## Supplementary information



Figure S1) Detailed pulse sequences used for the 2D full-volume images. a) represents the experiment with linear inversion  $\beta$  sweeps during detection and HS inversion sweeps for the Hadamard encoding (truncation  $\beta$ =6), which was used for 2D full-volume imaging in ref S1. b) shows the pulse sequence with lorentzian inversion sweeps used for Hadamard encoding and detection.

The basic scheme of the MRFM pulse sequence was described <sup>10</sup> in detail in ref S1. Here we focus on the differences in implementing the 2D full-volume pulse sequence with linear inversion sweeps or Lorentzian sweeps.

Figure S1 shows the detailed pulse sequences used for the 2D full-volume images. Figure S1a represents the experiment

- <sup>15</sup> with linear inversion sweeps during detection and HS inversion sweeps for the Hadamard quadrature encoding<sup>S2</sup> (truncation  $\beta$ =6). Figure S1b shows the pulse sequence with Lorentzian inversion sweeps used for Hadamard encoding and detection are presented. The truncation factor ( $\beta$ =40) was
- <sup>20</sup> optimized experimentally, schematically indicated in the AM line of figure S1b with higher maximal amplitude and sharper shaped inversion sweeps. For both experiments the average B<sub>1</sub> amplitude over the time period of one inversion cycle was kept at 11mT, the maximal average amplitude supported by
- <sup>25</sup> our micro rf coil. Although conventional Hadamard quadrature detection<sup>S2</sup> is not possible anymore with the Lorentzian inversion sweeps the SNR gain per unit time is ca. a factor six.
- The MRFM probe, spectrometer and software have been <sup>30</sup> described before.<sup>S3</sup> The probe works at room temperature under vacuum conditions (10<sup>-3</sup> mbar) in a 6.0 T wide-bore NMR magnet. The cantilever is oriented orthogonal to the main field and static gradient. Boltzmann polarization is detected in the amplitude mode.<sup>S4</sup> To allow the B<sub>1</sub> gradient to <sup>35</sup> encode the sample independently of the static B<sub>0</sub> gradient we
- used a piezo bender actor to move the static  $B_0$  gradient 2 mm

away from the sample during encoding pulses. The technical piezo bender details are described in <sup>S1</sup>

The polymer blend sample was cut out manually with a  $_{40}$  scalpel from a 30  $\mu$ m thick polymer film. And then glued to the tip of a custom-made cantilever  $^{19}$  (k=420  $\mu$ N/m, f<sub>c</sub>=900 Hz and Q~8k).

As a gradient source for the  $B_0$  field we used an iron cobalt (saturation magnetization of 2.3 T) cylinder of ~380  $\mu$ m <sup>45</sup> diameter and 10 mm length. The  $B_1$  field was generated with a ~220  $\mu$ m inner diameter solenoid coil. The mathematical description of the two fields can be found in <sup>S1</sup>.

The 2D image taken with the Lorentz Hadamard 8 was resolved by recording 32 and 96 image points in x and z <sup>50</sup> direction for both nuclei. The quadrature Hadamard 8 <sup>S2</sup> image was recorded with 32 and 98 points in x and z direction for both nuclei. The 32 points in the x direction were encoded by incrementing the hard nutation pulse in 32 steps of 600  $\mu$ s. The nutation frequency is  $\omega_1/2\pi \approx 400$  kHz at the sample scentre, with a B<sub>1</sub> gradient ranging from 200-600 kHz over the x-position in the sample. The acquisition period was set to 0.5 s. The mean value of rf amplitude over the sample during signal detection was ~75 kHz. Finally the x dimension was zero filled up to 512 points and Fourier transformed. Full <sup>60</sup> details of the image reconstruction are given in ref. S1.

The scans were repeated four times for the PEEK phase with the Lorentz Hadamard 8 and six times with the quadrature Hadamard 8,<sup>S2</sup> resulting in 200 h and 180 h measurement time respectively. This long acquisition time is

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mainly caused by the long proton  $T_1$  relaxation of PEEK in vacuum (ca. 25 s). For the PTFE phase twice as many averages than in the PEEK case were taken for both detection methods. Resulting in a measurement time of ca. 60 h and 56

s h for the Lorentz Hadamard and quadrature Hadamard technique  $^{\mathrm{S2}},$  respectively. The larger number of averages is needed due to the fast  $T_1$  relaxation of the PTFE (1.3 s), resulting in half of the signal amplitude being relaxed during the waiting time for the piezo bender movement.

## **10 References**

- S1 K. W. Eberhardt et al., Phys Rev. B, 2008, 78, 214401.
- S2 R. Joss, I. T. Tomka, J. D. van Beek, K. W. Eberhardt and B. H. Meier, *Phys Rev B*, 2011, **84**, 104408. 15 S3 Q. Lin et al., *Phys Rev. Lett..*, 2006, **96**, 137604.
- S4 D. Rugar et al. Science, 1994, 264, 1560.