**Supplementary Information**

**Superionic hcp-Fe alloys and their seismic velocities in Earth’s inner core**

Yu He1,2,#,\*, Shichuan Sun1,3,#, Duck Young Kim2, Bo Gyu Jang2, Heping Li1, Ho-kwang Mao2

1Key Laboratory of High-Temperature and High-Pressure Study of the Earth’s Interior, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, Guizhou, China

2Center for High Pressure Science and Technology Advanced Research, Shanghai 201203, China

3University of Chinese Academy of Sciences, Beijing 100049, China

\*E-mail: [heyu@mail.gyig.ac.cn](mailto:heyu@mail.gyig.ac.cn)

# These authors contribute equally to this work

S1. Structure construction and solid-superionic phase transition.

S2. The estimation of melting temperature using the two-phase method.

S3. Diffusion coefficients and ionic conductivities of superionic Fe alloys

S4. DFT + DMFT calculations and electronic conductivities of Fe alloys

S5. AIMD simulations on the elastic properties of superionic Fe alloys

S6. Convergence tests

S7. References

**S1. Structure construction and solid-superionic-liquid phase transition.**

The stable phase of Fe at Earth’s inner core (IC) conditions is still controversial. Both hexagonal-close-packed (hcp) Fe and body-center-cubic (bcc) Fe are considered as the candidate of the main phase in the IC1,2. Here, we chose hcp-Fe alloys as the simulation models because recent experimental and computational studies suggest hcp-Fe alloys show seismic velocities close to the Preliminary Reference Earth Model (PREM) under the IC conditions3-9, and the results of this study are compared with the results of previous researches.

The light-element impurities may take substitutional and/or interstitial sites in hcp-Fe. Therefore, the relative thermodynamic stability of H, O, C, S, and Si at interstitial site are investigated at 360 GPa and 0 K. We consider the stability of interstitial defect in binary (Fe-X) Fe alloys (X = H, O, C, S, and Si). A 4×4×2 supercell containing 64 Fe atoms was employed for the calculation. A substitutional defect (Xs) was generated by replacing a Fe atom, and an interstitial defect (Xi) was generated by adding an X atom at the octahedral interstitial site. When Xs and Xi are coexisted (Fe-Xs-Xi), both correlated (dimer) and separated (sub. + inter.) configurations are considered. Geometry optimizations were performed at 360 GPa using conjugate gradients minimization until all of the forces acting on the ions were less than 0.01 eV/Å per atom. A 3×3×4 K-point mesh was adopted. The chemical potential of distinctive element X in hcp-Fe are calculated by

where N and M are the number of Fe and light-element atoms in the structure. As shown in Supplementary Fig. 1, H at interstitial site is energetically much more favorable, which is consistent with previous experimental and computational results8,10. Substitutional structures are more stable for Fe-S and Fe-Si alloys. For C and O, the dimer structure is the most stable one indicating the coexisting of both substitutional and interstitial defects. Li et al.11 investigated the thermodynamic stability of Fe-C alloys at high temperature by quasiharmonic approximation (QHA) method, and they found the stability of the dimer structure is weakened and its relative free energy is slight higher than that of the substitutional structure at 6500 K. Nevertheless, the interstitial defects for C and O may still exist in Earth’s inner core due to the slight energy difference and complicated thermodynamic conditions in Earth’s inner core with different pressures, temperatures, compositions. Thus, it is still valuable to study the influence of C and O interstitial defects on the properties of Fe alloys and compare with geophysical observations. In addition, we evaluated the elastic stability of Fe-H, Fe-O and Fe-C alloys with H, C, and O at interstitial sites by performing *ab-initio* molecular dynamics (AIMD) simulations, which confirm the elastic stability of these structures at 360 GPa and 3000-6000 K (Supplementary S5). Thus, the properties of light elements at the interstitial sites are studied in this approach.

C:\Users\He Yu\AppData\Local\Microsoft\Windows\INetCache\Content.Word\Fig_0K_new.tif

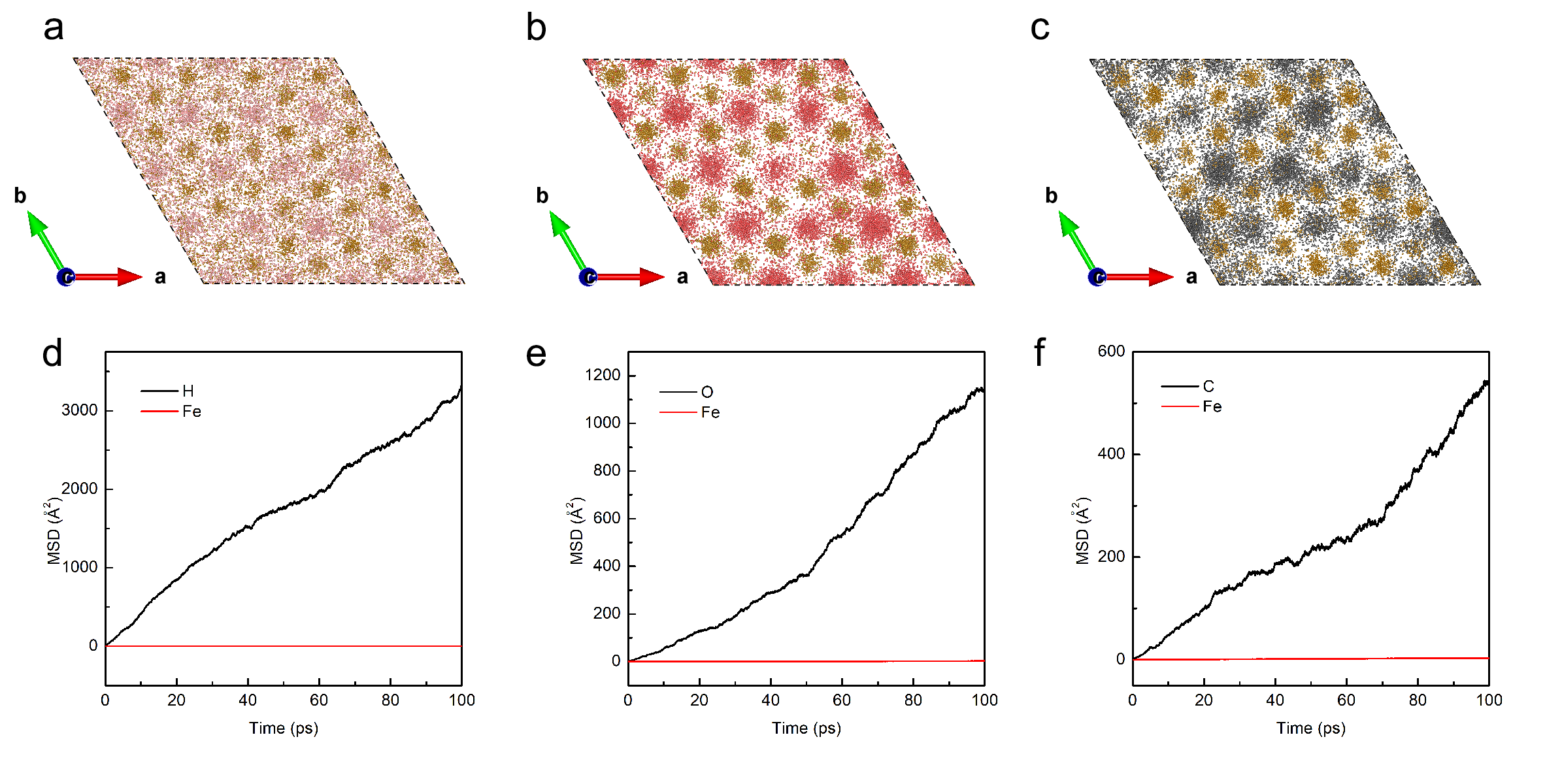
**Supplementary Figure 1 | Calculated chemical potentials of X (X = H, O, C, S, and Si) in hcp-Fe with different configurations at 360 GPa and 0 K.**

Regarding the concentration of light element defects, H is known to be soluble in solid Fe at high *P-T* with the concentration over ~1 wt.%12,13. The solubility of C in hcp-Fe is about 1 wt. % at ~211 GPa14, while the O concentration in solid Fe can be negligible15. On the other hand, the Si and S can form solid solution in hcp-Fe with high concentrations up to ~16% for Si and ~5% for S7,16-18. Based on the solubility, light-element bearing hcp-Fe structures were constructed by locating one H, O, C, Si, and S atom at the central site of hcp-Fe supercells (2×1×1 supercell for H and 2×2×1 supercell for O, C, Si and S). The light-element concentration in the Fe4H, Fe16O, Fe16C, Fe16Si, and Fe16S structures is 0.45, 1.75, 1.33, 3.04, and 3.46 wt. %, respectively. Although the solubility of O in hcp-Fe might be very low under the IC conditions, we still study the Fe16O structure and the result will benefit the understanding on O defects in hcp-Fe. The structure relaxations were performed using the PBE exchange-correlation functional and projector augmented wave (PAW) method19 as implemented in the Vienna Ab Inito Simulaton Package (VASP)20. In our calculations, a plane wave representation for the wave function with a cutoff energy of 800 eV was adopted. Geometry optimizations were performed at pressures from ~250 to ~360 GPa using conjugate gradients minimization until all of the forces acting on the ions were less than 0.01 eV/Å per atom. K-point mesh with a spacing of ca. 0.03 Å-1 was adopted.

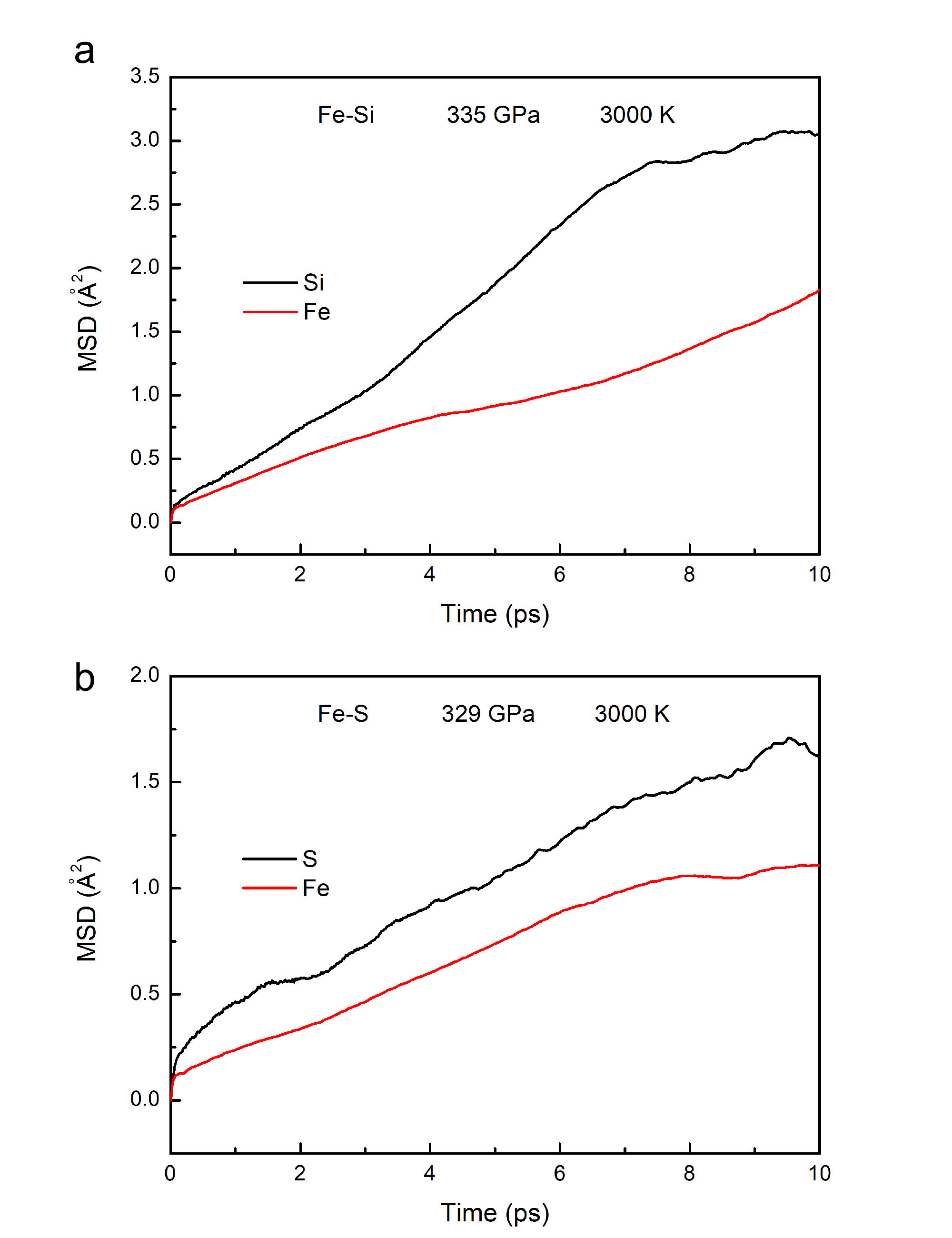
The solid-superionic-liquid phase transitions for these structures under the IC conditions were investigated by *ab-initio* molecular dynamics (AIMD). AIMD method has been widely used to simulate ionic conductivity21,22, melting temperature23,24, phase transition25-27, elastic property3-5,8,9,28 of materials at high *P-T*. Here, enlarged supercells for Fe64H16, and Fe64X4 (X = O, C, Si and S) were adopted for AIMD simulation. It is worth to note that a bigger supercells containing 128 Fe atoms and 32 H atoms or 8 X (X = O, C, Si and S) atoms did not change the simulation results. The simulations used the canonical ensemble (NVT：N-number of particles, V-volume, T-temperature) with a time step of 1 fs. The simulations lasted ~10-100 ps at temperatures from ~2000 to ~7000 K and pressure from ~250 to ~400 GPa. The mean square displacement (MSD):

,

is averaged over specific ions, and is the displacement of the ith ion at time *t*, and *N* is the total number of specific ions in the system. At the solid state, the MSDs of Fe and light elements are close to zero and do not show obvious increase over simulation time. The solid-superionic phase transition take place at temperatures over 3000 K. As shown in Supplementary Fig. 2, the MSD of H, O and C increases monotonically over simulation time, and the MSD of Fe is still stable, demonstrating that the materials are at the superionic state. The highly diffusive light-element defects can also be visualized by their trajectories. A further increase in temperature leads to melting of the lattice and the particles diffuse away from their equilibrium positions and their MSDs increase with simulation time. For Fe64Si4 and Fe64S4 alloys, we did not observed a superionic state, the structures transfer directly to a liquid state at ~3000 K and ~ 330 GPa (Supplementary Fig. 3). It suggests the solid Fe64Si4 and Fe64S4 hcp-Fe alloys,with Si and S at the interstitial sites, is not stable under the IC conditions.



**Supplementary Figure 2 | Trajectories and mean square displacements (MSDs) of H, O, C, and Fe in Fe-light-element alloys.** Trajectories of **a,** H and Fe in FeH0.25; **b,** O and Fe in FeO0.0625; **c,** C and Fe in FeC0.0625 at the superionic state under the IC conditions (~360 GPa and ~5000 K). Small pink, red, black and brown spheres represent the trajectories of H, O, C and Fe, respectively. MSDs of **d,** H and Fe in FeH0.25; **e,** O and Fe in FeO0.0625; **f**, C and Fe in FeC0.0625. The MSDs of H, O and C ions increase obviously with simulation time, while the MSDs of Fe remain close to zero indicating a superionic state.

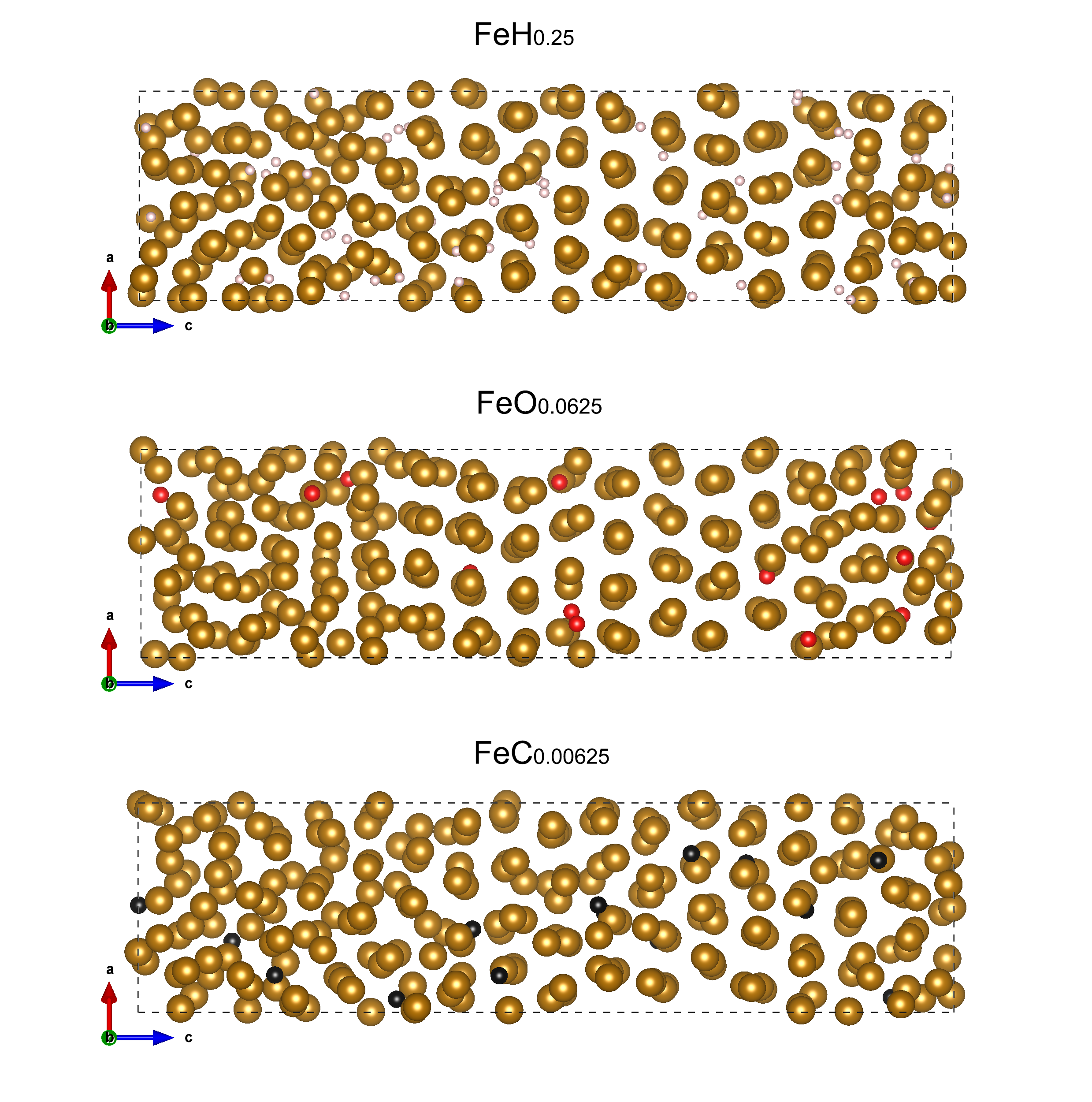


**Supplementary Figure 3 | MSDs of Si, S, and Fe in FeSi0.0625 and FeS0.0625 at ~330 GPa and 3000 K.** MSDs of **a,** Si and Fe in FeSi0.0625; **b,** S and Fe in FeS0.0625. The MSDs of Si, S and Fe increase obviously with simulation time indicating a liquid state.

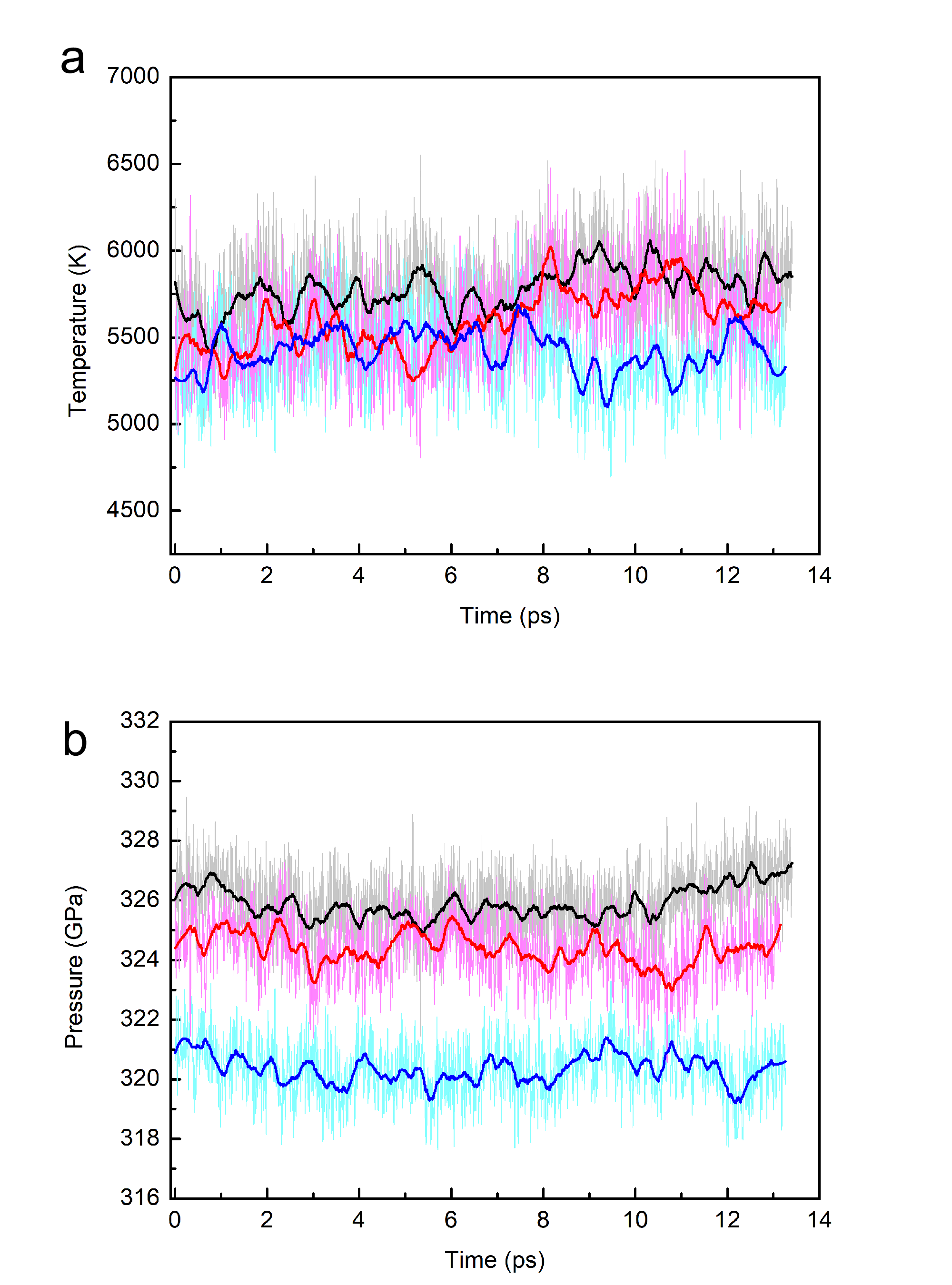
**S2. The estimation of melting temperature using the two-phase method.**

During AIMD simulations, a superheating solid state is obtained above melting temperature (Tm)29,30.In order to exclude the influence of superheating state, the melting temperatures of these alloys should be estimated. Considering the influence of light element partition between solid and liquid phase31, we used the solid and liquid coexisting system, which is also known as two-phase method32-36, to predict the melting temperature. In addition, the computational results are in agreement with the results of previous experimental studies12,14,37. Accordingly, the estimated melting temperatures of FeH0.25, FeO0.0625, and FeC0.0625 are approximately within 5500-6000 K, and simulation temperatures of our further studies on elastic properties are constrained below the melting temperatures.

The two-phase method has been widely used to predict the melting temperatures of different materials such as metals32,34, hydrogen35, lithium hydride36, and oxides33. Under a constant internal energy, coexisted two-phase system can maintain for a long time, and the average temperature and pressure of the coexisted system denotes a melting point. A simulated structure was generated by combining a solid and liquid Fe-light-element structure. The solid structures were obtained from the structures of FeH0.25, FeO0.0625, and FeC0.0625 at 360 GPa and 6000, 5500, and 5500 K, respectively. The liquid structures were generated by heating the solid structures at ~9000 K for ~2 ps, and then slowly cooling down to ~6000 K. The simulations were performed in the NVE (N-number of particles, V-volume, E-internal energy) ensemble using supercells with different volumes containing 256 Fe atoms and 64, 16, and 16 of H, O, and C atoms, respectively. The cutoff energy was set to 400 eV with Γ point for k-space sampling. The time step was set at 1 fs with total simulation of over 12 ps. The two-phase coexisting states are maintained during our simulation (Supplementary Fig. 4). The pressures and temperatures in these simulations are presented in Supplementary Fig. 5, and averaged melting temperatures for FeH0.25, FeO0.0625, and FeC0.0625 are approximately 5770 (± 200), 5595 (± 200), and 5413 (± 200) K at 324.4 (± 1.0), 320.3 (± 1.0), and 324.3 (± 1.0) GPa, respectively. Compared with previous experimental38,39 and computational studies24,34,40-42, these superionic impurities also reduce the melting temperature by about 500-1000 K. In the simulated systems, we also observed the diffusion of H, O, and C in solid phases indicting the superionic state. Interestingly, we observed the inter-diffusion of the light elements between the liquid and solid phases, thus the superionic state of Fe-alloys should be considered when calculating the light element partition at ICB.



**Supplementary Figure 4 | The structures of two-phase systems of Fe-H, Fe-O and Fe-C after the AIMD simulations.** These structures suggest the coexistence of solid and liquid Fe alloys. Pink, red, black and brown spheres represent H, O, C and Fe atoms, respectively.



**Supplementary Figure 5 | The evolution of temperature and pressure with respect to simulation time in the two-phase coexisting systems for Fe-H, Fe-O and Fe-C alloys. a.** temperatures and, **b.** pressures for Fe-H, Fe-O and Fe-C alloys are shown with light grey, pink and cyan curves, and the averaged data over a 0.5 ps period are shown with thick black, red, and blue curves.

The melting behavior of Fe-H, Fe-C, and Fe-O binary systems has also been studied by previous high pressure experiments12,14,37. The presence of these light elements depresses the melting temperature. However, the eutectic melting experiments were conducted at relatively lower pressures. Thus, Simon–Glatzel equation was used to extrapolate the melting curve to the inner core conditions. The melting temperatures of Fe-rich side Fe binary alloys can be roughly estimated from eutectic temperature and composition as well as the melting point of pure Fe38. Based on the extrapolation of experimental data, the estimated melting temperatures are obtained (green regions in Supplementary Fig. 6), which are basically agree with our simulation results. Therefore, the elastic properties of the Fe alloys at 360 GPa are calculated at temperatures below 6000 K for FeH0.25, and 5500 K for FeO0.0625 and FeC0.0625.

C:\Users\He Yu\AppData\Local\Microsoft\Windows\INetCache\Content.Word\melt_exp_new.tif

**Supplementary Figure 6 | Estimated melting temperatures of Fe-H, Fe-O, and Fe-C alloys with different light element contents at 330 and 360 GPa.** The melting temperatures are estimated from the extrapolated melting curves of Fe-H, Fe-O, and Fe-C systems measured by high pressure experiments12,14,37. The solid and dash lines represent the melting temperatures at 360 and 330 GPa. The dot-dash lines exhibit the light element content in our simulation modes, with corresponding melting temperatures shown by green regions.

**S3. Diffusion coefficients and ionic conductivities of superionic Fe alloys**

To study superionic transport rigorously, we calculated the diffusion coefficient for these defects using the MSD () of the ionic positions at superionic state. The diffusion coefficient is defined as:

,

where *d* is the dimension of the lattice on which ion hopping takes place. The value of D obtained at various temperatures can be fitted with an Arrhenius equation:

,

where is the activation enthalpy, *A* is a pre-exponential factor, *k* is the Boltzmann constant, and *T* is the temperature. The ionic conductivity was calculated using the diffusion coefficients and the Nernst-Einstein equation:

,

in which *σ* is the ionic conductivity, *f* is a numerical factor approximately equal to unity, *D* is the diffusion coefficient, *c* is the concentration of light-elements defects, *q* is the electrical charge, *k* is the Boltzmann constant, and *T* is the temperature. The ionic conductivity of H, O and C ions are shown in Supplementary Fig. 7.

C:\Users\He Yu\AppData\Local\Microsoft\Windows\INetCache\Content.Word\C_check.tif

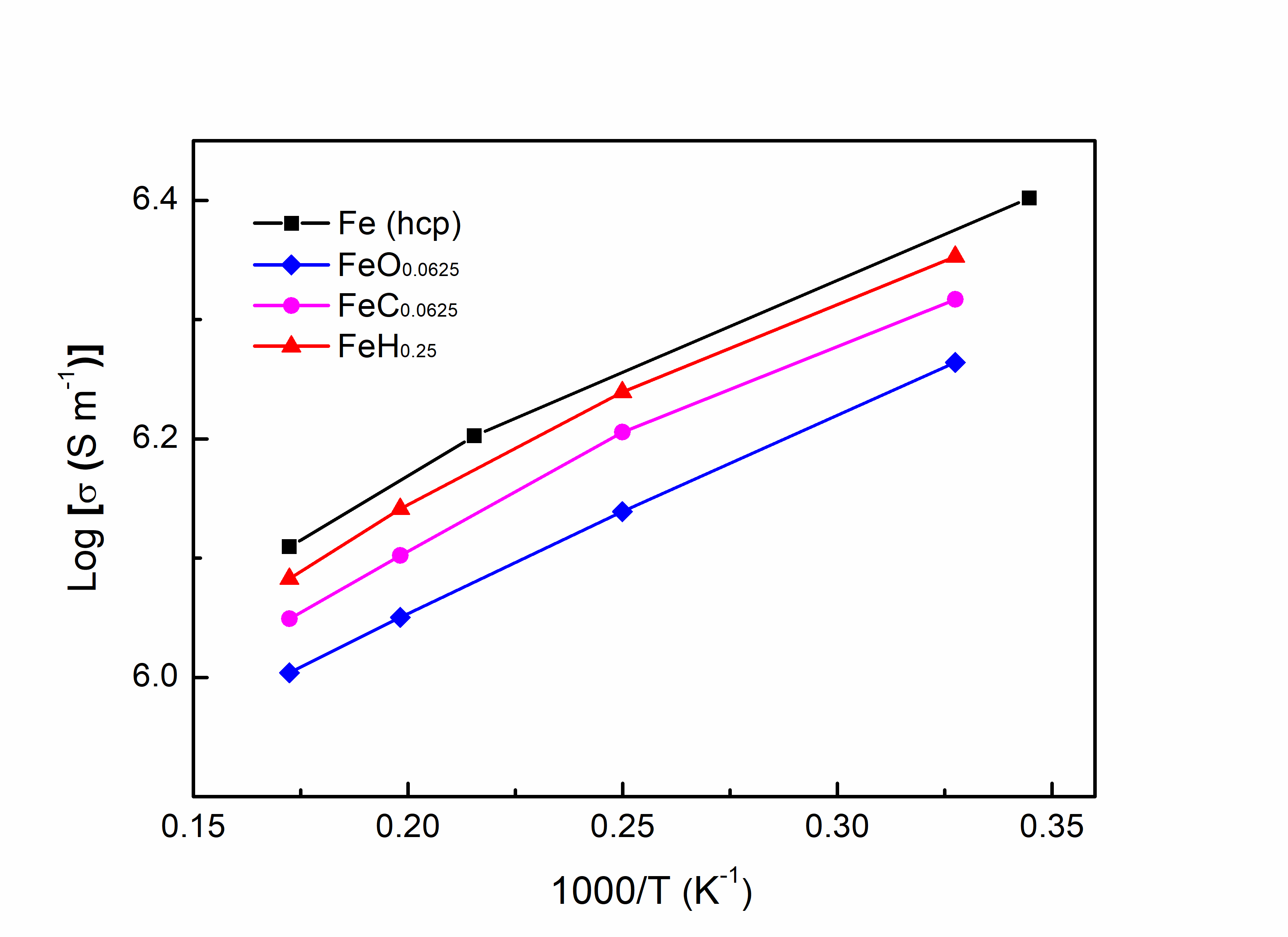
**Supplementary Figure 7 | Ionic conductivities of superionic Fe alloys at the core conditions.** Diffusion coefficients calculated using 4 × 4 × 2 supercells and 10 ps simluaiton time are labelled by open symbols. Blue squares: FeH0.25 at ~260 GPa; red squares: FeH0.25 at ~360 GPa; cyan triangles: FeO0.0625 at ~260 GPa; orange triangles: FeO0.0625 at ~360 GPa; green circles: FeC0.0625 at ~260 GPa; pink circles: FeC0.0625 at ~360 GPa. The convergence test results using 4 × 4 × 6 supercell and 100 ps simluaiton time are labeled by crosses and bars. The results of convergence test are presented with yellow, magenta, and cyan symbols for FeH0.25, FeO0.0625, and FeC0.0625, respectively.

**S4. DFT + DMFT calculations and electronic conductivities of Fe alloys**

To estimate the electronic conductivity of Fe, FeH0.25, FeO0.0625, and FeC0.0625, we considered the resistivity from electron-electron scattering (ρe-e) and electron-phonon scattering (ρe-ph). ρe-e of Fe and Fe alloys were calculated by using the combination of density functional theory with dynamical mean field theory (DFT+DMFT) as implemented in the WIEN2k+eDMFT package (<http://hauleweb.rutgers.edu/tutorials/>)43. We used the volume obtained from AIMD calculation at ~5500-6000 K in DFT+DMFT calculation to include the volume expansion effect on the electrionic conductivity. Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) was used for the exchange correlation functional44. The correlation effect of Fe 3d orbital was treated dynamically by local self-energy in DMFT part on the top of an effective one-electron Hamiltonian generated from DFT part. A 1000 k-points mesh was used in DFT+DMFT self-consistent calculation. We used the Coulomb interaction U=5 eV and Hund’s coupling J=0.943 eV, which is consistent with the previous theoretical study on hcp-Fe45. The local impurity problem was solved by using continuous time quantum Monte Carlo (CTQMC) impurity solver. A 7000 k-point mesh was used for the conductivity calculation based on the self-energy obtained from the self-consistent calculation with 1000 k-points mesh.

Since the unit cells of Fe alloys are too large to calculate ρe-ph in first principle manner, we just took previous calculated result on hcp-Fe by using the density functional perturbation theory (DFPT) and Boltzmann transport theory45. Therefore, the total resistivity was given by a sum of ρe-e calculated from DFT+DMFT and reported ρe-ph of hcp-Fe. Then, the electronic conductivity of Fe and Fe alloys were obtained. Considering the alloy effect on ρe-ph part, the electrionic conductivity of Fe alloys (FeH0.25, FeO0.0625, and FeC0.0625) can decrease further by ~10%46,47.

We found that the incorporation of light element leads to obvious decrease in electronic conductivity compared with pure Fe45,48, and the electronic conductivities decrease with temperature. However, they are 2-3 orders of magnitude higher than the ionic conductivities of superionic Fe alloys. Therefore, the significant increase in ionic conductivity upon superionic transition (Supplementary Fig. 7) has little influence on the total conductivities of these Fe alloys.



**Supplementary Figure 8 | Electronic conductivities of Fe and Fe alloys at 360 GPa with increasing temperature.** The electronic conductivities of Fe, FeH0.25, FeO0.0625, and FeC0.0625 calculated by DFT + DMFT method are shown by black, blue, red, and magenta symbols.

**S5. AIMD simulations on the elastic properties of superionic Fe alloys**

We constructed structures for Fe-hcp with C, H and O light elements in 4 × 4 × 2 supercells at 2000-6000 K and 360 GPa. We determined the equilibrium volume and cell parameters at different temperatures by conducting a grid of NVT ensemble simulations over volumes and temperatures using a Nosé thermostat49 For each equilibrium structure of different temperature, a 10,000 time-steps (10 ps) simulation was calculated to check for sure that the stress field is hydrostatic. The elastic constant Cij was calculated by distorting the equilibrium structure and solving the stress-strain relations.

The elastic properties of crystal are expressed as the relationship between stress and strain:

(6)

where refers to stress tensor, refers to strain tensor, and represents fourth order elastic modulus. Considering of the symmetry of , the equation is simplified as below:

(7)

The equation could be expanded as follow:

= . (8)

For the hexagonal system investigated in this work, we have calculated the three non-equivalent elastic constants, , , , and . , and could be obtained by a strain tensor:

(9)

we obtained using a strain tensor:

(10)

and we obtained using a strain tensor:

(11)

where is the magnitude of distortion. For in , and 0, five group of strains were added by:

(12)

where a represents a 3 cell parameter matrix, and ε represents added strain, represents a identity matrix. Then, we obtained elastic moduli trough dealing with the stress-strain relationship listed in equation (8). In order to ensure the reliability and full convergence of the results, 10 000 time steps of simulations was carried out for each direction of deformation. The final results of strain-stress data show very good linear relationship and were fitted employing central difference method. For each optimizing result, the calculated correlation coefficient R2 were larger than 99%. The calculated results satisfy with stability conditions:

, ， , (13)

We calculated the bulk modulus B and shear modulus G using Voigt average scheme which is proved more appropriate and accurate in calculating the seismic wave properties50.

And further calculated the primary wave velocity ，shear wave velocity , and bulk sound velocity :

, ,  (14)

The Possion’s ratio is calculated by:

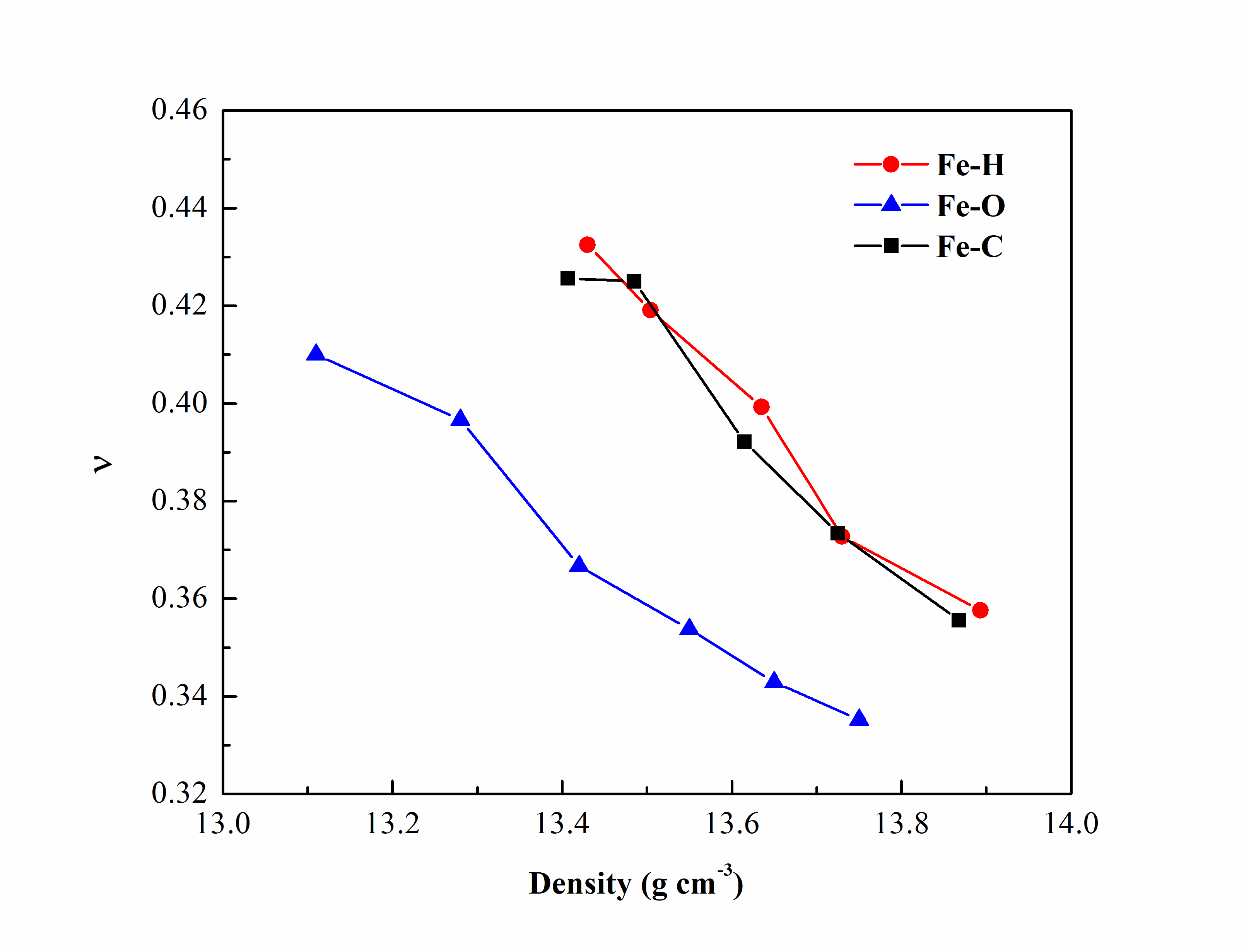
(15),

and shown in Supplementary Fig. 9.

Using the same method, the elastic constants for hcp-Fe at 360 GPa were calculated and compared with the results of the previous report (Supplementary Fig. 10)3. Our results show good consistence with the previous report at temperatures below 7000 K.

**Supplementary Table 1** | Densities (ρ), Elastic constants (Cij), sound velocities (VΦ, VP and VS), moduli (B and G) and Poisson’s ratio of FeH0.25, FeC0.0625 and FeO0.0625 at various temperatures and 360 GPa.

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
|  | T(K) | ρ | C11 | C12 | C13 | C33 | C44 | VΦ | VP | VS | B | G | ν |
|  |  | (g cm-3) | (GPa) | (GPa) | (GPa) | (GPa) | (GPa) | (km s-1) | (km s-1) | (km s-1) | (GPa) | (GPa) |  |
| FeH0.25 | 0 | 13.75 | 2390.3 | 1201.6 | 1107.3 | 2529.3 | 508 | 10.69 | 12.51 | 6.5 | 1571.23 | 581.7 | 0.335 |
|  | 2000 | 13.65 | 2272.9 | 1177.9 | 1085.4 | 2372.7 | 458.2 | 10.53 | 12.22 | 6.23 | 1512.88 | 530.77 | 0.343 |
|  | 3000 | 13.55 | 2194.4 | 1183.6 | 1070.1 | 2194.8 | 394.8 | 10.42 | 11.98 | 5.93 | 1470.13 | 476.32 | 0.354 |
|  | 4000 | 13.42 | 2050.3 | 1158.5 | 1073.3 | 2059 | 339.6 | 10.28 | 11.69 | 5.56 | 1418.87 | 415.32 | 0.367 |
|  | 5000 | 13.28 | 1868.8 | 1214.2 | 1110.8 | 1829.7 | 247.5 | 10.2 | 11.28 | 4.81 | 1381.1 | 306.56 | 0.397 |
|  | 6000 | 13.11 | 1732.7 | 1177.9 | 1054.2 | 1652.1 | 177.8 | 9.95 | 10.86 | 4.35 | 1298.9 | 248.68 | 0.410 |
| FeC0.0625 | 0 | 13.87 | 2275.1 | 1257 | 1199.6 | 2349.9 | 466.4 | 10.67 | 12.26 | 6.03 | 1579.17 | 504.64 | 0.356 |
|  | 3000 | 13.74 | 1940.7 | 1168.1 | 1113.8 | 2041 | 361.9 | 10.15 | 11.46 | 5.33 | 1412.64 | 390.47 | 0.373 |
|  | 4000 | 13.62 | 1986.8 | 1370.5 | 1053.2 | 1897.3 | 274.9 | 10.23 | 11.36 | 4.93 | 1424.97 | 331.18 | 0.392 |
|  | 5000 | 13.49 | 1732 | 1260.4 | 1222.7 | 1705.7 | 189.9 | 10.18 | 10.96 | 4.05 | 1397.92 | 220.71 | 0.425 |
|  | 5500 | 13.41 | 1687.5 | 1256.8 | 1079.7 | 1590.6 | 147 | 9.89 | 10.63 | 3.91 | 1310.89 | 205.17 | 0.427 |
| FeO0.0625 | 0 | 13.89 | 2250.7 | 1257.1 | 1216.8 | 2365.3 | 467.7 | 10.67 | 12.24 | 5.99 | 1583.12 | 498.17 | 0.358 |
|  | 3000 | 13.73 | 2095.7 | 1123.7 | 1209.9 | 2038.9 | 338.1 | 10.38 | 11.74 | 5.47 | 1479.7 | 411.56 | 0.373 |
|  | 4000 | 13.64 | 1902.3 | 1219.3 | 1228.6 | 1907.2 | 273.6 | 10.32 | 11.38 | 4.79 | 1451.64 | 313.43 | 0.400 |
|  | 5000 | 13.50 | 1807.3 | 1307 | 1117.2 | 1652.6 | 174.3 | 10.08 | 10.91 | 4.17 | 1372.22 | 234.81 | 0.419 |
|  | 5500 | 13.43 | 1675.6 | 1226.5 | 1227 | 1692.5 | 147.7 | 10.13 | 10.8 | 3.81 | 1378.3 | 194.86 | 0.432 |



**Supplementary Figure 9 | Calculated Poisson’s ratios of FeH0.25, FeC0.0625 and FeO0.0625 at various temperatures and 360 GPa.** Increasing temperature leads to obvious increases in Poisson’s ratios of Fe alloys approaching the Poisson’s ratio of Earth’s inner core (~0.44).

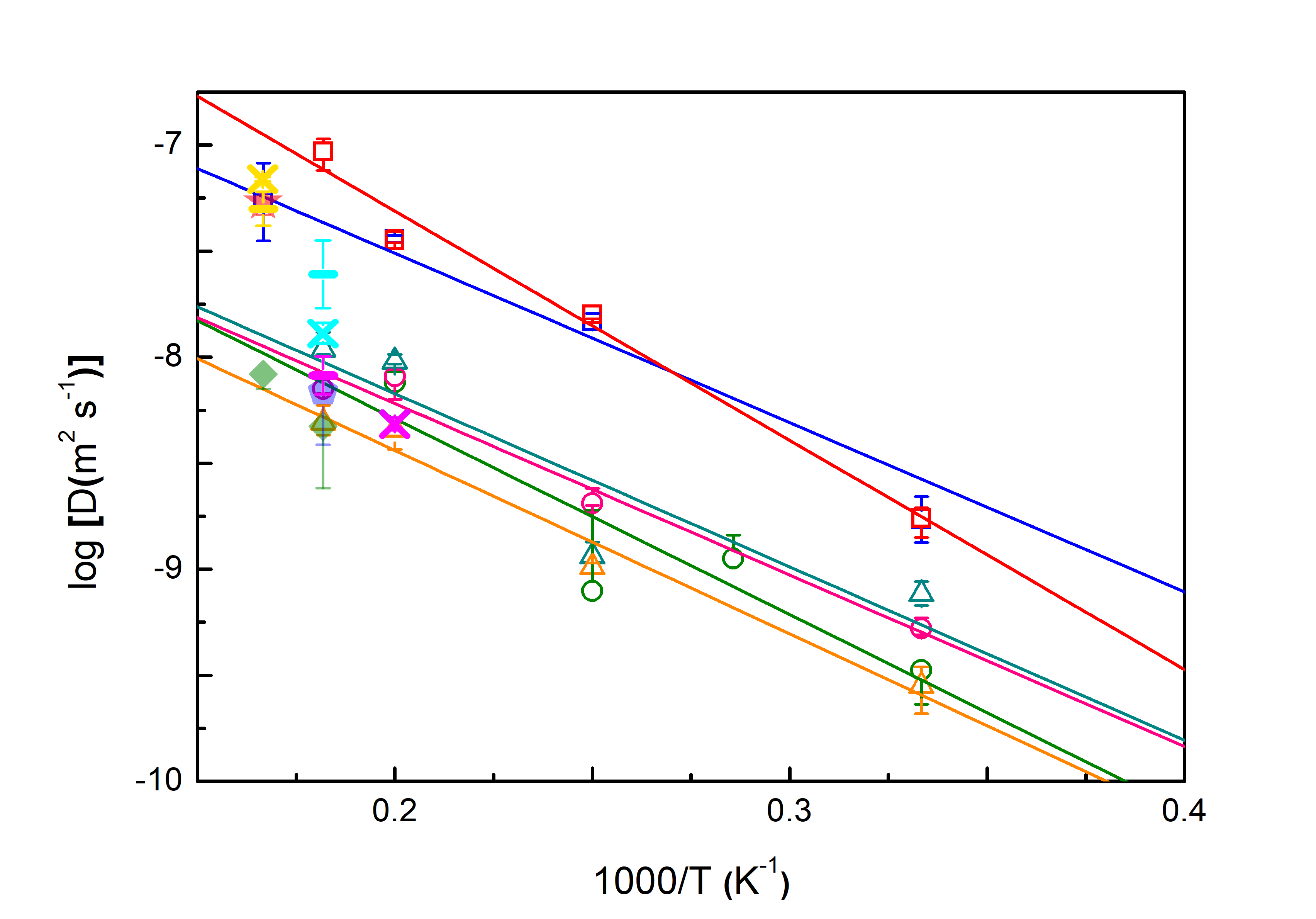
Fe_elastic_compare

**Supplementary Figure 10 | Calculated elastic constants for hcp-Fe as the function of simulation temperature at 360 GPa.** Our data represented by open symbols is compared with the results of a previous report represented by solid symbols. The elastic constants C11, C12, C13, C33 and C44 are shown with black, red, blue, green and orange symbols, respectively.

**S6. Convergence tests**

Convergence tests were performed on FeH0.25, FeO0.0625, and FeC0.0625 at 360 GPa and 5000-6000 K with supercells containing over 200 atoms and simulation time above 100 ps. 4 × 4 × 6 supercells containing 240, 204, and 204 atoms for FeH0.25, FeO0.0625, and FeC0.0625 are used for AIMD simulations. The simulations clearly confirm the superionic state of these Fe alloys. Also, the calculated diffusion coefficients and ionic conductivities are well converged (Supplementary Fig. 11).

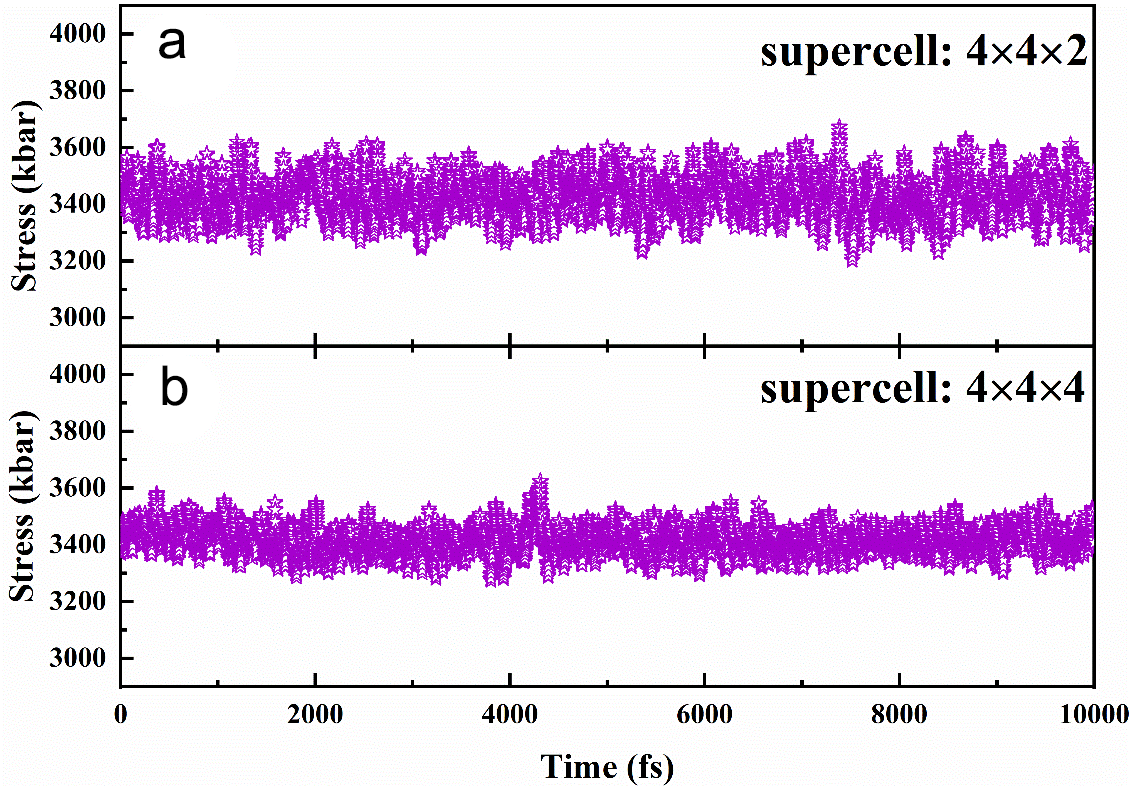
As the calculation results of the elastic constants are also very important in this work, we carried out convergence tests on the stresses of FeH0.25 with different supercells. For each test, we made a gird of calculations to ensure the hydrostatic state. Then, we tested the stress with 1% (δ=0.01) strain in the [001] direction and all the simulations lasted ~10000 time steps. Our convergence tests are shown in Supplementary Table 2 and Supplementary Fig. 12. It suggests a larger supercell does not change our results on elastic constants.



**Supplementary Figure 11 | Comparison of calculated diffusion coefficients with different supercells and simulation time.** Diffusion coefficients calculated using 4 × 4 × 2 supercells and 10 ps simluaiton time are labelled by different open symbols: FeH0.25 at ~260 GPa; red squares: FeH0.25 at ~360 GPa; cyan triangles: FeO0.0625 at ~260 GPa; orange triangles: FeO0.0625 at ~360 GPa; green circles: FeC0.0625 at ~260 GPa; pink circles: FeC0.0625 at ~360 GPa. The convergence test results using 4 × 4 × 6 supercell and 100 ps simluaiton time are labelled by crosses and bars. The results of convergence test are presented with yellow, magenta, and cyan symbols for FeH0.25, FeO0.0625, and FeC0.0625, respectively. The solid red star, green diamonds and blue pentagon represent the diffusion coefficients of H, O and C in liquid Fe at ~330 GPa.

**Supplementary Table 2** | Convergence tests on supercell size.

|  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- |
|  | (GPa) |  |  |  |  |  |
|  | 360 | 5000 |  | 80 | 0.01 | 342.582 |
|  | 360 | 5000 |  | 160 | 0.01 | 341.360 |



**Supplementary Figure 12 |** Convergence tests of supercell size on FeH0.25. (a), (b) exhibited the simulated stress, induced by 0.01 strain in , of [001] direction changed with time of a 4×4×2 supercell and 4×4×4 supercell, respectively.

**S7. References**

1. Tateno, S., Hirose, K., Ohishi, Y. & Tatsumi, Y. The Structure of Iron in Earth’s Inner Core. Science **330**, 359 (2010).
2. Belonoshko, A. B., Rajeev, A. & Johansson, B. Stability of the body-centred-cubic phase of iron in the Earth’s inner core. Nature **424**, 1032 (2003).
3. Martorell, B., Vočadlo, L., Brodholt, J. & Wood, I. G. Strong Premelting Effect in the Elastic Properties of hcp-Fe Under Inner-Core Conditions. Science **342**, 466 (2016).
4. Martorell, B., Wood, I.G., Brodholt, J. & Vočadlo, L. The elastic properties of hcp-Fe1-xSix at Earth’s inner-core conditions. Earth Planet. Sci. Lett. **451**, 89–96 (2016).
5. Li, Y., Vočadlo, L. & Brodholt J. B. The elastic properties of hcp-Fe alloys under the conditions of the Earth's inner core. Earth Planet. Sci. Lett. **493**, 118 (2018).
6. Sakamaki T. et al., Constraints on Earth’s inner core composition inferred from measurements of the sound velocity of hcp-iron in extreme conditions. Sci. Adv. **2**, e1500802, (2016).
7. Antonangeli, D. et al., Sound velocities and density measurements of solid hcp-Fe and hcp-Fe–Si (9 wt.%) alloy at high pressure: Constraints on the Si abundance in the Earth’s inner core. Earth Planet. Sci. Lett. **482**, 446-453 (2018).
8. Caracas, R. The influence of hydrogen on the seismic properties of solid iron, Geophys. Res. Lett., **42**, 3780–3785 (2015).
9. Caracas, R. The influence of carbon on the seismic properties of solid iron. Geophys. Res. Lett. **44**, 128-134 (2017).
10. Tagawa, S., Ohta, K., Hirose, K, Kato, C. & Ohishi, Y. Compression of Fe–Si–H alloys to core pressures. Geophys. Res. Lett. **43**, 3686-3692, (2016).
11. Li, Y., Vočadlo, L., Alfè, D. & Brodholt J. B. Carbon Partitioning Between the Earth's Inner and Outer Core. J. Geophys. Res.: Solid Earth **124**, 12812-12824.
12. Shibazaki, Y., et al. Sound velocity measurements in dhcp-FeH up to 70 GPa with inelastic X-ray scattering: implications for the composition of the Earth’s core. Earth Planet. Sci. Lett. **313–314**, 79–85 (2012).
13. Terasaki, H. et al., Stability of Fe–Ni hydride after the reaction between Fe–Ni alloy and hydrous phase (δ-AlOOH) up to 1.2 Mbar: possibility of H contribution to the core density deficit. Phys. Earth Planet. Inter. **194–195**, 18–24 (2012).
14. Mashino, I., Miozzi, F., Hirose, K., Morard, G. & Sinmyo, R. Melting experiments on the Fe–C binary system up to 255 GPa: Constraints on the carbon content in the Earth’s core. Earth Planet. Sci. Lett. **515**, 135-144 (2019).
15. Ozawa, H., Hirose, K., Tateno, S., Sata, N. & Ohishi, Y. Phase transition boundary between B1 and B8 structures of FeO up to 210 GPa. Phys. Earth Planet. Inter. **179**, 157–163 (2010).
16. Tateno, S., Kuwayama, Y., Hirose, K. & Ohishi, Y. The structure of Fe–Si alloy in Earth’s inner core. Earth Planet. Sci. Lett. **418**, 11-19 (2015).
17. Mori, Y. et al. Melting experiments on Fe-Fe3S system to 254 GPa. Earth Planet. Sci. Lett. **464**, 135-141 (2017).
18. Fischer, R. A., Campbell, A. J., Caracas, R., Reaman, D. M., Dera, P. & Prakapenka, V. B. Equation of state and phase diagram of Fe–16Si alloy as a candidate component of Earth’s core. **357-358**, 268-276 (2012).
19. Blöchl, P. E. Projector augmented-wave method. Phys. Rev. B **50**, 17953–17979 (1994).
20. Kresse, G. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. Phys. Rev. B **54**, 11169–11186 (1996).
21. Alfè, D. & Gillan, M. J. First-Principles Calculation of Transport Coefficients. Phys. Rev. Lett. **81**, 5161 (1998).
22. He, Y. et al. First-principles prediction of fast migration channels of potassium ions in KAlSi3O8 hollandite: Implications for high conductivity anomalies in subduction zones. Geophys. Res. Lett. **43**, 6228-6233 (2016).
23. Alfè, D. Temperature of the inner-core boundary of the Earth: Melting of iron at high pressure from first-principles coexistence simulations. Phys. Rev. B **79** 060101 (2009).
24. Belonoshko, A. B., Ahuja, R., & Johansson B. Quasi–Ab Initio Molecular Dynamic Study of Fe Melting. Phys. Rev. Lett. **84**, 3638-3641 (2000).
25. Mookherjee, M., Stixrude, L. & Karki, B. Hydrous silicate melt at high pressure. Nature **452**, 983-986 (2008).
26. Hernandez, J. A. & Caracas, R. Superionic-Superionic Phase transitions in body-centered cubic H2O ice. Phys. Rev. Lett. **117**, 135503 (2016).
27. Belonoshko, A. B. et al. Stabilization of body-centred cubic iron under inner-core conditions. Nature Geosci. **10**, 312-316 (2017).
28. Steinle-Neumann, G., Stixrude, L., Cohen, R. E., & Gülseren O., Elasticity of iron at the temperature of the Earth’s inner core. Nature **413**, 57 (2001).
29. Belonoshko, A. B., Skorodumova, N. V., Rosengren, A., & Johansson, B., Melting and critical superheating. Phys. Rev. B **73**, 012201 (2006).
30. Alfè, D., Cazorla, C. & Gillan, M. J. The kinetics of homogeneous melting beyond the limit of superheating. J. Chem. Phys. **135,** 024102 (2011).
31. Alfè, D., Gillan, M. J. & Price, G. D. Temperature and composition of the Earth’s core. Contemporary Physics **48**, 63–80 (2007)
32. Alfè, D. First-principles simulations of direct coexistence of solid and liquid aluminum. Phys. Rev. B **68**, 064423 (2003).
33. Alfè, D. Melting Curve of MgO from First-Principles Simulations. Phys. Rev. Lett. **94**, 235701 (2005).
34. Alfè, D. Temperature of the inner-core boundary of the Earth: Melting of iron at high pressure from first-principles coexistence simulations. Phys. Rev. B **79**, 060101 (2009).
35. Bonev, S. A., Schwegler, E., Ogitsu, T. & Galli, G. A quantum fluid of metallic hydrogen suggested by first-principles calculations. Nature **204**, 669-672 (2004).
36. Ogitsu, T., Schwegler, E., Gygi, F. & Galli, G. Melting of Lithium Hydride under Pressure. Phys. Rev. Lett. **91**, 175502 (2003).
37. Morard, G. et al. Fe–FeO and Fe–Fe3C melting relations at Earth’s core–mantle boundary conditions: Implications for a volatile-rich or oxygen-rich core. Earth Planet. Sci. Lett. **473**, 94-103 (2017).
38. Anzellini, S., Dewaele, A., Mezouar, M., Loubeyre, P. & Morard, G. Melting of Iron at Earth’s Inner Core Boundary Based on Fast X-ray Diffraction. Science **340**, 464–466 (2013).
39. C. S. Yoo, N. C. Holmes, M. Ross, D. J. Webb, C. Pike, Shock temperatures and melting of iron at Earth core conditions. Phys. Rev. Lett. **70**, 3931 (1993).
40. Belonoshko, A. B., Rosengren, A., Burakovsky, L., Preston, D. L. & Johansson, B. Melting of Fe and Fe0.9375Si0.0625 at Earth’s core pressures studied using ab initio molecular dynamics. Phys. Rev. B **79**, 220102(R) (2009).
41. Alfè, D., Price, G. D. & Gillan, M. J. Iron under Earth's core conditions: Liquid-state thermodynamics and high-pressure melting curve from ab initio calculations. Phys. Rev. B **65**, 165118 (2002).
42. Sola, E. & Alfè, D. Melting of iron under Earth's core conditions from diffusion Monte Carlo free energy calculations. Phys. Rev. Lett. **103**, 078501 (2009).
43. Haule, K., Yee, C.-H. & Kim, K. Dynamical mean-field theory within the full-potential methods: Electronic structure of CeIrIn5, CeCoIn5, and CeRhIn5. Phys. Rev. B **81**, 195107 (2010).
44. Perdew, J. P., Burke, K. & Ernzerhof, M. Generalized gradient approximation made simple. Phys. Rev. Lett. **78**, 1396 (1997).
45. Xu, J. et al. Thermal Conductivity and Electrical Resistivity of Solid Iron at Earth’s Core Conditions from First Principles. Phys. Rev. Lett. **121**, 096601 (2018).
46. H. Gomi et al. The high conductivity of iron and thermal evolution of the Earth’s core. Phys. Earth Planet. Inter. **224**, 88 (2013).
47. de Koker, N., Steinle-Neumann, G., & Vlček, V. Electrical resistivity and thermal conductivity of liquid Fe alloys at high P and T, and heat flux in Earth’s core. Proc. Natl. Acad. Sci. U.S.A. **109**, 4070 (2012).
48. Pozzo, M., Davies, C., Gubbins, D. & Alfè D. Thermal and electrical conductivity of iron at Earth’s core conditions. Nature **485**, 355, (2012).
49. Nosé, S. A unified formulation of the constant temperature molecular dynamics methods, J. Chem. Phys. **81**, 511 (1984).
50. Voigt, W. (1928) Lehrbuch der kristallphysik[M]. Leipzig, Teubner, pp 962.