### Table S1. Lattice parameters of szomolnokite at ambient pressure and temperature conditions.

| a (Å)    | b (Å)    | c (Å)    | β (degrees) | Volume (Å³) | ref.                        |
|----------|----------|----------|-------------|-------------|-----------------------------|
| 7.0823(2)| 7.5525(2)| 7.7786(5)| 118.631(3)  | 365.226(30) | Meusberger et al. (2019)    |
| 7.084    | 7.550    | 7.779    | 118.63      | 395.16      | Giester et al. (1994)       |
| 7.078(3) | 7.549(3) | 7.773(3) | 118.65(2)   | 364.48      | Wildner and Giester (1991)  |
| 7.123(9) | 7.469(9) | 7.837(9) | 118.65(2)   | 364.48      | Pistorius (1960)            |
| 7.09     | 7.55     | 7.78     | 118.65(2)   | 365.2(1)    | This study                  |
Table S2. Best fit hyperfine parameters for szomolnokite according to Model 1.

| Pressure (GPa) | 0° | 0 | 0.40(4) | 1.6(3) | 2.9(4) | 6.4(9) | 12(1) | 19(1) | 45(3) | 56(1) | 62(1) | 73.4(1) | 95(4) |
|---------------|----|---|---------|--------|--------|-------|-------|-------|-------|-------|-------|--------|-------|
| QS (mm/s)     |     |   |         |        |        |       |       |       |       |       |       |        |       |
| Site 1        | 2.670(3) | 2.642(3) | 2.64 | 2.79 | 2.79 | 2.83(2) | 2.795(2) | 2.838(6) | 2.96(4) | 2.843(9) | 2.89(2) | 3.08(3) | 3.05(2) |
| Site 2        | 2.07(3) | 2.306(13) | 1.95 | 1.51 | 1.51 | 2.178(8) | 2.01(7) | 2.187(4) | 2.41(3) | 2.10(2) | 2.18(4) | 2.23(2) | 2.24(2) |
| Site 3        | -     | -         | 1.16 | 1.27 | 1.27 | 1.06(19) | 1.216(32) | 2.76(3) | 1.78(2) | 1.841(5) | 1.88(1) | 1.80(2) | 1.718(7) |
| Site 4        | -     | -         | -    | -    | -    | -       | -       | 0.34(1) | -     | -     | -     | -     | -     |
| IS°, δIS1 (mm/s) |     |   |         |        |        |       |       |       |       |       |       |        |       |
| Site 1        | 1.231(7) | -         | -    | -    | -    | -       | -       | -     | -     | -     | -     | -     | -     |
| Site 2        | 0.98(2) | -0.339(12) | -0.16 | -0.16 | -0.16 | -0.069(6) | 0.02(1) | -0.061(6) | 0.048(1) | -0.054(4) | -0.071(5) | -0.042(7) | -0.102(6) |
| Site 3        | -     | -         | -0.35 | -0.4 | -0.4 | -0.241(5) | 0.32(1) | -0.1(1) | -0.72(2) | -0.796(3) | -0.794(4) | -0.74(2) | -0.84(1) |
| Site 4        | -     | -         | -    | -    | -    | -       | -       | -1.35(9) | -     | -     | -     | -     | -     |
| Weight Fraction |   |   |         |        |        |       |       |       |       |       |       |        |       |
| Site 1        | 0.83(6) | 0.91(66) | 0.647(9) | 0.625(7) | 0.612(5) | 0.643 | 0.643 | 0.51 | 0.463 | 0.522 | 0.475 | 0.409 | 0.368 |
| Site 2        | 0.17 | 0.09      | 0.144 | 0.153 | 0.159 | 0.2 | 0.2 | 0.27 | 0.21(4) | 0.15(1) | 0.21(2) | 0.30(2) | 0.28(2) |
| Site 3        | -     | -         | 0.209 | 0.221 | 0.229 | 0.157 | 0.157 | 0.16 | 0.24(1) | 0.242(6) | 0.2(1) | 0.223(7) | 0.29(7) |
| Site 4        | -     | -         | -    | -    | -    | -       | -       | 0.06 | -     | -     | -     | -     | -     |
| FWHM (mm/s)   |     |   |         |        |        |       |       |       |       |       |       |        |       |
| Site 1        | 0.116(4) | 0.061(2) | 0.085(3) | 0.093(4) | 0.119(4) | 0.095 | 0.095 | 0.15 | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 |
| Site 2        | 0.008(4) | 0.03(4) | 0.16(1) | 0.27(2) | 0.18(2) | 0.05 | 0.05 | 0.05 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |
| Site 3        | -     | -         | 0.5 | 0.5 | 0.5 | 0.2 | 0.2 | 0.2 | 0.3 | 0.3 | 0.3 | 0.3 | 0.3 |
| Site 4        | -     | -         | -    | -    | -    | -       | -       | -0.1 | -     | -     | -     | -     | -     |
| χ²            | 1.52 | 1.25      | 1.44 | 1.23 | 1.60 | 1.42 | 1.42 | 1.04 | 1.62 | 1.48 | 1.36 | 1.25 | 1.64 |

Notes: Best-fit values using CONUSS version 2.2.0 (Sturhahn, 2000; www.nrixs.com). Uncertainties are given at the 68% level. Values without reported errors were fixed. Pressures were determined using ruby fluorescence and the Dewaele et al. 2008 scale. Thickness was fixed at 3 µm and the Lamb-Mössbauer factor was fixed at 0.6 at all compression points. The isomer shifts of Sites 2 to 4 are given relative to Site 1 (see text for details), with the exception of the values for 0° GPa. Values from ambient pressure determined from a dual fit with a 10 µm thick naturally enriched stainless steel foil in the x-ray beam path with the sample, and isomer shift values are relative to α-iron.
Table S3. Best fit hyperfine parameters for szomolnokite according to Model 2.

| Pressure (GPa) | 0°  | 0    | 0.40(4) | 1.6(3) | 2.9(4) | 6.4(9) | 12(1) | 19(1) | 45(3) | 56(1) | 62(1) | 73.4(1) | 95(4) |
|---------------|-----|------|----------|--------|--------|--------|-------|-------|-------|-------|-------|---------|-------|
| QS (mm/s)     |     |      |          |        |        |        |       |       |       |       |       |         |       |
| Site 1        | 2.670(3) | 2.642(3) | 2.64    | 2.79   | 2.79   | 2.834(2) | 2.795(2) | 2.838(7) | 3.125(5) | 3.152(6) | 3.177(5) | 3.223(9) | 3.179(6) |
| Site 2        | 2.07(3)  | 2.31(1) | 1.95    | 1.51   | 1.51   | 2.177(8) | 2.009(7) | 2.187(4) | 2.64(5)  | 2.508(6) | 2.451(8) | 2.548(8) | 2.519(9)  |
| Site 3        | -     | -     | 1.16    | 1.27   | 1.27   | 1.06(2)  | 1.22(3)  | 2.76(3)  | 0       | 0      | 0       | 0       | 0       |
| Site 4        | -     | -     | -      | -      | -      | -       | -      | 0.34(1) | 1.12(1) | 0.983(9) | 1.12(1) | 1.086(8) | 1       |
| IS, ΔIS1 (mm/s) |     |      |        |        |        |        |       |       |       |       |       |         |       |
| Site 1        | 1.231(7) | -     | -      | -      | -      | -       | -      | -      | -      | -      | -      | -       | -      |
| Site 2        | 0.98(2)  | -0.34 | -0.16  | -0.16  | -0.16  | -0.069(5) | 0.03(1) | -0.061(6) | -0.07(5) | -0.066(6) | 0.02(6) | 0.042(6) | -0.13   |
| Site 3        | -     | -     | -0.35  | -0.4   | -0.4   | -0.241(5) | -0.32(1) | -0.13(2) | -0.449(5) | -0.456(4) | -0.429(8) | -0.473(5) | -0.435(4) |
| Site 4        | -     | -     | -      | -      | -      | -       | -      | -       | -       | -135(9) | -52(1) | -53(8)  | -47(1)  | -51(1)  | -463(7) |
| Weight Fraction |     |      |        |        |        |        |       |       |       |       |       |         |       |
| Site 1        | 0.83(6)  | 0.91(7) | 0.64(8) | 0.62(7) | 0.61(5) | 0.643   | 0.643  | 0.51   | 0.387  | 0.387  | 0.387  | 0.387  | 0.371   |
| Site 2        | 0.17    | 0.090  | 0.144   | 0.153  | 0.159  | 0.2     | 0.2    | 0.27   | 0.258  | 0.258  | 0.258  | 0.258  | 0.222   |
| Site 3        | -     | -     | 0.208   | 0.221  | 0.230  | 0.157   | 0.157  | 0.16   | 0.29   | 0.29   | 0.29   | 0.29   | 0.30(6) |
| Site 4        | -     | -     | -      | -      | -      | -       | -      | 0.06   | 0.065  | 0.065  | 0.065  | 0.065  | 0.111   |
| FWHM (mm/s)   |     |      |        |        |        |        |       |       |       |       |       |         |       |
| Site 1        | 0.116(4) | 0.061(1) | 0.085(3) | 0.093(3) | 0.119(4) | 0.095   | 0.095  | 0.15   | 0.4    | 0.4    | 0.4    | 0.5     | 0.5     |
| Site 2        | 0.008(4) | 0.02(4) | 0.16(1) | 0.27(2) | 0.18(2) | 0.05    | 0.05   | 0.05   | 0.3    | 0.3    | 0.3    | 0.3     | 0.3     |
| Site 3        | -     | -     | 0.5     | 0.5    | 0.5    | 0.2     | 0.2    | 0.2    | 0.6    | 0.6    | 0.6    | 0.6     | 0.47(6) |
| Site 4        | -     | -     | -      | -      | -      | -       | -      | 0.1    | 0.05   | 0.05   | 0.05   | 0.05   | 0.1     |

χ^2: 1.52 1.25 1.44 1.23 1.60 1.40 1.41 1.04 1.75 1.65 1.69 1.59 1.18

Notes: Best-fit values using CONUSS version 2.2.0 (Sturhahn, 2000; www.nrixs.com). Uncertainties are given at the 68% level. Values without reported errors were fixed. Pressures were determined using ruby fluorescence and the Dewaele et al. 2008 scale. Thickness was fixed at 3 µm and the Lamb-Mössbauer factor was fixed at 0.6 at all compression points. The isomer shift of Sites 2 to 4 are given relative to Site 1 (see text for details), with the exception of the values for 0° GPa. *Values from ambient pressure determined from a dual fit with a 10 µm thick naturally enriched stainless steel foil in the x-ray beam path with the sample, and isomer shift values are relative to α-iron.
Table S4. Best fit hyperfine parameters for szomolnokite according to Model 3.

| Pressure (GPa) | 0a | 0 | 0.40(4) | 1.6(3) | 2.9(4) | 6.4(9) | 12(1) | 19(1) | 45(3) | 56(1) | 62(1) | 73.4(1) | 95(4) |
|---------------|----|----|----------|---------|---------|--------|-------|-------|-------|-------|-------|---------|-------|
| QS (mm/s)     |     |    |          |         |         |        |       |       |       |       |       |         |       |
| Site 1        | 2.670(3) | 2.642(3) | 2.64 | 2.79 | 2.79 | 2.83(2) | 2.795(2) | 2.838(6) | 3.0(1) | 3.058(9) | 3.057(2) | 3.175(8) | 3.21(1) |
| Site 2        | 2.07(3) | 2.31(1) | 1.95 | 1.51 | 1.51 | 2.177(8) | 2.009(7) | 2.187(4) | 2.69(2) | 2.521(7) | 2.482(2) | 2.40(2) | 2.35(1) |
| Site 3        | - | - | 1.16 | 1.27 | 1.27 | 1.06(2) | 1.22(3) | 2.76(3) | 2.71(2) | 2.73(2) | 2.75(3) | 2.64(3) | 2.50(5) |
| Site 4        | - | - | - | - | - | - | - | 0.34(1) | 0.388(7) | 0.41(2) | 0.39(2) | 0.40(1) | 0.32(8) |
| Site 5        | - | - | - | - | - | - | - | 0 | 0 | 0 | 0 | 0 | 0 |
| IS, ΔIS1 (mm/s) |     |    |          |         |         |        |       |       |       |       |       |         |       |
| Site 1        | 1.23(1) | - | - | - | - | - | - | - | - | - | - | - | - |
| Site 2        | 0.98(2) | -0.34 | -0.16 | -0.16 | -0.16 | -0.069(5) | 0.025(10) | -0.061(6) | -0.07(1) | -0.019(7) | 0.01(1) | -0.03 | -0.029 |
| Site 3        | - | -0.35 | -0.4 | -0.4 | -0.241(5) | -0.32(1) | -0.13(1) | 0.32(1) | 0.37(1) | 0.41(2) | 0.32 | 0.29 |
| Site 4        | - | - | -0.35 | -0.4 | -0.393(7) | -0.42(4) | -0.59 | -0.603 |
| Site 5        | - | - | -0.245(5) | -0.02(1) | -0.03(2) | -0.07(2) | -0.074 | -0.078 |
| Weight Fraction |     |    |          |         |         |        |       |       |       |       |       |         |       |
| Site 1        | 0.83(6) | 0.91(7) | 0.64(8) | 0.62(7) | 0.61(5) | 0.643 | 0.643 | 0.51 | 0.406 | 0.406 | 0.401 | 0.406 | 0.406 |
| Site 2        | 0.17 | 0.090 | 0.144 | 0.153 | 0.159 | 0.2 | 0.2 | 0.27 | 0.297 | 0.297 | 0.297 | 0.297 |
| Site 3        | - | - | 0.208 | 0.221 | 0.230 | 0.157 | 0.157 | 0.16 | 0.158 | 0.129 | 0.12(9) | 0.099 | 0.05 |
| Site 4        | - | - | - | - | - | - | 0.06 | 0.099 | 0.099 | 0.098 | 0.099 | 0.099 |
| Site 5        | - | - | - | - | - | - | 0.04 | 0.069 | 0.088 | 0.099 | 0.149 |
| FWHM (mm/s)   |     |    |          |         |         |        |       |       |       |       |       |         |       |
| Site 1        | 0.116(4) | 0.061(1) | 0.085(3) | 0.093(3) | 0.119(4) | 0.095 | 0.095 | 0.15 | 0.4 | 0.4 | 0.4 | 0.4 | 0.42 |
| Site 2        | 0.008(4) | 0.02(4) | 0.161(1) | 0.27(2) | 0.18(2) | 0.05 | 0.05 | 0.05 | 0.3 | 0.3 | 0.3 | 0.3 | 0.32 |
| Site 3        | - | - | 0.5 | 0.5 | 0.5 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.22 |
| Site 4        | - | - | - | - | - | - | - | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | 0.12 |
| Site 5        | - | - | - | - | - | - | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.22 |
| $\chi^2$      | 1.52 | 1.25 | 1.44 | 1.23 | 1.60 | 1.42 | 1.41 | 1.04 | 1.38 | 1.48 | 1.43 | 1.49 | 1.28 |

Notes: Best-fit values using CONUSS version 2.2.0 (Sturhahn, 2000; www.nrixs.com). Uncertainties are given at the 68% level. Values without reported errors were fixed. Pressures were determined using ruby fluorescence and the Dewaele et al. 2008 scale. Thickness was fixed at 3 µm and the Lamb-Mössbauer factor was fixed at 0.6 at all compression points. The isomer shift of sites 2 to 5 are given relative to site 1 (see text for details), with the exception of the values for 0a GPa. Values from ambient pressure determined from a dual fit with a 10 µm thick naturally enriched stainless steel foil in the x-ray beam path with the sample, and isomer shift values are relative to $\alpha$-iron.
Table S5. Hydrated iron-bearing sulfates and their experimentally determined quadrupole and isomer shift values at ambient pressure and temperature.

| Mineral      | Formula       | Number (Fig. 2) | Sample          | IS (mm/s) | QS (mm/s) | Weight % |
|--------------|---------------|-----------------|-----------------|-----------|-----------|----------|
| Szomolnokite | FeSO₄·H₂O     | 1a              | This study      | 1.23      | 2.64      | 91       |
| Szomolnokite | FeSO₄·H₂O     | 1b              | This study      | 0.89      | 2.31      | 9        |
| Szomolnokite | FeSO₄·H₂O     | Blue Square     | Alboom et al. 2009 | 1.263     | 2.231     | 100      |
| Szomolnokite | FeSO₄·H₂O     | Orange Square   | Giester et al. 1994 | 1.26      | 2.71      | 100      |
| Szomolnokite | FeSO₄·H₂O     | Pink Square     | Russel & Montano 1978 | 1.18      | 2.69      | 100      |
| Szomolnokite | FeSO₄·H₂O     | 2               | ML-S77          | 1.26      | 2.73      | 94       |
| Szomolnokite | FeSO₄·H₂O     | 3               | 92942           | 1.26      | 2.73      | 96       |
| Szomolnokite | FeSO₄·H₂O     | 4a              | 136685-2        | 1.29      | 2.74      | 82       |
| Szomolnokite | FeSO₄·H₂O     | 4b              | 136685-2        | 0.23      | 0.69      | 12       |
| Szomolnokite | FeSO₄·H₂O     | 4c              | 136685-2        | 0.54      | 0.14      | 7        |
| Szomolnokite | FeSO₄·H₂O     | 5               | 104276          | 1.28      | 2.76      | 100      |
| Szomolnokite | FeSO₄·H₂O     | 6a              | ML-S60          | 1.31      | 2.89      | 57       |
| Szomolnokite | FeSO₄·H₂O     | 6b              | ML-S60          | 1.31      | 3.42      | 31       |
| Szomolnokite | FeSO₄·H₂O     | 6c              | 159098          | 1.24      | 2.78      | 43       |
| Szomolnokite | FeSO₄·H₂O     | 7a              | 159098          | 1.31      | 2.69      | 42       |
| Szomolnokite | FeSO₄·H₂O     | 7b              | 159098          | 0.49      | 0.18      | 12       |
| Szomolnokite | FeSO₄·H₂O     | 8a              | ML-S103         | 1.27      | 2.74      | 93       |
| Szomolnokite | FeSO₄·H₂O     | 8b              | ML-S103         | 0.43      | 0.42      | 7        |
| Szomolnokite | FeSO₄·H₂O     | 9a              | 159266          | 0.46      | 0.36      | 72       |
| Szomolnokite | FeSO₄·H₂O     | 9b              | 159266          | 0.14      | 0.56      | 28       |
| Szmikite     | MnSO₄·H₂O     | 10a             | 159189          | 0.52      | 0.93      | 18       |
| Szmikite     | MnSO₄·H₂O     | 10b             | 159189          | 1.24      | 2.59      | 82       |
| Gunningite   | ZnSO₄·H₂O     | 11a             | 56925           | 0.26      | 0.4       | 32       |
| Gunningite   | ZnSO₄·H₂O     | 11b             | 56925           | 0.37      | 1.26      | 10       |
| Gunningite   | ZnSO₄·H₂O     | 11c             | 56925           | 1.25      | 2.74      | 58       |
| Chalcanthite | CuSO₄·5H₂O    | 12a             | DD100           | 0.1       | 0.51      | 29       |
| Chalcanthite | CuSO₄·5H₂O    | 12b             | DD100           | 0.26      | 0.76      | 30       |
| Chalcanthite | CuSO₄·5H₂O    | 12c             | DD100           | 1.26      | 2.32      | 22       |
| Chalcanthite | CuSO₄·5H₂O    | 12d             | DD100           | 1.3       | 2.88      | 19       |
| Pentahydrite | MgSO₄·5H₂O    | 13a             | VZO121          | 0.5       | 0.83      | 8        |
| Pentahydrite | MgSO₄·5H₂O    | 13b             | VZO121          | 1.26      | 3         | 53       |
| Pentahydrite | MgSO₄·5H₂O    | 13c             | VZO121          | 1.27      | 3.68      | 38       |
| Jokokuite    | MnSO₄·5H₂O    | 14a             | G3536           | 0.15      | 0.56      | 26       |
| Mineral      | Formula               | Code | Rm  | Rm' | Ref. |
|--------------|-----------------------|------|-----|-----|------|
| Jokokute     | MnSO₄·5H₂O            | 14b  | 1.13| 2.63| 6    |
| Jokokute     | MnSO₄·5H₂O            | 14c  | 1.26| 2.51| 65   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 15a  | 1.27| 3.29| 92   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 16a  | 0.13| 0.49| 10   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 16b  | 0.36| 0.31| 14   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 16c  | 1.45| 1.56| 13   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 16d  | 1.28| 2.76| 43   |
| Halotrichite | FeAl₂(SO₄)₄·22H₂O     | 16e  | 1.3 | 3.29| 20   |
| Melanterite  | Fe²⁺(H₂O)₆SO₄·H₂O     | 16   | 1.27| 3.21| 100  |
| Rozenite     | FeSO₄·4H₂O            | 17a  | 1.33| 2.97| 59   |
| Rozenite     | FeSO₄·4H₂O            | 17b  | 1.33| 3.39| 34   |
| Rozenite     | FeSO₄·4H₂O            | 18a  | 0.39| 0.95| 8    |
| Rozenite     | FeSO₄·4H₂O            | 18b  | 1.27| 3.21| 92   |
| Starkeyite   | MgSO₄·4H₂O            | 19a  | 0   | 0.57| 9    |
| Starkeyite   | MgSO₄·4H₂O            | 19b  | 1.14| 2.65| 91   |
Figure S1. Fitted hyperfine parameters determined using CONUSS: The relative isomer shift values of sites 2 to 5 are given with respect to site 1 ($\Delta IS_1$), and the full width at half maximum (FWHM) describes the distribution of field gradients in mm/s for a particular site. (A) Low pressure values, as all models are identical from 0 to 19 GPa. (B) Values from Model 1 applied to 19-95 GPa. Sites 2 and 3 have FWHM fixed to the same value so they overlap completely in the figure. (C) Values from Model 2 applied to 19-95 GPa. In this model, site 3 (green) undergoes a high to low spin transition between 19 and 45 GPa. (D) Values from Model 3 (gradual spin transition) applied to 19-95 GPa. Sites 3 and 5 have FWHM fixed to the same value so they overlap completely in the figure. See text for more details on Models 1, 2, and 3. Values, including the $\chi^2$ for each fit, are reported in Tables S2, S3, and S4.