**Supplementary material**

Table S1. Structure information for monoclinic ferrosilite with the *C*2/*c* space group at 17.8(3) GPa. Unit-cell parameters are: *a* = 9.256(2), *b* = 8.631(2), *c* = 4.8402(6) and *β* = 100.88(2). Unit-cell volume is 379.7(9).

|  |  |  |  |
| --- | --- | --- | --- |
|  | *x* | *y* | *z* |
| Fe1 | 0.500 | 0.224 | –0.250 |
| Fe2 | 0.500 | 0.411 | 0.250 |
| Si1 | 0.802 | 0.591 | 0.214 |
| O1 | 0.874 | 0.739 | 0.363 |
| O2 | 0.629 | 0.592 | 0.151 |
| O3 | 0.859 | 0.558 | –0.084 |

Table S2. Structure information for monoclinic ferrosilite with the *P*21/*c* space group at 37.9(5) GPa. Unit-cell parameters are: *a* = 9.171(2), *b* = 7.924(2), *c* = 4.6041(5) and *β* = 99.11(1). Unit-cell volume is 330.4(1).

|  |  |  |  |
| --- | --- | --- | --- |
|  | *x* | *y* | *z* |
| Fe1 | 0.275 | 0.515 | 0.414 |
| Fe2 | 0.538 | 0.321 | 0.906 |
| Si1 | 0.481 | 0.667 | -0.003 |
| Si2 | 0.063 | 0.652 | 0.828 |
| O1 | 0.890 | 0.647 | 0.798 |
| O2 | 0.128 | 0.679 | 1.180 |
| O3 | 0.130 | 0.501 | 0.682 |
| O4 | 0.396 | 0.492 | 0.100 |
| O5 | 0.387 | 0.697 | -0.331 |
| O6 | 0.598 | 0.825 | -0.139 |

Table S3. Correlation matrix for the fitted equation of state parameters for the *C*2/*c* structure in this study using the MINUTI software. The Dewaele et al. (2008) ruby pressure scale was used for pressure measurements.

|  |  |  |  |
| --- | --- | --- | --- |
|  | This study | | |
|  | *V*0 | *K*0T | *K*'0T |
| V0 | 1.000 | –0.887 | 0.766 |
| K0T | –0.887 | 1.000 | –0.961 |
| *K*'0T | 0.766 | –0.961 | 1.000 |

Table S4. Correlation matrix for the fitted equation of state parameters for the *C*2/*c* structure in Pakhomova et al. (2017) study using the MINUTI software. The Dewaele et al. (2008) ruby pressure scale was used for ruby pressure measurements.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
|  | Pakhomova et al. (2017) | | | |
|  | V0 | *K*0T | | *K*'0T |
| V0 | 1.000 | –0.944 | | 0.862 |
| *K*0T | –0.944 | | 1.000 | –0.977 |
| *K*'0T | 0.862 | | –0.977 | 1.000 |

Table S5. Correlation matrix of the fitted hyperfine parameters for the low-pressure *P*21/*c* ferrosilite, FeSiO3 phase at 0 GPa. The quadrupole splitting distributions were fixed to 0.01 mm/s and the relative weight of the M1 to M2 sites was fixed to 50:50. Texture, thickness and the canting angle were determined from Monte Carlo searches.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
|  | QS M1 | QS M2 | IS  M1A–M1B | IS  M1A–M2A | IS  M1A–M2B |
| QS M1 | 1.000 | –0.291 | –0.151 | –0.642 | –0.085 |
| QS M1 | –0.291 | 1.000 | –2 | 0.461 | –0.220 |
| IS  M1A–M1B | –0.871 | 0.432 | 1.000 | 0.833 | 0.752 |
| IS  M1A–M2A | –0.570 | 0.461 | 0.833 | 1.000 | 0.556 |
| IS  M1A–M2B | –0.927 | –0.220 | 0.752 | 0.556 | 1.000 |

Table S6. Correlation matrix of the fitted hyperfine parameters for the *C*2/*c* ferrosilite, FeSiO3 phase at 9.3 GPa. The quadrupole splitting distributions were fixed to 0.01 mm/s, texture was fixed to 100% and the relative weight of the M1 to M2 sites was fixed to 50:50.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
|  | Thickness | QS M1 | QS M2 | IS  M1A–M2 | IS  M1A–M1B | M1A/(M1A+M1B) | Canting angle |
| Thickness | 1.000 | –0.566 | 0.386 | 0.791 | 0.076 | 0.355 | –0.575 |
| QS M1 | –0.566 | 1.000 | –0.908 | –0.816 | 0.444 | 0.019 | 0.348 |
| QS M2 | 0.386 | –0.908 | 1.000 | 0.714 | –0.607 | –0.250 | –0.318 |
| IS  M1A–M2 | 0.791 | –0.816 | 0.714 | 1.000 | –0.254 | 0.038 | –0.453 |
| IS  M1A–M1B | 0.076 | 0.444 | –0.607 | –0.254 | 1.000 | 0.601 | –0.125 |
| M1A/(M1A+M1B) | 0.355 | 0.019 | –0.250 | 0.038 | 0.601 | 1.000 | –0.201 |
| Canting angle | –0.575 | 0.348 | –0.318 | –0.453 | –0.125 | –0.201 | 1.000 |

Table S7. Correlation matrix of the fitted hyperfine parameters for the high-pressure *P*21/*c* ferrosilite, FeSiO3 phase at 48.0 GPa. The thickness, quadrupole splitting distribution, texture, canting angle and weight fractions of the different sites were determined from Monte Carlo searches and fixed to those values during the fitting. The quadrupole splitting of the low-spin site was fixed to 0.

|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | QS M1A | QS M1B | QS M2A | QS M2B | IS  M1A–M1B | IS  M1A–M2A | IS  M1A–M2B | IS  M1A–LS |
| QS M1A | 1.000 | 0.240 | –0.870 | 0.709 | 0.525 | 0.844 | 0.803 | 0.682 |
| QS M1B | 0.240 | 1.000 | –0.577 | 0.580 | 0.780 | 0.625 | 0.603 | 0.729 |
| QS M2A | –0.870 | –0.577 | 1.000 | –0.820 | –0.793 | –0.985 | –0.914 | –0.837 |
| QS M2B | 0.709 | 0.580 | –0.820 | 1.000 | 0.872 | 0.874 | 0.941 | 0.895 |
| IS  M1A–M1B | 0.525 | 0.780 | –0.793 | 0.872 | 1.000 | 0.874 | 0.917 | 0.957 |
| IS  M1A–M2A | 0.844 | 0.625 | –0.985 | 0.874 | 0.874 | 1.000 | 0.964 | 0.913 |
| IS  M1A–M2B | 0.803 | 0.603 | –0.914 | 0.941 | 0.917 | 0.964 | 1.000 | 0.964 |
| IS  M1A–LS | 0.682 | 0.729 | –0.837 | 0.895 | 0.957 | 0.913 | 0.964 | 1.000 |