plot of Inversion Recovery Data at 1.5 Equivalents \((n\text{-Bu})_4\text{NCl}\).

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/T1's

No. 0 = 0.8000

No. 1 = 0.2300

M(\text{inf})'s

No. 2 = 0.9000

No. 3 = 1.0000

M(0)−M(\text{inf})'s

No. 4 = −1.4000

No. 5 = 0.5000

and M(0)'s for reference

No. 4 = −0.5000

No. 5 = 1.5000

Rates

No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value   | Uncertainty |
|----|---------|-------------|
| 0  | 6.288211 | ±0.223150   |
| 1  | 5.761302 | ±0.266559   |
| 2  | 0.950538 | ±0.001076   |
| 3  | 1.037666 | ±0.001136   |
| 4  | −0.830596| ±0.002272   |
| 5  | −0.178808| ±0.002763   |
| 6  | 22.257770| ±0.514013   |
5) Data for 2.0 Equivalents \((n{-}Bu)_2\text{NCl}\):

![Stacked Spectra of Inversion Recovery Data at 2.0 Equivalents \((n{-}Bu)_2\text{NCl}\)](image)

Figure S-IX.5. Stacked Spectra of Inversion Recovery Data at 2.0 Equivalents \((n{-}Bu)_2\text{NCl}\).

Table S-IX.5. CIFIT Plot File.

| \(d_2\) (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | \(\sigma\)M1 | \(\sigma\)M2 |
|-------------|-------------|-------------|--------|--------|-------------|-------------|
| 0.001       | 0.3691      | 0.8144      | 0.3623 | 0.8105 | 0.0068      | 0.0038      |
| 0.002       | 0.3836      | 0.8067      | 0.3672 | 0.7953 | 0.0165      | 0.0114      |
| 0.003       | 0.3976      | 0.7995      | 0.4004 | 0.8041 | -0.0028     | -0.0047     |
| 0.004       | 0.4111      | 0.7927      | 0.4122 | 0.7912 | -0.0011     | 0.0015      |
| 0.005       | 0.424       | 0.7865      | 0.4243 | 0.7848 | -0.0003     | 0.0017      |
| 0.006       | 0.4365      | 0.7807      | 0.442  | 0.7865 | -0.0055     | -0.0058     |
| 0.007       | 0.4485      | 0.7752      | 0.4452 | 0.7722 | 0.0033      | 0.003       |
| 0.008       | 0.46        | 0.7702      | 0.4622 | 0.7708 | -0.0021     | -0.0006     |
| 0.009       | 0.4711      | 0.7656      | 0.476  | 0.7712 | -0.0048     | -0.0056     |
| 0.01        | 0.4819      | 0.7613      | 0.4851 | 0.7561 | -0.0033     | 0.0051      |
| 0.015       | 0.5299      | 0.7445      | 0.5356 | 0.7417 | -0.0056     | 0.0028      |
| 0.02        | 0.5703      | 0.7342      | 0.5649 | 0.7308 | 0.0054      | 0.0034      |
| 0.025       | 0.6045      | 0.7288      | 0.601  | 0.7368 | 0.0035      | -0.008      |
| 0.03        | 0.6338      | 0.7271      | 0.6414 | 0.733  | -0.0076     | -0.0059     |
| 0.035       | 0.6592      | 0.7282      | 0.6609 | 0.7332 | -0.0018     | -0.005      |
| 0.04        | 0.6813      | 0.7314      | 0.6867 | 0.7346 | -0.0054     | -0.0032     |
| 0.045       | 0.7009      | 0.7362      | 0.7027 | 0.7303 | -0.0019     | 0.0059      |
| t  | r(t)   | g(t)   | r(t)  | g(t)  | r(t)  | g(t)  |
|----|--------|--------|-------|-------|-------|-------|
| 0.05 | 0.7183 | 0.742  | 0.7255 | 0.7464 | -0.0072 | -0.0044 |
| 0.06 | 0.7483 | 0.7558 | 0.7438 | 0.7604 | 0.0045 | -0.0046 |
| 0.07 | 0.7733 | 0.7709 | 0.7663 | 0.7741 | 0.007 | -0.0031 |
| 0.08 | 0.7948 | 0.7863 | 0.7924 | 0.7851 | 0.0024 | 0.0013 |
| 0.09 | 0.8136 | 0.8014 | 0.8097 | 0.8043 | 0.0039 | -0.003 |
| 0.1  | 0.8303 | 0.8157 | 0.83  | 0.8175 | 0.0003 | -0.0018 |
| 0.15 | 0.8926 | 0.8744 | 0.8921 | 0.8704 | 0.0004 | 0.004  |
| 0.2  | 0.9322 | 0.9135 | 0.9243 | 0.9104 | 0.0079 | 0.0031 |
| 0.25 | 0.9579 | 0.939  | 0.954 | 0.9287 | 0.0039 | 0.0103 |
| 0.3  | 0.9747 | 0.9556 | 0.9736 | 0.9501 | 0.0011 | 0.0056 |
| 0.35 | 0.9856 | 0.9664 | 0.9929 | 0.9727 | -0.0073 | -0.0062 |
| 0.4  | 0.9927 | 0.9735 | 0.9908 | 0.9746 | 0.0019 | -0.0011 |
| 0.45 | 0.9973 | 0.9781 | 1.0027 | 0.98  | -0.0054 | -0.002 |
| 0.5  | 1.0003 | 0.9811 | 1.0049 | 0.9864 | -0.0046 | -0.0053 |
| 0.6  | 1.0035 | 0.9843 | 1.004 | 0.9859 | -0.0005 | -0.0016 |
| 0.7  | 1.0049 | 0.9856 | 1.0136 | 0.9928 | -0.0087 | -0.0072 |
| 0.8  | 1.0055 | 0.9862 | 0.998 | 0.9851 | 0.0075 | 0.0011 |
| 0.9  | 1.0057 | 0.9865 | 1.0078 | 0.9797 | -0.0021 | 0.0067 |
| 1    | 1.0058 | 0.9866 | 1.0019 | 0.9833 | 0.0039 | 0.0032 |
| 1.5  | 1.0059 | 0.9866 | 1.006 | 0.9818 | -0.0001 | 0.0048 |
| 2    | 1.0059 | 0.9866 | 1.0118 | 0.9909 | -0.0059 | -0.0043 |
| 2.5  | 1.0059 | 0.9866 | 1.0074 | 0.987 | -0.0015 | -0.0003 |
| 3    | 1.0059 | 0.9866 | 1.0059 | 0.9903 | 0 | -0.0037 |
| 4    | 1.0059 | 0.9866 | 1.0048 | 0.9851 | 0.0011 | 0.0015 |
| 6    | 1.0059 | 0.9866 | 1.0063 | 0.9865 | -0.0004 | 0.0002 |
| 8    | 1.0059 | 0.9866 | 0.9969 | 0.9811 | 0.009 | 0.0056 |
| 10   | 1.0059 | 0.9866 | 1.0124 | 0.9903 | -0.0066 | -0.0037 |
| 12   | 1.0059 | 0.9866 | 1.0007 | 0.9795 | 0.0052 | 0.0071 |
| 16   | 1.0059 | 0.9866 | 1.0011 | 0.9878 | 0.0048 | -0.0011 |
| 20   | 1.0059 | 0.9866 | 1.0137 | 0.9876 | -0.0078 | -0.0009 |
**Chart S-IX.5. Plot of Inversion Recovery Data at 2.0 Equivalents \((n-\text{Bu})_4\text{NCl}\).**

CIFIT Guesses and Fit Parameters

Initial values of parameters:
- 1/T1's
  - No. 0 = 0.8000
  - No. 1 = 0.2300
- M(\text{inf})'s
  - No. 2 = 0.9000
  - No. 3 = 1.0000
- M(0)–M(\text{inf})'s
  - No. 4 = -1.4000
  - No. 5 = 0.5000
  - and M(0)'s for reference
  - No. 4 = -0.5000
  - No. 5 = 1.5000
- Rates
  - No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
- # 0 = 8.441348 +/- 0.364276
- # 1 = 8.730979 +/- 0.482589
- # 2 = 1.005886 +/- 0.001133
- # 3 = 0.986632 +/- 0.001185
- # 4 = -0.651955 +/- 0.002527
- # 5 = -0.164032 +/- 0.003049
- # 6 = 20.394057 +/- 0.832915
6) Data for 6 Equivalents LiCl:

![Stacked Spectra of Inversion Recovery Data at 6 Equivalents LiCl.](image)

**Figure S-IX.6.** Stacked Spectra of Inversion Recovery Data at 6 Equivalents LiCl.

**Table S-IX.6.** CIFT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.3495     | 0.8316      | -0.3649| 0.8199 | 0.0154 | 0.0117 |
| 0.002  | -0.3239     | 0.8068      | -0.3343| 0.7999 | 0.0104 | 0.0068 |
| 0.003  | -0.2994     | 0.7831      | -0.3082| 0.7751 | 0.0087 | 0.008  |
| 0.004  | -0.2761     | 0.7606      | -0.2817| 0.7563 | 0.0055 | 0.0042 |
| 0.005  | -0.2539     | 0.7391      | -0.2619| 0.7329 | 0.008  | 0.0062 |
| 0.006  | -0.2326     | 0.7186      | -0.2369| 0.7128 | 0.0043 | 0.0058 |
| 0.007  | -0.2123     | 0.6991      | -0.2189| 0.6898 | 0.0066 | 0.0092 |
| 0.008  | -0.193      | 0.6805      | -0.1977| 0.6748 | 0.0048 | 0.0057 |
| 0.009  | -0.1745     | 0.6627      | -0.1785| 0.6557 | 0.004  | 0.0071 |
| 0.01   | -0.1568     | 0.6458      | -0.159 | 0.6409 | 0.0022 | 0.0049 |
| 0.015  | -0.0797     | 0.5725      | -0.079 | 0.5695 | -0.0008| 0.003  |
| 0.02   | -0.0185     | 0.515       | -0.0162| 0.5135 | -0.0022| 0.0015 |
| 0.025  | 0.0303      | 0.4699      | 0.0325 | 0.4693 | -0.0022| 0.0006 |
| 0.03   | 0.0692      | 0.4347      | 0.0721 | 0.4357 | -0.0028| -0.001 |
| 0.035  | 0.1004      | 0.4073      | 0.1043 | 0.4097 | -0.0039| -0.0025 |
| 0.04   | 0.1253      | 0.3859      | 0.1292 | 0.3904 | -0.0038| -0.0045 |
| 0.045  | 0.1454      | 0.3695      | 0.1474 | 0.3757 | -0.0019| -0.0062 |
| 0.05   | 0.1617      | 0.3569      | 0.1641 | 0.3623 | -0.0023| -0.0055 |
|   |      |      |      |      |      |      |      |
|---|------|------|------|------|------|------|------|
| 0.055 | 0.1749 | 0.3473 | 0.1765 | 0.3529 | -0.0016 | -0.0056 |
| 0.06  | 0.1857 | 0.3401 | 0.1875 | 0.3464 | -0.0018 | -0.0063 |
| 0.065 | 0.1947 | 0.3347 | 0.1973 | 0.342  | -0.0027 | -0.0072 |
| 0.07  | 0.2021 | 0.3309 | 0.205  | 0.3402 | -0.0029 | -0.0093 |
| 0.075 | 0.2083 | 0.3283 | 0.2109 | 0.3339 | -0.0025 | -0.0057 |
| 0.08  | 0.2136 | 0.3265 | 0.217  | 0.3356 | -0.0033 | -0.0091 |
| 0.09  | 0.2221 | 0.3251 | 0.2245 | 0.3345 | -0.0023 | -0.0094 |
| 0.1   | 0.2288 | 0.3256 | 0.2298 | 0.3333 | -0.001  | -0.0077 |
| 0.15  | 0.2508 | 0.3383 | 0.251  | 0.3478 | -0.0002 | -0.0095 |
| 0.2   | 0.2682 | 0.3548 | 0.2694 | 0.3647 | -0.0012 | -0.0099 |
| 0.25  | 0.2848 | 0.3714 | 0.2858 | 0.3801 | -0.001  | -0.0088 |
| 0.3   | 0.3009 | 0.3875 | 0.3019 | 0.3958 | -0.001  | -0.0083 |
| 0.35  | 0.3167 | 0.4032 | 0.3185 | 0.4091 | -0.0018 | -0.0059 |
| 0.4   | 0.332  | 0.4186 | 0.3333 | 0.4256 | -0.0014 | -0.007  |
| 0.45  | 0.3469 | 0.4336 | 0.347  | 0.4394 | -0.0001 | -0.0058 |
| 0.5   | 0.3615 | 0.4482 | 0.3592 | 0.4495 | 0.0024  | -0.0013 |
| 0.6   | 0.3896 | 0.4763 | 0.3868 | 0.4789 | 0.0028  | -0.0026 |
| 0.7   | 0.4163 | 0.503  | 0.4132 | 0.5028 | 0.0031  | 0.0002  |
| 0.8   | 0.4417 | 0.5284 | 0.4366 | 0.527  | 0.0051  | 0.0015  |
| 0.9   | 0.4658 | 0.5526 | 0.4633 | 0.5504 | 0.0025  | 0.0021  |
| 1     | 0.4888 | 0.5756 | 0.4829 | 0.5707 | 0.0058  | 0.0049  |
| 1.5   | 0.5877 | 0.6746 | 0.5821 | 0.6692 | 0.0056  | 0.0054  |
| 2     | 0.6646 | 0.7515 | 0.6628 | 0.7442 | 0.0018  | 0.0073  |
| 2.5   | 0.7243 | 0.8114 | 0.7272 | 0.8094 | -0.0029 | 0.002   |
| 3     | 0.7707 | 0.8579 | 0.7732 | 0.8531 | -0.0025 | 0.0047  |
| 4     | 0.8349 | 0.9221 | 0.847  | 0.9237 | -0.0121 | -0.0017 |
| 6     | 0.8971 | 0.9843 | 0.9065 | 0.9833 | -0.0095 | 0.001   |
| 8     | 0.9197 | 1.007  | 0.9226 | 0.9999 | -0.0029 | 0.0072  |
| 10    | 0.928  | 1.0153 | 0.9313 | 1.0043 | -0.0032 | 0.011   |
| 12    | 0.931  | 1.0183 | 0.9355 | 1.0093 | -0.0044 | 0.009   |
| 16    | 0.9326 | 1.0198 | 0.9381 | 1.0124 | -0.0056 | 0.0075  |
| 20    | 0.9328 | 1.02   | 0.9439 | 1.018  | -0.0112 | 0.002   |

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Chart S-IX.6. Plot of Inversion Recovery Data at 6 Equivalents LiCl.

CIFIT Guesses and Fit Parameters

| Initial values of parameters: | Final Values of Fitted Parameters and Uncertainties: |
|--------------------------------|-----------------------------------------------------|
| 1/T1's                         |                                                     |
| No. 0 = 0.8000 No. 1 = 0.2300  | # 0 = 0.530935 +/- 0.112951                         |
| M(inf)'s                       | # 1 = 0.477127 +/- 0.114632                         |
| No. 2 = 0.9000 No. 3 = 1.0000  | # 2 = 0.932782 +/- 0.002180                         |
| M(0)-M(inf)'s                  | # 3 = 1.020079 +/- 0.002294                         |
| No. 4 = -1.4000 No. 5 = 0.5000 | # 4 = -1.309037 +/- 0.002993                        |
| and M(0)'s for reference       | # 5 = -0.162456 +/- 0.003797                        |
| No. 4 = -0.5000 No. 5 = 1.5000 | # 6 = 23.315415 +/- 0.250699                        |
| Rates                          |                                                     |
| No. 6 = 20.0000                |                                                     |
7) Data for 12 Equivalents LiCl:

![Figure S-IX.7. Stacked Spectra of Inversion Recovery Data at 12 Equivalents LiCl.](image)

Table S-IX.7. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.2804     | 0.8935      | -0.2947| 0.8817 | 0.0143| 0.0117|
| 0.002  | -0.2562     | 0.8705      | -0.268 | 0.8617 | 0.0118| 0.0088|
| 0.003  | -0.2331     | 0.8487      | -0.2424| 0.8416 | 0.0093| 0.007 |
| 0.004  | -0.211      | 0.8278      | -0.2179| 0.824  | 0.0069| 0.0039|
| 0.005  | -0.1899     | 0.8079      | -0.1924| 0.8033 | 0.0025| 0.0047|
| 0.006  | -0.1698     | 0.789       | -0.1728| 0.7868 | 0.003 | 0.0022|
| 0.007  | -0.1506     | 0.7709      | -0.154 | 0.7651 | 0.0034| 0.0058|
| 0.008  | -0.1322     | 0.7537      | -0.1334| 0.7506 | 0.0012| 0.0031|
| 0.009  | -0.1147     | 0.7373      | -0.1157| 0.7335 | 0.001 | 0.0038|
| 0.01   | -0.0979     | 0.7216      | -0.0985| 0.7183 | 0.0006| 0.0033|
| 0.015  | -0.0247     | 0.6537      | -0.0223| 0.6513 | -0.0024| 0.0023|
| 0.02   | 0.0336      | 0.6003      | 0.0364 | 0.6008 | -0.0029| -0.0006|
| 0.025  | 0.08        | 0.5584      | 0.0806 | 0.5547 | -0.0006| 0.0037|
| 0.03   | 0.1172      | 0.5256      | 0.1175 | 0.5232 | -0.0003| 0.0024|
| 0.035  | 0.1469      | 0.5         | 0.1484 | 0.5018 | -0.0014| -0.0018|
| 0.04   | 0.1708      | 0.4802      | 0.1708 | 0.4817 | 0    | -0.0016|
| 0.045  | 0.1901      | 0.4648      | 0.1906 | 0.4666 | -0.0005| -0.0018|
| 0.05   | 0.2057      | 0.453       | 0.208 | 0.458  | -0.0023| -0.005 |
|       | 0.055 | 0.2184 | 0.444  | 0.2181 | 0.4484 | 0.0003 | -0.0043 |
|-------|-------|--------|--------|--------|--------|--------|---------|
| 0.06  | 0.2288| 0.4373 | 0.2299 | 0.4419 | -0.0011| -0.0046|
| 0.065 | 0.2374| 0.4323 | 0.2371 | 0.4379 | 0.0003 | -0.0055|
| 0.07  | 0.2445| 0.4288 | 0.2442 | 0.4356 | 0.0003 | -0.0068|
| 0.075 | 0.2505| 0.4263 | 0.2499 | 0.4316 | 0.0006 | -0.0053|
| 0.08  | 0.2556| 0.4247 | 0.2563 | 0.4323 | -0.0007| -0.0076|
| 0.09  | 0.2638| 0.4234 | 0.2655 | 0.432  | -0.0017| -0.0086|
| 0.1   | 0.2702| 0.4239 | 0.273  | 0.4326 | -0.0029| -0.0087|
| 0.15  | 0.2912| 0.4363 | 0.2926 | 0.4431 | -0.0014| -0.0068|
| 0.2   | 0.3077| 0.4523 | 0.3109 | 0.4589 | -0.0032| -0.0066|
| 0.25  | 0.3233| 0.4682 | 0.3243 | 0.4735 | -0.001  | -0.0053|
| 0.3   | 0.3385| 0.4838 | 0.3382 | 0.4906 | 0.0003 | -0.0069|
| 0.35  | 0.3533| 0.4989 | 0.3564 | 0.5058 | -0.0031| -0.0069|
| 0.4   | 0.3677| 0.5137 | 0.3675 | 0.5195 | 0.0001 | -0.0059|
| 0.45  | 0.3817| 0.528  | 0.382  | 0.532  | -0.0003| -0.004  |
| 0.5   | 0.3953| 0.542  | 0.3965 | 0.5475 | -0.0012| -0.0055|
| 0.6   | 0.4215| 0.5688 | 0.4212 | 0.5724 | 0.0003 | -0.0036|
| 0.7   | 0.4463| 0.5942 | 0.4452 | 0.5973 | 0.001  | -0.0031|
| 0.8   | 0.4697| 0.6182 | 0.4679 | 0.6181 | 0.0018 | 0.0001 |
| 0.9   | 0.492 | 0.641  | 0.4892 | 0.6418 | 0.0028 | -0.0008|
| 1     | 0.5131| 0.6626 | 0.5099 | 0.6618 | 0.0032 | 0.0008 |
| 1.5   | 0.6029| 0.7547 | 0.6009 | 0.7512 | 0.002  | 0.0035 |
| 2     | 0.6715| 0.8249 | 0.6707 | 0.8207 | 0.0007 | 0.0042 |
| 2.5   | 0.7238| 0.8785 | 0.7252 | 0.8731 | -0.0014| 0.0054 |
| 3     | 0.7637| 0.9194 | 0.7639 | 0.9137 | -0.0002| 0.0057 |
| 4     | 0.8174| 0.9744 | 0.8181 | 0.9662 | -0.0007| 0.0082 |
| 6     | 0.8668| 1.025  | 0.8715 | 1.0186 | -0.0047| 0.0065 |
| 8     | 0.8836| 1.0422 | 0.8859 | 1.0354 | -0.0023| 0.0068 |
| 10    | 0.8893| 1.048  | 0.8942 | 1.0436 | -0.005  | 0.0044 |
| 12    | 0.8912| 1.05   | 0.8983 | 1.0422 | -0.0071| 0.0078 |
| 16    | 0.8921| 1.0509 | 0.9014 | 1.0486 | -0.0093| 0.0023 |
| 20    | 0.8922| 1.051  | 0.9022 | 1.052  | -0.0101| -0.001 |
**Chart S-IX.7. Plot of Inversion Recovery Data at 12 Equivalents LiCl.**

![Inversion Recovery Data for 12 Equiv. LiCl](chart.png)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

- $1/T_1$'s
  - No. 0 = 0.8000
  - No. 1 = 0.2300

- M($\infty$)'s
  - No. 2 = 0.9000
  - No. 3 = 1.0000

- M(0)–M($\infty$)'s
  - No. 4 = -1.4000
  - No. 5 = 0.5000

And M(0)'s for reference

- No. 4 = -0.5000
- No. 5 = 1.5000

Rates

- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value          | Uncertainty     |
|----|----------------|-----------------|
| 0  | 1.106666       | ±0.103311       |
| 1  | -0.011124      | ±0.100364       |
| 2  | 0.892200       | ±0.001773       |
| 3  | 1.051018       | ±0.001881       |
| 4  | -1.197991      | ±0.002480       |
| 5  | -0.133519      | ±0.003141       |
| 6  | 23.126912      | ±0.221502       |
8) **Data for 24 Equivalents LiCl:**

![Stacked Spectra](image)

**Figure S-IX.8.** Stacked Spectra of Inversion Recovery Data at 24 Equivalents LiCl.

**Table S-IX.8.** CIFIT Plot File.

| d2  | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-----|-------------|-------------|--------|--------|------|------|
| (s) |             |             |        |        |      |      |
| 0.001 | -0.219     | 0.9198      | -0.2408 | 0.9027 | 0.0218 | 0.017 |
| 0.002 | -0.1956    | 0.8975      | -0.2137 | 0.885  | 0.018  | 0.0124 |
| 0.003 | -0.1733    | 0.8762      | -0.1917 | 0.8645 | 0.0184 | 0.0116 |
| 0.004 | -0.152     | 0.8558      | -0.163  | 0.8454 | 0.011  | 0.0104 |
| 0.005 | -0.1316    | 0.8364      | -0.1434 | 0.8257 | 0.0118 | 0.0108 |
| 0.006 | -0.1121    | 0.8179      | -0.1183 | 0.8095 | 0.0062 | 0.0084 |
| 0.007 | -0.0935    | 0.8003      | -0.0991 | 0.7915 | 0.0057 | 0.0087 |
| 0.008 | -0.0756    | 0.7834      | -0.0781 | 0.7765 | 0.0024 | 0.0069 |
| 0.009 | -0.0586    | 0.7673      | -0.06   | 0.7595 | 0.0014 | 0.0079 |
| 0.01  | -0.0423    | 0.752       | -0.0425 | 0.7453 | 0.0002 | 0.0067 |
| 0.015 | 0.029      | 0.6852      | 0.0303  | 0.6822 | -0.0013 | 0.003 |
| 0.02  | 0.0861     | 0.6325      | 0.0905  | 0.6313 | -0.0044 | 0.0011 |
| 0.025 | 0.1318     | 0.5909      | 0.1354  | 0.5904 | -0.0035 | 0.0005 |
| 0.03  | 0.1685     | 0.5582      | 0.1717  | 0.5603 | -0.0031 | -0.002 |
| 0.035 | 0.1981     | 0.5326      | 0.2013  | 0.5347 | -0.0033 | -0.0021 |
| 0.04  | 0.2219     | 0.5126      | 0.2259  | 0.518  | -0.004  | -0.0054 |
| 0.045 | 0.2412     | 0.497       | 0.244   | 0.5044 | -0.0028 | -0.0074 |
| 0.05  | 0.2569     | 0.485       | 0.2606  | 0.4923 | -0.0037 | -0.0073 |
|   |   |   |   |   |   |   |
|---|---|---|---|---|---|---|
| 0.055 | 0.2698 | 0.4759 | 0.2752 | 0.4842 | -0.0054 | -0.0083 |
| 0.06 | 0.2803 | 0.4689 | 0.2827 | 0.4788 | -0.0024 | -0.0099 |
| 0.065 | 0.289 | 0.4637 | 0.294 | 0.4764 | -0.005 | -0.0126 |
| 0.07 | 0.2963 | 0.46 | 0.2994 | 0.4717 | -0.003 | -0.0117 |
| 0.075 | 0.3025 | 0.4573 | 0.3073 | 0.473 | -0.0048 | -0.0157 |
| 0.08 | 0.3077 | 0.4556 | 0.3114 | 0.4688 | -0.0037 | -0.0132 |
| 0.09 | 0.3162 | 0.4541 | 0.3194 | 0.4665 | -0.0033 | -0.0124 |
| 0.1 | 0.3227 | 0.4544 | 0.3251 | 0.4672 | -0.0023 | -0.0128 |
| 0.15 | 0.3443 | 0.4664 | 0.3441 | 0.4803 | 0.0002 | -0.0139 |
| 0.2 | 0.3609 | 0.4823 | 0.3632 | 0.4939 | -0.0023 | -0.0116 |
| 0.25 | 0.3767 | 0.4982 | 0.3771 | 0.5086 | -0.0004 | -0.0104 |
| 0.3 | 0.3919 | 0.5137 | 0.3904 | 0.522 | 0.0015 | -0.0083 |
| 0.35 | 0.4068 | 0.5287 | 0.4042 | 0.5376 | 0.0025 | -0.0089 |
| 0.4 | 0.4212 | 0.5433 | 0.419 | 0.551 | 0.0022 | -0.0077 |
| 0.45 | 0.4352 | 0.5575 | 0.4327 | 0.5641 | 0.0025 | -0.0065 |
| 0.5 | 0.4488 | 0.5713 | 0.4457 | 0.5757 | 0.0031 | -0.0044 |
| 0.6 | 0.4749 | 0.5978 | 0.4692 | 0.6014 | 0.0057 | -0.0036 |
| 0.7 | 0.4995 | 0.6228 | 0.4939 | 0.6234 | 0.0056 | -0.0006 |
| 0.8 | 0.5228 | 0.6464 | 0.5195 | 0.6468 | 0.0033 | -0.0004 |
| 0.9 | 0.5448 | 0.6687 | 0.5392 | 0.6675 | 0.0056 | 0.0012 |
| 1 | 0.5655 | 0.6897 | 0.5583 | 0.6866 | 0.0072 | 0.0031 |
| 1.5 | 0.6533 | 0.7787 | 0.65 | 0.7721 | 0.0033 | 0.0066 |
| 2 | 0.7192 | 0.8456 | 0.719 | 0.839 | 0.0002 | 0.0065 |
| 2.5 | 0.7687 | 0.8958 | 0.7729 | 0.8905 | -0.0041 | 0.0053 |
| 3 | 0.806 | 0.9336 | 0.8103 | 0.9253 | -0.0043 | 0.0083 |
| 4 | 0.855 | 0.9833 | 0.8637 | 0.9801 | -0.0087 | 0.0032 |
| 6 | 0.8983 | 1.0272 | 0.9115 | 1.0217 | -0.0132 | 0.0055 |
| 8 | 0.9121 | 1.0412 | 0.9233 | 1.033 | -0.0102 | 0.0082 |
| 10 | 0.9165 | 1.0457 | 0.9238 | 1.0304 | -0.0073 | 0.0153 |
| 12 | 0.9179 | 1.0471 | 0.927 | 1.0355 | -0.0091 | 0.0116 |
| 16 | 0.9185 | 1.0477 | 0.9302 | 1.0384 | -0.0117 | 0.0093 |
| 20 | 0.9186 | 1.0478 | 0.9311 | 1.0404 | -0.0125 | 0.0074 |
**Chart S-IX.8. Plot of Inversion Recovery Data at 24 Equivalents LiCl.**

![Inversion Recovery Data for 24 Equiv. LiCl](image)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/T1's
- No. 0 = 0.8000
- No. 1 = 0.2300

M(inf)'s
- No. 2 = 0.9000
- No. 3 = 1.0000

M(0)–M(inf)'s
- No. 4 = -1.4000
- No. 5 = 0.5000

and M(0)'s for reference
- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

|   |       | ±       |       |
|---|--------|---------|-------|
| 0 | 0.894  | 0.178   | 0.179 |
| 1 | 0.253  | 0.177   | 0.177 |
| 2 | 0.919  | 0.003   | 0.003 |
| 3 | 1.048  | 0.003   | 0.003 |
| 4 | -1.162 | 0.004   | 0.004 |
| 5 | -0.105 | 0.005   | 0.005 |
| 6 | 22.650 | 0.372   | 0.372 |
9) **Data for 48 Equivalents LiCl:**

![Stacked Spectra of Inversion Recovery Data at 48 Equivalents LiCl.](image)

**Figure S–IX.9.** Stacked Spectra of Inversion Recovery Data at 48 Equivalents LiCl.

**Table S–IX.9.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.272      | 0.9168      | -0.2888| 0.9072 | 0.0168| 0.0096|
| 0.002  | -0.2492     | 0.8953      | -0.2633| 0.8838 | 0.0141| 0.0115|
| 0.003  | -0.2274     | 0.8748      | -0.2389| 0.866  | 0.0115| 0.0088|
| 0.004  | -0.2065     | 0.8552      | -0.2151| 0.8474 | 0.0086| 0.0078|
| 0.005  | -0.1864     | 0.8364      | -0.1928| 0.8293 | 0.0064| 0.0072|
| 0.006  | -0.1671     | 0.8184      | -0.1744| 0.8114 | 0.0072| 0.0071|
| 0.007  | -0.1487     | 0.8012      | -0.1534| 0.7953 | 0.0047| 0.0059|
| 0.008  | -0.131      | 0.7847      | -0.1339| 0.7792 | 0.0029| 0.0055|
| 0.009  | -0.114      | 0.7689      | -0.114 | 0.7642 | 0    | 0.0048|
| 0.01   | -0.0977     | 0.7538      | -0.0974| 0.7485 | -0.0004| 0.0053|
| 0.015  | -0.0258     | 0.6874      | -0.0246| 0.6813 | -0.0013| 0.0061|
| 0.02   | 0.0325      | 0.6341      | 0.0327 | 0.6297 | -0.0002| 0.0044|
| 0.025  | 0.08        | 0.5914      | 0.0805 | 0.592 | -0.0005| -0.0006|
| 0.03   | 0.1187      | 0.5572      | 0.1201 | 0.5564 | -0.0015| 0.0008|
| 0.035  | 0.1502      | 0.5299      | 0.1519 | 0.5284 | -0.0018| 0.0015|
| 0.04   | 0.1759      | 0.5082      | 0.1783 | 0.5092 | -0.0024| -0.0011|
| 0.045  | 0.1971      | 0.4909      | 0.1985 | 0.4964 | -0.0015| -0.0054|
| 0.05   | 0.2144      | 0.4773      | 0.2151 | 0.4825 | -0.0007| -0.0052|

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|   |   |   |   |   |   |   |
|---|---|---|---|---|---|---|
| 0.055 | 0.2287 | 0.4667 | 0.2324 | 0.4716 | -0.0037 | -0.005 |
| 0.06 | 0.2406 | 0.4584 | 0.2422 | 0.4655 | -0.0016 | -0.0071 |
| 0.065 | 0.2505 | 0.452 | 0.2502 | 0.4579 | 0.0003 | -0.0059 |
| 0.07 | 0.2588 | 0.4471 | 0.2598 | 0.4546 | -0.0009 | -0.0074 |
| 0.075 | 0.2658 | 0.4435 | 0.2657 | 0.4509 | 0.0001 | -0.0074 |
| 0.08 | 0.2718 | 0.4409 | 0.2735 | 0.452 | -0.0017 | -0.0111 |
| 0.09 | 0.2813 | 0.438 | 0.2834 | 0.4484 | -0.0021 | -0.0104 |
| 0.1 | 0.2886 | 0.4372 | 0.2883 | 0.4486 | 0.0003 | -0.0114 |
| 0.15 | 0.311 | 0.4466 | 0.3119 | 0.457 | -0.0009 | -0.0105 |
| 0.2 | 0.327 | 0.4614 | 0.3292 | 0.4735 | -0.0023 | -0.0121 |
| 0.25 | 0.3418 | 0.4765 | 0.343 | 0.487 | -0.0012 | -0.0105 |
| 0.3 | 0.3561 | 0.4912 | 0.3568 | 0.5 | -0.0007 | -0.0088 |
| 0.35 | 0.3701 | 0.5057 | 0.3723 | 0.5147 | -0.0022 | -0.009 |
| 0.4 | 0.3837 | 0.5197 | 0.3836 | 0.5288 | 0.0002 | -0.009 |
| 0.45 | 0.397 | 0.5334 | 0.398 | 0.5394 | -0.001 | -0.006 |
| 0.5 | 0.4099 | 0.5467 | 0.41 | 0.5542 | -0.0001 | -0.0075 |
| 0.6 | 0.4348 | 0.5724 | 0.4339 | 0.5777 | 0.0009 | -0.0053 |
| 0.7 | 0.4584 | 0.5968 | 0.46 | 0.6011 | -0.0016 | -0.0044 |
| 0.8 | 0.4808 | 0.6199 | 0.4793 | 0.6222 | 0.0015 | -0.0023 |
| 0.9 | 0.5021 | 0.6419 | 0.4997 | 0.642 | 0.0024 | -0.0001 |
| 1 | 0.5223 | 0.6627 | 0.5204 | 0.6628 | 0.0019 | -0.0001 |
| 1.5 | 0.609 | 0.7521 | 0.605 | 0.7485 | 0.004 | 0.0037 |
| 2 | 0.6759 | 0.8211 | 0.6723 | 0.8146 | 0.0036 | 0.0066 |
| 2.5 | 0.7274 | 0.8744 | 0.725 | 0.8655 | 0.0024 | 0.0089 |
| 3 | 0.7672 | 0.9154 | 0.767 | 0.9063 | 0.0002 | 0.0091 |
| 4 | 0.8216 | 0.9715 | 0.824 | 0.9624 | -0.0024 | 0.0092 |
| 6 | 0.8732 | 1.0247 | 0.8802 | 1.0162 | -0.007 | 0.0085 |
| 8 | 0.8915 | 1.0436 | 0.8986 | 1.0372 | -0.0071 | 0.0064 |
| 10 | 0.8979 | 1.0503 | 0.9093 | 1.0453 | -0.0114 | 0.005 |
| 12 | 0.9002 | 1.0526 | 0.909 | 1.0446 | -0.0088 | 0.008 |
| 16 | 0.9013 | 1.0538 | 0.9119 | 1.0464 | -0.0106 | 0.0074 |
| 20 | 0.9014 | 1.0539 | 0.9139 | 1.049 | -0.0125 | 0.0049 |
Chart S-IX.9. Plot of Inversion Recovery Data at 48 Equivalents LiCl.

CIFIT Guesses and Fit Parameters

Initial values of parameters:
1/T1's
No. 0 = 0.8000 No. 1 = 0.2300

M(inf)'s
No. 2 = 0.9000 No. 3 = 1.0000

M(0)-M(inf)'s
No. 4 = -1.4000 No. 5 = 0.5000
and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000
Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value       | Uncertainty |
|----|-------------|-------------|
| 0  | 0.894011    | +/−0.178277 |
| 1  | 0.253202    | +/−0.177138 |
| 2  | 0.918561    | +/−0.002971 |
| 3  | 1.047780    | +/−0.003142 |
| 4  | -1.161982   | +/−0.004162 |
| 5  | -0.104642   | +/−0.005296 |
| 6  | 22.650877   | +/−0.371693 |
X. Inversion Recovery Data for Section VI (Tl(OTf) Addition)

1) Data 1 h after addition of 2 equiv. Tl(OTf):

![Stacked Spectra of Inversion Recovery Data 1 h After Tl(OTf) Addition.](image)

**Figure S–X.1.** Stacked Spectra of Inversion Recovery Data 1 h After Tl(OTf) Addition.

**Table S–X.1.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|-----|-----|
| 0.001  | -0.0773     | 0.7513      | -0.0894| 0.7429 | 0.0121| 0.0084|
| 0.002  | -0.0608     | 0.7358      | -0.0611| 0.7359 | 0.0003| -0.0001|
| 0.003  | -0.0451     | 0.721       | -0.0489| 0.7187 | 0.0038| 0.0023|
| 0.004  | -0.0301     | 0.7069      | -0.0346| 0.7059 | 0.0045| 0.001 |
| 0.005  | -0.0157     | 0.6935      | -0.0182| 0.6914 | 0.0025| 0.002 |
| 0.006  | -0.002      | 0.6806      | -0.0068| 0.6792 | 0.0047| 0.0015|
| 0.007  | 0.0111      | 0.6684      | 0.0101 | 0.665  | 0.001 | 0.0034|
| 0.008  | 0.0236      | 0.6568      | 0.0194 | 0.6437 | 0.0042| 0.0131|
| 0.009  | 0.0355      | 0.6457      | 0.0321 | 0.6384 | 0.0034| 0.0072|
| 0.01   | 0.0469      | 0.6351      | 0.0413 | 0.6313 | 0.0056| 0.0038|
| 0.015  | 0.0968      | 0.5892      | 0.0936 | 0.581  | 0.0032| 0.0083|
| 0.02   | 0.1366      | 0.5532      | 0.1425 | 0.5531 | -0.0058| 0.0001|
| 0.025  | 0.1684      | 0.525       | 0.1697 | 0.5258 | -0.0013| -0.0008|
| 0.03   | 0.1939      | 0.503       | 0.1927 | 0.5052 | 0.0012| -0.0022|
| 0.035  | 0.2144      | 0.4859      | 0.2156 | 0.4909 | -0.0012| -0.0049|
| 0.04   | 0.2309      | 0.4727      | 0.2313 | 0.4662 | -0.0004| 0.0066|
| Value | 0.045 | 0.2443 | 0.4625 | 0.2375 | 0.4612 | 0.0067 | 0.0014 |
|-------|-------|--------|--------|--------|--------|--------|--------|
| Value | 0.05  | 0.2551 | 0.4548 | 0.2582 | 0.465  | -0.0031| -0.0102|
| Value | 0.06  | 0.2714 | 0.4447 | 0.2743 | 0.4526 | -0.0029| -0.0079|
| Value | 0.07  | 0.2827 | 0.4394 | 0.2845 | 0.4504 | -0.0019| -0.011 |
| Value | 0.08  | 0.2908 | 0.4372 | 0.2915 | 0.4428 | -0.0007| -0.0056|
| Value | 0.09  | 0.2969 | 0.4369 | 0.3075 | 0.4435 | -0.0106| -0.0065|
| Value | 0.1   | 0.3018 | 0.4378 | 0.3051 | 0.4463 | -0.0033| -0.0085|
| Value | 0.15  | 0.3189 | 0.449  | 0.3272 | 0.4594 | -0.0083| -0.0104|
| Value | 0.2   | 0.3328 | 0.4626 | 0.3358 | 0.4668 | -0.003 | -0.0041|
| Value | 0.25  | 0.3461 | 0.4761 | 0.3399 | 0.4778 | 0.0062 | -0.0016|
| Value | 0.3   | 0.359  | 0.4893 | 0.3565 | 0.4988 | 0.0025 | -0.0095|
| Value | 0.35  | 0.3716 | 0.5022 | 0.3721 | 0.5047 | -0.0006| -0.0026|
| Value | 0.4   | 0.3838 | 0.5147 | 0.3856 | 0.5242 | -0.0018| -0.0095|
| Value | 0.45  | 0.3957 | 0.5269 | 0.3915 | 0.5238 | 0.0042 | 0.0031 |
| Value | 0.5   | 0.4073 | 0.5387 | 0.4066 | 0.54   | 0.0008 | -0.0013|
| Value | 0.6   | 0.4297 | 0.5615 | 0.4315 | 0.5615 | -0.0018| 0.0001 |
| Value | 0.7   | 0.4509 | 0.5832 | 0.4543 | 0.5862 | -0.0034| -0.003 |
| Value | 0.8   | 0.471  | 0.6037 | 0.4682 | 0.6037 | 0.0027 | 0.0001 |
| Value | 0.9   | 0.49   | 0.6232 | 0.4964 | 0.6245 | -0.0064| -0.0013|
| Value | 1     | 0.5081 | 0.6417 | 0.4995 | 0.6324 | 0.0086 | 0.0093 |
| Value | 1.5   | 0.5855 | 0.7208 | 0.583  | 0.7178 | 0.0025 | 0.003  |
| Value | 2     | 0.645  | 0.7815 | 0.6428 | 0.7769 | 0.0022 | 0.0046 |
| Value | 2.5   | 0.6907 | 0.8282 | 0.6879 | 0.8223 | 0.0028 | 0.0059 |
| Value | 3     | 0.7258 | 0.8641 | 0.7356 | 0.8631 | -0.0098| 0.001  |
| Value | 4     | 0.7735 | 0.9128 | 0.7758 | 0.9142 | -0.0023| -0.0014|
| Value | 6     | 0.8182 | 0.9585 | 0.8232 | 0.957  | -0.0049| 0.0015 |
| Value | 8     | 0.8338 | 0.9744 | 0.8292 | 0.972  | 0.0046 | 0.0025 |
| Value | 10    | 0.8393 | 0.98   | 0.8368 | 0.9761 | 0.0025 | 0.0039 |
| Value | 12    | 0.8412 | 0.9819 | 0.8472 | 0.9758 | -0.0061| 0.0061 |
| Value | 16    | 0.8421 | 0.9828 | 0.8491 | 0.9839 | -0.007 | -0.0011|
| Value | 20    | 0.8422 | 0.983  | 0.8485 | 0.9794 | -0.0063| 0.0036 |
Chart S-X.1. Plot of Inversion Recovery Data 1 h After TlOTf Addition.

CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0 = 0.8000 No. 1 = 0.2300

M(\text{inf})'s
No. 2 = 0.9000 No. 3 = 1.0000

M(0)−M(\text{inf})'s
No. 4 = -1.4000 No. 5 = 0.5000

and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000

Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 1.027168 +/- 0.142993
# 1 = 0.037824 +/- 0.139149
# 2 = 0.842176 +/- 0.001980
# 3 = 0.982968 +/- 0.002091
# 4 = -0.936670 +/- 0.002771
# 5 = -0.215392 +/- 0.003395
# 6 = 23.109811 +/- 0.361239
Data 4 d after addition of 2 equiv. of Tl(OTf):

Figure S-X.2. Stacked Spectra of Inversion Recovery Data 4 d After Tl(OTf) Addition.

Table S-X.2. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|------|------|
| 0.001 | 0.4471      | 0.7782      | 0.4416 | 0.7739 | 0.0054 | 0.0043 |
| 0.002 | 0.452       | 0.7733      | 0.4498 | 0.7713 | 0.0022 | 0.002 |
| 0.003 | 0.4567      | 0.7687      | 0.454  | 0.7677 | 0.0027 | 0.0009 |
| 0.004 | 0.4612      | 0.7642      | 0.46   | 0.7628 | 0.0012 | 0.0015 |
| 0.005 | 0.4655      | 0.7611      | 0.4681 | 0.7611 | -0.0026 | -0.0011 |
| 0.006 | 0.4696      | 0.756       | 0.4669 | 0.754  | 0.0027 | 0.002 |
| 0.007 | 0.4735      | 0.7522      | 0.4776 | 0.7539 | -0.0041 | -0.0017 |
| 0.008 | 0.4773      | 0.7486      | 0.4778 | 0.75   | -0.0005 | -0.0013 |
| 0.009 | 0.4808      | 0.7452      | 0.4823 | 0.7465 | -0.0015 | -0.0013 |
| 0.01  | 0.4842      | 0.7419      | 0.489  | 0.7449 | -0.0048 | -0.003 |
| 0.015 | 0.4992      | 0.7277      | 0.4975 | 0.727  | 0.0017 | 0.0007 |
| 0.02  | 0.5111      | 0.7167      | 0.5031 | 0.7108 | 0.0079 | 0.0059 |
| 0.025 | 0.5206      | 0.7082      | 0.5201 | 0.7069 | 0.0006 | 0.0013 |
| 0.03  | 0.5283      | 0.7016      | 0.5293 | 0.7005 | -0.001 | 0.0011 |
| 0.035 | 0.5345      | 0.6966      | 0.5333 | 0.6948 | 0.0012 | 0.0018 |
| 0.04  | 0.5395      | 0.6928      | 0.539  | 0.6933 | 0.0006 | -0.0005 |
| 0.045 | 0.5437      | 0.6899      | 0.5437 | 0.6898 | -0.0001 | 0.0001 |
| 0.05  | 0.5471      | 0.6878      | 0.5407 | 0.6848 | 0.0064 | 0.003 |
|   | Value1 | Value2 | Value3 | Value4 | Value5 | Value6 | Value7 |
|---|--------|--------|--------|--------|--------|--------|--------|
| 0.06 | 0.5523 | 0.6852 | 0.5559 | 0.689 | -0.0036 | -0.0037 |
| 0.07 | 0.5561 | 0.6842 | 0.5553 | 0.6843 | 0.0008 | -0.0002 |
| 0.08 | 0.559 | 0.684 | 0.5597 | 0.6834 | -0.0008 | 0.0007 |
| 0.09 | 0.5613 | 0.6845 | 0.5625 | 0.6863 | -0.0012 | -0.0018 |
| 0.1 | 0.5633 | 0.6852 | 0.562 | 0.6839 | 0.0013 | 0.0013 |
| 0.15 | 0.5712 | 0.691 | 0.572 | 0.6924 | -0.0009 | -0.0014 |
| 0.2 | 0.578 | 0.6975 | 0.5806 | 0.6999 | -0.0026 | -0.0025 |
| 0.25 | 0.5847 | 0.7038 | 0.588 | 0.7065 | -0.0033 | -0.0027 |
| 0.3 | 0.5911 | 0.7099 | 0.5907 | 0.7101 | 0.0004 | -0.0002 |
| 0.35 | 0.5974 | 0.7159 | 0.601 | 0.7183 | -0.0036 | -0.0024 |
| 0.4 | 0.6035 | 0.7217 | 0.6047 | 0.7231 | -0.0012 | -0.0014 |
| 0.45 | 0.6094 | 0.7274 | 0.6103 | 0.7286 | -0.0009 | -0.0013 |
| 0.5 | 0.6152 | 0.7328 | 0.6157 | 0.7351 | -0.0005 | -0.0022 |
| 0.6 | 0.6262 | 0.7434 | 0.6273 | 0.7446 | -0.001 | -0.0012 |
| 0.7 | 0.6367 | 0.7533 | 0.6383 | 0.7564 | -0.0016 | -0.003 |
| 0.8 | 0.6466 | 0.7628 | 0.6512 | 0.7673 | -0.0046 | -0.0046 |
| 0.9 | 0.6559 | 0.7717 | 0.6584 | 0.7734 | -0.0025 | -0.0018 |
| 1 | 0.6647 | 0.7801 | 0.6626 | 0.7792 | 0.0022 | 0.0009 |
| 1.5 | 0.7022 | 0.8158 | 0.6983 | 0.8127 | 0.0039 | 0.003 |
| 2 | 0.7304 | 0.8427 | 0.7244 | 0.8393 | 0.0061 | 0.0034 |
| 2.5 | 0.7518 | 0.8631 | 0.7494 | 0.8617 | 0.0023 | 0.0014 |
| 3 | 0.7679 | 0.8784 | 0.7678 | 0.8775 | 0.0001 | 0.0009 |
| 4 | 0.7893 | 0.8988 | 0.7891 | 0.8994 | 0.0002 | -0.0006 |
| 6 | 0.8085 | 0.9171 | 0.8067 | 0.9138 | 0.0018 | 0.0033 |
| 8 | 0.8147 | 0.9231 | 0.8123 | 0.9201 | 0.0024 | 0.0029 |
| 10 | 0.8167 | 0.925 | 0.8207 | 0.9264 | -0.004 | -0.0014 |
| 12 | 0.8174 | 0.9256 | 0.8186 | 0.9246 | -0.0012 | 0.001 |
| 16 | 0.8177 | 0.9259 | 0.8226 | 0.9274 | -0.0049 | -0.0015 |
| 20 | 0.8177 | 0.9259 | 0.8189 | 0.9267 | -0.0012 | -0.0008 |
**Chart S-X.2. Plot of Inversion Recovery Data 4 d After Ti(OTf) Addition.**

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

1/T1's
- No. 0 = 0.8000
- No. 1 = 0.2300

M(\text{inf})'s
- No. 2 = 0.9000
- No. 3 = 1.0000

M(0)–M(\text{inf})'s
- No. 4 = -1.4000
- No. 5 = 0.5000

and M(0)'s for reference
- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #   | Value      | Uncertainty  |
|-----|------------|--------------|
| 0   | -0.538128  | 0.177407     |
| 1   | 1.714098   | 0.196291     |
| 2   | 0.817723   | 0.001004     |
| 3   | 0.925923   | 0.001027     |
| 4   | -0.375819  | 0.001400     |
| 5   | -0.142635  | 0.001666     |
| 6   | 23.527460  | 0.617042     |
XI. Inversion Recovery Data for Section VII (Rotation Eyring)

1) Rotation Eyring Data at -61.4 °C:

![Stacked Spectra of Inversion Recovery Data at -61.4 °C.]

**Figure S–XI.1.** Stacked Spectra of Inversion Recovery Data at -61.4 °C.

**Table S–XI.1.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|-----|-----|
| 0.001 | -0.2647     | 0.921       | -0.2866| 0.9234 | 0.0219 | -0.0024 |
| 0.002 | -0.2624     | 0.9191      | -0.2727| 0.9201 | 0.0103 | -0.001  |
| 0.003 | -0.2601     | 0.9171      | -0.2707| 0.9179 | 0.0106 | -0.0008 |
| 0.004 | -0.2579     | 0.9152      | -0.268 | 0.9235 | 0.0101 | -0.0083 |
| 0.005 | -0.2556     | 0.9134      | -0.2554| 0.9146 | -0.0002| -0.0012 |
| 0.006 | -0.2534     | 0.9115      | -0.2566| 0.9154 | 0.0032 | -0.0039 |
| 0.007 | -0.2511     | 0.9096      | -0.2536| 0.9068 | 0.0025 | 0.0028  |
| 0.008 | -0.2489     | 0.9077      | -0.2539| 0.9077 | 0.005  | 0.0001  |
| 0.009 | -0.2467     | 0.9059      | -0.2441| 0.9052 | -0.0025| 0.0007  |
| 0.01  | -0.2444     | 0.904       | -0.2457| 0.9047 | 0.0012 | -0.0007 |
| 0.015 | -0.2335     | 0.8949      | -0.2342| 0.8863 | 0.0007 | 0.0086  |
| 0.02  | -0.2228     | 0.8861      | -0.2189| 0.8831 | -0.0038| 0.003   |
| 0.025 | -0.2122     | 0.8774      | -0.2061| 0.8728 | -0.0061| 0.0046  |
| 0.03  | -0.2019     | 0.869       | -0.2021| 0.8612 | 0.0002 | 0.0078  |
| 0.035 | -0.1917     | 0.8607      | -0.1848| 0.8628 | -0.0069| -0.0021 |
| 0.04  | -0.1818     | 0.8527      | -0.1756| 0.8566 | -0.0062| -0.0039 |
| 0.045 | -0.172 | 0.8449 | -0.1655 | 0.8406 | -0.0065 | 0.0043 |
| 0.05  | -0.1625 | 0.8373 | -0.1547 | 0.8331 | -0.0077 | 0.0041 |
| 0.06  | -0.1438 | 0.8226 | -0.1363 | 0.8181 | -0.0075 | 0.0044 |
| 0.07  | -0.1259 | 0.8086 | -0.1225 | 0.8027 | -0.0034 | 0.0059 |
| 0.08  | -0.1087 | 0.7953 | -0.1019 | 0.7945 | -0.0068 | 0.0008 |
| 0.09  | -0.092 | 0.7827 | -0.0866 | 0.7834 | -0.0055 | -0.0007 |
| 0.1   | -0.076 | 0.7708 | -0.0665 | 0.77 | -0.0096 | 0.0008 |
| 0.15  | -0.0042 | 0.7196 | -0.0038 | 0.7192 | -0.0004 | 0.0003 |
| 0.2   | 0.0558 | 0.6806 | 0.0561 | 0.6842 | -0.0003 | -0.0036 |
| 0.25  | 0.1063 | 0.6514 | 0.1085 | 0.6601 | -0.0022 | -0.0087 |
| 0.3   | 0.1491 | 0.63 | 0.1489 | 0.6367 | 0.0002 | -0.0067 |
| 0.35  | 0.1856 | 0.6148 | 0.1842 | 0.6206 | 0.0014 | -0.0058 |
| 0.4   | 0.217 | 0.6045 | 0.2168 | 0.6091 | 0.0003 | -0.0046 |
| 0.45  | 0.2443 | 0.5981 | 0.2404 | 0.6053 | 0.0039 | -0.0071 |
| 0.5   | 0.2682 | 0.5948 | 0.2618 | 0.6001 | 0.0065 | -0.0053 |
| 0.6   | 0.3083 | 0.5949 | 0.3084 | 0.5955 | -0.0002 | -0.0006 |
| 0.7   | 0.3407 | 0.6011 | 0.3402 | 0.6039 | 0.0005 | -0.0028 |
| 0.8   | 0.3679 | 0.6108 | 0.3663 | 0.6112 | 0.0017 | -0.0005 |
| 0.9   | 0.3915 | 0.6224 | 0.386 | 0.6208 | 0.0055 | 0.0017 |
| 1     | 0.4123 | 0.6351 | 0.4111 | 0.6334 | 0.0012 | 0.0017 |
| 1.5   | 0.4927 | 0.6981 | 0.4905 | 0.6896 | 0.0022 | 0.0085 |
| 2     | 0.5507 | 0.7497 | 0.5518 | 0.7468 | -0.0011 | 0.0029 |
| 2.5   | 0.5946 | 0.7895 | 0.5936 | 0.789 | 0.001 | 0.0004 |
| 3     | 0.628 | 0.8198 | 0.6334 | 0.8168 | -0.0054 | 0.0031 |
| 4     | 0.6728 | 0.8606 | 0.673 | 0.8621 | -0.0002 | -0.0015 |
| 6     | 0.714 | 0.898 | 0.7183 | 0.9016 | -0.0043 | -0.0036 |
| 8     | 0.7279 | 0.9107 | 0.7289 | 0.9095 | -0.0009 | 0.0012 |
| 10    | 0.7326 | 0.9149 | 0.7312 | 0.9122 | 0.0014 | 0.0027 |
| 12    | 0.7342 | 0.9164 | 0.7351 | 0.919 | -0.0009 | -0.0026 |
| 16    | 0.7349 | 0.917 | 0.7358 | 0.9179 | -0.0009 | -0.0009 |
| 20    | 0.735 | 0.9171 | 0.7369 | 0.9082 | -0.0019 | 0.0088 |
Chart S-XI.1. Plot of Inversion Recovery Data at -61.4 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:
1/T1's
No. 0= 0.8000 No. 1= 0.2300
M(inf)'s
No. 2= 0.9000 No. 3= 1.0000
M(0)-M(inf)'s
No. 4= -1.4000 No. 5= 0.5000
and M(0)'s for reference
No. 4= -0.5000 No. 5= 1.5000
Rates
No. 6= 20.0000

Final Values of Fitted Parameters and Uncertainties:
# 0 = 0.368664 +/- 0.016620
# 1 = 0.734924 +/- 0.029447
# 2 = 0.735023 +/- 0.001894
# 3 = 0.917094 +/- 0.002057
# 4 = -1.002029 +/- 0.002121
# 5 = 0.005814 +/- 0.002590
# 6 = 1.915320 +/- 0.023656
2) **Rotation Eyring Data at -53.7 °C:**

![Figure S-XI.2. Stacked Spectra of Inversion Recovery Data at -53.7 °C.](image)

**Table S-XI.2. CiFiT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|------|-------------|-------------|--------|--------|-----|-----|
| 0.001 | -0.4474     | 0.977       | -0.4553 | 0.9798 | 0.0079 | -0.0028 |
| 0.002 | -0.4394     | 0.9696      | -0.4495 | 0.9712 | 0.01  | -0.0017 |
| 0.003 | -0.4316     | 0.9622      | -0.4409 | 0.9651 | 0.0094 | -0.003 |
| 0.004 | -0.4238     | 0.9549      | -0.4292 | 0.9544 | 0.0054 | 0.0005 |
| 0.005 | -0.4161     | 0.9477      | -0.4136 | 0.9504 | -0.0025 | -0.0027 |
| 0.006 | -0.4085     | 0.9406      | -0.4063 | 0.94   | -0.0022 | 0.0005 |
| 0.007 | -0.401      | 0.9336      | -0.4006 | 0.9343 | -0.0004 | -0.0007 |
| 0.008 | -0.3936     | 0.9267      | -0.3915 | 0.924  | -0.0021 | 0.0026 |
| 0.009 | -0.3863     | 0.9198      | -0.3855 | 0.9129 | -0.0008 | 0.0069 |
| 0.01  | -0.379      | 0.9131      | -0.3738 | 0.9066 | -0.0052 | 0.0065 |
| 0.015 | -0.3441     | 0.8807      | -0.3418 | 0.8824 | -0.0023 | -0.0017 |
| 0.02  | -0.3112     | 0.8504      | -0.309  | 0.8489 | -0.0021 | 0.0015 |
| 0.025 | -0.2802     | 0.8221      | -0.2758 | 0.8173 | -0.0043 | 0.0048 |
| 0.03  | -0.2509     | 0.7955      | -0.2428 | 0.7964 | -0.0082 | -0.0008 |
| 0.035 | -0.2234     | 0.7707      | -0.2198 | 0.7703 | -0.0036 | 0.0004 |
| 0.04  | -0.1974     | 0.7475      | -0.1934 | 0.7528 | -0.004  | -0.0053 |
| 0.045 | -0.1729     | 0.7259      | -0.1702 | 0.7272 | -0.0027 | -0.0014 |
| 0.05  | -0.1498     | 0.7056      | -0.1466 | 0.7043 | -0.0031 | 0.0013 |
| x  | z   | y   | z   | y   | z   | y   | z   | y   | z   |
|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| 0.06| -0.1074| 0.669| -0.1026| 0.6708| -0.0048| -0.0017 |
| 0.07| -0.0696| 0.6372| -0.066| 0.6383| -0.0036| -0.0011 |
| 0.08| -0.0358| 0.6094| -0.0392| 0.6126| 0.0034| -0.0031 |
| 0.09| -0.0057| 0.5854| -0.0038| 0.5903| -0.0019| -0.0049 |
| 0.1 | 0.0213| 0.5646| 0.0233| 0.5725| -0.002| -0.0079 |
| 0.15| 0.1204| 0.4969| 0.1203| 0.5046| 0.0001| -0.0077 |
| 0.2 | 0.1809| 0.4682| 0.1738| 0.4752| 0.0071| -0.007 |
| 0.25| 0.2206| 0.46| 0.2206| 0.4606| -0.0001| -0.0006 |
| 0.3 | 0.2489| 0.4627| 0.2518| 0.4646| -0.0029| -0.0019 |
| 0.35| 0.271| 0.4708| 0.2696| 0.47| 0.0015| 0.0008 |
| 0.4 | 0.2896| 0.4818| 0.2844| 0.4808| 0.0052| 0.001 |
| 0.45| 0.3062| 0.4941| 0.3053| 0.4868| 0.0009| 0.0072 |
| 0.5 | 0.3214| 0.5068| 0.3173| 0.4976| 0.0042| 0.0092 |
| 0.6 | 0.3496| 0.5325| 0.3455| 0.5211| 0.0041| 0.0114 |
| 0.7 | 0.3757| 0.5573| 0.3716| 0.5526| 0.0041| 0.0047 |
| 0.8 | 0.4003| 0.5809| 0.3989| 0.5764| 0.0014| 0.0045 |
| 0.9 | 0.4235| 0.6033| 0.4253| 0.6004| -0.0018| 0.0029 |
| 1  | 0.4454| 0.6245| 0.4405| 0.6184| 0.0049| 0.0062 |
| 1.5| 0.5383| 0.7146| 0.5386| 0.7095| -0.0002| 0.0051 |
| 2  | 0.6087| 0.7827| 0.6166| 0.7841| -0.0079| -0.0013 |
| 2.5 | 0.662| 0.8344| 0.6644| 0.838| -0.0024| -0.0036 |
| 3  | 0.7024| 0.8735| 0.7142| 0.8795| -0.0118| -0.006 |
| 4  | 0.7561| 0.9255| 0.7606| 0.9384| -0.0045| -0.013 |
| 6  | 0.8046| 0.9724| 0.8039| 0.9772| 0.0006| -0.0048 |
| 8  | 0.8205| 0.9878| 0.8238| 0.9899| -0.0033| -0.0021 |
| 10 | 0.8257| 0.9929| 0.8252| 0.997| 0.0006| -0.0041 |
| 12 | 0.8275| 0.9946| 0.8234| 0.9922| 0.004| 0.0024 |
| 16 | 0.8282| 0.9953| 0.8194| 0.9879| 0.0088| 0.0074 |
| 20 | 0.8283| 0.9954| 0.8211| 0.9924| 0.0072| 0.003 |
**Chart S-XI.2.** Plot of Inversion Recovery Data at –53.7 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's  
No. 0= 0.8000 No. 1= 0.2300

M(Inf)'s  
No. 2= 0.9000 No. 3= 1.0000

M(0)–M(Inf)'s  
No. 4= -1.4000 No. 5= 0.5000

and M(0)'s for reference  
No. 4= -0.5000 No. 5= 1.5000

Rates  
No. 6= 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value   | Uncertainty |
|----|---------|-------------|
| 0  | 0.366672| 0.029155    |
| 1  | 0.751950| 0.035598    |
| 2  | 0.828296| 0.001760    |
| 3  | 0.995370| 0.001903    |
| 4  | -1.283774| 0.002117   |
| 5  | -0.010744| 0.002631    |
| 6  | 6.003374| 0.044677    |
3) **Rotation Eyring Data at -42.0 °C:**

![Figure S-XI.3: Stacked Spectra of Inversion Recovery Data at -42.0 °C.](image)

**Table S-XI.3. CIFIT Plot File.**

| d2  | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-----|-------------|-------------|--------|--------|-----|-----|
| 0.001 | -0.1303 | 0.8252 | -0.1429 | 0.8224 | 0.0127 | 0.0028 |
| 0.002 | -0.1113 | 0.8065 | -0.1191 | 0.7996 | 0.0078 | 0.0068 |
| 0.003 | -0.0931 | 0.7886 | -0.0965 | 0.7873 | 0.0034 | 0.0013 |
| 0.004 | -0.0757 | 0.7716 | -0.0795 | 0.7673 | 0.0038 | 0.0043 |
| 0.005 | -0.0591 | 0.7554 | -0.0607 | 0.7526 | 0.0016 | 0.0028 |
| 0.006 | -0.0433 | 0.7399 | -0.045 | 0.7339 | 0.0017 | 0.006  |
| 0.007 | -0.0282 | 0.7251 | -0.0333 | 0.7207 | 0.0051 | 0.0045 |
| 0.008 | -0.0138 | 0.7111 | -0.0137 | 0.7107 | -0.0001 | 0.0004 |
| 0.009 | 0 | 0.6977 | 0.0003 | 0.6924 | -0.0002 | 0.0053 |
| 0.01 | 0.0132 | 0.6849 | 0.0131 | 0.68 | 0.0001 | 0.0049 |
| 0.015 | 0.0707 | 0.6295 | 0.0749 | 0.6285 | -0.0042 | 0.001  |
| 0.02 | 0.1164 | 0.586 | 0.1176 | 0.5827 | -0.0012 | 0.0033 |
| 0.025 | 0.1529 | 0.5519 | 0.1515 | 0.5584 | 0.0014 | -0.0065 |
| 0.03 | 0.182 | 0.5253 | 0.1894 | 0.5264 | -0.0074 | -0.0011 |
| 0.035 | 0.2054 | 0.5045 | 0.2068 | 0.5108 | -0.0014 | -0.0063 |
| 0.04 | 0.2241 | 0.4884 | 0.2296 | 0.4978 | -0.0055 | -0.0094 |
| 0.045 | 0.2393 | 0.4759 | 0.2424 | 0.4828 | -0.0031 | -0.0069 |
| 0.05 | 0.2515 | 0.4664 | 0.256 | 0.472 | -0.0045 | -0.0056 |
| x   | y   | x   | y   | y   | x   |
|-----|-----|-----|-----|-----|-----|
| 0.06| 0.2697 | 0.4537 | 0.271 | 0.4611 | -0.0013 | -0.0074 |
| 0.07| 0.2822 | 0.4468 | 0.2817 | 0.4504 | 0.0005 | -0.0035 |
| 0.08| 0.291 | 0.4436 | 0.29 | 0.4465 | 0.001 | -0.003 |
| 0.09| 0.2976 | 0.4426 | 0.2997 | 0.4458 | -0.0021 | -0.0033 |
| 0.1 | 0.3027 | 0.443 | 0.3017 | 0.4462 | 0.001 | -0.0033 |
| 0.15| 0.3202 | 0.4531 | 0.3215 | 0.4596 | -0.0014 | -0.0065 |
| 0.2 | 0.334 | 0.4661 | 0.3315 | 0.4693 | 0.0026 | -0.0032 |
| 0.25| 0.3472 | 0.479 | 0.351 | 0.4848 | -0.0037 | -0.0057 |
| 0.3 | 0.3601 | 0.4917 | 0.3579 | 0.4925 | 0.0022 | -0.0009 |
| 0.35| 0.3726 | 0.504 | 0.3729 | 0.5069 | -0.0004 | -0.0029 |
| 0.4 | 0.3847 | 0.516 | 0.3851 | 0.5165 | -0.0004 | -0.0005 |
| 0.45| 0.3966 | 0.5276 | 0.3956 | 0.5274 | 0.001 | 0.0002 |
| 0.5 | 0.4081 | 0.539 | 0.4038 | 0.5395 | 0.0043 | -0.0005 |
| 0.6 | 0.4303 | 0.5608 | 0.428 | 0.5624 | 0.0022 | -0.0016 |
| 0.7 | 0.4513 | 0.5815 | 0.4509 | 0.5786 | 0.0003 | 0.0029 |
| 0.8 | 0.4712 | 0.6011 | 0.4656 | 0.5956 | 0.0056 | 0.0055 |
| 0.9 | 0.4901 | 0.6197 | 0.4871 | 0.6165 | 0.003 | 0.0032 |
| 1 | 0.508 | 0.6374 | 0.505 | 0.6327 | 0.003 | 0.0047 |
| 1.5 | 0.5844 | 0.7127 | 0.5834 | 0.7093 | 0.001 | 0.0034 |
| 2 | 0.643 | 0.7704 | 0.6432 | 0.7762 | -0.0002 | -0.0058 |
| 2.5 | 0.6878 | 0.8145 | 0.689 | 0.8127 | -0.0012 | 0.0019 |
| 3 | 0.7221 | 0.8484 | 0.7266 | 0.8449 | -0.0045 | 0.0034 |
| 4 | 0.7685 | 0.8941 | 0.7753 | 0.9002 | -0.0068 | -0.0061 |
| 6 | 0.8117 | 0.9366 | 0.8202 | 0.9355 | -0.0085 | 0.0011 |
| 8 | 0.8265 | 0.9512 | 0.8328 | 0.9509 | -0.0063 | 0.0003 |
| 10 | 0.8316 | 0.9562 | 0.8353 | 0.952 | -0.0037 | 0.0042 |
| 12 | 0.8334 | 0.958 | 0.837 | 0.9543 | -0.0036 | 0.0037 |
| 16 | 0.8342 | 0.9588 | 0.8335 | 0.9509 | 0.0007 | 0.0079 |
| 20 | 0.8343 | 0.9589 | 0.8286 | 0.9548 | 0.0057 | 0.0041 |
Chart S-XI.3. Plot of Inversion Recovery Data at -42.0 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:

- 1/T1's
  No. 0= 0.8000 No. 1= 0.2300

- M(\text{inf})'s
  No. 2= 0.9000 No. 3= 1.0000

- M(0)-M(\text{inf})'s
  No. 4= -1.4000 No. 5= 0.5000
  and M(0)'s for reference
  No. 4= -0.5000 No. 5= 1.5000

- Rates
  No. 6= 20.0000

Final Values of Fitted Parameters and Uncertainties:

- No. 0 = 0.190781 +/- 0.113610
- No. 1 = 0.882260 +/- 0.119240
- No. 2 = 0.834290 +/- 0.001591
- No. 3 = 0.958876 +/- 0.001670
- No. 4 = -0.984521 +/- 0.002220
- No. 5 = -0.114023 +/- 0.002792
- No. 6 = 23.226432 +/- 0.247924
4) **Rotation Eyring Data at -31.8 °C:**

![Figure S-XI.4. Stacked Spectra of Inversion Recovery Data at -31.8 °C.](image)

**Table S-XI.4. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | 0.111       | 0.4656      | 0.1221 | 0.4743 | -0.0111 | -0.0087 |
| 0.002  | 0.1409      | 0.4335      | 0.1504 | 0.444  | -0.0095 | -0.0105 |
| 0.003  | 0.1663      | 0.4063      | 0.1759 | 0.4165 | -0.0096 | -0.0103 |
| 0.004  | 0.1879      | 0.3832      | 0.2011 | 0.3954 | -0.0133 | -0.0122 |
| 0.005  | 0.2062      | 0.3637      | 0.2223 | 0.3758 | -0.0161 | -0.012 |
| 0.006  | 0.2219      | 0.3472      | 0.2335 | 0.3602 | -0.0116 | -0.013 |
| 0.007  | 0.2352      | 0.3333      | 0.2505 | 0.3461 | -0.0153 | -0.0128 |
| 0.008  | 0.2465      | 0.3215      | 0.2573 | 0.3302 | -0.0108 | -0.0087 |
| 0.009  | 0.2562      | 0.3116      | 0.2655 | 0.3197 | -0.0093 | -0.0081 |
| 0.01   | 0.2645      | 0.3032      | 0.2721 | 0.3073 | -0.0076 | -0.0041 |
| 0.015  | 0.2911      | 0.2776      | 0.2996 | 0.2867 | -0.0085 | -0.0092 |
| 0.02   | 0.3038      | 0.2673      | 0.3104 | 0.2749 | -0.0066 | -0.0076 |
| 0.025  | 0.3103      | 0.2636      | 0.3145 | 0.272  | -0.0041 | -0.0084 |
| 0.03   | 0.3142      | 0.263      | 0.3156 | 0.2676 | -0.0013 | -0.0046 |
| 0.035  | 0.317       | 0.2636      | 0.3196 | 0.2696 | -0.0026 | -0.006 |
| 0.04   | 0.3191      | 0.2648      | 0.3185 | 0.27   | 0.0007  | 0.0053 |
| 0.045  | 0.3211      | 0.2662      | 0.3143 | 0.2627 | 0.0068  | 0.0035 |
| 0.05   | 0.323       | 0.2677      | 0.3198 | 0.2643 | 0.0032  | 0.0034 |
| $r$  | $x$     | $y$     | $z$     | $w$     | $u$     | $v$     |
|------|---------|---------|---------|---------|---------|---------|
| 0.06 | 0.3266  | 0.2709  | 0.3218  | 0.2681  | 0.0048  | 0.0028  |
| 0.07 | 0.3301  | 0.2741  | 0.3235  | 0.2662  | 0.0066  | 0.0079  |
| 0.08 | 0.3336  | 0.2773  | 0.3281  | 0.2695  | 0.0056  | 0.0077  |
| 0.09 | 0.3371  | 0.2805  | 0.328   | 0.2683  | 0.0092  | 0.0122  |
| 0.1  | 0.3406  | 0.2836  | 0.3319  | 0.2721  | 0.0087  | 0.0115  |
| 0.15 | 0.3577  | 0.2992  | 0.3426  | 0.2827  | 0.0152  | 0.0165  |
| 0.2  | 0.3744  | 0.3143  | 0.3638  | 0.299   | 0.0106  | 0.0153  |
| 0.25 | 0.3907  | 0.3291  | 0.3795  | 0.3144  | 0.0111  | 0.0146  |
| 0.3  | 0.4065  | 0.3434  | 0.3926  | 0.3279  | 0.0139  | 0.0155  |
| 0.35 | 0.4219  | 0.3574  | 0.4108  | 0.3417  | 0.0112  | 0.0158  |
| 0.4  | 0.4369  | 0.3711  | 0.4276  | 0.3568  | 0.0093  | 0.0143  |
| 0.45 | 0.4516  | 0.3844  | 0.4432  | 0.3695  | 0.0084  | 0.0149  |
| 0.5  | 0.4658  | 0.3973  | 0.4544  | 0.381   | 0.0114  | 0.0163  |
| 0.6  | 0.4932  | 0.4222  | 0.4857  | 0.4085  | 0.0075  | 0.0137  |
| 0.7  | 0.5193  | 0.4458  | 0.515   | 0.4366  | 0.0043  | 0.0092  |
| 0.8  | 0.5439  | 0.4682  | 0.5399  | 0.4562  | 0.004   | 0.012   |
| 0.9  | 0.5674  | 0.4895  | 0.5633  | 0.4846  | 0.0041  | 0.0049  |
| 1    | 0.5896  | 0.5097  | 0.5869  | 0.5054  | 0.0027  | 0.0043  |
| 1.5  | 0.6849  | 0.5962  | 0.6914  | 0.6024  | -0.0065 | -0.0063 |
| 2    | 0.7582  | 0.6627  | 0.7703  | 0.678   | -0.0121 | -0.0153 |
| 2.5  | 0.8146  | 0.7139  | 0.8329  | 0.7364  | -0.0184 | -0.0224 |
| 3    | 0.858   | 0.7534  | 0.8738  | 0.7755  | -0.0158 | -0.0222 |
| 4    | 0.9171  | 0.807   | 0.9285  | 0.8232  | -0.0115 | -0.0162 |
| 6    | 0.9728  | 0.8576  | 0.9727  | 0.8675  | 0.0002  | -0.0098 |
| 8    | 0.9924  | 0.8754  | 0.9861  | 0.8775  | 0.0063  | -0.0021 |
| 12   | 1.0016  | 0.8838  | 0.9893  | 0.877   | 0.0123  | 0.0067  |
| 16   | 1.0028  | 0.8848  | 0.9884  | 0.878   | 0.0144  | 0.0068  |
| 20   | 1.0029  | 0.885   | 0.9933  | 0.879   | 0.0096  | 0.0059  |
Chart S-XI.4. Plot of Inversion Recovery Data at $-31.8 \, ^\circ\text{C}$.

**CIFIT Guesses and Fit Parameters**

**Initial values of parameters:**

1/T1's
- No. 0 = 0.8000
- No. 1 = 0.2300

M(inf)'s
- No. 2 = 0.9000
- No. 3 = 1.0000

M(0)–M(inf)'s
- No. 4 = -1.4000
- No. 5 = 0.5000

and M(0)'s for reference
- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

**Final Values of Fitted Parameters and Uncertainties:**

| #  | Value     | Uncertainty |
|----|-----------|-------------|
| 0  | -6.995422 | 1.016564    |
| 1  | 8.805540  | 1.205582    |
| 2  | 1.002937  | 0.004595    |
| 3  | 0.884974  | 0.009043    |
| 4  | -0.927070 | 0.009043    |
| 5  | -0.381346 | 0.010430    |
| 6  | 81.684411 | 4.608239    |
XII. Inversion Recovery Data for Section VIII (Ring Flip Eyring).

1) Ring Flip Eyring Data in CD$_2$Cl$_2$ at 13.7 °C:

![Stacked Spectra of Inversion Recovery Data at 13.7 °C in CD$_2$Cl$_2$.](image)

**Figure S–XII.1.** Stacked Spectra of Inversion Recovery Data at 13.7 °C in CD$_2$Cl$_2$.

**Table S–XII.1.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.4817     | 0.79        | -0.485 | 0.7887 | 0.0033 | 0.0012 |
| 0.002  | -0.4755     | 0.7863      | -0.4795| 0.7847 | 0.004  | 0.0015 |
| 0.003  | -0.4693     | 0.7826      | -0.4738| 0.7804 | 0.0045 | 0.0022 |
| 0.004  | -0.4631     | 0.779       | -0.4678| 0.7775 | 0.0046 | 0.0014 |
| 0.005  | -0.457      | 0.7754      | -0.4633| 0.7731 | 0.0063 | 0.0023 |
| 0.007  | -0.4449     | 0.7683      | -0.448 | 0.7668 | 0.0031 | 0.0015 |
| 0.01   | -0.427      | 0.758       | -0.4314| 0.7559 | 0.0045 | 0.0021 |
| 0.015  | -0.3978     | 0.7415      | -0.3991| 0.74   | 0.0013 | 0.0015 |
| 0.02   | -0.3695     | 0.7259      | -0.3708| 0.7252 | 0.0013 | 0.0007 |
| 0.025  | -0.342      | 0.7111      | -0.3412| 0.7105 | -0.0008| 0.0006 |
| 0.03   | -0.3153     | 0.6972      | -0.3119| 0.6971 | -0.0035| 0.0001 |
| 0.035  | -0.2894     | 0.684       | -0.2883| 0.6841 | -0.0011| -0.0001|
| 0.04   | -0.2642     | 0.6716      | -0.2637| 0.67   | -0.0006| 0.0015 |
| 0.045  | -0.2398     | 0.6598      | -0.2376| 0.661  | -0.0022| -0.0012|
| 0.055  | -0.1929     | 0.6383      | -0.2005| 0.6339 | 0.0076 | 0.0044 |
| 0.06   | -0.1704     | 0.6285      | -0.1685| 0.6291 | -0.0019| -0.0006|
| 0.065  | -0.1485     | 0.6193      | -0.147 | 0.6199 | -0.0015| -0.0006|
| 0.07   | -0.1273     | 0.6107      | -0.1283| 0.6104 | 0.001  | 0.0003 |
0.075 -0.1066  0.6026 -0.0988  0.6063 -0.0078 -0.0038
0.08  -0.0865  0.595  -0.079  0.5974 -0.0074 -0.0024
0.085 -0.0669  0.5879 -0.0616  0.59 -0.0053 -0.0022
0.09  -0.0478  0.5812 -0.0413  0.5839 -0.0065 -0.0026
0.095 -0.0292  0.5751 -0.0222  0.578 -0.0071 -0.003  
0.1   -0.0112  0.5693 -0.0067  0.5706 -0.0044 -0.0013
0.15  0.1462   0.5316  0.153   0.5366 -0.0068 -0.005
0.2   0.2695   0.5198  0.2777  0.5266 -0.0083 -0.0068
0.25  0.3679   0.5239  0.3691  0.5284 -0.0012 -0.0045
0.3   0.4479   0.5371  0.4448  0.5405  0.003   -0.0034
0.35  0.5139   0.5551  0.4977  0.551  0.0162   0.0041
0.4   0.5691   0.5754  0.5702  0.5802 -0.0011 -0.0048
0.45  0.6159   0.5961  0.6036  0.5945  0.0123   0.0017
0.5   0.6558   0.6164  0.6484  0.6167  0.0074 -0.0003
0.6   0.7199   0.6533  0.711   0.6516  0.009   0.0017
0.7   0.7685   0.6844  0.7716  0.6873 -0.0031 -0.0029
0.8   0.8057   0.7096  0.8112  0.7135 -0.0056 -0.0039
0.9   0.8344   0.7298  0.8281  0.7265  0.0063   0.0033
1     0.8568   0.7457  0.8535  0.7437  0.0033   0.002
1.5   0.913    0.7865  0.9107  0.7853  0.0023   0.0012
2     0.9294   0.7985  0.9381  0.8011 -0.0087 -0.0026
2.5   0.9342   0.802   0.9277  0.7968  0.0064   0.0052
3     0.9356   0.803   0.9261  0.7962  0.0095   0.0068
4     0.9361   0.8034  0.9489  0.8073 -0.0128 -0.004
5     0.9361   0.8034  0.9516  0.809  -0.0155 -0.0056
6     0.9361   0.8034  0.9332  0.8001  0.003   0.0033
7     0.9361   0.8034  0.9497  0.8091 -0.0136 -0.0057
8     0.9361   0.8034  0.9321  0.8002  0.004   0.0032
9     0.9361   0.8034  0.9484  0.8084 -0.0123 -0.0005
10    0.9361   0.8034  0.932   0.7999  0.0042   0.0035
12    0.9361   0.8034  0.9493  0.8088 -0.0132 -0.0054
14    0.9361   0.8034  0.9364  0.8017 -0.0002  0.0017
15    0.9361   0.8034  0.9123  0.7908  0.0238  0.0126
16    0.9361   0.8034  0.9406  0.8043 -0.0045 -0.0009
18    0.9361   0.8034  0.9327  0.8009  0.0035   0.0025
19    0.9361   0.8034  0.9248  0.7961  0.0113  0.0073
20    0.9361   0.8034  0.9465  0.8065 -0.0103 -0.0031
Chart S-XII.1. Plot of Inversion Recovery Data at 13.68 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:
- 1/T1's
  No. 0 = 0.8000 No. 1 = 0.2300
- M(inf)'s
  No. 2 = 0.9000 No. 3 = 1.0000
- M(0)-M(inf)'s
  No. 4 = -1.4000 No. 5 = 0.5000
- Rates
  No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
- # 0 = 1.742447 +/- 0.035463
- # 1 = 3.456408 +/- 0.098012
- # 2 = 0.936134 +/- 0.001407
- # 3 = 0.803401 +/- 0.001447
- # 4 = -1.424087 +/- 0.002192
- # 5 = -0.009672 +/- 0.002731
- # 6 = 2.688728 +/- 0.041685
2) Ring Flip Eyring Data in CD$_2$Cl$_2$ at 18.8 °C:

Figure S-XII.2. Stacked Spectra of Inversion Recovery Data at 18.8 °C.

Table S-XII.2. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.661      | 0.8814      | -0.659 | 0.8801 | -0.0019 | 0.0013 |
| 0.002  | -0.6513     | 0.8749      | -0.6503 | 0.8741 | -0.001 | 0.0008 |
| 0.003  | -0.6417     | 0.8685      | -0.6428 | 0.8682 | 0.0011 | 0.0003 |
| 0.004  | -0.6322     | 0.8622      | -0.6323 | 0.863  | 0.0001 | -0.0008 |
| 0.005  | -0.6228     | 0.856       | -0.6242 | 0.8554 | 0.0015 | 0.0006 |
| 0.007  | -0.6042     | 0.8438      | -0.6045 | 0.8439 | 0.0003 | -0.0001 |
| 0.01   | -0.5769     | 0.8262      | -0.5785 | 0.8258 | 0.0016 | 0.0004 |
| 0.015  | -0.5331     | 0.7984      | -0.5374 | 0.796  | 0.0043 | 0.0025 |
| 0.02   | -0.4911     | 0.7727      | -0.4916 | 0.773  | 0.0005 | -0.0004 |
| 0.025  | -0.451      | 0.7487      | -0.4517 | 0.7493 | 0.0007 | -0.0006 |
| 0.03   | -0.4126     | 0.7265      | -0.4128 | 0.7271 | 0.0002 | -0.0006 |
| 0.035  | -0.3758     | 0.706       | -0.3742 | 0.707  | -0.0016 | -0.001 |
| 0.04   | -0.3406     | 0.6869      | -0.3422 | 0.6857 | 0.0016 | 0.0012 |
| 0.045  | -0.3068     | 0.6694      | -0.3064 | 0.6703 | -0.0004 | -0.0009 |
| 0.055  | -0.2434     | 0.6383      | -0.2465 | 0.6355 | 0.0031 | 0.0027 |
| 0.06   | -0.2136     | 0.6246      | -0.2139 | 0.6237 | 0.0003 | 0.0009 |
| 0.065  | -0.1849     | 0.612       | -0.1825 | 0.6132 | -0.0024 | -0.0012 |
| 0.07   | -0.1575     | 0.6005      | -0.1558 | 0.601  | -0.0016 | -0.0005 |
| 0.075  | -0.1311     | 0.59        | -0.1321 | 0.5884 | 0.0011 | 0.0016 |
| 0.08   | -0.1057     | 0.5805      | -0.1019 | 0.5825 | -0.0038 | -0.002 |
|    |    |    |    |    |    |    |    |
|---|---|---|---|---|---|---|---|
| 0.085 | -0.0812 | 0.5718 | -0.0801 | 0.5723 | -0.0011 | -0.0005 |
| 0.09 | -0.0577 | 0.564 | -0.0567 | 0.565 | -0.0011 | -0.001 |
| 0.095 | -0.0351 | 0.557 | -0.0328 | 0.5583 | -0.0023 | -0.0014 |
| 0.1 | -0.0133 | 0.5507 | -0.0111 | 0.5521 | -0.0022 | -0.0014 |
| 0.15 | 0.1668 | 0.5196 | 0.1682 | 0.5213 | -0.0014 | -0.0017 |
| 0.2 | 0.2971 | 0.5258 | 0.2985 | 0.528 | -0.0014 | -0.0022 |
| 0.25 | 0.3959 | 0.5503 | 0.3918 | 0.5483 | 0.0041 | 0.002 |
| 0.3 | 0.4739 | 0.5827 | 0.4737 | 0.5835 | 0.0002 | -0.0008 |
| 0.35 | 0.5374 | 0.6174 | 0.5369 | 0.6188 | 0.0005 | -0.0014 |
| 0.4 | 0.5904 | 0.6515 | 0.5894 | 0.6516 | 0.001 | -0.0002 |
| 0.45 | 0.6356 | 0.6835 | 0.6316 | 0.6819 | 0.0039 | 0.0016 |
| 0.5 | 0.6744 | 0.7128 | 0.6741 | 0.7139 | 0.0003 | -0.001 |
| 0.6 | 0.7374 | 0.763 | 0.7365 | 0.7638 | 0.0009 | -0.0008 |
| 0.7 | 0.7857 | 0.8028 | 0.7855 | 0.8026 | 0.0002 | 0.0002 |
| 0.8 | 0.823 | 0.834 | 0.8222 | 0.8339 | 0.0008 | 0.0001 |
| 0.9 | 0.8519 | 0.8583 | 0.8512 | 0.858 | 0.0007 | 0.0003 |
| 1 | 0.8743 | 0.8772 | 0.875 | 0.8785 | -0.0007 | -0.0013 |
| 1.5 | 0.9303 | 0.9245 | 0.9302 | 0.9244 | 0.0001 | 0.0001 |
| 2 | 0.9462 | 0.9378 | 0.9443 | 0.9365 | 0.0018 | 0.0014 |
| 2.5 | 0.9506 | 0.9416 | 0.9517 | 0.9426 | -0.0011 | -0.001 |
| 3 | 0.9519 | 0.9427 | 0.9533 | 0.9426 | -0.0014 | 0.0001 |
| 4 | 0.9524 | 0.9431 | 0.9543 | 0.9446 | -0.002 | -0.0015 |
| 5 | 0.9524 | 0.9431 | 0.9498 | 0.9413 | 0.0026 | 0.0019 |
| 6 | 0.9524 | 0.9431 | 0.9524 | 0.9432 | 0 | -0.0001 |
| 7 | 0.9524 | 0.9431 | 0.9549 | 0.9437 | -0.0025 | -0.0005 |
| 8 | 0.9524 | 0.9431 | 0.9545 | 0.9434 | -0.0021 | -0.0003 |
| 9 | 0.9524 | 0.9431 | 0.9557 | 0.9444 | -0.0033 | -0.0013 |
| 10 | 0.9524 | 0.9431 | 0.9547 | 0.945 | -0.0023 | -0.0018 |
| 12 | 0.9524 | 0.9431 | 0.9548 | 0.9443 | -0.0024 | -0.0012 |
| 14 | 0.9524 | 0.9431 | 0.9435 | 0.9359 | 0.0089 | 0.0072 |
| 15 | 0.9524 | 0.9431 | 0.9506 | 0.9415 | 0.0018 | 0.0016 |
| 16 | 0.9524 | 0.9431 | 0.9526 | 0.9423 | -0.0002 | 0.0008 |
| 18 | 0.9524 | 0.9431 | 0.9547 | 0.9434 | -0.0023 | -0.0003 |
| 19 | 0.9524 | 0.9431 | 0.9534 | 0.9431 | -0.001 | 0 |
| 20 | 0.9524 | 0.9431 | 0.9531 | 0.9429 | -0.0007 | 0.0002 |
**Chart S-XII.2. Plot of Inversion Recovery Data at 18.8 °C.**

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

- 1/T1's
  - No. 0 = 0.8000
  - No. 1 = 0.2300

- M(inf)'s
  - No. 2 = 0.9000
  - No. 3 = 1.0000

- M(0)–M(inf)'s
  - No. 4 = -1.4000
  - No. 5 = 0.5000

and M(0)'s for reference

- No. 4 = -0.5000
- No. 5 = 1.5000

Rates
- No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

- No. 0 = 1.848960 +/− 0.012173
- No. 1 = 3.324287 +/− 0.024308
- No. 2 = 0.952404 +/− 0.000430
- No. 3 = 0.943127 +/− 0.000445
- No. 4 = -1.623167 +/− 0.000714
- No. 5 = -0.055117 +/− 0.000875
- No. 6 = 4.347268 +/− 0.015079
3) **Ring Flip Eyring Data at 28.1 °C in CD$_2$Cl$_2$:**

![Stacked Spectra of Inversion Recovery Data at 28.1 °C.](image)

**Figure S–XII.3.** Stacked Spectra of Inversion Recovery Data at 28.1 °C.

**Table S–XII.3.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|-----|-----|
| 0.001 | -0.4737     | 0.7392      | -0.4749| 0.7373 | 0.0013 | 0.0019 |
| 0.002 | -0.4584     | 0.7275      | -0.4603| 0.7247 | 0.002 | 0.0028 |
| 0.003 | -0.4434     | 0.716       | -0.4455| 0.715  | 0.0021 | 0.001 |
| 0.004 | -0.4287     | 0.7048      | -0.4311| 0.7036 | 0.0025 | 0.0013 |
| 0.005 | -0.4142     | 0.694       | -0.4178| 0.692  | 0.0035 | 0.002 |
| 0.007 | -0.3863     | 0.6731      | -0.3873| 0.6731 | 0.0011 | 0 |
| 0.01  | -0.3463     | 0.6438      | -0.347 | 0.643  | 0.0007 | 0.0009 |
| 0.015 | -0.2847     | 0.6001      | -0.2835| 0.5999 | -0.0012 | 0.0002 |
| 0.02  | -0.2287     | 0.5622      | -0.2295| 0.5602 | 0.0007 | 0.0019 |
| 0.025 | -0.1778     | 0.5293      | -0.1786| 0.5301 | 0.0008 | -0.0008 |
| 0.03  | -0.1314     | 0.5009      | -0.133 | 0.4996 | 0.0016 | 0.0013 |
| 0.035 | -0.0891     | 0.4765      | -0.0877| 0.4771 | -0.0014 | -0.0006 |
| 0.04  | -0.0504     | 0.4557      | -0.0481| 0.4554 | -0.0023 | 0.0003 |
| 0.045 | -0.0149     | 0.438       | -0.0132| 0.4386 | -0.0018 | -0.0006 |
| 0.055 | 0.0476      | 0.4108      | 0.0486 | 0.4107 | -0.001 | 0.0002 |
| 0.06  | 0.0752      | 0.4007      | 0.0768 | 0.4008 | -0.0015 | -0.0001 |
| 0.065 | 0.1008      | 0.3925      | 0.1042 | 0.3952 | -0.0034 | -0.0027 |
| 0.07  | 0.1244      | 0.386       | 0.1259 | 0.3878 | -0.0014 | -0.0018 |
| 0.075 | 0.1464      | 0.3811      | 0.1478 | 0.3825 | -0.0015 | -0.0014 |
| 0.08  | 0.1668      | 0.3775      | 0.1675 | 0.3774 | -0.0007 | 0.0001 |
| X  | Y1   | Y2   | Y3   | Y4   | Y5   | Y6   | Y7   | Y8   | Y9   | Y10  |
|----|------|------|------|------|------|------|------|------|------|------|
| 0.085 | 0.1859 | 0.3751 | 0.1865 | 0.3775 | -0.0006 | -0.0024 |
| 0.09 | 0.2038 | 0.3738 | 0.2034 | 0.3756 | 0.0003 | -0.0018 |
| 0.095 | 0.2205 | 0.3734 | 0.2218 | 0.3766 | -0.0013 | -0.0032 |
| 0.1 | 0.2363 | 0.3738 | 0.2375 | 0.3756 | -0.0012 | -0.0018 |
| 0.15 | 0.3558 | 0.4052 | 0.3548 | 0.4078 | 0.0009 | -0.0025 |
| 0.2 | 0.4371 | 0.4559 | 0.4363 | 0.4597 | 0.0007 | -0.0038 |
| 0.25 | 0.5004 | 0.5072 | 0.4985 | 0.51 | 0.0018 | -0.0028 |
| 0.3 | 0.5529 | 0.554 | 0.5503 | 0.5561 | 0.0027 | -0.0022 |
| 0.35 | 0.5977 | 0.5951 | 0.5959 | 0.5961 | 0.0018 | -0.0009 |
| 0.4 | 0.6362 | 0.631 | 0.6346 | 0.6323 | 0.0016 | -0.0013 |
| 0.45 | 0.6694 | 0.662 | 0.6681 | 0.663 | 0.0013 | -0.001 |
| 0.5 | 0.6981 | 0.6889 | 0.6961 | 0.6891 | 0.002 | -0.0002 |
| 0.6 | 0.7444 | 0.7323 | 0.7428 | 0.7324 | 0.0016 | -0.0001 |
| 0.7 | 0.779 | 0.7648 | 0.7762 | 0.7629 | 0.0028 | 0.0018 |
| 0.8 | 0.8049 | 0.7891 | 0.8027 | 0.7872 | 0.0022 | 0.0019 |
| 0.9 | 0.8243 | 0.8072 | 0.8211 | 0.8048 | 0.0031 | 0.0025 |
| 1 | 0.8387 | 0.8208 | 0.836 | 0.8172 | 0.0028 | 0.0036 |
| 1.5 | 0.8717 | 0.8517 | 0.8698 | 0.8492 | 0.0018 | 0.0025 |
| 2 | 0.8794 | 0.8589 | 0.8781 | 0.8561 | 0.0013 | 0.0028 |
| 2.5 | 0.8812 | 0.8606 | 0.8793 | 0.8578 | 0.0019 | 0.0028 |
| 3 | 0.8816 | 0.861 | 0.8795 | 0.8587 | 0.0021 | 0.0023 |
| 4 | 0.8817 | 0.8611 | 0.8802 | 0.8572 | 0.0015 | 0.0039 |
| 5 | 0.8817 | 0.8611 | 0.8813 | 0.86 | 0.0004 | 0.0012 |
| 6 | 0.8817 | 0.8611 | 0.8823 | 0.8593 | -0.0006 | 0.0018 |
| 7 | 0.8817 | 0.8611 | 0.8834 | 0.8607 | -0.0016 | 0.0005 |
| 8 | 0.8817 | 0.8611 | 0.8838 | 0.8606 | -0.002 | 0.0005 |
| 9 | 0.8817 | 0.8611 | 0.8831 | 0.8604 | -0.0014 | 0.0007 |
| 10 | 0.8817 | 0.8611 | 0.8845 | 0.8612 | -0.0028 | -0.0001 |
| 12 | 0.8817 | 0.8611 | 0.8834 | 0.8613 | -0.0017 | -0.0002 |
| 14 | 0.8817 | 0.8611 | 0.8838 | 0.862 | -0.0021 | -0.0009 |
| 15 | 0.8817 | 0.8611 | 0.8857 | 0.8635 | -0.004 | -0.0023 |
| 16 | 0.8817 | 0.8611 | 0.885 | 0.8623 | -0.0033 | -0.0012 |
| 18 | 0.8817 | 0.8611 | 0.8858 | 0.8637 | -0.0041 | -0.0026 |
| 19 | 0.8817 | 0.8611 | 0.886 | 0.8625 | -0.0042 | -0.0013 |
| 20 | 0.8817 | 0.8611 | 0.8859 | 0.8634 | -0.0042 | -0.0022 |
Chart S–XII.3. Plot of Inversion Recovery Data at 28.1 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0= 0.8000 No. 1= 0.2300

M(∞)'s
No. 2= 0.9000 No. 3= 1.0000

M(0)–M(∞)'s
No. 4= -1.4000 No. 5= 0.5000

and M(0)'s for reference
No. 4= -0.5000 No. 5= 1.5000

Rates
No. 6= 20.0000

Final Values of Fitted Parameters and Uncertainties:

| #  | Value           | Uncertainty |
|----|-----------------|-------------|
| 0  | 2.280800        | 0.026497    |
| 1  | 3.567518        | 0.037314    |
| 2  | 0.881739        | 0.000438    |
| 3  | 0.861147        | 0.000455    |
| 4  | -1.371021       | 0.000821    |
| 5  | -0.109826       | 0.001018    |
| 6  | 10.024921       | 0.041309    |
4) Ring Flip Eyring Data at 38.2 °C in CD$_2$Cl$_2$:

![Figure S-XII.4. Stacked Spectra of Inversion Recovery Data at 38.2 °C.](image)

**Table S-XII.4. CIFIT Plot File.**

| $d_2$ (s) | Observed $M_1$ | Observed $M_2$ | Fit $M_1$ | Fit $M_2$ | $\sigma M_1$ | $\sigma M_2$ |
|-----------|----------------|----------------|------------|------------|--------------|--------------|
| 0.001     | -0.3031        | 0.6618         | -0.3085    | 0.6593     | 0.0054       | 0.0025       |
| 0.002     | -0.277         | 0.6402         | -0.282     | 0.6367     | 0.005        | 0.0035       |
| 0.003     | -0.252         | 0.6198         | -0.2552    | 0.6157     | 0.0032       | 0.0041       |
| 0.004     | -0.2281        | 0.6006         | -0.2304    | 0.5992     | 0.0023       | 0.0014       |
| 0.005     | -0.2053        | 0.5824         | -0.2105    | 0.5775     | 0.0052       | 0.0048       |
| 0.007     | -0.1626        | 0.549          | -0.165     | 0.5453     | 0.0025       | 0.0037       |
| 0.01      | -0.1051        | 0.5056         | -0.1048    | 0.5036     | -0.0003      | 0.002        |
| 0.015     | -0.0244        | 0.4486         | -0.024     | 0.4458     | -0.0003      | 0.0027       |
| 0.02      | 0.0412         | 0.4068         | 0.0417     | 0.4062     | -0.0005      | 0.0006       |
| 0.025     | 0.0949         | 0.3769         | 0.0974     | 0.3793     | -0.0025      | -0.0024      |
| 0.03      | 0.1393         | 0.3562         | 0.1438     | 0.3576     | -0.0045      | -0.0014      |
| 0.035     | 0.1764         | 0.3426         | 0.1784     | 0.3446     | -0.002       | -0.002       |
| 0.04      | 0.2079         | 0.3345         | 0.2109     | 0.3391     | -0.003       | -0.0047      |
| 0.045     | 0.2348         | 0.3305         | 0.236      | 0.3348     | -0.0012      | -0.0043      |
| 0.05      | 0.2788         | 0.3315         | 0.2804     | 0.3335     | -0.0015      | -0.0019      |
| 0.06      | 0.2972         | 0.3351         | 0.2993     | 0.3387     | -0.0021      | -0.0035      |
| 0.065     | 0.3138         | 0.3402         | 0.316      | 0.3435     | -0.0021      | -0.0033      |
| 0.07      | 0.329          | 0.3462         | 0.3309     | 0.3498     | -0.0019      | -0.0036      |
| 0.075     | 0.3431         | 0.3531         | 0.3438     | 0.357      | -0.0008      | -0.0039      |
| 0.08      | 0.3562         | 0.3605         | 0.3553     | 0.3636     | 0.0008       | -0.0031      |
| 0.085     | 0.3685         | 0.3684         | 0.3673     | 0.3713     | 0.0011       | -0.0029      |
| x  | y1   | y2   | y3   | y4   | y5   | y6   | y7   | y8   | y9   | y10  |
|----|------|------|------|------|------|------|------|------|------|------|
| 0.09| 0.3802| 0.3765| 0.3808| 0.3808| -0.0006| -0.0044|
| 0.095| 0.3913| 0.3847| 0.3916| 0.3886| -0.0003| -0.0038|
| 0.1 | 0.402 | 0.3931| 0.4019| 0.3963| 0.0001| -0.0032|
| 0.15| 0.4928| 0.4742| 0.4906| 0.4742| 0.0022| 0.0001|
| 0.2 | 0.5654| 0.5435| 0.5629| 0.5447| 0.0025| -0.0012|
| 0.25| 0.6251| 0.6007| 0.6231| 0.6009| 0.002| -0.0002|
| 0.3 | 0.6744| 0.6479| 0.6724| 0.6472| 0.0019| 0.0007|
| 0.35| 0.7149| 0.6869| 0.7122| 0.6855| 0.0027| 0.0014|
| 0.4 | 0.7484| 0.719 | 0.7469| 0.7191| 0.0014| -0.0001|
| 0.45| 0.776 | 0.7455| 0.7739| 0.7449| 0.002| 0.0006|
| 0.5 | 0.7987| 0.7673| 0.7941| 0.7633| 0.0046| 0.004 |
| 0.6 | 0.8329| 0.8001| 0.8316| 0.7986| 0.0013| 0.0015|
| 0.7 | 0.8562| 0.8225| 0.8533| 0.8202| 0.0029| 0.0022|
| 0.8 | 0.872 | 0.8376| 0.8699| 0.8348| 0.0021| 0.0028|
| 0.9 | 0.8827| 0.848 | 0.8796| 0.845 | 0.0031| 0.0029|
| 1  | 0.89  | 0.855 | 0.8888| 0.8538| 0.0012| 0.0012|
| 1.5| 0.9033| 0.8677| 0.9004| 0.8643| 0.0029| 0.0034|
| 2  | 0.9052| 0.8696| 0.9017| 0.8651| 0.0035| 0.0045|
| 2.5| 0.9055| 0.8698| 0.9049| 0.8679| 0.0006| 0.0019|
| 3  | 0.9055| 0.8699| 0.9044| 0.867 | 0.0011| 0.0029|
| 4  | 0.9056| 0.8699| 0.9039| 0.8649| 0.0017| 0.005 |
| 5  | 0.9056| 0.8699| 0.9068| 0.8685| -0.0012| 0.0013|
| 6  | 0.9056| 0.8699| 0.9059| 0.8689| -0.0004| 0.001 |
| 7  | 0.9056| 0.8699| 0.9064| 0.8683| -0.0008| 0.0016|
| 8  | 0.9056| 0.8699| 0.9082| 0.8707| -0.0026| -0.0008|
| 9  | 0.9056| 0.8699| 0.9083| 0.8695| -0.0027| 0.0003|
| 10 | 0.9056| 0.8699| 0.9077| 0.87 | -0.0021| -0.0002|
| 12 | 0.9056| 0.8699| 0.9081| 0.8701| -0.0025| -0.0003|
| 14 | 0.9056| 0.8699| 0.9088| 0.8711| -0.0032| -0.0012|
| 15 | 0.9056| 0.8699| 0.9106| 0.872 | -0.0051| -0.0022|
| 16 | 0.9056| 0.8699| 0.91 | 0.872 | -0.0045| -0.0021|
| 18 | 0.9056| 0.8699| 0.9114| 0.8725| -0.0058| -0.0026|
| 19 | 0.9056| 0.8699| 0.9107| 0.873 | -0.0051| -0.0031|
| 20 | 0.9056| 0.8699| 0.9115| 0.8721| -0.0059| -0.0023|
Chart S-XII.4. Plot of Inversion Recovery Data at 38.2 °C.

CIFIT Guesses and Fit Parameters

Initial values of parameters:
1/T1's
No. 0 = 0.8000 No. 1 = 0.2300
M(inf)'s
No. 2 = 0.9000 No. 3 = 1.0000
M(0) - M(inf)'s
No. 4 = -1.4000 No. 5 = 0.5000
and M(0)'s for reference
No. 4 = -0.5000 No. 5 = 1.5000
Rates
No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
# 0 = 2.929290 +/- 0.070971
# 1 = 4.827882 +/- 0.085781
# 2 = 0.905556 +/- 0.000590
# 3 = 0.869870 +/- 0.000610
# 4 = -1.235982 +/- 0.001361
# 5 = -0.185295 +/- 0.001655
# 6 = 23.164390 +/- 0.176035
5) Ring Flip Eyring Data at 7.0 °C in C₆D₆:

![Figure S-XII.5. Stacked Spectra of Inversion Recovery Data at 7.0 °C in C₆D₆.](image)

**Table S-XII.5. CIFIT Plot File.**

| d² (s) | Observed | Observed | Fit | Fit | σM1 | σM2 |
|-------|----------|----------|-----|-----|-----|-----|
| 0.001 | -0.1049  | 0.7551   | -0.1389 | 0.7522 | 0.0339 | 0.0029 |
| 0.002 | -0.1019  | 0.7543   | -0.1328 | 0.7519 | 0.0309 | 0.0024 |
| 0.003 | -0.0989  | 0.7534   | -0.1245 | 0.7505 | 0.0256 | 0.0029 |
| 0.004 | -0.0959  | 0.7525   | -0.1177 | 0.7488 | 0.0218 | 0.0037 |
| 0.005 | -0.0929  | 0.7516   | -0.1093 | 0.7482 | 0.0164 | 0.0034 |
| 0.007 | -0.087   | 0.7499   | -0.0986 | 0.7453 | 0.0116 | 0.0046 |
| 0.01  | -0.0782  | 0.7474   | -0.0868 | 0.7427 | 0.0086 | 0.0046 |
| 0.015 | -0.0637  | 0.7433   | -0.0705 | 0.7398 | 0.0068 | 0.0035 |
| 0.02  | -0.0495  | 0.7394   | -0.0541 | 0.7377 | 0.0046 | 0.0017 |
| 0.025 | -0.0355  | 0.7357   | -0.0353 | 0.7349 | -0.0002 | 0.0008 |
| 0.03  | -0.0218  | 0.7322   | -0.0193 | 0.7301 | -0.0025 | 0.0021 |
| 0.035 | -0.0083  | 0.7288   | -0.0027 | 0.7292 | -0.0057 | -0.0004 |
| 0.04  | 0.0049   | 0.7257   | 0.012   | 0.7249 | -0.0071 | 0.0008 |
| 0.045 | 0.0179   | 0.7226   | 0.0264  | 0.7229 | -0.0086 | -0.0003 |
| 0.05  | 0.0306   | 0.7197   | 0.042   | 0.7207 | -0.0113 | -0.001 |
| 0.055 | 0.0431   | 0.717    | 0.055   | 0.7169 | -0.0118 | 0.0001 |
| 0.06  | 0.0554   | 0.7144   | 0.0703  | 0.7145 | -0.0149 | -0.0001 |
| 0.065 | 0.0675   | 0.7119   | 0.0851  | 0.7141 | -0.0176 | -0.0022 |
| 0.07  | 0.0793   | 0.7096   | 0.0968  | 0.7126 | -0.0174 | -0.003 |
| 0.075 | 0.091    | 0.7073   | 0.1091  | 0.7105 | -0.0181 | -0.0031 |
| 0.08  | 0.1024   | 0.7053   | 0.1218  | 0.707 | -0.0194 | -0.0018 |
| 0.085 | 0.1136 | 0.7033 | 0.1344 | 0.7064 | -0.0208 | -0.0031 |
|-------|--------|--------|--------|--------|---------|---------|
| 0.09  | 0.1247 | 0.7014 | 0.1468 | 0.7033 | -0.0221 | -0.0019 |
| 0.095 | 0.1355 | 0.6997 | 0.1587 | 0.7039 | -0.0232 | -0.0043 |
| 0.1   | 0.1461 | 0.698  | 0.1703 | 0.7009 | -0.0242 | -0.0029 |
| 0.15  | 0.2428 | 0.6865 | 0.2532 | 0.6928 | -0.0103 | -0.0062 |
| 0.2   | 0.324  | 0.6821 | 0.326  | 0.6905 | -0.002  | -0.0084 |
| 0.25  | 0.3925 | 0.6824 | 0.3875 | 0.6907 | 0.005   | -0.0084 |
| 0.3   | 0.4504 | 0.6857 | 0.4405 | 0.6929 | 0.0099  | -0.0072 |
| 0.35  | 0.4994 | 0.6908 | 0.4863 | 0.6968 | 0.0131  | -0.006  |
| 0.4   | 0.5412 | 0.6971 | 0.5245 | 0.7013 | 0.0166  | -0.0042 |
| 0.45  | 0.5767 | 0.7038 | 0.5607 | 0.7077 | 0.016   | -0.004  |
| 0.5   | 0.6071 | 0.7105 | 0.5892 | 0.712  | 0.0179  | -0.0015 |
| 0.6   | 0.6553 | 0.7234 | 0.6371 | 0.7231 | 0.0183  | 0.0003  |
| 0.7   | 0.6909 | 0.7346 | 0.6724 | 0.7316 | 0.0185  | 0.003   |
| 0.8   | 0.7171 | 0.7439 | 0.7008 | 0.7393 | 0.0163  | 0.0046  |
| 0.9   | 0.7367 | 0.7513 | 0.7233 | 0.7471 | 0.0134  | 0.0043  |
| 1     | 0.7512 | 0.7572 | 0.7401 | 0.7526 | 0.0111  | 0.0047  |
| 1.5   | 0.7842 | 0.7716 | 0.7832 | 0.7671 | 0.001   | 0.0045  |
| 2     | 0.792  | 0.7752 | 0.7961 | 0.7722 | -0.0041 | 0.0031  |
| 2.5   | 0.7939 | 0.7761 | 0.7996 | 0.7729 | -0.0057 | 0.0032  |
| 4     | 0.7945 | 0.7764 | 0.8023 | 0.7747 | -0.0078 | 0.0017  |
| 6     | 0.7945 | 0.7764 | 0.8029 | 0.7757 | -0.0084 | 0.0007  |
| 8     | 0.7945 | 0.7764 | 0.8037 | 0.777  | -0.0093 | -0.0006 |
| 10    | 0.7945 | 0.7764 | 0.8029 | 0.7765 | -0.0084 | -0.0001 |
| 12    | 0.7945 | 0.7764 | 0.8044 | 0.7774 | -0.0099 | -0.001  |
| 14    | 0.7945 | 0.7764 | 0.8018 | 0.7742 | -0.0073 | 0.0022  |
| 16    | 0.7945 | 0.7764 | 0.8012 | 0.7743 | -0.0067 | 0.002   |
| 18    | 0.7945 | 0.7764 | 0.8011 | 0.7744 | -0.0067 | 0.0019  |
| 20    | 0.7945 | 0.7764 | 0.8003 | 0.7743 | -0.0058 | 0.0021  |
Chart S-XII.5. Plot of Inversion Recovery Data at 7.0 °C in C$_6$D$_6$.

CIFIT Guesses and Fit Parameters
Initial values of parameters:
1/T1's
  No. 0 = 0.8000 No. 1 = 0.2300
M(inf)'s
  No. 2 = 0.9000 No. 3 = 1.0000
M(0)−M(inf)'s
  No. 4 = −1.4000 No. 5 = 0.5000
and M(0)'s for reference
  No. 4 = −0.5000 No. 5 = 1.5000
Rates
  No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
  # 0 = 2.263373 +/- 0.104782
  # 1 = 4.118448 +/- 0.578497
  # 2 = 0.794467 +/- 0.003048
  # 3 = 0.776389 +/- 0.003163
  # 4 = −0.902448 +/- 0.004006
  # 5 = −0.020334 +/- 0.005169
  # 6 = 1.126540 +/- 0.111701
6) **Ring Flip Eyring Data at 13.8 °C in C₆D₆:**

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**Figure S–XII.6.** Stacked Spectra of Inversion Recovery Data at 13.8 °C in C₆D₆.

**Table S–XII.6.** CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.1791     | 1.0251      | -0.1815| 1.0267 | 0.0024| -0.0016|
| 0.002  | -0.1746     | 1.0235      | -0.1782| 1.0244 | 0.0035| -0.0009|
| 0.003  | -0.1702     | 1.022       | -0.1731| 1.0209 | 0.0029| 0.0011 |
| 0.004  | -0.1657     | 1.0204      | -0.1684| 1.0     | 0.0027| 0.0004 |
| 0.005  | -0.1613     | 1.0189      | -0.1639| 1.0185 | 0.0026| 0.0004 |
| 0.007  | -0.1525     | 1.0159      | -0.1549| 1.0148 | 0.0023| 0.0011|
| 0.01   | -0.1395     | 1.0115      | -0.1416| 1.01   | 0.0021| 0.0015 |
| 0.015  | -0.1181     | 1.0045      | -0.1193| 1.0032 | 0.0012| 0.0013|
| 0.02   | -0.0972     | 0.9978      | -0.0983| 0.998  | 0.0011| -0.0002|
| 0.025  | -0.0767     | 0.9915      | -0.0775| 0.9903 | 0.0008| 0.0011|
| 0.03   | -0.0566     | 0.9855      | -0.0559| 0.985  | -0.0007| 0.0004|
| 0.035  | -0.0369     | 0.9798      | -0.0363| 0.9794 | -0.0006| 0.0004|
| 0.04   | -0.0176     | 0.9744      | -0.017 | 0.9746 | -0.0006| -0.0002|
| 0.045  | 0.0013      | 0.9693      | 0.0023 | 0.9698 | -0.001 | -0.0004|
| 0.05   | 0.0198      | 0.9646      | 0.0215 | 0.9641 | -0.0016| 0.0005 |
| 0.055  | 0.038       | 0.9601      | 0.0403 | 0.9611 | -0.0023| -0.0011|
| 0.06   | 0.0558      | 0.9558      | 0.0573 | 0.9544 | -0.0015| 0.0014 |
| 0.065  | 0.0732      | 0.9518      | 0.0754 | 0.9518 | -0.0021| 0.0001 |
| 0.07   | 0.0904      | 0.9481      | 0.0931 | 0.9482 | -0.0027| -0.0001|
| 0.075  | 0.1071      | 0.9446      | 0.1096 | 0.9437 | -0.0025| 0.0009 |
| 0.08   | 0.1236      | 0.9413      | 0.126  | 0.9413 | -0.0024| 0      |
| Value | Column 1 | Column 2 | Column 3 | Column 4 | Column 5 | Column 6 | Column 7 | Column 8 | Column 9 | Column 10 |
|-------|----------|----------|----------|----------|----------|----------|----------|----------|----------|-----------|
| 0.085 | 0.1397   | 0.9383   | 0.142    | 0.9393   | -0.0023  | -0.001   |
| 0.09  | 0.1555   | 0.9354   | 0.1578   | 0.9354   | -0.0023  | 0        |
| 0.095 | 0.1711   | 0.9328   | 0.1732   | 0.9332   | -0.0022  | -0.0004  |
| 0.1   | 0.1863   | 0.9303   | 0.1881   | 0.9307   | -0.0018  | -0.0004  |
| 0.15  | 0.3235   | 0.915    | 0.3254   | 0.9162   | -0.0019  | -0.0012  |
| 0.2   | 0.4376   | 0.9121   | 0.4389   | 0.9138   | -0.0013  | -0.0016  |
| 0.25  | 0.5329   | 0.9173   | 0.5334   | 0.9201   | -0.0005  | -0.0028  |
| 0.3   | 0.6131   | 0.9274   | 0.613    | 0.93     | 0.0001   | -0.0026  |
| 0.35  | 0.681    | 0.9402   | 0.6802   | 0.9424   | 0.0008   | -0.0022  |
| 0.4   | 0.7387   | 0.9544   | 0.7366   | 0.9571   | 0.0021   | -0.0027  |
| 0.45  | 0.788    | 0.9688   | 0.7864   | 0.9709   | 0.0016   | -0.0021  |
| 0.5   | 0.8302   | 0.983    | 0.8286   | 0.984    | 0.0016   | -0.0009  |
| 0.6   | 0.8977   | 1.0092   | 0.8955   | 1.0093   | 0.0022   | -0.0001  |
| 0.7   | 0.9481   | 1.0315   | 0.9455   | 1.0302   | 0.0026   | 0.0013   |
| 0.8   | 0.986    | 1.0496   | 0.9831   | 1.0473   | 0.0029   | 0.0023   |
| 0.9   | 1.0146   | 1.0642   | 1.0113   | 1.0611   | 0.0034   | 0.0031   |
| 1     | 1.0363   | 1.0756   | 1.0319   | 1.0715   | 0.0044   | 0.004    |
| 1.5   | 1.0879   | 1.104    | 1.0836   | 1.0983   | 0.0043   | 0.0056   |
| 2     | 1.1013   | 1.1116   | 1.0991   | 1.107    | 0.0023   | 0.0046   |
| 2.5   | 1.1048   | 1.1135   | 1.1029   | 1.11     | 0.0019   | 0.0035   |
| 4     | 1.1061   | 1.1142   | 1.1057   | 1.111    | 0.0004   | 0.0033   |
| 6     | 1.1061   | 1.1143   | 1.1059   | 1.1128   | 0.0002   | 0.0014   |
| 8     | 1.1061   | 1.1143   | 1.1065   | 1.1144   | -0.0004  | -0.0002  |
| 10    | 1.1061   | 1.1143   | 1.1079   | 1.1154   | -0.0018  | -0.0011  |
| 12    | 1.1061   | 1.1143   | 1.11     | 1.1164   | -0.0039  | -0.0022  |
| 14    | 1.1061   | 1.1143   | 1.1098   | 1.1164   | -0.0037  | -0.0021  |
| 16    | 1.1061   | 1.1143   | 1.1102   | 1.1171   | -0.0042  | -0.0028  |
| 18    | 1.1061   | 1.1143   | 1.1093   | 1.1184   | -0.0032  | -0.0041  |
| 20    | 1.1061   | 1.1143   | 1.1111   | 1.119    | -0.005   | -0.0048  |
Chart S-XII.6. Plot of Inversion Recovery Data at 7.0 °C in C₆D₆.

CIFIT Guesses and Fit Parameters

Initial values of parameters:
- 1/T1's
  - No. 0 = 0.8000
  - No. 1 = 0.2300
- M(inf)'s
  - No. 2 = 0.9000
  - No. 3 = 1.0000
- M(0)−M(inf)'s
  - No. 4 = -1.4000
  - No. 5 = 0.5000
- and M(0)'s for reference
  - No. 4 = -0.5000
  - No. 5 = 1.5000
- Rates
  - No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
- # 0 = 1.985377 +/- 0.016838
- # 1 = 3.902108 +/- 0.063284
- # 2 = 1.106079 +/- 0.000643
- # 3 = 1.114254 +/- 0.000666
- # 4 = -1.289669 +/- 0.000852
- # 5 = -0.087541 +/- 0.001092
- # 6 = 1.614867 +/- 0.019787
7) Ring Flip Eyring Data at 24.0 °C in C₆D₆:

Figure S–XII.7. Stacked Spectra of Inversion Recovery Data at 24.0 °C in C₆D₆.

Table S–XII.7. CIFIT Plot File.

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | -0.1408     | 0.8095      | -0.1478| 0.8082 | 0.0069| 0.0013|
| 0.002  | -0.1328     | 0.8063      | -0.1398| 0.8038 | 0.007 | 0.0025|
| 0.003  | -0.1248     | 0.8031      | -0.1311| 0.8008 | 0.0063| 0.0023|
| 0.004  | -0.1168     | 0.8         | -0.1231| 0.7958 | 0.0063| 0.0042|
| 0.005  | -0.1089     | 0.797       | -0.1152| 0.7939 | 0.0063| 0.0031|
| 0.007  | -0.0934     | 0.7911      | -0.0988| 0.7885 | 0.0054| 0.0026|
| 0.01   | -0.0706     | 0.7826      | -0.0743| 0.78    | 0.0037| 0.0026|
| 0.015  | -0.0338     | 0.7696      | -0.0356| 0.7677 | 0.0018| 0.0018|
| 0.02   | 0.0014      | 0.7578      | 0.0021 | 0.756  | -0.0007| 0.0017|
| 0.025  | 0.0352      | 0.7471      | 0.0366 | 0.7451 | -0.0014| 0.002 |
| 0.03   | 0.0676      | 0.7376      | 0.0701 | 0.7363 | -0.0025| 0.0012|
| 0.035  | 0.0987      | 0.729       | 0.1014 | 0.7294 | -0.0026| -0.0004|
| 0.04   | 0.1286      | 0.7214      | 0.1325 | 0.7205 | -0.0039| 0.0009|
| 0.045  | 0.1573      | 0.7147      | 0.1617 | 0.7149 | -0.0044| -0.0002|
| 0.05   | 0.1849      | 0.7088      | 0.1899 | 0.7098 | -0.005 | -0.001 |
| 0.055  | 0.2114      | 0.7037      | 0.2165 | 0.7059 | -0.0051| -0.0022|
| 0.06   | 0.2369      | 0.6993      | 0.2418 | 0.7005 | -0.005 | -0.0012|
| 0.065  | 0.2614      | 0.6955      | 0.2654 | 0.6977 | -0.004 | -0.0022|
| 0.07   | 0.285       | 0.6924      | 0.2901 | 0.6948 | -0.0051| -0.0024|
| 0.075  | 0.3077      | 0.6898      | 0.3128 | 0.6925 | -0.0052| -0.0027|
| 0.08   | 0.3296      | 0.6878      | 0.3342 | 0.6912 | -0.0046| -0.0034|
|   |     |     |     |     |     |     |
|---|-----|-----|-----|-----|-----|-----|
| 0.085 | 0.3507 | 0.6863 | 0.3561 | 0.6904 | -0.0055 | -0.0041 |
| 0.09 | 0.371 | 0.6852 | 0.3761 | 0.6887 | -0.0051 | -0.0035 |
| 0.095 | 0.3906 | 0.6845 | 0.3944 | 0.688 | -0.0039 | -0.0035 |
| 0.1 | 0.4095 | 0.6843 | 0.4125 | 0.6889 | -0.0031 | -0.0046 |
| 0.15 | 0.5666 | 0.6977 | 0.5661 | 0.7036 | 0.0005 | -0.006 |
| 0.2 | 0.6804 | 0.7272 | 0.6762 | 0.7342 | 0.0042 | -0.0071 |
| 0.25 | 0.7656 | 0.7615 | 0.7592 | 0.7668 | 0.0064 | -0.0053 |
| 0.3 | 0.8308 | 0.7953 | 0.8241 | 0.7998 | 0.0068 | -0.0045 |
| 0.35 | 0.8819 | 0.8261 | 0.8739 | 0.829 | 0.0079 | -0.0029 |
| 0.4 | 0.9223 | 0.8531 | 0.9153 | 0.8533 | 0.0071 | -0.0002 |
| 0.45 | 0.9548 | 0.8762 | 0.9458 | 0.8754 | 0.009 | 0.0007 |
| 0.5 | 0.981 | 0.8956 | 0.9721 | 0.8929 | 0.0089 | 0.0027 |
| 0.6 | 1.0195 | 0.9253 | 1.012 | 0.9216 | 0.0074 | 0.0037 |
| 0.7 | 1.0451 | 0.9456 | 1.0395 | 0.9398 | 0.0057 | 0.0058 |
| 0.8 | 1.0623 | 0.9594 | 1.0563 | 0.9534 | 0.006 | 0.006 |
| 0.9 | 1.0738 | 0.9687 | 1.0691 | 0.9617 | 0.0047 | 0.007 |
| 1 | 1.0816 | 0.975 | 1.0789 | 0.9682 | 0.0027 | 0.0068 |
| 1.5 | 1.0953 | 0.9861 | 1.0933 | 0.98 | 0.002 | 0.006 |
| 2 | 1.0972 | 0.9876 | 1.0958 | 0.9814 | 0.0014 | 0.0062 |
| 2.5 | 1.0974 | 0.9878 | 1.0969 | 0.983 | 0.0006 | 0.0048 |
| 4 | 1.0975 | 0.9878 | 1.0991 | 0.9836 | -0.0016 | 0.0042 |
| 6 | 1.0975 | 0.9878 | 1.1 | 0.9862 | -0.0025 | 0.0016 |
| 8 | 1.0975 | 0.9878 | 1.1017 | 0.9875 | -0.0042 | 0.0004 |
| 10 | 1.0975 | 0.9878 | 1.1047 | 0.9902 | -0.0072 | -0.0024 |
| 12 | 1.0975 | 0.9878 | 1.1048 | 0.9901 | -0.0073 | -0.0022 |
| 14 | 1.0975 | 0.9878 | 1.1059 | 0.9922 | -0.0084 | -0.0043 |
| 16 | 1.0975 | 0.9878 | 1.1061 | 0.992 | -0.0086 | -0.0041 |
| 18 | 1.0975 | 0.9878 | 1.1067 | 0.9943 | -0.0092 | -0.0065 |
| 20 | 1.0975 | 0.9878 | 1.1064 | 0.9929 | -0.0089 | -0.0051 |
**Chart S-XII.7. Plot of Inversion Recovery Data at 24.0 °C in C₆D₆**

![Inversion Recovery Data at 24.0 °C in C₆D₆](image)

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:

| Parameter   | No. 0 | No. 1 | No. 2 | No. 3 | No. 4 | No. 5 | No. 6 |
|-------------|-------|-------|-------|-------|-------|-------|-------|
| $1/T_1$'s   | 0.800 | 0.230 | 0.900 | 1.000 | -1.400| -0.500| 20.000|
| M(inf)'s    |       |       |       |       |       |       |       |
| No. 2       | 0.900 | 1.000 |       |       |       |       |       |
| M(0)−M(inf)'s |     |       |       |       |       |       |       |
| No. 4       | -1.400| 0.500 |       |       |       |       |       |
| and M(0)'s for reference | | | | | | | |
| No. 4       | -0.500| 1.500 |       |       |       |       |       |

Rates

No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:

| # | Value       | Uncertainty |
|---|-------------|-------------|
| 0 | 3.226847    | 0.060591    |
| 1 | 4.875314    | 0.138039    |
| 2 | 1.097482    | 0.001221    |
| 3 | 0.987835    | 0.001297    |
| 4 | -1.246472   | 0.001904    |
| 5 | -0.175058   | 0.002376    |
| 6 | 3.883901    | 0.078210    |
8) **Ring Flip Eyring Data at 34.2 °C in C₆D₆:**

![Figure S-XII.8. Stacked Spectra of Inversion Recovery Data at 34.2 °C in C₆D₆.](image)

**Table S-XII.8. CIFIT Plot File.**

| d2 (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|-------|-------------|-------------|--------|--------|------|------|
| 0.001 | -0.0071     | 0.7384      | -0.0072| 0.7416 | 0.0001| -0.0032|
| 0.002 | 0.0078      | 0.7322      | 0.0032 | 0.7305 | 0.0045| 0.0017 |
| 0.003 | 0.0224      | 0.7263      | 0.0144 | 0.7224 | 0.008 | 0.0039 |
| 0.004 | 0.0367      | 0.7206      | 0.0294 | 0.7156 | 0.0073| 0.005  |
| 0.005 | 0.0508      | 0.7153      | 0.0469 | 0.7115 | 0.0038| 0.0038 |
| 0.007 | 0.078       | 0.7053      | 0.0736 | 0.6994 | 0.0044| 0.0059 |
| 0.01  | 0.1168      | 0.6921      | 0.1155 | 0.687  | 0.0013| 0.0052 |
| 0.015 | 0.1765      | 0.6746      | 0.1755 | 0.6711 | 0.0011| 0.0035 |
| 0.02  | 0.2308      | 0.6618      | 0.2348 | 0.6611 | -0.0041| 0.0007 |
| 0.025 | 0.2802      | 0.6531      | 0.283  | 0.6525 | 0.0028| 0.0005 |
| 0.03  | 0.3254      | 0.6477      | 0.3294 | 0.6492 | -0.004| 0.0015 |
| 0.035 | 0.3669      | 0.6453      | 0.3703 | 0.6468 | -0.0034| 0.0014 |
| 0.04  | 0.4051      | 0.6453      | 0.41   | 0.649  | -0.005 | 0.0037 |
| 0.045 | 0.4403      | 0.6474      | 0.4448 | 0.6507 | -0.0044| 0.0033 |
| 0.05  | 0.473       | 0.6511      | 0.4769 | 0.655  | -0.0038| 0.0038 |
| 0.055 | 0.5035      | 0.6563      | 0.5085 | 0.6624 | -0.0051| 0.0006 |
| 0.06  | 0.5318      | 0.6627      | 0.5358 | 0.6677 | -0.004 | 0.005  |
| 0.065 | 0.5584      | 0.67       | 0.5612 | 0.6742 | -0.0028| 0.0041 |
| 0.07  | 0.5833      | 0.6781      | 0.5853 | 0.6822 | -0.002 | 0.0004 |
| 0.075 | 0.6068      | 0.6869      | 0.6089 | 0.6911 | -0.0021| 0.0042 |
| 0.08  | 0.6289      | 0.6961      | 0.6305 | 0.6997 | -0.0016| 0.0036 |
| 0.085 | 0.6499 | 0.7057 | 0.6487 | 0.7076 | 0.0011 | -0.0019 |
| 0.09  | 0.6698 | 0.7156 | 0.6679 | 0.7177 | 0.0018 | -0.0021 |
| 0.095 | 0.6886 | 0.7257 | 0.689  | 0.7276 | -0.0003 | -0.0019 |
| 0.1   | 0.7066 | 0.7359 | 0.7043 | 0.738  | 0.0023 | -0.0021 |
| 0.15  | 0.8502 | 0.8357 | 0.8464 | 0.8367 | 0.0038 | -0.001 |
| 0.2   | 0.9507 | 0.9191 | 0.9455 | 0.9182 | 0.0052 | 0.001  |
| 0.25  | 1.0246 | 0.9839 | 1.0193 | 0.9815 | 0.0053 | 0.0024 |
| 0.3   | 1.0799 | 1.033  | 1.075  | 1.0305 | 0.0049 | 0.0026 |
| 0.35  | 1.1214 | 1.0702 | 1.116  | 1.0667 | 0.0054 | 0.0034 |
| 0.4   | 1.1526 | 1.0981 | 1.1484 | 1.0951 | 0.0042 | 0.003 |
| 0.45  | 1.1761 | 1.1192 | 1.1709 | 1.1162 | 0.0052 | 0.0029 |
| 0.5   | 1.1937 | 1.135  | 1.1882 | 1.1315 | 0.0055 | 0.0035 |
| 0.6   | 1.217  | 1.1559 | 1.2129 | 1.1527 | 0.0041 | 0.0032 |
| 0.7   | 1.2302 | 1.1677 | 1.2293 | 1.1647 | 0.0009 | 0.0031 |
| 0.8   | 1.2377 | 1.1744 | 1.2353 | 1.1724 | 0.0024 | 0.002  |
| 0.9   | 1.2419 | 1.1782 | 1.2409 | 1.1767 | 0.0011 | 0.0016 |
| 1     | 1.2443 | 1.1804 | 1.2434 | 1.1775 | 0.0009 | 0.0029 |
| 1.5   | 1.2473 | 1.183  | 1.2459 | 1.1816 | 0.0014 | 0.0014 |
| 2     | 1.2475 | 1.1832 | 1.2461 | 1.1808 | 0.0013 | 0.0024 |
| 2.5   | 1.2475 | 1.1832 | 1.2469 | 1.1832 | 0.0005 | 0 |
| 4     | 1.2475 | 1.1832 | 1.2483 | 1.1826 | -0.0008 | 0.0006 |
| 6     | 1.2475 | 1.1832 | 1.2517 | 1.1847 | -0.0042 | -0.0015 |
| 8     | 1.2475 | 1.1832 | 1.2505 | 1.1842 | -0.003 | -0.001 |
| 10    | 1.2475 | 1.1832 | 1.2526 | 1.1851 | -0.0052 | -0.0019 |
| 12    | 1.2475 | 1.1832 | 1.2535 | 1.1847 | -0.0061 | -0.0015 |
| 14    | 1.2475 | 1.1832 | 1.2535 | 1.185  | -0.006 | -0.0018 |
| 16    | 1.2475 | 1.1832 | 1.2526 | 1.185  | -0.0051 | -0.0018 |
| 18    | 1.2475 | 1.1832 | 1.253  | 1.1845 | -0.0055 | -0.0013 |
| 20    | 1.2475 | 1.1832 | 1.2539 | 1.1855 | -0.0064 | -0.0023 |
Chart S-XII.8. Plot of Inversion Recovery Data at 34.2 °C in C₆D₆

CIFIT Guesses and Fit Parameters

Initial values of parameters:

1/T1's
No. 0= 0.8000 No. 1= 0.2300

M(inf)'s
No. 2= 0.9000 No. 3= 1.0000

M(0)−M(inf)'s
No. 4= -1.4000 No. 5= 0.5000

and M(0)'s for reference
No. 4= -0.5000 No. 5= 1.5000

Rates
No. 6= 20.0000

Final Values of Fitted Parameters and Uncertainties:

# 0 = 4.474118 +/- 0.091534
# 1 = 7.032864 +/- 0.133538
# 2 = 1.247476 +/- 0.000842
# 3 = 1.183201 +/- 0.000876
# 4 = -1.269876 +/- 0.001608
# 5 = -0.438303 +/- 0.001956
# 6 = 11.707086 +/- 0.216313
9) **Ring Flip Eyring Data at 44.2 °C in C₆D₆:**

**Figure S-XII.9.** Stacked Spectra of Inversion Recovery Data at 44.2 °C in C₆D₆.

**Table S-XII.9.** CIFIT Plot File.

| d² (s) | Observed M1 | Observed M2 | Fit M1 | Fit M2 | σM1 | σM2 |
|--------|-------------|-------------|--------|--------|------|------|
| 0.001  | 0.4714      | 1.8291      | 0.4653 | 1.8277 | 0.0061 | 0.0014 |
| 0.002  | 0.5473      | 1.8131      | 0.531  | 1.7995 | 0.0163 | 0.0136 |
| 0.003  | 0.6199      | 1.8001      | 0.6017 | 1.7857 | 0.0182 | 0.0144 |
| 0.004  | 0.6894      | 1.7898      | 0.6673 | 1.7702 | 0.0221 | 0.0196 |
| 0.005  | 0.7561      | 1.7821      | 0.7378 | 1.7617 | 0.0183 | 0.0204 |
| 0.007  | 0.8816      | 1.7734      | 0.8687 | 1.7592 | 0.0129 | 0.0142 |
| 0.01   | 1.0525      | 1.7751      | 1.0614 | 1.7741 | -0.0089 | 0.001 |
| 0.015  | 1.2999      | 1.8079      | 1.3068 | 1.8088 | -0.0069 | -0.0009 |
| 0.02   | 1.5113      | 1.8666      | 1.5175 | 1.8769 | -0.0061 | -0.0103 |
| 0.025  | 1.6961      | 1.9418      | 1.7123 | 1.9592 | -0.0163 | -0.0173 |
| 0.03   | 1.8605      | 2.0271      | 1.8    | 2.0393 | -0.0095 | -0.0121 |
| 0.035  | 2.0093      | 2.1179      | 2.0231 | 2.137  | -0.0138 | -0.0191 |
| 0.04   | 2.1457      | 2.2113      | 2.1697 | 2.2307 | -0.024  | -0.0195 |
| 0.045  | 2.272       | 2.305       | 2.2835 | 2.317  | -0.0115 | -0.012 |
| 0.05   | 2.39        | 2.3978      | 2.4021 | 2.4218 | -0.0121 | -0.024 |
| 0.055  | 2.5008      | 2.4887      | 2.5117 | 2.4919 | -0.0109 | -0.0032 |
| 0.06   | 2.6053      | 2.5772      | 2.6061 | 2.5852 | -0.0007 | -0.008 |
| 0.065  | 2.7044      | 2.6629      | 2.713  | 2.6737 | -0.0086 | -0.0108 |
| 0.07   | 2.7985      | 2.7456      | 2.805  | 2.7475 | -0.0066 | -0.0019 |
| 0.075  | 2.888       | 2.8252      | 2.8936 | 2.8407 | -0.0056 | -0.0155 |
| 0.08   | 2.9732      | 2.9017      | 2.9771 | 2.9075 | -0.0039 | -0.0058 |
| x   | y     | x     | y     | y     | y     | y     | y     |
|-----|-------|-------|-------|-------|-------|-------|-------|
| 0.085 | 3.0546 | 2.9751 | 3.0549 | 2.9785 | -0.0004 | -0.0034 |
| 0.09  | 3.1322 | 3.0455 | 3.1244 | 3.0419 | 0.0078  | 0.0036  |
| 0.095 | 3.2063 | 3.1129 | 3.1953 | 3.1093 | 0.011   | 0.0036  |
| 0.1   | 3.2771 | 3.1775 | 3.2726 | 3.1734 | 0.0045  | 0.0041  |
| 0.15  | 3.8349 | 3.6885 | 3.8277 | 3.679  | 0.0072  | 0.0095  |
| 0.2   | 4.1919 | 4.0162 | 4.178  | 3.9979 | 0.0139  | 0.0182  |
| 0.25  | 4.4206 | 4.226  | 4.4011 | 4.2038 | 0.0195  | 0.0223  |
| 0.3   | 4.5671 | 4.3605 | 4.5409 | 4.3362 | 0.0262  | 0.0243  |
| 0.35  | 4.6609 | 4.4466 | 4.6363 | 4.4229 | 0.0246  | 0.0237  |
| 0.4   | 4.721  | 4.5018 | 4.6876 | 4.4675 | 0.0334  | 0.0342  |
| 0.45  | 4.7595 | 4.5371 | 4.7276 | 4.5034 | 0.0319  | 0.0338  |
| 0.5   | 4.7842 | 4.5598 | 4.759  | 4.5398 | 0.0252  | 0.02    |
| 0.6   | 4.8101 | 4.5836 | 4.7815 | 4.5527 | 0.0286  | 0.0309  |
| 0.7   | 4.8208 | 4.5933 | 4.7984 | 4.5638 | 0.0224  | 0.0295  |
| 0.8   | 4.8251 | 4.5973 | 4.7927 | 4.5775 | 0.0324  | 0.0199  |
| 0.9   | 4.8269 | 4.599  | 4.8028 | 4.5726 | 0.0241  | 0.0263  |
| 1     | 4.8276 | 4.5996 | 4.8052 | 4.5756 | 0.0224  | 0.024   |
| 1.5   | 4.8282 | 4.6001 | 4.7936 | 4.5792 | 0.0346  | 0.0209  |
| 2     | 4.8282 | 4.6001 | 4.819  | 4.5918 | 0.0092  | 0.0083  |
| 2.5   | 4.8282 | 4.6001 | 4.8155 | 4.591  | 0.0126  | 0.0091  |
| 4     | 4.8282 | 4.6001 | 4.8273 | 4.6027 | 0.0009  | -0.0026 |
| 6     | 4.8282 | 4.6001 | 4.848  | 4.6157 | -0.0198 | -0.0156 |
| 8     | 4.8282 | 4.6001 | 4.8591 | 4.6378 | -0.031  | -0.0376 |
| 10    | 4.8282 | 4.6001 | 4.8633 | 4.6253 | -0.0352 | -0.0251 |
| 12    | 4.8282 | 4.6001 | 4.8708 | 4.635  | -0.0427 | -0.0349 |
| 14    | 4.8282 | 4.6001 | 4.8774 | 4.6424 | -0.0493 | -0.0423 |
| 16    | 4.8282 | 4.6001 | 4.8735 | 4.6442 | -0.0453 | -0.0441 |
| 18    | 4.8282 | 4.6001 | 4.8743 | 4.6438 | -0.0461 | -0.0436 |
| 20    | 4.8282 | 4.6001 | 4.8993 | 4.6413 | -0.0711 | -0.0412 |
Chart S-XII.9. Plot of Inversion Recovery Data at 44.2 °C in C₆D₆.

**CIFIT Guesses and Fit Parameters**

Initial values of parameters:
- \(1/T1's\)
  - No. 0 = 0.8000 No. 1 = 0.2300
- \(M(\infty)'s\)
  - No. 2 = 0.9000 No. 3 = 1.0000
- \(M(0)-M(\infty)'s\)
  - No. 4 = -1.4000 No. 5 = 0.5000
- and \(M(0)'s\) for reference
  - No. 4 = -0.5000 No. 5 = 1.5000
- Rates
  - No. 6 = 20.0000

Final Values of Fitted Parameters and Uncertainties:
- \# 0 = 6.311334 +/- 0.328134
- \# 1 = 11.736610 +/- 0.387678
- \# 2 = 4.828156 +/- 0.005139
- \# 3 = 4.600111 +/- 0.005177
- \# 4 = -4.436135 +/- 0.013904
- \# 5 = -2.751749 +/- 0.015276
- \# 6 = 31.590672 +/- 1.895127
XIII. Evans Method Study of Complex 4.

An Evans method magnetic susceptibility experiment revealed no paramagnetic contact shift to dichloromethane solvent. We attempted to detect paramagnetism in complex 4 using the Evans method (equation 1). In the dry box, a solution of complex 4 (35.0 mg, 63.4 μmol) in 0.5 mL of CD$_2$Cl$_2$ was placed into an insert tube and sealed. The insert tube was placed into an NMR tube containing only CD$_2$Cl$_2$. The $^1$H NMR spectrum (600 MHz) was obtained at 25 °C (Figure S–XIII.1). No shift in the solvent peak could be detected; this signal was Lorentzian with half–height width of 4 Hz. Thus, we could not calculate the magnetic susceptibility of complex 4.

![Figure S–XIII.1. H NMR Spectrum of complex 4 (127 mM) in CD$_2$Cl$_2$.](image)

\[
\chi_m = \left( \frac{-3 \Delta f}{4 \pi fm} \right) + \chi_o + \left( \frac{\chi_o (d_o - d_s)}{m} \right) \quad \text{(eq. 1)}
\]

where:
- $\chi_m$ = mass magnetic susceptibility of the solute (cm$^3$ g$^{-1}$)
- $\Delta f$ = observed frequency shift of reference resonance (Hz)
- $f$ = spectrometer frequency (Hz)
- $m$ = mass of substance per cm$^3$ of solution (g)
- $\chi_o$ = mass magnetic susceptibility of solvent ($5.4 \times 10^{-7}$ cm$^3$ g$^{-1}$; calculated from molar magnetic solubility of dichloromethane)
- $d_o$ = density of solution (1.362 g cm$^{-3}$)
- $d_s$ = density of solvent (1.3606 g cm$^{-3}$)
XIV. B3LYP geometries and energies for all optimized minima and transition structures

Species A
HF energy = $-1691.10366444$
No imaginary frequency
Zero–point correction = 0.347339 (Hartree/Particle)
Thermal correction to Energy = 0.371984
Thermal correction to Enthalpy = 0.372928
Thermal correction to Gibbs Free Energy = 0.295084
Sum of electronic and zero–point Energies = $-1690.756325$
Sum of electronic and thermal Energies = $-1690.731681$
Sum of electronic and thermal Enthalpies = $-1690.730736$
Sum of electronic and thermal Free Energies = $-1690.808580$

Coordinates: A

|   |   |   |   |
|---|---|---|---|
| Cl | 8.57840000 | 3.96570000 | 6.39970000 |
| Ni | 10.09160000 | 3.12510000 | 7.78110000 |
| N  | 11.46530000 | 2.26390000 | 8.76260000 |
| P  | 11.33100000 | 4.87390000 | 7.23970000 |
| N  | 8.86800000  | 1.76460000 | 8.43260000 |
| C  | 8.02800000  | 1.28560000 | 7.58010000 |
| H  | 12.57110000 | 1.81010000 | 8.11610000 |
| H  | 12.60110000 | 1.98670000 | 7.03440000 |
| C  | 13.60580000 | 1.15530000 | 8.78050000 |
| H  | 14.47510000 | 0.79780000 | 8.21920000 |
| C  | 13.48910000 | 0.97980000 | 10.17040000|
| H  | 14.28810000 | 0.48570000 | 10.73690000|
| C  | 12.33100000 | 1.42400000 | 11.88860000|
| H  | 12.20180000 | 1.27390000 | 11.88860000|
| C  | 11.27980000 | 2.05630000 | 10.10280000|
| B  | 9.82950000  | 2.43330000 | 10.76000000|
| C  | 9.73430000  | 1.95220000 | 12.32250000|
| H  | 9.90600000  | 0.86790000 | 12.48920000|
| H  | 8.74100000  | 2.18300000 | 12.75780000|
| H  | 10.46530000 | 2.48940000 | 12.96110000|
| C  | 8.79510000  | 1.60090000 | 9.78890000 |
| C  | 7.14590000  | 0.13180000 | 8.04130000 |
| H  | 6.53350000  | -0.42230000| 7.32250000 |
| C  | 7.01610000  | -0.04930000| 9.42670000 |
| H  | 6.28350000  | -0.76000000| 9.82920000 |
| C  | 7.83880000  | 0.68480000 | 10.28560000|
| H  | 7.76140000  | 0.55920000 | 11.37020000|
| C  | 9.51970000  | 4.05380000 | 10.64360000|
Species B
HF energy = -1691.08876337
no imaginary frequency
Zero-point correction = 0.345759 (Hartree/Particle)
Thermal correction to Energy = 0.371132
Thermal correction to Enthalpy = 0.372076
Thermal correction to Gibbs Free Energy = 0.290622
Sum of electronic and zero-point Energies = -1690.743004
Sum of electronic and thermal Energies = -1690.717631
Sum of electronic and thermal Enthalpies = -1690.716687
Sum of electronic and thermal Free Energies = -1690.798141

Coordinates: B
Cl  9.87520000  2.74130000  5.67950000
Ni 10.08950000  3.20370000  7.91860000
N  11.54140000  2.04430000  8.63670000
P  11.18320000  5.19120000  7.36300000
N  8.65950000  1.98410000  8.65350000
C  7.61730000  1.55820000  7.90130000
H  7.61040000  1.90860000  6.86250000
C 12.55820000  1.61570000  7.85250000
H 12.51750000  1.93520000  6.80450000
C 13.57530000  0.79700000  8.34440000
H 14.38260000  0.46770000  7.68200000
C 13.51810000  0.41440000  9.69470000
H 14.30030000  0.22450000 10.12350000
C 12.44380000  0.84380000  10.47990000
H 12.36890000  0.54220000  11.52970000
C 11.42460000  1.66870000  9.94710000
|   |           |           |           |
|---|-----------|-----------|-----------|
| B | 10.10470000 | 2.16330000 | 10.78600000 |
| C | 10.13530000 | 1.64820000 | 12.33830000 |
| H | 10.17600000 | 0.54470000 | 12.45390000 |
| H | 9.24080000  | 1.98630000 | 12.89980000 |
| H | 11.00710000 | 2.05190000 | 12.89280000 |
| C | 8.81300000  | 1.58550000 | 9.95590000  |
| C | 6.63200000  | 0.70970000 | 8.40920000  |
| H | 5.80230000  | 0.39100000 | 7.76970000  |
| C | 6.74960000  | 0.28280000 | 9.74070000  |
| H | 5.99970000  | -0.38920000 | 10.17700000 |
| C | 7.84040000  | 0.71840000 | 10.50100000 |
| H | 7.95750000  | 0.39330000 | 11.54100000 |
| C | 10.00530000 | 3.81500000 | 10.73850000 |
| H | 10.90520000 | 4.30950000 | 11.15530000 |
| H | 9.12570000  | 4.17720000 | 11.30740000 |
| H | 9.87020000  | 4.25970000 | 9.71290000  |
| C | 10.09010000 | 6.34340000 | 6.41870000  |
| H | 9.23840000  | 6.65060000 | 7.05060000  |
| H | 10.63940000 | 7.24360000 | 6.08720000  |
| H | 9.69650000  | 5.79980000 | 5.54290000  |
| C | 12.62100000 | 4.96950000 | 6.23600000  |
| H | 12.29760000 | 4.37470000 | 5.36760000  |
| H | 13.02380000 | 5.94430000 | 5.89740000  |
| H | 13.42700000 | 4.41730000 | 6.76050000  |
| C | 11.87120000 | 6.26770000 | 8.70950000  |
| H | 11.06420000 | 6.55750000 | 9.40410000  |
| H | 12.62290000 | 5.70070000 | 9.28580000  |
| H | 12.34210000 | 7.17830000 | 8.29610000  |

Species TSBB'

HF energy = -1691.08394573
Imaginary frequency: 83i
Zero-point correction = 0.345215 (Hartree/Particle)
Thermal correction to Energy = 0.370008
Thermal correction to Enthalpy = 0.370952
Thermal correction to Gibbs Free Energy = 0.291136
Sum of electronic and zero-point Energies = -1690.738731
Sum of electronic and thermal Energies = -1690.713938
Sum of electronic and thermal Enthalpies = -1690.712994
Sum of electronic and thermal Free Energies = -1690.792810

Coordinates: TSBB'
Cl  2.25980000  1.65030000  0.00000000
Ni  0.18750000  0.64680000  0.00000000
N   0.38820000  -0.72730000  1.42600000
| Atom | X       | Y       | Z       |
|------|---------|---------|---------|
| P    | -0.7881 | 2.8298  | 0.0000  |
| N    | 0.3882  | -0.7273 | -1.4260 |
| C    | 1.2229  | -0.5111 | -2.4710 |
| H    | 1.7901  | 0.4261  | -2.4388 |
| C    | 1.2229  | -0.5111 | 2.4710  |
| H    | 1.7901  | 0.4261  | 2.4388  |
| C    | 1.3690  | -1.4380 | 3.5038  |
| H    | 2.0523  | -1.2284 | 4.3332  |
| C    | 0.6294  | -3.7681 | 0.0000  |
| H    | -0.7909 | -3.6810 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
| H    | 2.0523  | -1.2284 | -4.3332 |
| C    | 0.6294  | -2.6290 | -3.4320 |
| H    | 0.7181  | -3.3861 | 4.2213  |
| C    | -0.2119 | -2.8427 | 2.3356  |
| H    | -0.7909 | -3.7681 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
| H    | 2.0523  | -1.2284 | -4.3332 |
| C    | 0.6294  | -2.6290 | -3.4320 |
| H    | 0.7181  | -3.3861 | 4.2213  |
| C    | -0.2119 | -2.8427 | 2.3356  |
| H    | -0.7909 | -3.7681 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
| H    | 2.0523  | -1.2284 | -4.3332 |
| C    | 0.6294  | -2.6290 | -3.4320 |
| H    | 0.7181  | -3.3861 | 4.2213  |
| C    | -0.2119 | -2.8427 | 2.3356  |
| H    | -0.7909 | -3.7681 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
| H    | 2.0523  | -1.2284 | -4.3332 |
| C    | 0.6294  | -2.6290 | -3.4320 |
| H    | 0.7181  | -3.3861 | 4.2213  |
| C    | -0.2119 | -2.8427 | 2.3356  |
| H    | -0.7909 | -3.7681 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
| H    | 2.0523  | -1.2284 | -4.3332 |
| C    | 0.6294  | -2.6290 | -3.4320 |
| H    | 0.7181  | -3.3861 | 4.2213  |
| C    | -0.2119 | -2.8427 | 2.3356  |
| H    | -0.7909 | -3.7681 | 2.2514  |
| C    | -0.3402 | -1.8797 | 1.3084  |
| H    | -1.3122 | -2.0542 | 0.0000  |
| C    | -2.1102 | -3.4815 | 0.0000  |
| H    | -1.4505 | -4.3746 | 0.0000  |
| H    | -2.7733 | -3.5838 | -0.8831 |
| H    | -2.7733 | -3.5838 | 0.8831  |
| C    | -0.3402 | -1.8797 | -1.3084 |
| C    | 1.3690  | -1.4380 | -3.5038 |
XV. Comparison of Selected Bond Distances and Angles for Compounds 4/A

**Table S-XV.1. Comparison and Measured and Calculated Geometries.**

| Metric | Diffraction data | DFT data |
|--------|------------------|----------|
| Ni(1)–P(1) | 2.2260(9)        | 2.2108   |
| Ni(1)–Cl(1) | 2.1742(9)        | 2.2146   |
| Ni(1)–N(1) | 1.893(3)         | 1.895    |
| Ni(1)–N(2) | 1.945(2)         | 1.942    |
| P(1)–Ni(1)–N(2) | 170.48(8) | 171.65   |
| P(1)–Ni(1)–N(1) | 94.08(8)      | 94.58    |
| P(1)–Ni(1)–Cl(1) | 86.82(3)        | 85.99    |
| Cl(1)–Ni(1)–N(1) | 169.87(9) | 172.19   |
| Cl(1)–Ni(1)–N(2) | 92.31(8)       | 92.56    |
| N(1)–Ni(1)–N(2) | 88.44(10)       | 87.97    |

XVI. References

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