Electronic Supplementary Information (ESI)

# An integrated approach towards extracting structural characteristics of chlorosomes from a *bchQ* mutant of *Chlorobaculum tepidum*

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Table S1 $^{13}C$ chemical shifts for chlorosomes of *bchQ* from *Cba. tepidum* in ppm: liquid $\sigma_{liq}^{C}$, solid-state $\sigma_{s}^{C}$ and aggregation shifts $\Delta\sigma_{s}^{C} = \sigma_{s}^{C} - \sigma_{liq}^{C}$

| Position | $\sigma_{liq}^{C}$ (Balaban et al. 1995)[1] | $\sigma_{s}^{C}$ chlorosomes | $\Delta\sigma_{s}^{C}$ chlorosomes |
|---|---|---|---|
| 1-C | 153.79 | 154.1 | 0.3 |
| 2-C | 135.08 | 134.9 | -0.2 |
| 3-C | 145.13 | 139.0 | -6.2 |
| 4(I)-C | 145.37 | 145.3 | -0.1 |
| 4(II)-C | | 144.1 | |
| 5(I)-CH | 100.03 | 96.0 | -4.1 |
| 5(II)-CH | | 102.3 | |
| 6(I)-C | 150.72 | 150.8 | 0.1 |
| 6(II)-C | | 150.5 | |
| 7(I)-C | 133.45 | 130.2 | -3.2 |
| 7(II)-C | | 131.7 | |
| 8-C | 143.39 | 143.7 | 0.3 |
| 9-C | 146.01 | 147.3 | 1.2 |
| 10-CH | 105.57 | 106.4 | 0.8 |
| 11-C | 146.47 | 147.4 | 0.9 |
| 12-C | 140.65 | 140.6 | -0.1 |
| 13-C | 130.26 | 130.5 | 0.2 |
| 14-C | 161.24 | 161.8 | 0.6 |
| 15-C | 104.74 | 104.3 | -0.4 |
| 16-C | 153.98 | 154.5 | 0.5 |
| 17-CH | 50.07 | 50.0 | 0.0 |
| 18-CH | 47.64 | 48.3 | 0.6 |
| 19(I)-C | 167.76 | 171.0 | 3.2 |
| 19(II)-C | | 168.2 | |
| 20(I)-C | 104.74 | 105.6 | 0.9 |
| 20(II)-C | | 103.9 | |
| $2^1$-$CH_3$ | 16.76 | 14.4 | -2.3 |
| $3^1$(I)-CH | 64.95 | 63.9 | -1.1 |
| $3^1$(II)-CH | | 63.7 | |
| $3^2$(I)-$CH_3$ | 25.43 | 22.3 | -3.1 |
| $3^2$(II)-$CH_3$ | | 25.4 | |
| $7^1$(I)-$CH_3$ | 10.37 | 6.2 | -4.1 |
| $7^1$(II)-$CH_3$ | | 11.2 | |
| $8^1$-$CH_2$ | 19.14 | 19.9 | 0.7 |
| $8^2$-CH3 | 16.97 | 19.9 | 2.9 |

| | | | |
|---|---|---|---|
| $12^1$-$CH_2$ | 20.77 | 18.4 | -2.4 |
| $12^2$-$CH_3$ | 16.56 | 18.4 | 1.9 |
| $13^1$(I)-C | 197.49 | 196.2 | -1.3 |
| $13^1$(II)-C | | | |
| $13^2$-$CH_2$ | 48.28 | 48.7 | 0.4 |
| $17^1$-$CH_2$ | 29.6 | 29.9 | 0.3 |
| $17^2$-$CH_2$ | 30.6 | 29.9 | -0.7 |
| $17^3$-C | 173.62 | 173.2 | -0.5 |
| $18^1$-CH | 20.36 | 21.4 | 1.0 |
| $20^1$-$CH_3$ | 20.96 | 21.5 | 0.5 |
| F1-$CH_2$ | 61.14 | | |
| F2-CH | 117.5 | | |
| F3-C | 142.23 | 140.9 | -1.3 |
| F4-$CH_2$ | 39.22 | | |
| F5-$CH_2$ | 26.23 | 26.2 | 0.0 |
| F6-CH | 123.19 | 123.9 | 0.7 |
| F7-C | 134.98 | | |
| F8-$CH_2$ | 39.02 | 39.6 | 0.5 |
| F9-$CH_2$ | 25.7 | 24.9 | -0.8 |
| F10-CH | 123.87 | | |
| F11-C | 130.82 | | |
| F12-$CH_3$ | 25.04 | | |
| $F3^1$-$CH_3$ | 15.79 | 16.1 | 0.3 |
| $F7^1$-$CH_3$ | 15.35 | | |
| $F11^1$-$CH_3$ | 17.03 | | |

*I and II correspond to two components present in the system. A lower in chemical shift value is considered as component (I) and which has a higher chemical shift value is considered component (II)

**Blank space indicates that no chemical shift values could be derived from the experiment.

***Aggregation shifts that is > |1.5| ppm is seen for carbon resonances such as 3-C, 5-C, 7-C, 19-C, $2^1$-C, $3^2$-C, $7^1$-C, $8^2$-C, $12^1$-C, $12^2$-C, $13^1$-C, F3-C. These mostly originate from rings 1 and 3.

Table S2. $^1$H chemical shifts for chlorosomes of *bchQ* from *Cba. tepidum* in ppm: liquid $\sigma_{liq}^H$, solid-state $\sigma_s^H$ and aggregation shifts $\Delta\sigma_s^H = \sigma_s^H - \sigma_{liq}^H$.

| Position | $\sigma_{liq}^H$(van Rossum et al. 2001)[2] | $\sigma_s^H$ chlorosomes | $\Delta\sigma_s^H$ chlorosomes |
|---|---|---|---|
| $2^1$ (I) - $H_3$ | 3.3 | 0.7 | -2.6 |
| $2^1$ (II) - $H_3$ | | 1.4 | |
| $3^1$(I)- H | 6.25 | 3.4 | -2.8 |
| $3^1$(II) - H | | | |

| | | | |
|---|---|---|---|
| $3^2$(I) - $H_3$ | 2.01 | 1.1 | -0.9 |
| $3^2$(II) - $H_3$ | | | |
| 5(I) - H | 9.56 | 6.9 | -2.7 |
| 5(II) - H | 9.56 | 7.4 | -2.1 |
| $7^1$(I) - $H_3$ | 3.19 | -0.7 | -3.9 |
| $7^1$(II) - $H_3$ | 3.19 | 3.3 | 0.1 |
| $8^1$ - $H_2$ | 3.67 | 1.0 | -2.7 |
| $8^2$ - $H_3$ | 1.62 | | |
| 10 - H | 9.43 | 9.8 | 0.4 |
| $12^1$ - $H_2$ | 3.97 | 2.4 | -1.6 |
| $12^2$ - $H_3$ | 1.79 | | |
| $13^2$ - $H_2$ | 5.09 | 5.0* | -0.1 |
| 17 - H | 4.08 | 4.8 | 0.7 |
| $17^1$ - $H_2$ | 2.00/2.20 | 2.1 | 0.1/-0.1 |
| $17^2$ - $H_2$ | 2.35/2.42 | 2.1 | -0.2/-0.3 |
| 18(I) - H | 4.52 | 5.0* | 0.5 |
| 18(II)- H | 4.52 | | -4.5 |
| $18^1$(I) - $H_3$ | 1.43 | 3.8* | 0.1 |
| $18^1$(II) - $H_3$ | 1.43 | | -1.4 |
| $20^1$ - $H_3$ | 3.72 | 3.8* | |
| F1 - $H_2$ | 4.31 | 4.0 | -0.3 |
| F2 - H | 5.05 | 6.8 | 1.7 |
| F4 - $H_2$ | 1.88 | 2.9 | 1.0 |
| F5 - $H_2$ | 1.88 | | |
| F6 - H | 4.95 | | |
| $F7^1$- $H_3$ | 1.5 | | |
| F8 - $H_2$ | 1.88 | 1.6 | -0.3 |
| F9 - $H_2$ | 1.88 | | |
| F10 - H | 4.95 | | |
| $F11^1$ - $H_3$ | 1.5 | | |
| F12 - $H_3$ | 1.54 | 1.4 | -0.1 |

Asterisk (*) indicate that their chemical shifts for carbon resonances resonate at almost same frequency.

Table S3. Signals originating from chlorosomes in INEPT spectra

| Position | Chemical shift (ppm) |
|---|---|
| F10-CH | 124.0 |
| F1-$CH_2$ | 60.8 |
| 17-CH | 49.4 |
| 18-CH | 47.7 |
| F4-$CH_2$ | 39.4 |
| $17^2$-$CH_2$ | 31.7 |
| $17^1$-$CH_2$ | 29.9 |

| | |
|---|---|
| F5-$CH_2$ | 26.2 |
| $3^2$-$CH_2$ | 22.1 |
| $8^1$-$CH_2$ | 19.1 |
| $8^2$-$CH_3$ | 16.6 |
| $F3^1$-$CH_3$ | 15.6 |
| $2^1$-$CH_3$ | 13.7 |
| $7^1$-$CH_3$ | 10.8 |

Table S4 2D experimental parameters

| Experiment | Dipolar 2D hCH[3,4] |
|---|---|
| ns | 32 |
| ds | 8 |
| d1(s) | 1.2 |
| $^1$H 90$^o$ (kHz) | 105 |
| $^{13}$C 90$^o$ (kHz) | 51 |
| $^1$H 90$^o$ (kHz) Back CP | 114 |
| $^{13}$C 90$^o$ (kHz) Back CP | 44.25 |
| $^1$H dec (kHz) | 15 |
| $^{13}$C dec (kHz) | 15 |
| Water suppression (kHz) | 17 |
| Mississippi[5] | |
| MAS | 60 |
| Set T | 245 |
| Decoupling | PISSARRO[6] |

List of abbreviations:

| | |
|---|---|
| ns | Number of scans |
| ds | dummy scans |
| $^1$H/$^{13}$C 90$^o$ | hard 90$^o$ pulses applied on both channels |
| $^1$H/$^{13}$C dec | applied on both channels during decoupling |
| MAS | Magic Angle Spinning rate |
| Set T | set Temperature |
| d1 | recycle delay |

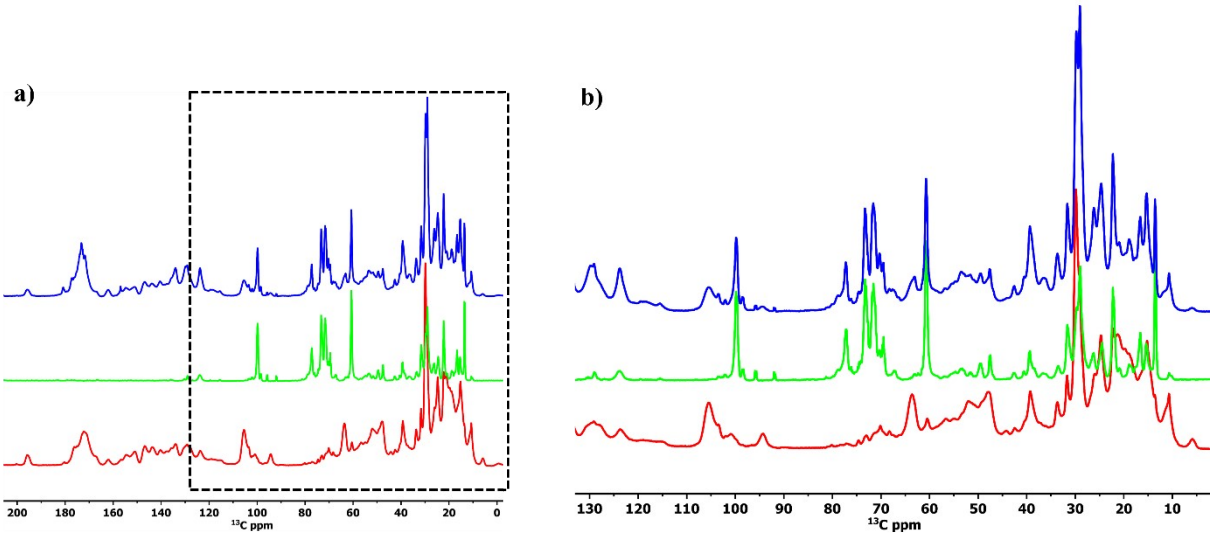


Figure S1: a) Overlaid CP (Red), DP(Blue) and INEPT(Green) spectra of chlorosomes of *bchQ* at 277K spinning at 20kHz. b) Enlarged image in boxed area of a).

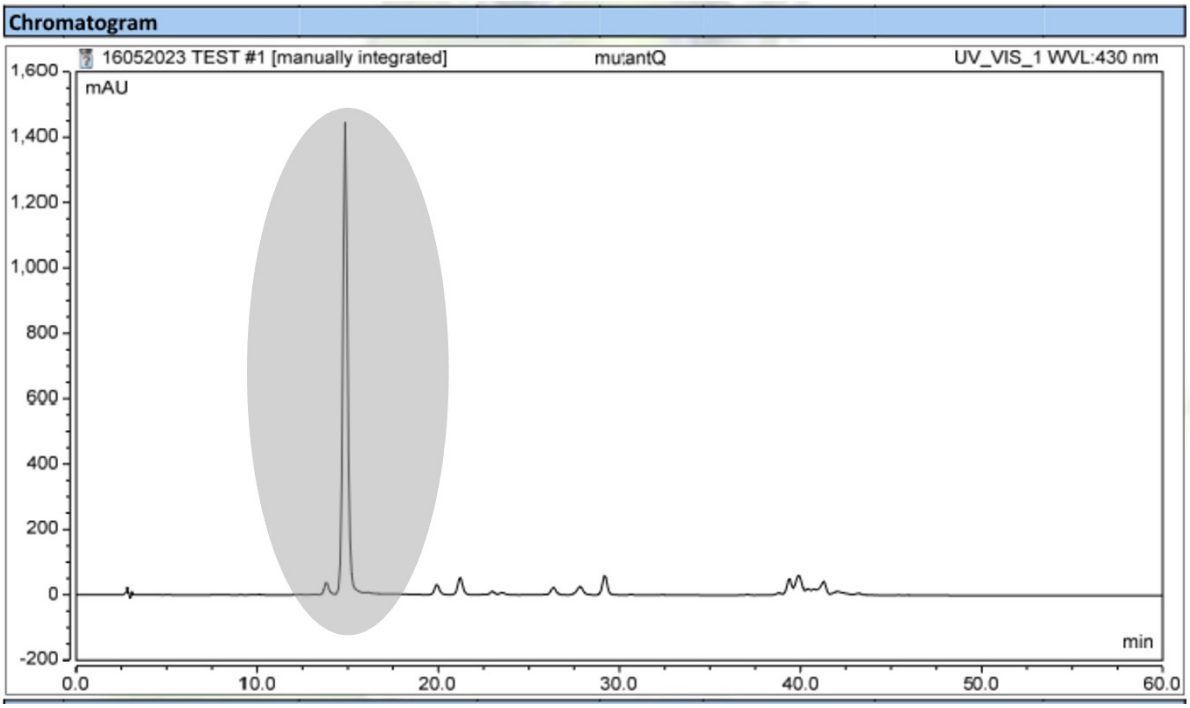


Fig S2: Chromatogram of *bchQ* which shows 2 signals between 10-20 minutes, highlighted in grey. Smaller peak corresponds to 5% [Mt, Et] and the larger peak corresponds to 95%[Et, Et]

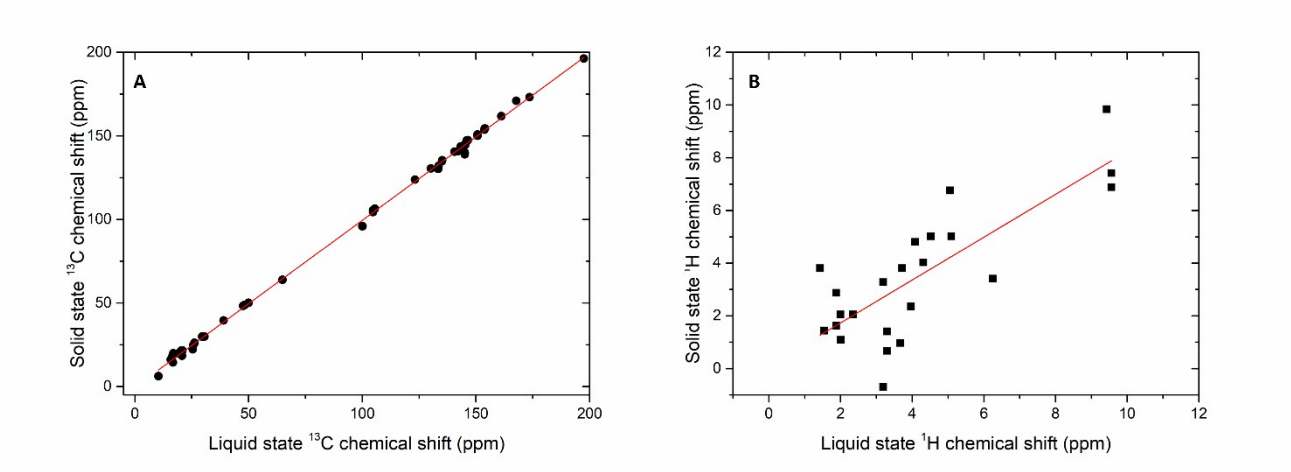


Figure S3: Chemical shift correlation plots of *bchQ* chlorosomes. The $^{13}$C and $^{1}$H shifts in the solid-state NMR data are plotted against the monomer shifts in the solution-state NMR data. The solid line represents the diagonal. The filled circles and squares representing $^{13}$C and $^{1}$H respectively deviating from the solid line correspond to the aggregation shifts due to neighboring BChl molecules.

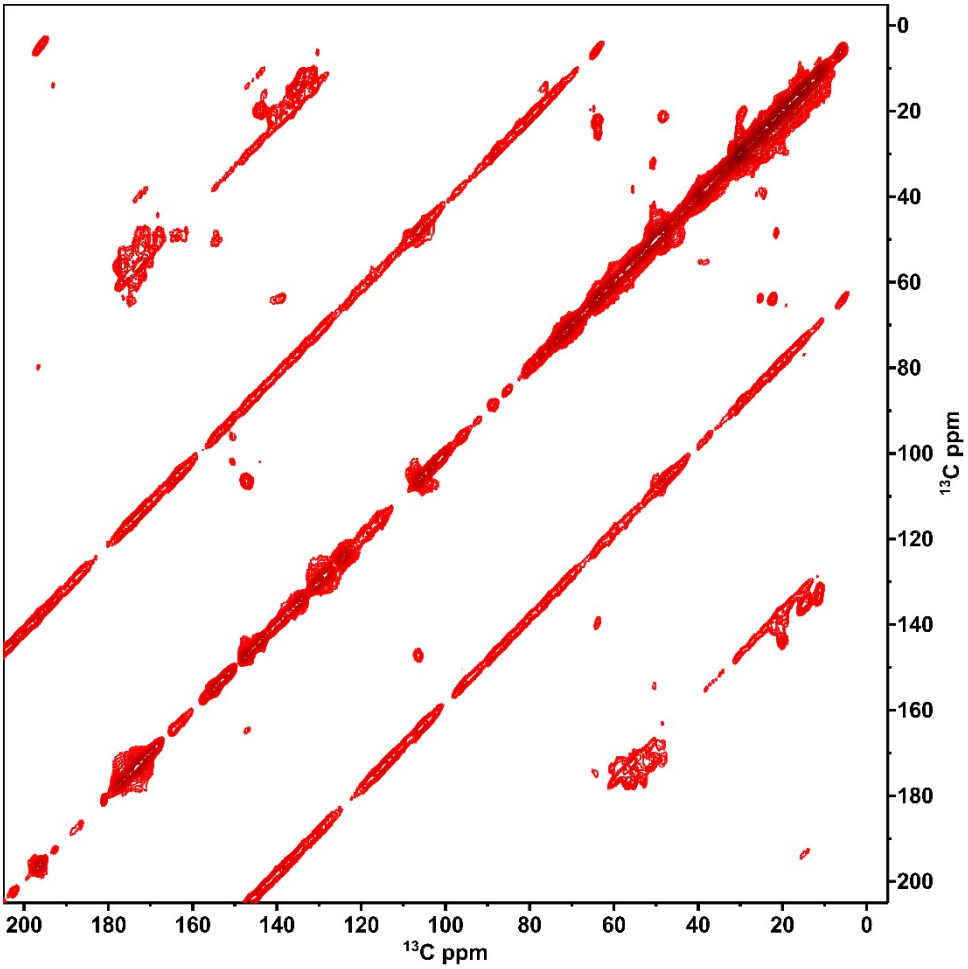


Figure S4 $^{13}$C-$^{13}$C RFDR spectra of *bchQ* recorded at 277K, 11kHz, 3.2 ms mixing time. The spectra show short range intramolecular correlations.

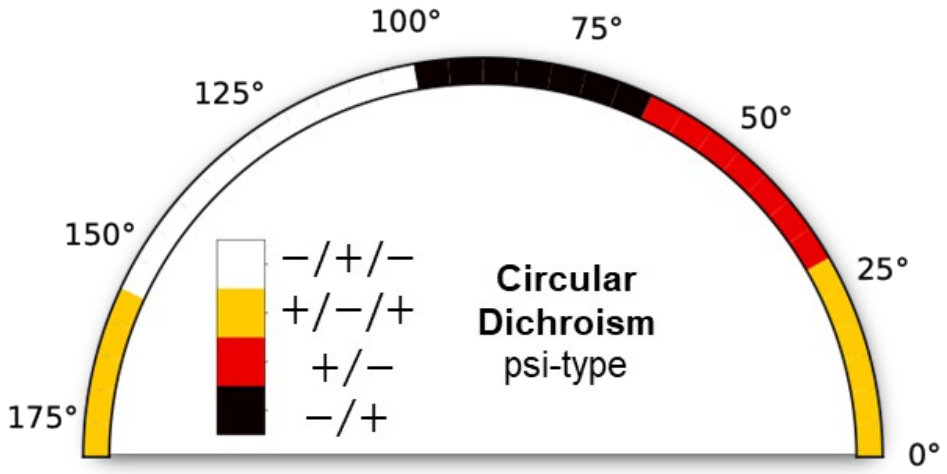


Figure S5: Polar plot of spectral features for CD as a function of chiral angle δ. The resolution for δ is 5$^{o}$. Figure is drawn according to Li *et al* taking into account the reversed signs in the direction of opposite curvature.[7]

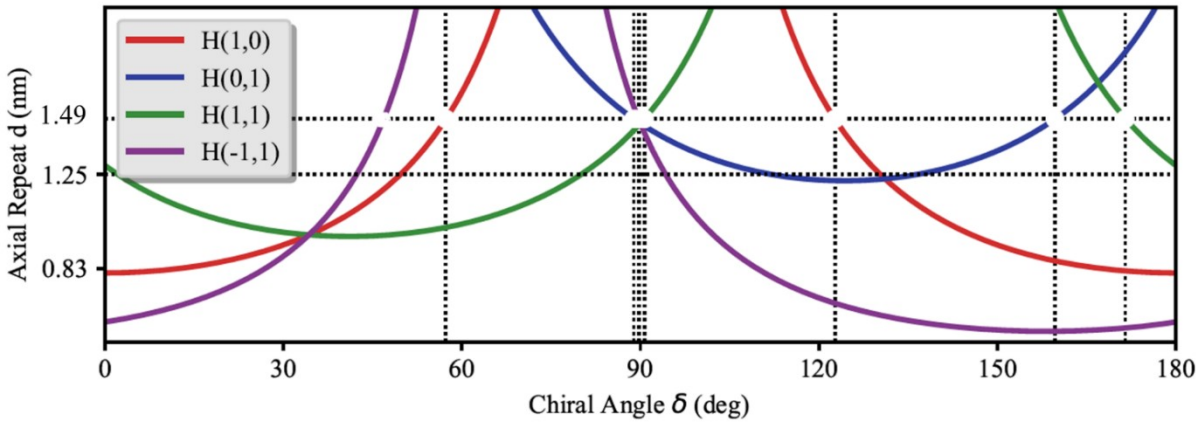


Figure S6 The axial repeat d as a function of the chiral angle δ ϵ [0, 180] corresponds to the H(1, 0), H(0, 1), H(1, 1) and H(-1, 1) helical families. Since we are targeting the experimentally determined axial repeat distance d-1.49 nm for *bchQ*, an additional dotted line was drawn. The lattice parameters are set to (1.48, 0.98, 124.3$^{o}$) as determined from MD simulations of BChl *c* tubular assemblies to evaluate the geometrical relations. Matching chiral angles are highlighted by white circles.

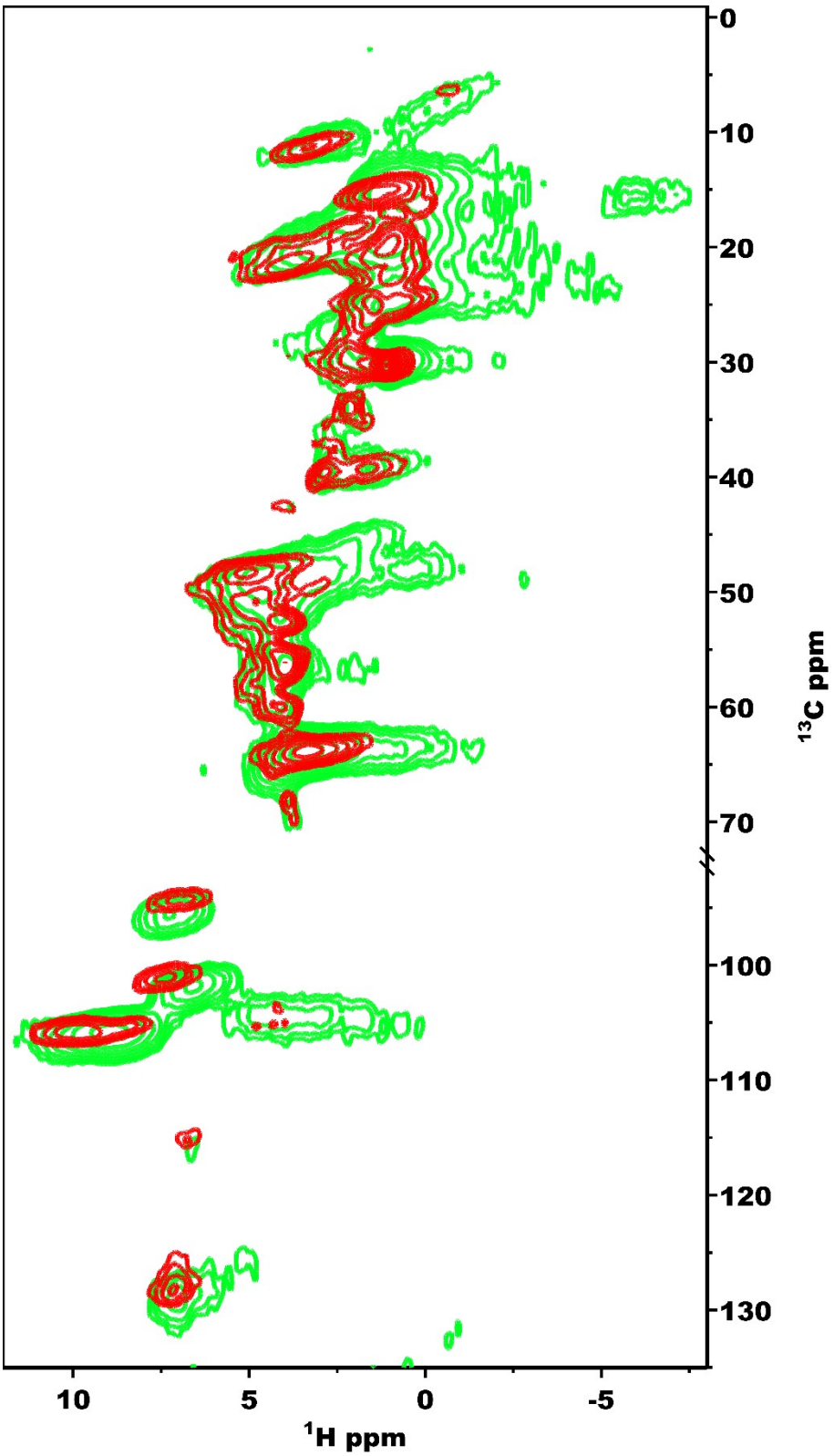


Figure S7 is a dipolar hCH spectra of WT (Green) and *bchQ* (Red), recorded in a 1.2 GHz magnet spinning at 60kHz. The cross peaks for the *bchQ* are well resolved in comparison to WT revealing the heterogeneity of chlorosomes of the WT

## References


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