Supporting Information

Structural Dynamics by NMR in the Solid-State: The Unified MOMD Perspective Applied to Organic Frameworks with Interlocked Molecules

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Figure S1. (a) Linkers 7-22, 7-24 and 7-B24, with synthesis depicted in Scheme 1 of ref 1. (b) View down the c axis of the X-ray structure of UWDM-1 with 22C6, depicting the hexagonal shaped channels (framework shown in blue, macrocycles in red, and Cu²⁺ metal ions in green. (c) 24C6 MIM pillaring inside UWDM-2 and UWDM-3. (d) Ball-and-stick representations of UWDM-2 (top) and UWDM-3 (bottom) showing the interpenetrating lattice frameworks (UWDM-2 is three-fold interpenetrated, and UWDM-3 is two-fold interpenetrated). Space-filling models show the position of the macrocycle relative to the square framework grid. Color-key: blue – aniline axle, red – 24C6 wheel, yellow – carboxylate linkers, green – zinc atoms. (parts a and b): Reproduced with permission from ref 1. Copyright 2015 American Chemical Society. (parts c and d): Reproduced with permission from ref 2: Copyright 2014 American Chemical Society.



Figure S2. (a) Experimental ²H lineshapes of UWDM-1 with 24C6 as a function of temperature. (b) Experimental ²H lineshapes of β -UWDM-3 with 24C6 as a function of temperature. (c) Experimental ²H lineshapes of α -UWDM-3 with 24C6 as a function of temperature. (d) Experimental ²H lineshapes of UWDM-2 with 24C6 as a function of temperature. Reproduced with permission from ref 2. Copyright 2014 American Chemical Society.



Figure S3. Experimental ²H lineshapes of UWDM-1 with 24C6 (**a**), 22C6 (**b**), and B246 (**c**) as a function of temperature. Reproduced with permission from ref 1. Copyright 2015 American Chemical Society.



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Figure S4. (row 1) Experimental ²H lineshapes of UWDM-1 with 24C6 at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. "FML" stands for "fast-motion limit". Reproduced with permission from ref 1. Copyright 2015 American Chemical Society. (row 2) Experimental ²H lineshapes of UWDM-5 with 24C6 at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. Reproduced with permission from ref 3. Copyright 2016 American Chemical Society. (row 3) Experimental ²H lineshapes of β -UWDM-3 with 24C6 at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. Reproduced with permission from ref 2. Copyright 2014 American Chemical Society. (row 4) Experimental ²H lineshapes of α -UWDM-3 with 24C6 at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. Reproduced with permission from ref 2. Copyright 2014 American Chemical Society. (row 5) Experimental ²H lineshapes of UWDM-2 with 24C6 at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. Reproduced with permission from ref 2. Copyright 2014 American Chemical Society.



Figure S5. Experimental ²H lineshapes of UWDM-1 with 24C6 (left), 22C6 (middle) and B24C6 (right) at the temperatures depicted, the motional modes used to reproduce them, and the corresponding bets-fit parameters. "FML" stands for "fast-motion limit". Reproduced with permission from ref 1. Copyright 2015 American Chemical Society.



Figure S6. **(A)** Experimental ²H lineshapes of UWDM-1 with 24C6, **(B)** corresponding best-fit MSM spectra, and **(C)** the following motional models used: (i) undetectably slow motions, (ii) two-site jumps with jump-axis tilted at 77° from the C–D bond, (iii) two-site jumps with jump-axis tilted at 60° from the C–D bond combined with partial rotation of the ring over 225° in 45° steps, and (iv) two-site jumps with jump-axis tilted at 70° from the C–D bond combined with permission from the C–D bond combined with full rotation of the ring. Reproduced with permission from ref 1. Copyright 2015 American Chemical Society.



Figure S7. Experimental (blue) and calculated with MSM (red) ²H lineshapes of UWDM5 with 24C6 as a function of temperature. Reproduced with permission from ref 3. Copyright 2016 American Chemical Society.

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