SUPPORTING INFORMATION

Detection of remote proton-nitrogen correlations by ¹H-detected ¹⁴N overtone solid-state NMR at fast MAS

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Figure S1. Hist-B: a-d) the 2D ¹H/¹⁴N *D*-HMQC spectra and e-h) the extracted ¹⁴N slices at ¹H chemical shift of 6.5 ppm (H6, vertical lines in a-d)). These spectra were acquired at τ_{mix} of 0.768 ms (4 loops of SR4), 1.152 ms (6 loops of SR4), 1.536 ms (8 loops of SR4), and 2.304 ms (12 loops of SR4) (from left to right). The other experimental conditions were the same as those in Fig. 2a except the number of scans were 24 for a-c and 128 for d. The positive contour signals are presented in black while the negative ones are presented in red.



Figure S2. Hist-B: the 2D ¹H-¹⁴N OT correlation map. The experimental conditions are identical to those in Fig. 3a except the ¹⁴N OT shift increment step of 5 ppm. On the far right is the skyline projection of ¹⁴N OT shift array with an increment step of 20 ppm from Fig. 3a for comparison. On the far left is the ¹⁴N OT shift at the central band (n = 0).



Figure S3. Hist-B: the 2D ¹H-¹⁴N OT correlation maps at τ_{CW} of 0.064 ms (a), 0.128 ms (b), 0.192 ms (c), and 0.256 ms (d). For d), the * denotes a sideband of ¹⁴NH₃ OT peak. Both S_0 and S' were alternatively obtained for each ¹⁴N OT frequency. The other experimental conditions are identical to those in Fig. 3a. The vertical lines denote the ¹H chemical shift where ¹⁴N OT shift arrays are extracted. On the far left is the ¹⁴N OT shift at the central band (n = 0).



Figure S4. Hist-B: the 2D ¹H-¹⁴N OT correlation maps with ¹⁴N OT rf-field of a) full strength (100 %: 125 kHz) and b) half strength (50 %: 63 kHz). The other experimental conditions are identical to those in Fig. 3a. On the far left is the ¹⁴N OT shift at the central band (n = 0).