Supplementary Information for
Oxygen-Driven Enhancement of Electron
Correlation in Hexagonal Iron at Earth’s Inner
Core Conditions

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DFT+DMFT calculation

One should note that the proper values of $U$ and $J$ are method-dependent. They sensitively depend on the screening by the bands included in the model. The DFT+DMFT with Wannierized correlated orbitals usually use a smaller $U$ value since the Wannierization energy window is usually smaller than the energy window used in our calculation (−10 to 10 eV with respect to the Fermi level), resulting in a smaller screening effect. In addition, the Wannier functions contain a substantial amount of uncorrelated orbital subsets. As a result, the proper $U$ value can be smaller. The more detailed explanation can be found in the supplementary material of Ref.\textsuperscript{1,2}

The inverse quasiparticle lifetime $\Gamma$ is given by $-Z \text{Im}(i\omega^+)$, where $Z^{-1} = 1 - \partial \text{Im}\Sigma(i\omega)/\partial \omega$. Here we used averaged imaginary part of self-energy ($\text{Im}\Sigma$) of five $d$ orbitals of Fe atom.

The $dc$ electrical resistivity $\rho_{e-e}$ can be obtained using the Kubo-Greenwood formula:

$$ (\rho_{e-e})^{-1} = \frac{\pi e^2}{V} \sum_k d\epsilon(\sum_{\mathbf{k}}\rho_k(\epsilon)\rho_k(\epsilon)v_k)^2 $$  \hspace{1cm} (S1)

where $\rho_k(\epsilon)$ is the spectral function at wave vector $k$ and is related to the Green’s function $G(\epsilon)$ by $\rho_k = (G^\dagger - G)/(2\pi i)$, $v_k$ is the velocity vector, and $f$ is the Fermi-Dirac distribution function.

AIMD calculation

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

$$ \sigma_{ij} = C_{ijkl}\epsilon_{kl} $$  \hspace{1cm} (S2)

where $\sigma_{ij}$ refers to stress tensor, $\epsilon_{kl}$ refers to strain tensor, and $C_{ijkl}$ represents fourth-order elastic modulus. Considering the symmetry of $C_{ijkl}$, the equation is simplified as follows:
$$\sigma_i = C_{ij}\epsilon_j$$  \hspace{1cm} (S3)

Non-equivalent elastic constants of hcp Fe alloys were calculated by applying following
distortion matrices to the structures:

$$\begin{pmatrix} \delta & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \delta \end{pmatrix}, \text{ and } \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & \delta/2 \\ 0 & \delta/2 & 0 \end{pmatrix}$$  \hspace{1cm} (S4)

where \( \delta \) is the magnitude of distortion. For \( \delta \) in \( \pm 0.01, \pm 0.005, \) and 0, five groups of
strains were added by

$$a' = a(I + \epsilon)$$  \hspace{1cm} (S5)

where \( a' \) represents third-order cell parameter matrix, and \( \epsilon \) represents added strain
\( \Delta \epsilon \), and \( I \) represents third-order identity matrix. Over 10000-time steps (10 ps) of NVT
simulations were carried out for each direction of deformation to ensure the reliability and
full convergence of the results. The final results of strain-stress data show a very good linear
relationship and were fitted to equation [2] employing central difference method.

The Voigt scheme was used to estimate the elastic properties of Fe alloys. The bulk and
shear modulus are defined as:

$$K_V = \frac{2(C_{11} + C_{12}) + 4C_{13} + C_{33}}{9}, \quad G_V = \frac{12C_{44} + 7C_{11} - 5C_{12} + 2C_{33} - 4C_{13}}{30}$$  \hspace{1cm} (S6)

With the calculated moduli and density, the compression \( (V_p) \) and shear \( (V_s) \) velocities
in Fe and Fe\(_9\)O are obtained.
\[ V_P = \sqrt{\frac{B + \frac{4G}{3}}{\rho}}, \quad V_S = \sqrt{\frac{G}{\rho}} \]  

(S7)

The calculated elastic constants, densities, moduli, and seismic velocities of Fe and Fe\(_9\)O are presented in Table S1 and seismic velocities are compared with the geophysical observed PREM.
Figure S1: Crystal structures of oxygen, FeO$_2$, and Fe$_2$O used in the convex hull plot of Fig.1 in the main text.

Figure S2: Crystal structures of FeO and the enthalpy difference between three different phases at 300 GPa as a function of $U$. At $U = 0$ eV (DFT 0 K), R3m phase is the most stable phase like the previous studies. As $U$ value increases in DFT+DMFT calculation, R3m phase becomes unstable and B2 phase becomes the most stable phase with $U = 10$ eV. The uncertainty denoted by error bar arises from the Monte Carlo sampling in DMFT calculations. This result clearly indicates the importance of electron correlation effect of Fe 3$d$ orbitals.
Figure S3: Partial density of states (PDOS) and Imaginary self-energy on real frequency for Fe$_9$O and hcp-Fe ($U = 5$ eV, $J = 0.943$ eV, $T = 7000$ K) (a) PDOS of Fe$_9$O and hcp-Fe. Due to the interaction and charge transfer between Fe1 and O atom, PDOS of Fe1 atom is different from those of other Fe atoms in Fe$_9$O. The peak at -12 eV indicates the hybridization between O 2p and Fe1 $E''$ orbitals. Density of states of Fe1 atom of Fe$_9$O at the Fermi level ($E_F$) is larger than those of hcp-Fe and other Fe atoms in Fe$_9$O. From Fe2 atom, PDOS is almost identical to that of hcp-Fe. (b) Imaginary self-energy ($\text{Im}\Sigma(\omega)$) of Fe$_9$O and hcp-Fe. Like PDOS, the self-energy is converged to that of hcp-Fe from Fe2 atom. However, $\text{Im}\Sigma(\omega)$ of Fe1 atom shows a clear difference. First, $-\text{Im}\Sigma(\omega)$ of Fe1 atom is larger than those of hcp-Fe and other Fe atoms in Fe$_9$O, indicating the enhanced electron-electron scattering. Second, $\text{Im}\Sigma(\omega)$ of $E''$, which is strongly hybridized with O 2p orbitals, shows a clear difference from others.
Figure S4: $k$-resolved spectral function obtained from DFT+DMFT calculations ($U = 5$ eV, $J = 0.943$ eV, $T = 7000$ K) Due to the charge transfer from Fe1 to O atom, the flat band located around 1 eV in the pure hcp-Fe moves toward $E_F$. This feature can be clearly seen from Fe$_7$O case and induces the larger density of state near $E_F$ as shown in Fig. S2 and Fig. 2c in the main text.

Figure S5: Directional resistivity obtained from DFT+DMFT calculations In the main text, the averaged value of $\rho_{xx}$ and $\rho_{zz}$ are shown for simplicity.
Figure S6: Calculated mean square displacements (MSDs) of Fe and O ions in hcp Fe$_9$O at 360 GPa and temperatures from 4000 to 6000 K. No ionic diffusion is observed in hcp Fe$_9$O indicating the stability of hcp Fe$_9$O under inner core conditions.
Table S1: The density and compression velocity ($V_P$) obtained from DFT calculations at 300 GPa and comparison with the previous theoretical results

| Phase  | hcp-Fe | Fe$_9$O |
|--------|--------|---------|
|        | Vočadlo et al. | This work | This work |
| Method | AIMD 5500 K, 300 GPa | AIMD 0 K, 300 GPa | DFT 300 GPa | DFT 300 GPa |
| Density (g/cm$^2$) | 13.16 | 13.70 | 13.78 | 13.28 |
| $V_P$ (km/s) | 11.17 | 12.59 | 12.66 | 12.80 |

Table S2: The calculated elastic constants, densities, moduli and seismic velocities of Fe and Fe$_9$O at high P-T

| Phase  | P (GPa) | T (K) | Density (g/cm$^2$) | $C_{11}$ (GPa) | $C_{12}$ (GPa) | $C_{13}$ (GPa) | $C_{33}$ (GPa) | $C_{44}$ (GPa) | $K_V$ (GPa) | $G_V$ (GPa) | $V_P$ (Km/s) | $V_S$ (km/s) |
|--------|---------|------|-------------------|----------------|--------------|--------------|--------------|--------------|-------------|-------------|-------------|-------------|
| Fe     | 359.5   | 4992.7 | 13.90             | 1996.3         | 1235.5       | 1085.6       | 2047.7       | 308.4        | 1428.2      | 375.0       | 11.78       | 5.19        |
| Fe     | 359.5   | 5990.0 | 13.77             | 1855.7         | 1287.0       | 1028.5       | 1928.9       | 225.8        | 1367.2      | 300.3       | 11.33       | 4.67        |
| Fe$_9$O | 359.6   | 4000.1 | 13.49             | 2042.2         | 1255.2       | 1115.8       | 2133.5       | 302.9        | 1465.7      | 381.9       | 12.10       | 5.32        |
| Fe$_9$O | 359.1   | 5000.4 | 13.36             | 1904.9         | 1290.7       | 1103.3       | 2037.8       | 242.1        | 1417.1      | 321.9       | 11.76       | 4.91        |
| Fe$_9$O | 360.8   | 6000.2 | 13.24             | 1769.6         | 1268.6       | 1170.3       | 205.7        | 205.4        | 1406.0      | 254.4       | 11.48       | 4.38        |
| Fe$_9$O | 330.9   | 6000.5 | 12.95             | 1684.7         | 1138.9       | 1027.7       | 1733.6       | 169.4        | 1276.8      | 249.6       | 11.15       | 4.39        |
Supplementary References

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(2) Kim, H.-S.; Haule, K.; Vanderbilt, D. Mott Metal-Insulator Transitions in Pressurized Layered Trichalcogenides. *Phys. Rev. Lett.* **2019**, *123*, 236401.

(3) Vočadlo, L.; Dobson, D. P.; Wood, I. G. Ab initio calculations of the elasticity of hcp-Fe as a function of temperature at inner-core pressure. *Earth Planet. Sci. Lett.* **2009**, *288*, 534–538.