

Collective motion in hcp-Fe at Earth's inner core conditions

Youjun Zhang^{a,c,1}, Yong Wang^{b,1}, Yuqian Huang^a, Junjie Wang^b, Zhixin Liang^b, Long Hao^d, Zhipeng Gao^d, Jun Li^{d,2}, Qiang Wu^d, Hong Zhang^e, Yun Liu^c, Jian Sun^{b,2}, Jung-Fu Lin^{f,2}

^aInstitute of Atomic and Molecular Physics, Sichuan University, Chengdu 610065, China.

^bNational Laboratory of Solid State Microstructures, School of Physics and Collaborative Innovation Center of Advanced Microstructures, Nanjing University, Nanjing, 210093, China.

^cInternational Center for Planetary Science, College of Earth Sciences, Chengdu University of Technology, Chengdu 610059, China.

^dNational Key Laboratory for Shock Wave and Detonation Physics, Institute of Fluid Physics, Mianyang 621900, China.

^eCollege of Physics, Sichuan University, Chengdu 610065, China.

^fDepartment of Geological Sciences, Jackson School of Geosciences, The University of Texas at Austin, Austin, TX 78712, USA.

¹Y. Z. and Y.W. contributed equally to this work.

²To whom correspondence may be addressed: afu@jsg.utexas.edu; jiansun@nju.edu.cn; lijun102@caep.cn;

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Main Text

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Abstract

Earth's inner core is predominantly composed of solid iron (Fe) and displays intriguing properties such as strong shear softening and an ultrahigh Poisson's ratio. Insofar, physical mechanisms to explain these features coherently remain highly debated. Here we have studied longitudinal and shear wave velocities of hcp-Fe (hexagonal close-packed iron) at relevant pressure-temperature (P-T) conditions of the inner core using *in-situ* shock experiments and machine learning molecular dynamics simulations. Our results demonstrate that the shear wave velocity of hcp-Fe along the Hugoniot in the premelting condition, defined as T/T_m (T_m : melting temperature of iron) above 0.96, is significantly reduced by ~30% while Poisson's ratio jumps to approximately 0.44. Machine learning molecular dynamics simulations at 230-330 GPa indicate that collective motion with fast diffusive atomic migration occurs in premelting hcp-Fe primarily along [100] or [010] crystallographic direction, contributing to its elastic softening and enhanced Poisson's ratio. Our study reveals that hcp-Fe atoms can diffusively migrate to neighboring positions, forming open-loop and close-loop clusters in the inner core conditions. Hcp-Fe with collective motion at the inner core conditions is thus not an ideal solid previously believed. The premelting hcp-Fe with collective motion behaves like an extremely soft solid with an ultralow shear modulus and an ultrahigh Poisson's ratio that are consistent with seismic observations of the region. Our findings indicate that premelting hcp-Fe with fast diffusive motion represents the underlying physical mechanism to help explain the unique seismic and geodynamic features of the inner core.

Significance

Earth's solid inner core exhibits intriguing characteristics such as an exceptionally low shear-wave velocity and an ultrahigh Poisson's ratio that significantly deviate from our understanding for normal solid metals. In this study, we have used high pressure-temperature experiments and machine learning calculations with extremely large supercells to examine the dynamics and sound velocities of iron at the extreme pressure-temperature conditions of the inner core. Our research results reveal a drastic reduction in shear wave velocity when hcp-Fe approaches the melting point induced by shock. Our advanced machine learning calculations show that iron atoms undergo collective motion with fast diffusion in the premelting regime, leading to the ultralow shear modulus and ultrahigh Poisson's ratio. These results are consistent with seismic observations and

67 geodynamics of the inner core. We thus propose that collective motion in hcp-Fe is the underlying
68 physical mechanism for the soft inner core very close to the melting curve of constituent iron alloy.
69

Introduction

Seismology and mineral physics studies over the past century reveal that Earth's inner core is mainly made of solid iron (approximately 96 wt% iron) (1, 2). The inner core only accounts for less than 1% of the planet's total volume but plays a key role in its dynamics, thermal evolution, core convection, and habitability. Of particular example is its nucleation and light element release to provide thermochemical energy sources powering the geodynamo, the generation of the magnetic field, and subsequent geological processes (3-5). Recent seismological investigations from modern J wave (6) and preliminary reference Earth model (1) have shown that the inner core has a shear wave velocity 30-40% lower than expected from solid hcp-Fe alloy at similar pressure, while the compressional wave velocity is overall consistent with that of the constituent Fe alloy (7, 8). This indicates an ultralow shear modulus in the inner core, with a Poisson's ratio (ν) as high as 0.44-0.45, analog to that of soft metals such as lead and thallium (9) and close to that of molten iron ($\nu = 0.5$). The Poisson's ratio of the inner core is much larger than that of hcp-Fe estimated at the core's pressure and room temperature ($\nu = \sim 0.35$) (10, 11). These seismic observations, therefore, suggest the existence of an extremely soft solid iron alloy with low rigidity in the inner core (12). This raises the fundamental question about the main underlying physical mechanism responsible for the unique seismic and geodynamic features in the region. Understanding the atomistic dynamics and its influence on the longitudinal (V_p), bulk (V_b), and shear (V_s) sound velocities of iron at inner core conditions can greatly enhance our knowledge of the inner working of our planet (13-15).

Driven by the continuous cooling of the planet, the solidification of the inner core occurs when its adiabat intersects the melting curve of the constituent iron alloy at the inner-core boundary (ICB) (16). Using the melting curve of pure iron as a reference, the melting temperature (T_m) at the ICB is determined to be $\sim 6200(300)$ K at 330 GPa by experiments (17-21) and theories (22, 23). Addition of ~ 4 wt.% light element(s) in iron is expected to depress the melting temperature by several hundreds of K (24). According to an isentropic modeling, the temperature gradient across the inner core radius (r), dT_i/dr , is expected to be approximately -0.17 K/km (16). This suggests that the temperature increase is only 100-200 K from the ICB to the center across the radius of ~ 1200 km. As a result, the inner core adiabat is expected to be within the temperature regime T_i/T_m exceeding 0.95. As the temperature approaches T_m , a previous study has suggested that solid metals can commonly show premelting behaviors with collective motion and fast atomic diffusion (25). Some simulations have predicted that unusual elastic and rheological properties occur in several

metals like Cu, Al, Ta, and Ca at the premelting temperature (26-28). This raises the possibility that premelting iron at T/T_m above 0.95 and inner-core pressures can exhibit unique elastic and rheological characteristics manifested in the aforementioned seismic observations.

To date, several hypotheses have been proposed to explain the observed seismic features in the inner core (7, 29-32). A pioneering study on the elasticity calculations of hcp-Fe predicted a strong shear softening at high P-T prior to melting (30), suggesting that premelting hcp-Fe may exhibit distinct behavior from ideal hcp-Fe. However, the simulation was conducted using relatively short simulation time and small cell size under homogeneous melting with superheating effects (> 7500 K at the ICB pressure where iron is expected to be molten) (30). On the other hand, first-principles DFT calculations showed that the shear modulus of hcp-Fe decreases quasi-linearly with temperature at a given density and agrees with the seismology and free oscillation data for the inner core without the requirement of premelting behavior (33, 34). Numerous high P-T experiments and modeling studies on the strength and plastic deformation have suggested that hcp-Fe is rheologically weak with a low shear strength due to pressure-induced slip and creep and its shear strength could be reduced by the elevated temperature (35-39). However, these experiments were conducted at P-T conditions far below from those of the inner core. Furthermore, some high-pressure experiments and molecular dynamic (MD) computations indicate that iron carbide (*e.g.*, Fe_7C_3 compound (31)) or iron-hydrogen superionic alloy (32, 40) can reduce the shear wave velocity of the inner core. However, it is still highly controversial whether C and/or H light elements exist in Earth's inner core (24). Bcc-Fe has also been proposed as a possible explanation for the origin of the low shear wave velocity in the inner core (41-43), but *in situ* x-ray diffraction (XRD) experiments did not observe its existence at the relevant P-T conditions (19, 20, 44). Possible presence of melt pockets in the inner core has been suggested as an explanation for the high Poisson's ratio (45), although the inner core growth models indicate that interstitial liquid would have been squeezed out during its complete solidification and compaction below the solidus (46). Insofar, these existing hypotheses could help address some aspects of the inner-core's unique properties, but a comprehensive physical model incorporating direct experimental elasticity data and theoretical simulations with large supercells remains lacking.

Iron is believed to be stable in the hcp structure under the inner core P-T conditions as revealed by both static (18, 44) and dynamic (19, 20) experiments. In previous elasticity studies of iron, several experimental investigations of the sound velocities and Poisson's ratio of hcp-Fe have been carried out (29, 47-49); however, some experiments were conducted at relatively low P-T

conditions (*e.g.*, up to ~ 73 GPa and 1700 K (29); up to ~ 300 GPa at room temperature (50, 51)) or did not reach to the temperature prior to melting points (*e.g.*, static (49) and shock (47, 48) experiments). In this study, we have performed direct shock-wave measurements and machine learning-enhanced simulations with supercell sizes of more than 10,000 atoms to investigate the V_p , V_b , and V_s of compressed hcp-Fe near the premelting temperature at the core pressure. Our theoretical results suggest that collective atomic motion occurs in premelting hcp-Fe, causing a strong sound velocity softening and enhanced Poisson's ratio. The collective motion-induced elastic softening in hcp-Fe is proposed to be the underlying physics to explain the unique seismic features of the inner core, including its ultralow shear wave velocity, low rigidity, and ultrahigh Poisson's ratio.

Results and Discussion

High P-T shock compression experiments were conducted on high purity polycrystalline iron ($>99.98\%$) using hypervelocity reverse-impact technique in two-stage light-gas guns (see [Materials and Methods](#) and [SI Appendix Fig. S1](#)). Iron was shock-compressed to pressures ranging from 82-231 GPa and temperatures ranging from 1600-5220 K along its Hugoniot P-T curve, where the shock Hugoniot temperature has been well determined by both experiments and simulations (17, 19, 23, 52) (see [SI Appendix Table S1 and Fig. S8](#)). The most recent shock compression experiments coupled with *in situ* XRD indicate that the hcp-Fe should be the only phase present at pressures between 200 and 1000 GPa prior to melting (19, 20). In this work, we targeted a maximum single shock pressure just below the pressure at which shock-induced melting occurs (~ 242 GPa) (19). Therefore, iron should be in the hcp structure at the present investigated P-T range. We then obtained the (Eulerian) sound velocities of hcp-Fe including V_p , V_b , and V_s under shock compression by solving the relationship among time interval, wave speed, and distance (see [SI Appendix Figs. S2-S4](#)).

Analysis of the shock experimental results indicates that the V_p of hcp-Fe linearly increases with increasing shock pressure (P_H) between 80-160 GPa ([Fig. 1a](#)). After further shock pressure increase, the rate of change in V_p with respect to P_H (dV_p/dP_H) gradually decreases and eventually changes to a negative slope above ~ 206 GPa. Additionally, the V_b in shock-compressed iron shows a monotonous increase from 82 to 231 GPa. Our measured V_p are generally consistent with those of shock-compressed iron in Refs. (48, 53), while we did not observe a sharp discontinuity of V_p at 200-220 GPa as reported earlier that was explained as a solid-solid phase transition from hcp to

a new phase (47). Our V_p results at relatively low P-T conditions agree well with an earlier report (49) on hcp-Fe up to ~ 163 and 3000 K using inelastic x-ray scattering (IXS) in laser-heated diamond anvil cells (LH-DACs) (Fig. 1a). Both sets of results support the notion that the V_p of hcp-Fe exhibits a quasi-linear relationship with density in this P-T range (49). However, our results suggest that as the P-T conditions approach the region of shock-induced melting, the sound velocity of hcp-Fe has a nonlinear relation with density and does not follow the quasi-linear sound velocity-density relationship (see SI Appendix Fig. S9). Our measured V_s of iron under shock compression shows a gradual increase with pressure and density from ~ 80 to 160 GPa but stops to increase above 160 GPa. At shock pressures above 200 GPa, the V_s of iron displays a dramatic drop from 4.2 km/s to zero as iron melts along the Hugoniot (Fig. 1a and SI Appendix Fig. S9).

Under shock compression, both P-T is simultaneously elevated along the Hugoniot, which could affect the sound velocities of hcp-Fe in a different way. Previous experiments using a LH-DAC has shown that the V_p and V_s of hcp-Fe generally increases with pressure but decreases with temperature (29, 54). Therefore, we have further investigated the effect of the shock Hugoniot temperature (T_H) on the sound velocity of hcp-Fe using the recently developed optical pyrometry technique (17). These results show that T_H in hcp-Fe increases monotonously from approximately 1600 to 5220 K as pressure increases from 82 to 231 GPa (see SI Appendix Fig. S8). Based on a consensual melting curve of hcp-Fe (17-21), we calculated its ratio of shock Hugoniot temperature to the melting point, T_H/T_m , which gradually increases from 0.44 to 0.96 with shock pressure from 82-231 GPa (see SI Appendix Table S1). We note that the V_s stops to increase at shock pressures above approximately 160 GPa, where the T_H/T_m approaches ~ 0.74 . At P-T conditions further up to ~ 230 GPa, with a solid Hugoniot temperature close to the melting point ($T_H/T_m \sim 0.96$), the V_s values drastically drops by approximately 30% (Fig. 1a). Our shock results reveal that the V_s of hcp-Fe at $T_H/T_m \sim 0.96$ -1.00, which we define as the premelting region, displays a strong temperature-dependent reduction. This dramatic drop in the measured V_s agrees with some previous theoretical predictions by *ab initio* molecular dynamics (AIMD) simulations (30), which shows a strong nonlinear elastic constant weakening in hcp-Fe just before melting.

Based on our measured V_p , V_b , and V_s along the Hugoniot, the derived Poisson's ratio of iron increases from ~ 0.34 to 0.43 as shock pressure increases from ~ 82 GPa (1600 K) to 231 GPa (5220 K), as shown in Fig. 1b. At conditions below ~ 160 GPa and 3450 K (with T_H/T_m less than ~ 0.74), the Poisson's ratio appears to be comparable to that of hcp-Fe at high pressure and room temperature (~ 0.35), as observed in DAC experiments (10, 11) (Fig. 1b), suggesting that

temperature has only a minor effect at relatively low values of T_H/T_m . Furthermore, static high-pressure experiments demonstrate that the Poisson's ratio of iron does not significantly change with increasing pressure to the core's at room temperature (10, 11), indicating that the rise in shock-compressed iron up to 230 GPa should be mainly due to shock-induced high temperatures. When compared to seismic observations, the Poisson's ratio of 0.43 in hcp-Fe at $T_H/T_m \sim 0.96$ and $P_H \sim 230$ GPa is generally consistent with the ratio of ~ 0.44 - 0.45 in the inner core (the striped rectangle area in Fig. 1b).

To gain insight into the atomistic behavior and lattice dynamics of premelting hcp-Fe, we developed a machine learning potential with density-functional-theory (DFT) accuracy, using the deep concurrent learning method (see *Materials and Methods* and *SI Appendix Figs. S5-S7*). Equilibrium and non-equilibrium molecular dynamics simulations (EMD and NEMD) were conducted at pressures ranging from 230-330 GPa and at various temperatures, up to the nanosecond simulation scale. At temperatures close to the melting point ($T/T_m > \sim 0.96$), we observed the spontaneous emergence of atomic collective motion in hcp-Fe, along its [100] or [010] crystallographic orientation, where Fe atoms diffuse collectively in both intralayer and interlayer directions (Fig. 2a and *SI Appendix Movie S1*). Specifically, most atoms exhibit a one-dimensional (1-D), longitudinal wave-like behavior in open-loop (chain-like) diffusions, wherein one atom jumps out of its equilibrium position and pushes its neighboring atoms along specific crystallographic directions, such as [100] or [010] in the a-b plane (Fig. 2b). This collective motion generates a wavefront of diffusion, moving randomly forward until it meets a vacancy and releases the compression. This phenomenon was detailed proposed in cubic Ca within the premelting regime under high pressure and has been indicated as a possibly prevalent high-temperature feature in metals under extreme conditions (28). The longitudinal-wave-like behavior could dissipate shear in the system at the atomistic level, which was also observed in the premelting bcc-Ta (55) and fcc-Al (56) at ambient pressure.

The distribution of iron atomic displacements is also investigated within a given lag time (up to 0.1 ns) using time-dependent van Hove self-correlation functions $P(\Delta r, t)$ (57, 58). At the V_s softening regime (e.g., $T/T_m \sim 0.98$), hcp-Fe atoms migrate collectively to their neighboring lattice points. This migration leads to a non-Gaussian multiple-peak distribution of $P(\Delta r, t)$ (horizontal dashed-line in Fig. 2c). However, in solid hcp-Fe at a relatively low temperature below the region (e.g., $T/T_m \sim 0.92$), $P(\Delta r, t)$ exhibits a stable Gaussian distribution over time in the nanosecond-scale simulation, indicating the absence of collective motion (see *SI Appendix Fig. S11*).

Furthermore, we computed the mean square displacements (MSD) for hcp-Fe at different temperatures and found that the MSD of premelting hcp-Fe at 230 GPa and 5300 K ($T/T_m \sim 0.98$) steadily increases throughout 250 picosecond (ps) simulation time. This means that collective motion contributes to significant diffusion of atoms (Fig. 3a). Notably, the average diffusion in the a-b planes of premelting hcp-Fe spreads faster than that along the c-axis. Conversely, at a relatively low temperature of 4000 K ($T/T_m \sim 0.74$) at 230 GPa, the MSD of hcp-Fe atoms remains nearly constant during the simulation time, indicating no observable diffusion (Fig. 3a).

For a deeper understanding of the collective atomic motion in premelting hcp-Fe, we have theoretically investigated its finite-temperature phonon dispersions including the anharmonicity and enthalpy at 230 GPa and various temperatures (Fig. 3b). Our calculations show abnormal softening in some high symmetry points at high P-T (Fig. 3b), corresponding to the relative vibrations between close-packed planes along the c-axis in hcp structures (Fig. 3c). These softening reflect temperature effects as a dynamical driving force for collective atomic motion, which mostly diffuses between the close-packed planes of hcp-Fe. From an energetic perspective, the mixture of solid and liquid near melting promotes disorders and increases the system's potential energy compared to an ideal hcp-Fe crystal. However, the directional priority of collective motion pushes atoms to diffuse along the lowest energy paths, which only slightly increases the system's enthalpy compared to ideal hcp-Fe without collective motion (Fig. 3d). Given the complexity of the potential energy surface at high temperatures and the large entropy induced by the collective motion, hcp-Fe with collective motion may be dynamically stable in the premelting P-T conditions.

To investigate how collective atomic motion affects physical properties of hcp-Fe, we conducted calculations on its elastic constants and Poisson's ratio at high P-T. It should be noted that traditional elastic modulus calculations using EMD method for single crystal hcp-Fe are susceptible to superheating effects. To overcome this, we employed a solid defective structure quenched from the two-phase model in NEMD simulations to mitigate the impact of superheating in order to accurately calculate the high P-T elastic constants (see Supplementary Material). Results from both the *ab initio* method and the constructed machine learning model show that a significant shear elastic softening occurs in hcp-Fe at 230 GPa as the temperature approaches the melting point, consistent with the present experimental observations (Fig. 4). The present calculations and experiments indicate that the shear modulus of hcp-Fe at 230 GPa and $T/T_m \sim 0.96-0.98$ agrees overall with the seismically observed low shear modulus of the inner core (Fig.

4). Furthermore, the calculated Poisson's ratio using NEMD increases from 0.36 to 0.42 as the temperature rises from 300 to 5000 K ($T/T_m \sim 0.92$) at 230 GPa (Fig. 1b). When the temperature continues to rise from 5000 K to the melting point at 5400 K, the Poisson's ratio of hcp-Fe increases dramatically from 0.42 to 0.5. Our computations reveal that the Poisson's ratio in hcp-Fe with collective motion is approximately 0.45 at 230 GPa and 5200 K ($T/T_m \sim 0.96$), which is overall consistent with our experimental value at similar P-T conditions (Fig. 1b).

We conducted more calculations at pressures ranging from 230 to 330 GPa and temperatures between 4000 and 7000 K to determine the P-T regime of hcp-Fe with collective motion at relevant inner core conditions. Our results reveal the dynamic phase boundary between ideal hcp-Fe and hcp-Fe with collective motion, as well as the boundary between hcp-Fe and liquid Fe (Fig. 5). We found that the temperature for hcp-Fe with collective motion (T_c) extends approximately 500 K below its T_m at inner core pressures. Comparison between our predicted dynamic phase of hcp-Fe with collective motion and a typical adiabat of iron core indicates that the inner core adiabat is slightly below the T_m and falls within the temperature region of premelting hcp-Fe with collective motion (Fig. 5 and SI Appendix Fig. S13). As mentioned in the Introduction, the T/T_m of the inner core is expected to be between 0.95-0.99 (16, 59). Our results indicate that the dynamics of collective motion in the premelting hcp-Fe at the conditions relevant to the inner core may be the primary physical mechanism behind the observed seismic properties, such as the ultralow shear wave velocity, low rigidity, and ultrahigh Poisson's ratio (6).

Earth's inner core also contains ~5 wt.% Ni and ~4 wt.% light element(s) alloyed with iron, though the exact identity of the major light element remains uncertain (24). Here we consider Si as a candidate light element as an example to understand light element effects on T_m depression and associated elasticity. Addition of 4 wt.% of Si in Fe or Fe-Ni alloy would lower the melting temperature to a T_m of ~5800 K (60) and overall increase the V_p by 3-5% (50, 54, 61, 62). Meanwhile, Fe alloyed with ~5 wt.% Ni and a few % of Si would retain the hcp structure at relevant inner-core P-T conditions (63-65). Using a temperature gradient of 100-200 K in the inner core, we estimated that the expected inner core adiabat for iron alloyed with a few wt.% light elements would have a T/T_m of ~0.97-0.98, which falls within the premelting region ($T_c/T_m > \sim 0.95$). These results suggest that such an Fe-Ni-light element alloy can also exhibit collective atomic motion, resulting in the ultralow shear wave velocity and ultrahigh Poisson's ratio of the inner core.

Our research into the effects of collective atomic motion on premelting hcp-Fe has yielded exciting results regarding its shear softening and ultrahigh Poisson's ratio. These findings provide

a novel physical mechanism to explain the unique seismic and geodynamic features of Earth's solid inner core. The ultralow shear modulus in premelting hcp-Fe suggests that the inner core may have relatively low shear viscosity, making it susceptible to deformation and convectively instability (66). Future exploration into the atomistic dynamic behavior and high pressure-temperature sound velocities of iron alloyed with approximately 5 wt.% nickel and 2-4 wt.% light elements at the inner-core conditions is necessary to shed more light on the geophysics and geodynamics of both the inner and outer cores. Together with previous research on premelting metals (26-28), a scenario beyond the perspective of classical nucleation theory together with its potential effects on material properties such as the elasticity need to be further clarified in future works.

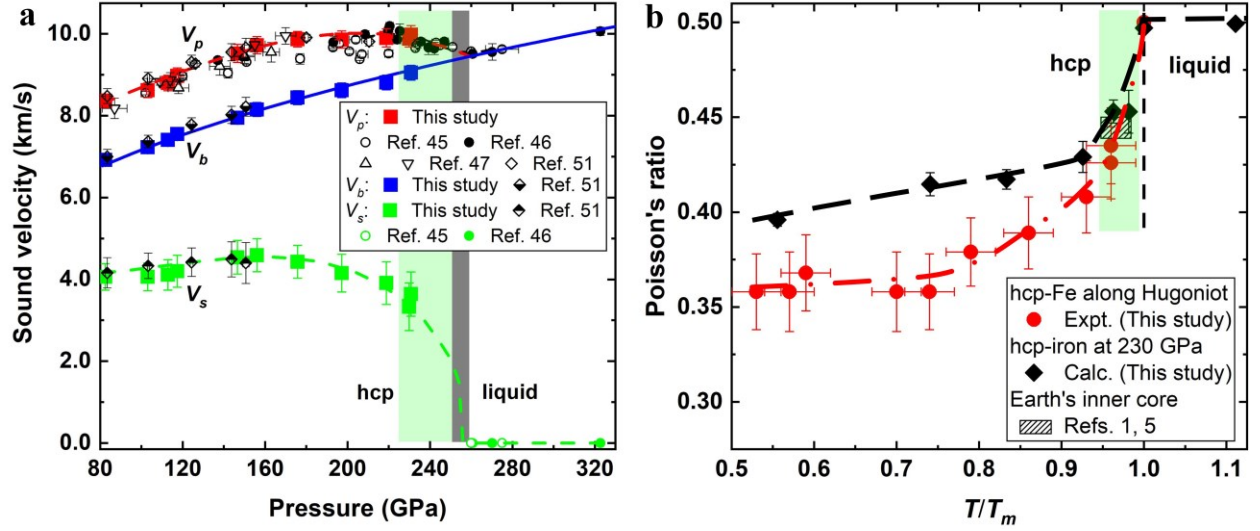


Fig. 1. Sound velocities and Poisson's ratio of iron along the shock Hugoniot conditions. (a) Measured sound velocities V_p (red squares), V_b (blue squares), and V_s (green squares) of iron under shock compression. Solid (48) and open (47) circles are the measured V_p under shock compression from the literature results. Diamonds, diamonds with bottom half black, and diamonds with top half black represent the measured V_p , V_b , and V_s , respectively, under shock compression by Ref. (53). The triangles and inverted triangles represent the measured V_p of hcp-Fe at given pressures and ~ 3000 and 2300 K, respectively, using IXS in LH-DACs (49). The blue line represents the calculated V_b of iron using a thermodynamic equation under shock compression. The red dashed line in V_p was fitted to the measured data using a third-order polynomial and is meant as a guide for the eye. The green dashed line through the experimental data in V_s are derived from the V_p and V_b . The vertical grey bar represents the pressure range for iron melting to occur along the shock Hugoniot. (b) Measured and calculated Poisson's ratio of hcp-Fe as a function of the ratio of temperature to the melting point of iron (T/T_m). By definition, the Poisson's ratio reaches 0.5 at T/T_m of 1.0 in molten iron (horizontal dashed black line). The Poisson's ratio of the inner core constrained by seismic observations falls within the range of 0.44-0.45 (striped rectangle region) (1, 6), which is located at $T/T_m \sim 0.96$ for hcp-Fe at high pressure. The difference in Poisson's ratio between experimental and computational results at relatively low T/T_m may be attributed to the pressure difference between Hugoniot conditions in experiments and isobaric conditions in calculations. The light-green region indicates a significant shear softening and ultrahigh Poisson's ratio induced by collective atomic motion in hcp-Fe at shock pressures above ~ 230 GPa and T/T_m greater than ~ 0.96 .

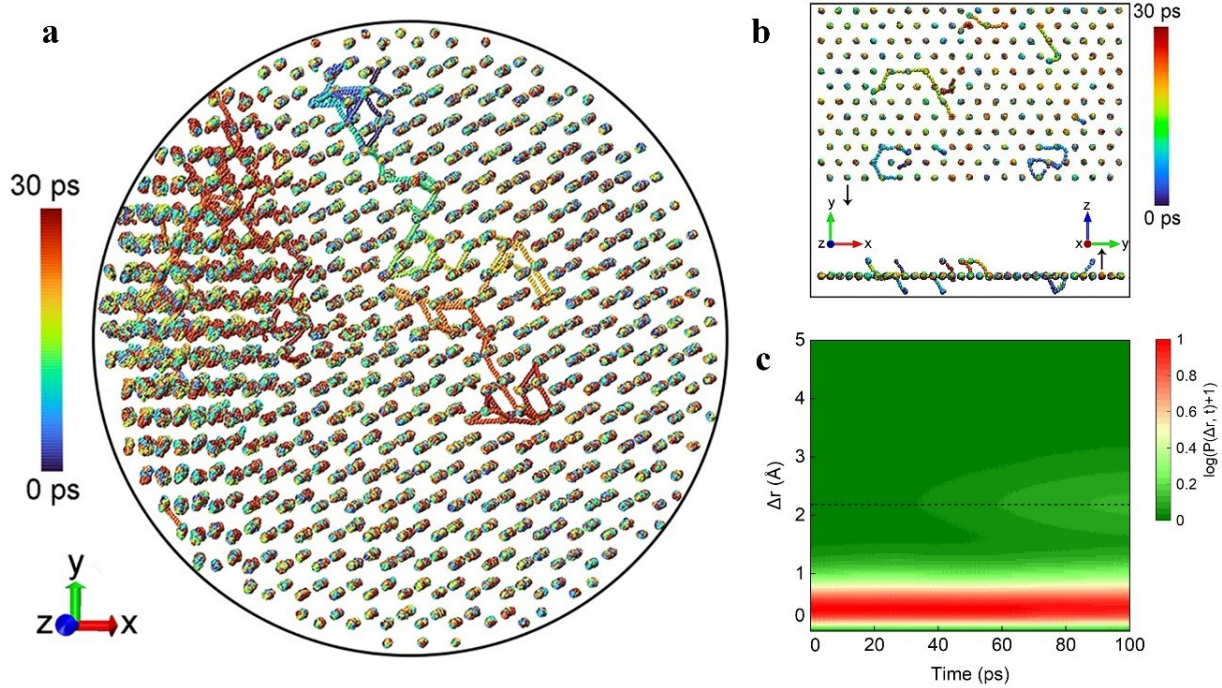


Fig. 2. Collective atomic motion of compressed hcp-Fe at near melting point and 230 GPa. (a) Dynamic behavior of hcp-Fe atoms at 230 GPa and near melting temperature ($T/T_m \sim 0.98$) using machine learning molecular dynamics. Iron atoms are colored with time over a 30 picosecond (ps) period in the snapshot. **(b)** Top- and side-view dynamical snapshots of a single close-packed a-b (x-y) plane during 30 ps in premelting hcp-Fe with collective motion. Iron atoms predominantly diffuse along [100] or [010] directions of the hcp structure. This diffusion leads to the formation of open-loop strings and a few closed-loop shapes such as triangular. The open-loop diffusion behavior propels iron atoms to form a 1-D longitudinal wave-like pattern. Snapshots in (a) and (b) were smoothed within a 1 ps time window to eliminate thermal noises. **(c)** Self van Hove correlation function $P(\Delta r, t)$ of premelting hcp-Fe. Iron atoms migrate to neighbors during collective atomic motion, which induces multiple-peak non-Gaussian distribution vs. time in the correlation functions. The $P(\Delta r, t)$ represents the probability of finding an atom at distance Δr after a time interval t . Vertical bar represents the logarithm form of the $P(\Delta r, t)$ to highlight the diffusion of collective motion (marked by the black dashed line as the nearest neighbor distance).

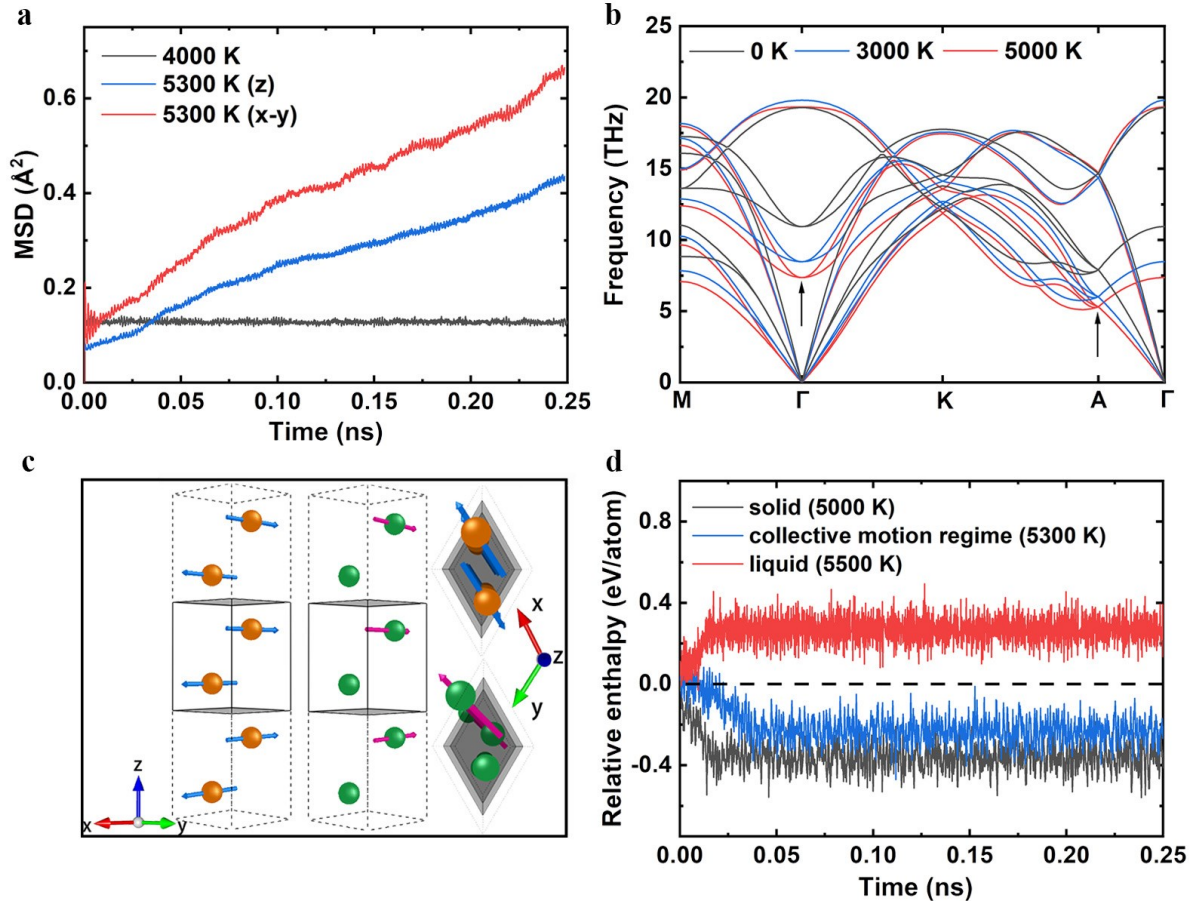


Fig. 3. Dynamical properties of hcp-Fe at ~230 GPa and representative temperatures. (a) Calculated mean square displacements (MSD) of hcp-Fe atoms at 230 GPa and varied temperatures. The atoms in premelting hcp-Fe diffuses with the simulation time. The diffusion of hcp-Fe atoms primarily occurs in the a - b planes as they migrate to neighboring atoms. Diffusion in the z -direction is usually triggered by the vacancies' migration between different a - b planes, of which vacancies were made by the collective motion in a - b planes. (b) Phonon spectra of hcp-Fe at 230 GPa and three given temperatures (0 K, 3000 K, and 5000 K). Anharmonicity is taken into consideration in the calculations. Black arrows highlight some modes in the phonon spectra that show strong softening as the temperature increases. (c) Schematic diagram of the softening direction of hcp-Fe at high P-T. Blue arrows through orange atoms show a strong phonon softening in the Γ point of hcp-Fe reduced from ~12 THz at 0 K to ~7 THz at 5000 K and at 230 GPa, where the vibrational direction is between adjacent a - b close-packed planes. Magenta arrows through green atoms show a similar softening direction in the A (0, 0, 0.5) point, where the softening happens between non-adjacent planes. (d) Enthalpy of iron in hcp, hcp with collective motion, and liquid states, respectively, at 230 GPa as a function of simulation time. Systems evolve from the initial solid-liquid mixture to hcp crystal at 5000 K ($T/T_m \sim 0.92$), to hcp with collective motion at 5300 K ($T/T_m \sim 0.98$), and to liquid at 5500 K ($T/T_m \sim 1.02$), respectively. The horizontal dashed line represents the initial enthalpy of the solid-liquid mixture (see [SI Appendix Fig. S7](#)).

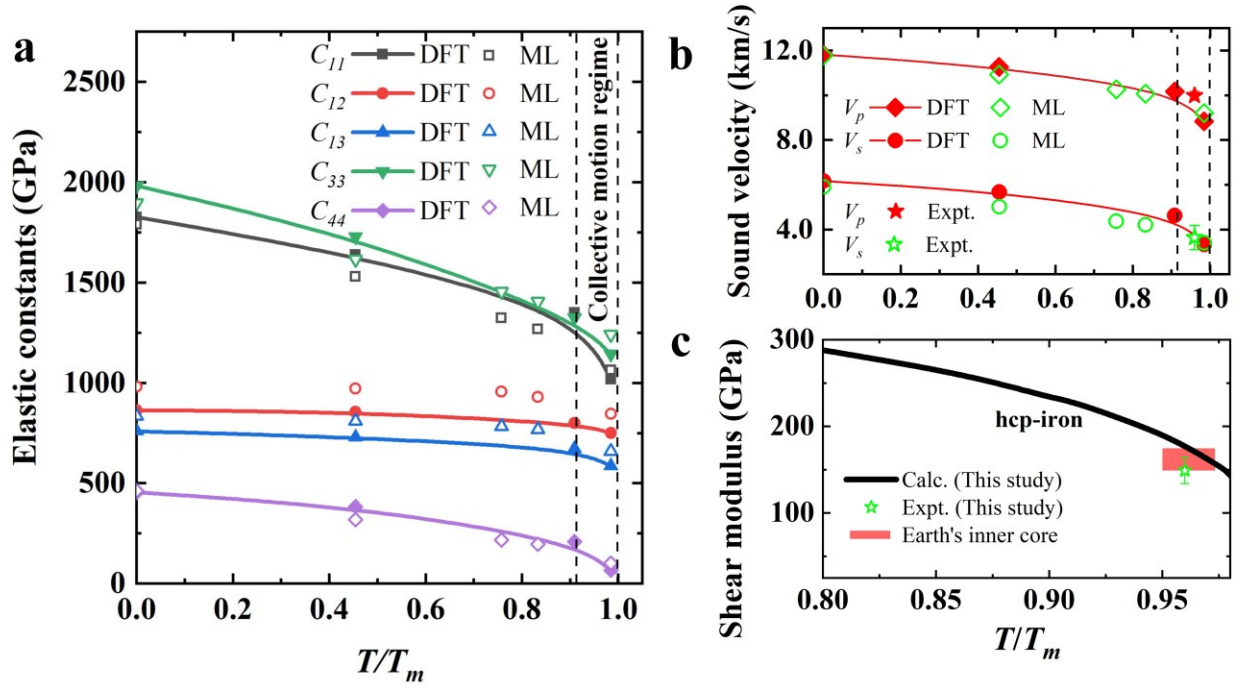


Fig. 4. Elastic constants, sound velocities, and shear modulus of hcp-Fe at high pressure-temperature conditions. (a) Calculated elastic constants of hcp-Fe as a function of the ratio of temperature to its melting point (T/T_m) at 230 GPa. The elastic constants were calculated using both the DFT-AIMD and machine learning (ML) method. (b) Comparisons of sound velocities of hcp-Fe between theoretical and experimental results at high temperatures and 230 GPa. Solid and open symbols represent the results from the DFT-AIMD and ML calculations, respectively, in this study. The melting point T_m of iron at 230 GPa is calculated to be ~ 5400 K by two-phase methods (see *SI Appendix Fig. S8*). The region between vertical dashed lines covers the premelting condition. The calculated longitudinal and shear wave velocities are consistent with our experimental results at ~ 230 GPa and $T/T_m \sim 0.96$. The shear softening emerges near melting, which accompanies by collective motion in the large spatiotemporal machine learning molecular dynamics (See *SI Appendix Movie S1*). (c) Calculated (black curve) and measured (open star) shear modulus of hcp-Fe at high temperatures and 230 GPa. The shear modulus of premelting hcp-Fe at the relevant conditions of the core can overall match with the seismic observation of 149-176 GPa (red region) in the inner core (red bar) (1, 6).

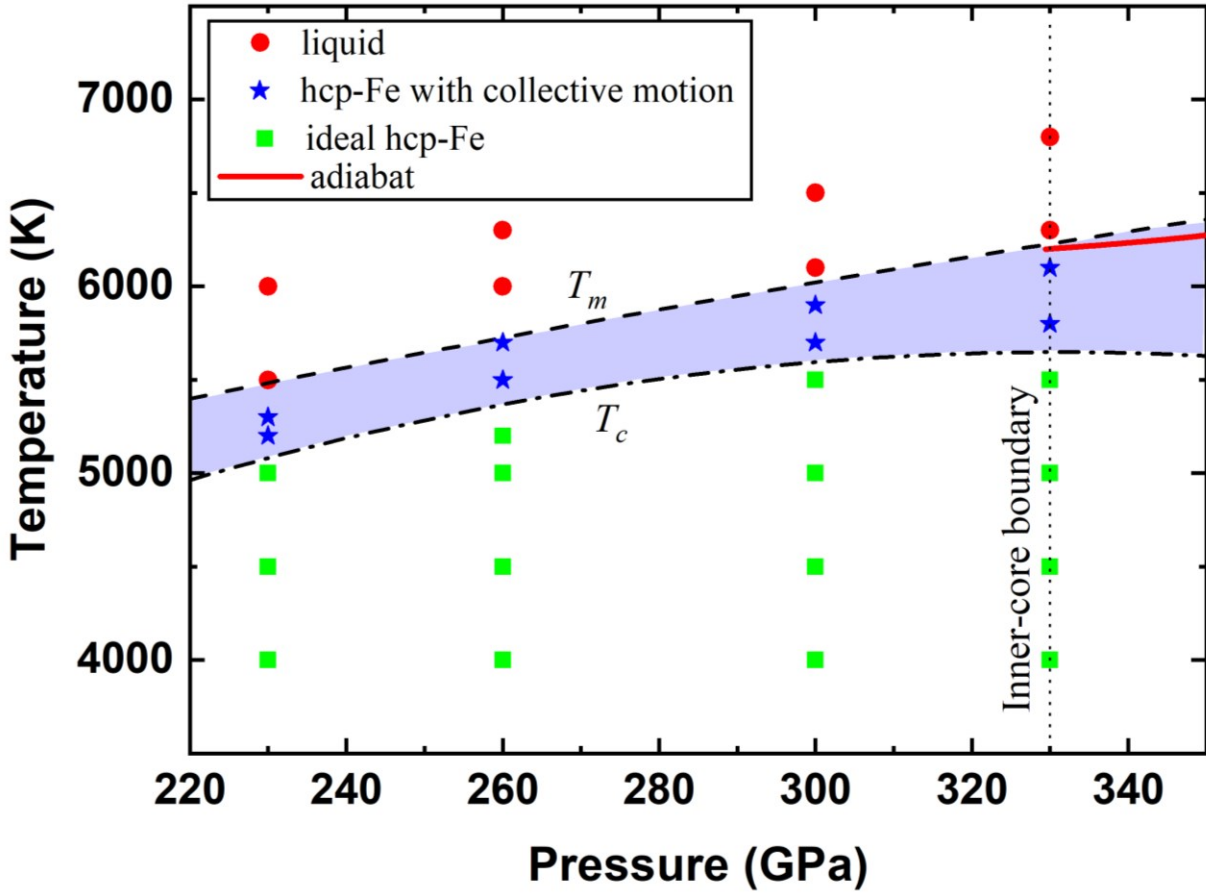


Fig. 5. Predicted dynamic phase boundary of hcp-Fe with collective atomic motion at high pressure-temperature conditions relevant to the Earth's core. The dashed line represents the melting curve (T_m) of iron at high pressures in this study fitted by Simon-Glatzel law with $T_m = T_0 (P/11.8+1)^{0.367}$, where $T_0 = 1811$ K is the melting point of iron at ambient pressure. Our calculated melting points of iron is generally consistent with recent experimental results as shown in *SI Appendix Fig. S8* (17-21), in which the melting point is ~6200 K at the pressure of the inner-core boundary (~330 GPa). The dash-dotted line represents the dynamic phase boundary between ideal hcp-Fe and hcp-Fe with collective motion (T_c) in this study fitted by a second-order polynomial. The light-blue area represents the predicted P-T conditions of hcp-Fe with collective motion. The red curve represents a typical adiabat of iron anchored at the inner core boundary. The adiabat gradient across the inner-core boundary is approximately 0.17 K/km based on isentropic modeling (16).

Materials and Methods

Starting Materials. High-purity polycrystalline bulk iron with a purity greater than 99.98% was used as the starting material. The iron sample were prepared into 0.662-1.480-mm-thick disks with a diameter of ~24 mm, and their surfaces were polished to a mirror finish with a roughness of ~15 nm (17).

Sound velocity measurements and Poisson's ratio of hcp-Fe at high P-T. Reverse-impact experiments (see [SI Appendix Fig. S1](#)) were carried out at both Sichuan University and Institute of Fluid Physics in China. We employed both lithium fluoride (LiF) crystal with <100> orientation and sapphire Al₂O₃ crystal with <0001> orientation as window materials, respectively. The interface particle velocity profile was measured using photon Doppler velocimetry (PDV) with a laser wavelength of 1550 nm and a time resolution of less than 1 nanosecond. The impact velocity was measured using an electromagnetic method with an uncertainty of ~0.5%. In the first series of runs, we used LiF crystals with <100> orientation as the transparent window, where the particle velocity profile was recorded at the sample/window interface. This allowed us to generate P-T conditions up to ~156 GPa and ~3400 K with the highest impact velocity being 7.472(37) km/s. To achieve higher pressures, we employed Al₂O₃ crystals with <0001> orientation as the window material in the second series of runs because it is denser than LiF crystal and can generate much higher P-T on reverse impact. Even after the phase transformation to CaIrO₃, Al₂O₃ crystal remained transparent up to at least 210 GPa, and its refractive index under shock compression was recently determined (67). Iron was shocked to Al₂O₃ crystal up to the impact velocity of 7.695(30) km/s, reaching in the highest P-T of ~230.8 GPa and ~5220 K in this study. The particle velocity wave profiles for these shots are shown in [SI Appendix Fig. S2](#).

To ensure experimental accuracy, we used four PDV probes to simultaneously measure the particle velocity profiles at four different locations of the sample in a shot (see [SI Appendix Fig. S3](#)): one probe was positioned at the center of the impact surface, while the other three were located at the outer ring with a diameter of 5 mm. The wave profiles recorded by multiple probes almost overlapped in one shot, indicating low uncertainty in the sound velocity measurements (< 3%). The Lagrangian longitudinal sound velocity of iron, C_l , can be directly obtained using the measured time interval (Δt) and the thickness of the sample (δ):

$$C_l = \frac{\delta}{(t_1 - t_0) - \delta/U_s} = \frac{\delta}{\Delta t - \delta/U_s} \quad (1)$$

Then, the Eulerian longitudinal sound velocity, V_p , which accounts for the change of sample thickness due to shock compression (68), is:

$$V_p = \frac{\rho_0}{\rho_H} C_l = \frac{\rho_0}{\rho_H} \frac{\delta}{(t_1 - t_0) - \delta/U_s} \quad (2)$$

where ρ_0 and ρ_H are the starting and compressed densities of iron, U_s represents the shock wave velocity in shock-compressed iron. The time interval is represented by $\Delta t = t_2 - t_1$. t_1 and t_2 is the time of impact and the arrival of the release wave at the impact interface, respectively, which can be obtained from the measured interface particle velocity profile (see [SI Appendix Figs. S2-S3](#)). The Lagrangian bulk sound velocity (C_b) at the Hugoniot state can be determined by linearly extrapolating the plastic unloading part of the C_l line to the Hugoniot state (see [SI Appendix Fig. S4](#)). The elastic-plastic transition point at time t_2 can be identified by calculating the derivative of particle velocity with respect to time (see [SI Appendix Fig. S3](#)). The Eulerian bulk sound velocity V_b is obtained by substituting t_1 with t_2 in equation (2) (53). By using V_p and V_b , the shear wave velocity, V_s , can be obtained using the following relation:

$$V_s^2 = \frac{3}{4}(V_p^2 - V_b^2) \quad (3)$$

The reverse-impact experiments (69-71) do not require the fitting of the overtake ratio of sample plate thickness to impactor plate thickness, which differs from symmetric impact experiments previously used (47, 48). Instead, this method directly measures the shock and rarefaction fronts in iron samples, which leads to a significant improvement in the accuracy of sound velocity measurements.

On the other hand, the bulk sound velocity (V_b) of iron under shock compression can be calculated using a thermodynamic equation:

$$V_b^2 = -V^2 \frac{dP_H}{dV} \left[1 - \left(\frac{\gamma}{V} \right) \frac{(V_0 - V)}{2} \right] + V^2 \left(\frac{\gamma}{V} \right) \frac{P_H}{2} \quad (4)$$

$$P_H = \rho_0 C_0^2 (1 - \rho_0/\rho) / [1 - \lambda(1 - \rho_0/\rho)]^2 \quad (5)$$

where $\rho_0(V_0)$ and $\rho(V)$ are the density (specific volumes) of iron at ambient and shock compression, respectively; γ is the Grüneisen parameter of iron along the Hugoniot state (17, 72); C_0 and λ are the Hugoniot parameters, where U_s (km/s) = 3.935 + 1.578 with $\rho_0 = 7.850(2)$ g cm⁻³ for iron (73). The V_p of hcp-Fe is significantly reduced by elevated temperature, especially above $T/T_m > \sim 0.74$ ($P_H \sim 160$ GPa). We used a third-order polynomial to fit the measured V_p with pressure as V_p (km/s) = 6.9 + 0.011 P_H + 1.15E-4 P_H^2 - 4.61E-7 P_H^3 in the investigated pressure range. The fitted curve gives a maximum V_p of ~ 10.02 km/s at ~ 206 GPa.

The Poisson's ratio of iron can be determined from the measured V_p , V_b , and V_s in the present work using the following equation:

$$\nu = \frac{1}{2} \left[1 - \frac{1}{(V_p/V_s)^2 - 1} \right] = 1 - \frac{2}{3(V_b/V_p)^2 + 1} \quad (6)$$

The uncertainty of the measured sound velocities under shock compression were estimated through the uncertainty propagation using:

$$\sigma_{V_p} = \sqrt{\left(\frac{\partial V_p}{\partial \delta} \sigma_\delta \right)^2 + \left(\frac{\partial V_p}{\partial \Delta t} \sigma_{\Delta t} \right)^2 + \left(\frac{\partial V_p}{\partial U_s} \sigma_{U_s} \right)^2 + \left(\frac{\partial V_p}{\partial \rho_H} \sigma_{\rho_H} \right)^2} \quad (7)$$

$$\sigma_{V_s} = \sqrt{\left(\frac{\partial V_s}{\partial V_p} \sigma_{V_p} \right)^2 + \left(\frac{\partial V_s}{\partial V_b} \sigma_{V_b} \right)^2} \quad (8)$$

The uncertainties in the sample thickness (δ) and the travel time of shock wave in the sample (Δt) were measured to be $\sim 0.1\%$ and $\sim 1\%$, respectively. As shown in [SI Appendix Fig. S3](#), the measured Δt has a generally small error of 1-2 ns, with a total time interval of 120-330 ns. Based on the equations (2) and (7), the uncertainties of V_p and V_b are estimated to be $\sim 2\%$. The uncertainty of V_s is estimated to be between 5% and 15% by using the equations (3) and (8).

DFT calculations at finite temperature and AIMDs. In this study, DFT calculations, including structure optimizations and AIMD simulations, are carried out by using the Vienna *ab initio* simulation (VASP) code (74). The projector augmented-wave (PAW) potentials (75) and the Perdew-Burke-Ernzerhof (PBE) Exchange-correlation functional (76) were employed in the calculations. Considering extreme P-T conditions, a 750-eV energy cutoff potential with 16 valence electrons was used, which sufficiently avoided any overlapping of core states. To ensure accuracy, all necessary tests were conducted and the electronic energy was converged within 1 meV per atom. The optimization of hcp-Fe at 0 K was achieved using a

primitive cell with a grid density of $0.025 \times 2\pi \text{ \AA}^{-1}$ for k point sampling in the Brillouin zone at 230 GPa. A series of *ab initio* NPT (constant number of atoms, pressure, and temperature) molecular dynamics (77) with Langevin thermostat (78) were performed with a time step of 1 fs to access the proper c/a ratio of hcp-Fe at 230 GPa and finite temperature up to melting (3000-10000 K). The NPT AIMD simulation cells consisted of a $4 \times 4 \times 3$ supercell for the 4-atom C-centered cell in hcp structure with orthogonal axes and used Γ -point sampling. These data were utilized to train the initial machine learning model during the Concurrent machine learning.

Concurrent machine learning. In this work, we utilized an end-to-end deep learning model called Deep MD (79, 80) to build a potential energy surface (PES) using a dataset generated by DFT calculations. This model has been applied to various systems and has demonstrated credible results, including high-pressure calcium (28) and water (81). In addition, it is important to note that a high-quality dataset is crucial for effectively training the model and achieving a more accurate representation of the phase space. Recently, a novel learning procedure with on-the-fly properties was proposed (82), which has been dubbed concurrent learning as the data is generated in real-time as the training progresses.

We have leveraged our workflow and DFT results to construct a deep learning potential. The construction process involves three parts, as illustrated in [SI Appendix Fig. S5](#). Initially, we employed data comprising energy, atomic force, and stress from AIMD simulations of perfect hcp-Fe and hcp-Fe with vacancies to train three initial models with different initialized parameters. Subsequently, we utilized these models to explore a much broader phase space, thereby expediting the process. We then applied an error indicator to identify configurations that were inadequately predicted by the initial models. Those configurations were subjected to DFT self-consistent calculations, and the results were integrated into the dataset. With the aid of this improved dataset, we developed new models with enhanced predictive capabilities that could better anticipate the phase space. This procedure was iterated several times (in our study, three times) until we had acquired a reliable potential with DFT accuracy, as demonstrated in [SI Appendix Fig. S6](#). Additionally, in step 2, we investigated various hcp-Fe structures, including perfect crystal, vacancies, twin crystals, grain boundaries, and a mixture of solid and liquid as depicted in [SI Appendix Fig. S7](#). This rigorous examination guarantees that our models are practical for a variety of applications.

Machine learning molecular dynamics. Machine Learning Molecular Dynamics (MLMD) were carried out using the LAMMPS code, employing periodic boundary conditions and a time step of 1 fs (83). The simulations comprised a range of particle number from 6912 to 80640 and were conducted for up to ns-scale durations. We tested the fluctuations in temperature and diagonal stress tensors of iron at 230 GPa and various temperatures using machine learning simulations with different supercell cell sizes ranging from 144 atoms (a typical size in *ab initio* simulations) to 31104 atoms (see [SI Appendix Fig. S14](#)).

Melting point calculation by the two-phase method using machine learning. Theoretical determination of the melting point typically involves comparing free energies of solid and liquid phases or employing direct molecular dynamics (MD). One method for determining the melting point of a substance involves directly heating it from the low-temperature phase until it transforms into a liquid. The temperature at which this transition occurs is typically considered to be the melting point. However, this method tends to overestimate the melting temperature due to the superheating effect. Using this one-phase method, the melting point of iron at 230 GPa was calculated to be $\sim 6600\text{K}$, which is much higher than the experimental value. Various factors may contribute to this superheating issue, such as the simulation cells with only a small number of atoms compared to real systems, particularly in DFT calculations. Consequently, in finite-time simulations, this method often lacks the full distribution of phase space, causing the melting of the

system to lag behind the temperature rise. Additionally, heating from a perfect solid to melting is an idealized process, with a transition barrier much higher than in real systems with interfaces.

To address this issue, an improved MD method was proposed, called the two-phase method or coexistence melting method. This method involves simulating a mixture of solid and liquid at different temperatures, allowing solid and liquid to compete with each other. The addition of liquid broadens the phase-space distribution of the system. As melting occurs, their free energy becomes equal, allowing the solid and liquid to coexist. However, this method requires a larger cell to accommodate two states and is not commonly used in AIMDs. With the aid of machine learning potential, it's now feasible to construct a large box and simulate the melting behavior of hcp-Fe in nanosecond scale. Initially, we heated the perfect hcp-Fe with orthogonal axes until it turned into a fully melted liquid. We then integrated it with an initial solid to form a larger supercell, containing up to 80640 atoms. To avoid any unreasonable structures, like the occurrence of atoms that are too close to each other at the solid-liquid interface, we added a vacuum layer with a thickness of 1 Å to the interface (as illustrated in *SI Appendix Fig. S7*), which could be relaxed during the following NPT simulations. Using this coexistence box, a series of NPT machine learning molecular dynamics simulations were conducted, and the length of the simulations was up to 0.5 ns to ensure proper systems convergence. We then calculated the free energy of the solid and liquid iron at constant pressure and different temperatures. The temperature at which the free energies of solid and liquid iron are equal is known as the temperature of solid-liquid coexistence or the melting point. The melting point of hcp-Fe at 230 GPa was determined to be 5350–5400 K at 230 GPa and 6200–6300 K at 330 GPa (asterisks in *SI Appendix Fig. S8*), below which the collective atomic motion occurs at $T/T_m \sim 0.96$. Our calculated melting point of high-pressure hcp-Fe are in close to the previous work using the similar two-phase method(23) and are also consistent with recent high P-T experiments (17–21).

Van Hove self-correlation functions. The van Hove self-correlation function $P(\Delta r, t)$ gives the probability of an atom at distance Δr after a time interval t . It is defined as,

$$P(\Delta r, t) = \frac{1}{N} \left\langle \sum_{i=1}^N \delta(r - |\vec{r}_i(t_0 + t) - \vec{r}_i(t_0)|) \right\rangle_{t_0} \quad (9)$$

where $\vec{r}_i(t_0)$ is the position of i th atom at a time t_0 , N is the number of atoms, and $\langle \dots \rangle_{t_0}$ is the average of time ensemble. In our simulations, we used 0.1 ns trajectories as the lag time t and 0.05 ns trajectories as the time window of ensemble average.

Finite-temperature lattice dynamics. To obtain the 0 K phonon dispersions, lattice dynamics calculations were performed using the phonopy package (84). A $4 \times 4 \times 3$ hcp-Fe supercell, comprising a 2-atom primitive cell and a grid of $0.025 \times 2\pi$ Å⁻¹ for k points was utilized in the simulation. The renormalized phonon spectra including anharmonicity and temperature-dependent phonon properties were determined at finite temperature using the dynaphopy package (85). The same iron supercell ($4 \times 4 \times 3$) was used to perform the MLMDs at varying temperatures with a 1-fs timestep and the 50-ps simulation time. The velocity-autocorrelation functions via MD were analyzed by projecting them onto the harmonic modes, which were calculated in the 0 K phonon calculations. Further theoretical details can be found in Ref. (85). The lattice dynamics calculated using DFT and ML at 0 K are compared in *SI Appendix Fig. S6e*.

Calculation for Poisson's ratio in hcp-Fe near melting. When stretching or compressing a material, it can undergo changes in both axial and transverse directions, which is referred to as the Poisson effect. Poisson's ratio (ν) quantifies this phenomenon and is defined as follows:

$$\epsilon_{axial} = dl_{axial}/l_{axial}$$

$$\varepsilon_{trans} = dl_{trans}/l_{trans}$$

$$\nu = \frac{-\varepsilon_{trans}}{\varepsilon_{axial}} \quad (10)$$

where l , dl , and ε are length, changes in length and strain along transverse and axial (subscript: trans and axial) directions under certain axial stress.

Non-equilibrium machine learning molecular dynamics (NEMLMD) calculations were conducted to directly simulate the Poisson effect. The NEMLMD simulations were performed using the LAMMPS code, employing periodic boundary conditions and a time step of 1 fs (ref. (83)). In these calculations, tensile stresses were incrementally applied to completely relaxed equilibrium states, derived from two-phase equilibrium molecular dynamics, to achieve a strain ε_{axial} of 0.05 (see *SI Appendix Fig. S10*). Throughout the deformation process, the temperature was regulated using a Nose-Hoover thermostat, while the pressure on those two dimensions perpendicular to the deformation direction remained fixed at 230 GPa. These deformations encompassed the Poisson effect, and the change in volume under the deformation is given by:

$$\frac{dV}{V} = \frac{l_{axial} \times l_{trans}^2 \times (1 + \varepsilon_{axial})(1 + \varepsilon_{trans})^2 - l_{axial} \times l_{trans}^2}{l_{axial} \times l_{trans}^2} \quad (11)$$

Combining the definition in equation (10), for a very small value of strain (here 0.05 scale), the first-order approximation yields:

$$\frac{dV}{V} \approx \varepsilon_{axial} + 2\varepsilon_{trans} = \varepsilon_{axial}(1 - 2\nu) = dl_{axial}/l_{axial}(1 - 2\nu) \quad (12)$$

When the Poisson's ratio ν equals 0.5, the volume remains unchanged by applied stress, rendering the system an incompressible state, such as a liquid. By examining the correlation between $\frac{dV}{V}$ and $\frac{dl_{axial}}{l_{axial}}$ at 230 GPa and various temperatures (see *SI Appendix Fig. S10*), we can ascertain the value of ν through a linear fitting. Utilizing this approach at room temperature, the Poisson's ratio of hcp-Fe at 230 GPa is approximately 0.36, aligning with some previous experimental findings (10, 11). At temperature T/T_m of approximately 0.96-0.98, the Poisson's ratio of hcp-Fe at 230 GPa is determined to be approximately 0.44, and the value derived from the calculated elastic constants using the Voigt average scheme (32) at $T/T_m \sim 0.98$ is 0.447.

Movies and Snapshots of deep learning simulations. Movies and snapshots of deep learning simulations are rendered using visual molecular dynamics (VMD) (86) and OVITO (87).

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Code availability. Six codes were used to obtain results: VASP for *ab initio* molecular dynamics (available at <https://www.vasp.at>); LAMMPS for classical molecular dynamics (available at <http://lammps.sandia.gov>); Deep-MD kit and DP-GEN for training machine learning potential (available at <https://github.com/deepmodeling/deepmd-kit> and <https://github.com/deepmodeling/dpgen>); Phonopy and Dynaphopy for lattice dynamics (available at <https://phonopy.github.io/phonopy> and <http://abelcarreras.github.io/DynaPhoPy>).

Data availability. All data in the paper are present in the article and/or Supplementary Materials.

Supplementary Information for

Collective atomic motion in solid iron at the conditions of Earth's inner core

Youjun Zhang, Yong Wang, Yuqian Huang, Junjie Wang, Zhixin Liang, Long Hao, Zhipeng Gao,
Jun Li, Qiang Wu, Hong Zhang, Yun Liu, Jian Sun, Jung-Fu Lin

This PDF file includes:

Text S1 to S5.

Fig. S1 to S14.

Table S1.

Movie S1.

Supplementary Text

Text S1. Solid to liquid phase transition of iron under shock compression. In previous studies, longitudinal sound velocity V_p of iron under shock compression had been reported to display sharp discontinuities at around 200 GPa and 220 GPa (47, 48). The discontinuity observed at 200 GPa was initially interpreted as indicative of an unknown solid-solid phase transition (47); however, subsequent experiments have refuted this observation (48). In this study, the measured V_p and V_s did not show any sharp discontinuity up to the shock pressure of 230 GPa. Instead, our new experiments showed significant sound velocity softening because the shock Hugoniot temperature gradually approaches the melting point of iron with increasing shock pressure above 200 GPa. This suggests that there was no first-order phase structural transition according to the changes in the measured sound velocity in the pressure range of 200-230 GPa, which further confirmed that only single hcp phase structure without obvious embedded melting was present in solid iron before shock-induced melting using *in situ* XRD measurements under well-characterized shock compression (19, 20). The gradual reduction in sound velocity observed with increasing shock pressure is thought to be caused by the collective atomic motion of premelting hcp-Fe through the atomistic simulations. Notably, when iron was shocked to ~ 260 GPa, the measured V_p drops down to V_b , indicating complete melting above this pressure (48). Furthermore, static high P-T experiments also suggest that hcp was the only phase observed in solid iron at conditions relevant to the inner core (44).

Text S2. Dynamical behaviors of iron in hcp solid and liquid state. At 230 GPa and 5000 K (with $T/T_m \sim 0.92$), iron has an ideal hcp structure without any collective motion, as revealed by our ns-scale two-phase simulations (Fig. S11a). Upon further heating to 5500 K using the two-phase method, the system transforms into a liquid state (Fig. S11b). In addition, the van Hove self-correlation functions $P(\Delta r, t)$ in hcp crystal at 230 GPa and 5000 K displays a stable Gaussian distribution over time, indicating that the atoms vibrate around their equilibrium positions (Fig. S11c). However, in liquid iron at 5500 K, $P(\Delta r, t)$ diverges over time due to the disorderly diffusion of atoms (Fig. S11d), and its mean square displacements (MSD) of atoms increases rapidly with the whole simulation time (Fig. S12). Furthermore, the collective motion of premelting hcp-Fe at Earth's inner core conditions (approximately 330 GPa and 6000 K with $T/T_m \sim 0.97$) was computed and illustrated in Fig. S13.

Text S3. Two-phase melting model and melting points of hcp-Fe at the core pressure. Collective atomic motion in premelting hcp-Fe can result in significant softening of the shear modulus and an ultrahigh Poisson's ratio. However, the temperature in the shear softening regime calculated using the traditional EMD method is ~ 1000 K higher than the determined melting point of hcp-Fe in experiments (30). This discrepancy arises from the fact that homogeneous melting in the one-phase model is often accompanied by superheating. The one-phase method does not take into account some subsistent impurities in solid iron such as defects, grain boundaries, and interfaces, which could contribute to initial nucleation on melting.

To accurately reproduce the measured melting points of typical d-block transition metals such as iron, tantalum, and vanadium at high P-T, a two-phase melting model is usually more appropriate (88). For instance, using machine learning simulations with the two-phase model, the melting point of hcp-Fe is estimated to be approximately 5400 K at 230 GPa and 6200-6300 K at 330 GPa, respectively. These values are consistent with experimental determinations of 5300-5500 K and 6000-6300 K obtained by both static (18) and shock (17, 19) experiments (Fig. S8). Consequently, we adopted the two-phase method to investigate the melting behavior and Poisson's ratio of hcp-Fe at the P-T conditions relevant to the inner core.

Text S4. Elastic modulus of hcp-Fe at high pressure-temperature. The elastic modulus of a single crystal hcp-Fe was calculated at different temperatures and a constant pressure of 230 GPa using *ab initio* molecular dynamics (AIMD) method, which was also used in Ref. (30). To create unit cells at different temperatures, the average lattice parameters from NPT (constant number of atoms, pressure, and temperature) AIMDs were employed., NVT (constant number of atoms, volume, and temperature) AIMDs were then carried out on these unit cells and their distortional duplicates of hcp symmetry (± 0.02 strain) to compute stresses and corresponding distortions. Through a non-linear 2nd fitting based on these outcomes, the elastic modulus of the system at finite temperatures can be determined. The NVT AIMD simulation cells were constructed of a $4 \times 2 \times 2$ supercell of the 4-atom C-centered cell in hcp structure with orthogonal axes and 4 irreducible k-points.

The calculated elastic modulus of hcp-Fe exhibited a significant reduction before reaching to the melting point ($T/T_m > \sim 0.95$) (Fig. 4), consistent with the results predicted in Ref. (30). To confirm the effect of collective motion on the shear modulus, we further employed the machine learning potential to calculate elastic constants of hcp-Fe. Our results indicate that collective atomic motion in premelting hcp-Fe provides a microscopic explanation of the temperature-induced strong shear softening. As a result, both two-phase and one-phase simulations simultaneously revealed the temperature-induced shear softening in premelting hcp-Fe with collective motion in this study.

Text S5. Effect of the supercell size on the calculation. To assess the influence of supercell size on our simulations, we conducted tests to examine the fluctuations in temperature and diagonal stress tensors of iron at 230 GPa and various temperatures using machine learning simulations. We employed cell sizes ranging from 144 atoms (a typical size in *ab initio* simulations) to 31104 atoms (Fig. S14). The results revealed substantial thermodynamic fluctuations in simulations with smaller cell sizes, such as 144 atoms, leading to relatively large errors in the calculations. However, as the number of atoms increased to approximately 10000, the systems exhibited convergence to the target temperature and pressure, with significantly reduced fluctuations. For instance, the standard deviations in temperature and pressure for simulations with 144 atoms and 18000 atoms were estimated to be 380 K and 31 K, 9.53 GPa and 0.91

GPa, respectively. As a result, the pressure and temperature fluctuations in our simulations were too small to induce iron encompassing between the solid and liquid phases in each run. As our machine learning simulations were based on cells with at least 10000 atoms, we are confident that our data and conclusions are both valid and physically meaningful.

Text S6. Effect of nickel and possible light elements on the collective motion of hcp-Fe in the inner core. Our computations show that the collective atomic motion of hcp-Fe occurs at 330 GPa and a temperature range of 5800-6200 K, which is slightly lower by 0-6% than its melting temperature. However, our study here does not account for the effect of the presence of nickel (Ni) at ~5wt.% and light elements at 2-4 wt.% in the inner core (2). Literature results have demonstrated that adding small quantities of light elements, such as Si and O, to iron can decrease its melting temperature by several hundreds of K at core pressures (60). It is important to note that the collective atomic motion happens in hcp-Fe at a temperature ratio of $T/T_m > \sim 0.95$. Given this, the incorporation of light elements (such as Si, S, O, and C) into iron should result in the occurrence of collective motion at a relatively lower temperature due to the reduced melting point. For example, collective motion in hcp-Fe alloys could take place at ~5200-5500 K. As a result, it is reasonable to expect that iron alloyed with ~5 wt.%Ni and 2-4 wt.% light elements could also display shear wave softening and collective motion nearing melting at relevant P-T conditions of the inner core.

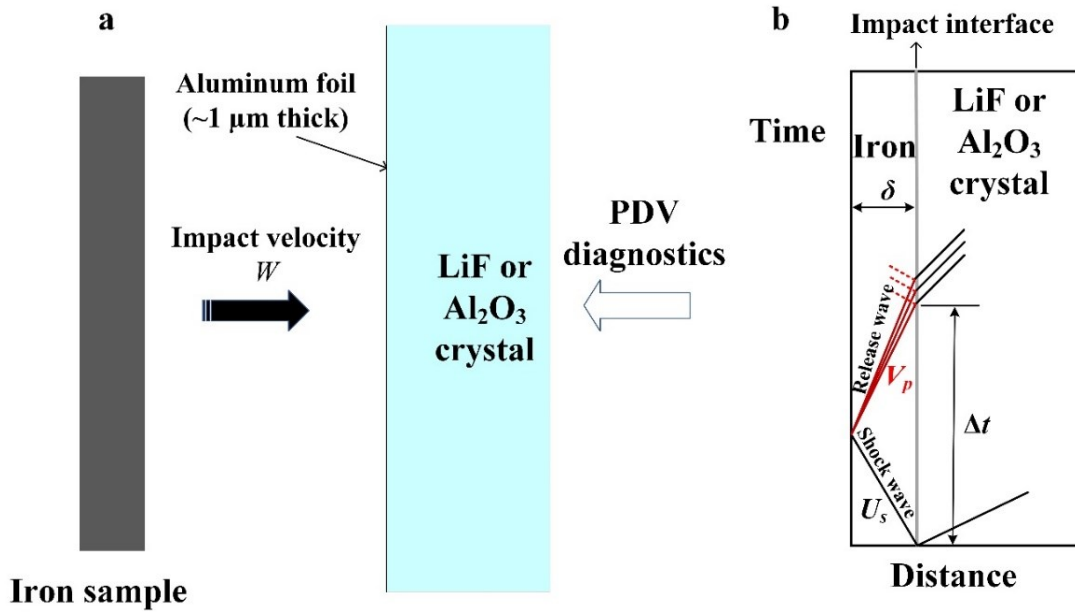


Fig. S1. Schematic diagram of the experimental setup for sound velocity measurements of iron at high pressure-temperature using the reverse-impact method in a two-stage light-gas gun. (a) The reverse-impact method was used to measure both the longitudinal and bulk sound velocities of iron. Photon Doppler velocimetry was used to obtain wave profiles at the iron/window interface. We used $\langle 100 \rangle$ LiF or $\langle 0001 \rangle$ Al_2O_3 crystals as transparent windows. W (km/s) represents the impact velocity. A thin aluminum film ($\sim 1 \mu\text{m}$) was vapor-deposited on the impact surface of the LiF and Al_2O_3 windows to efficiently reflect the laser light. **(b)** The relationship between time and Lagrangian distance shows the propagation of shock and release waves in the sample and window after impact.

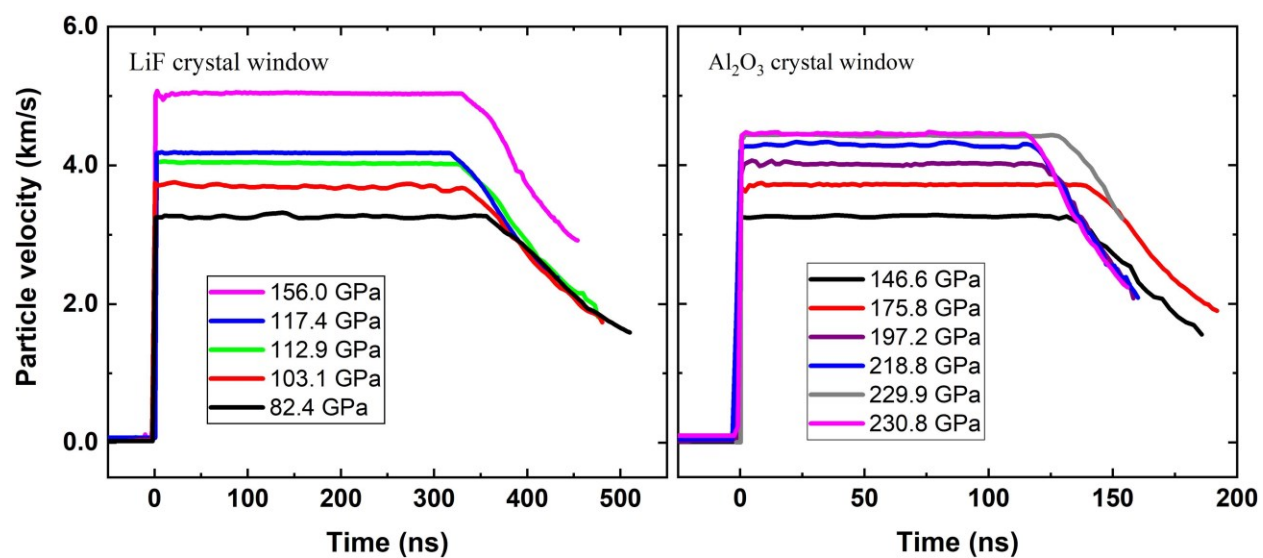


Fig. S2. Particle velocity profiles at the iron/window interface during shock compression. The wave profiles were measured using PDV with a time resolution of ~ 1 ns. The used windows were LiF (left) and Al_2O_3 crystals (right), respectively. The details of the reverse-impact conditions are listed in [Table S1](#).

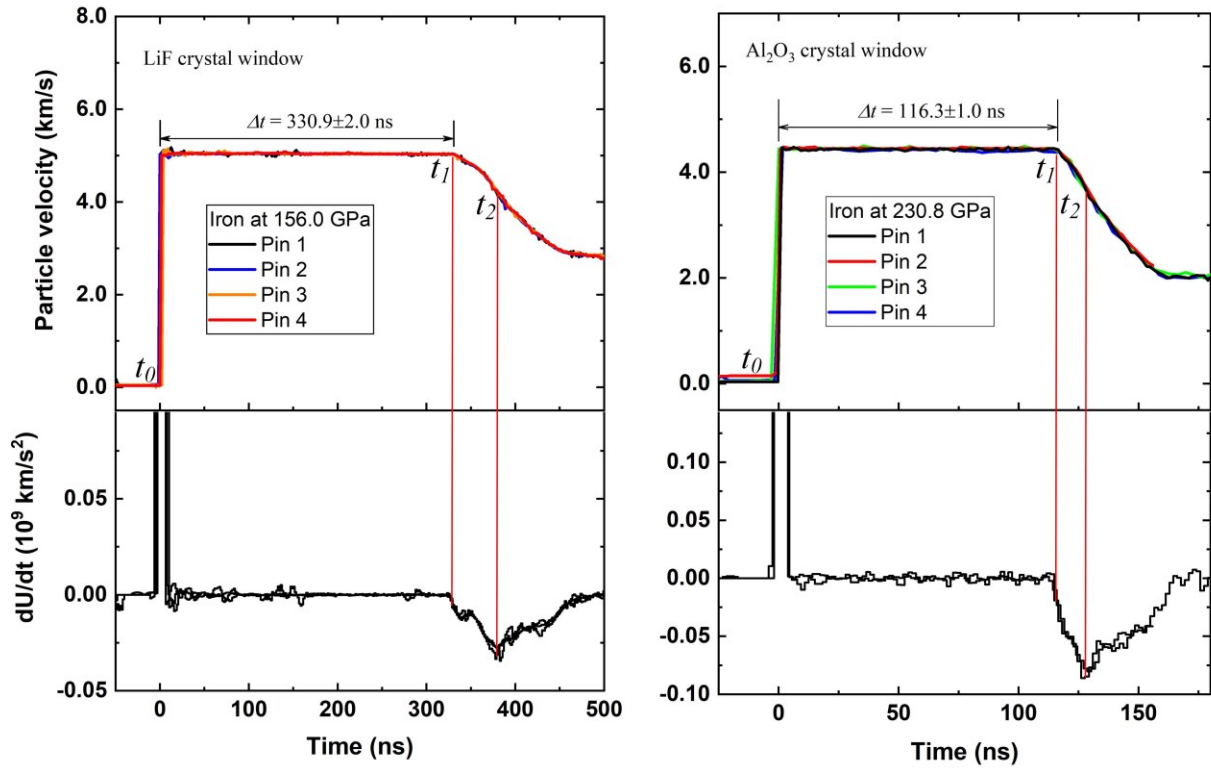


Fig. S3. Typical particle velocity profiles at the interface between iron and windows by multiple pins. The used windows were LiF crystal in shot No. 6 (left) and Al₂O₃ crystal in shot No. 11 (right). The derivative of particle velocity with respect to time, dU/dt , is used to help obtain the time t_0 , t_1 , and t_2 that correspond to the arrival times of shock wave front, rarefaction wave at the interface, and the elastic-plastic transition point.

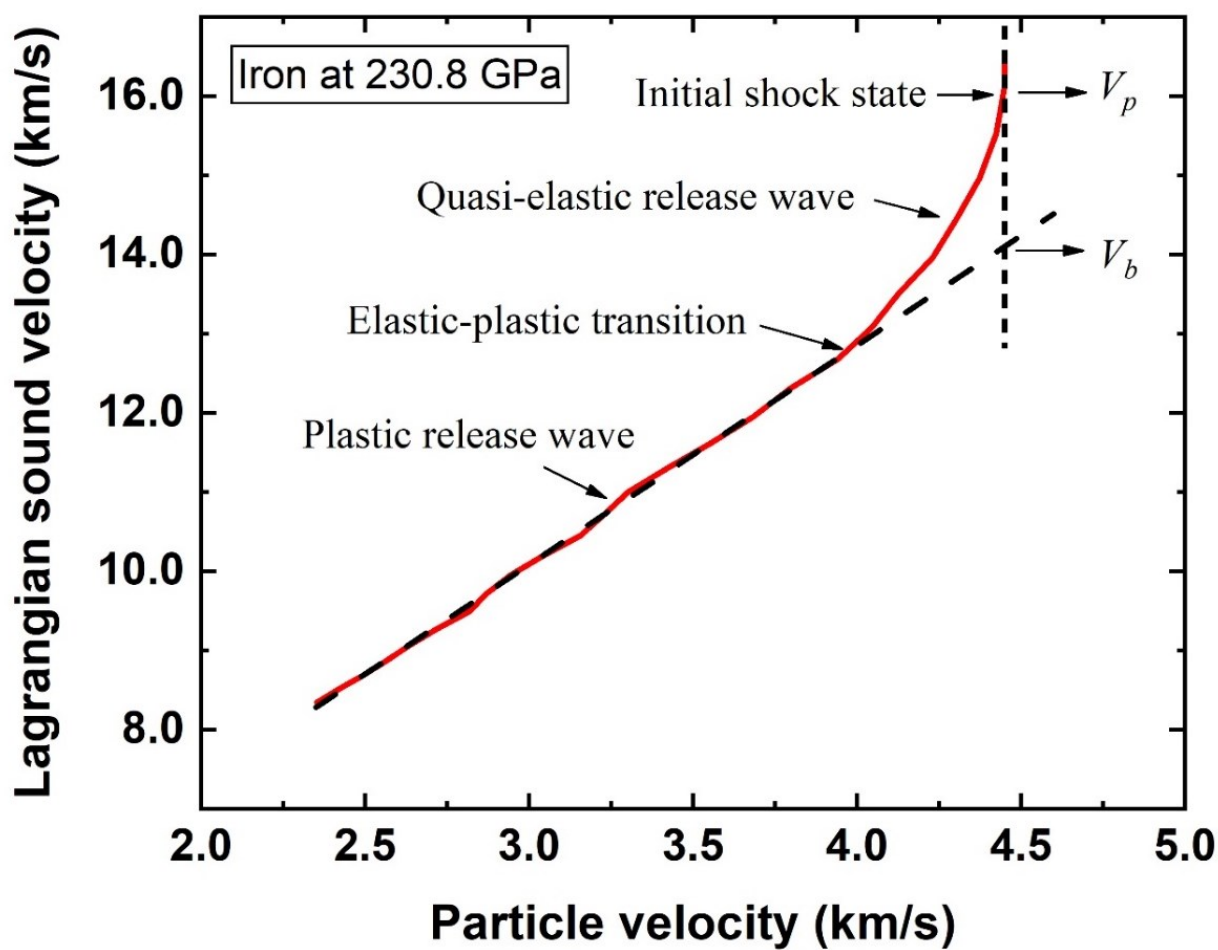


Fig. S4. Typical Lagrangian sound velocity in iron versus the particle velocity. The V_b of iron can be extrapolated via a linear fitting upon the plastic release wave. The shock pressure is ~ 230.8 GPa in Shot No. 11.

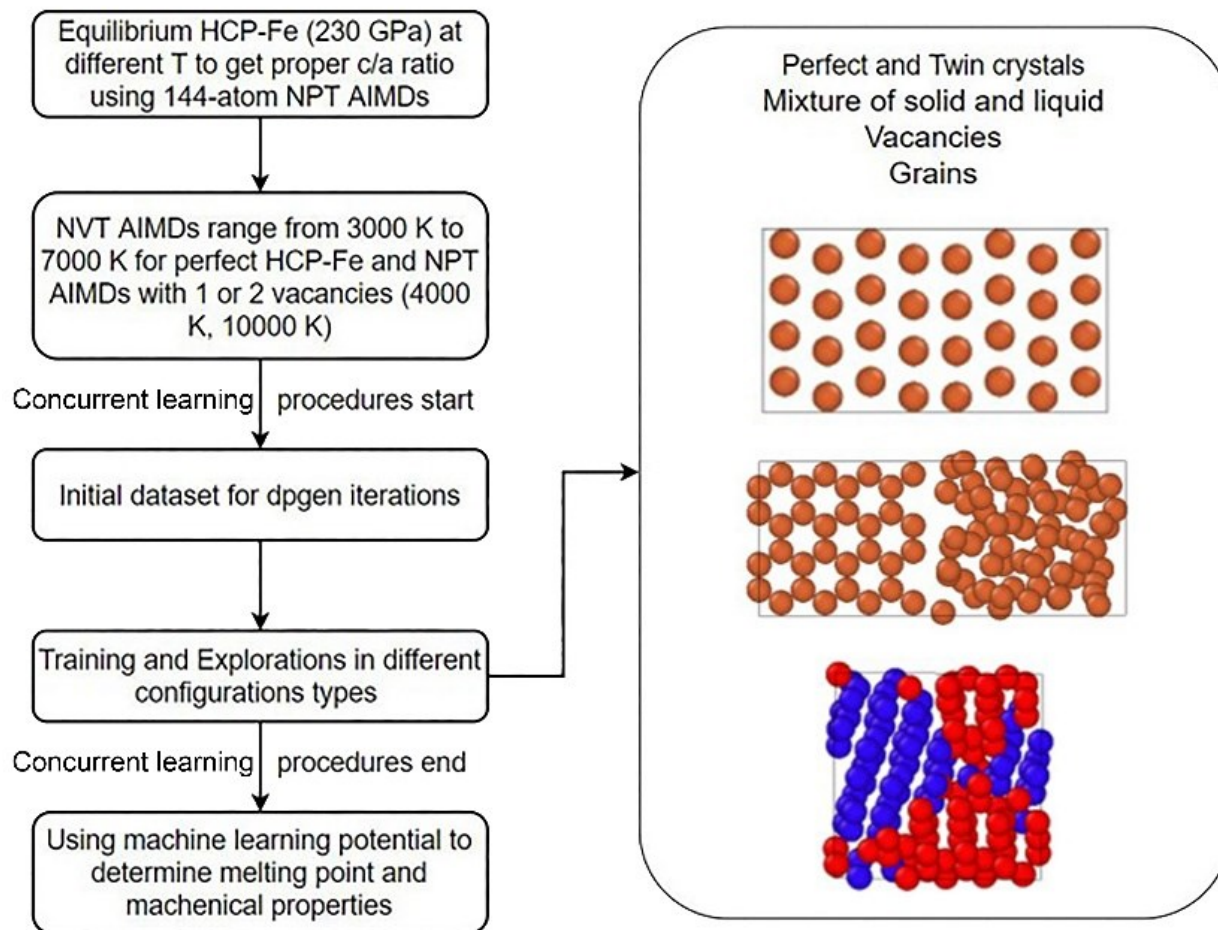


Fig. S5. Schematic illustration of concurrent machine learning procedure in our work. The learning procedure was separated into several parts including initialization, exploration, model deviation, etc. Different configuration types in phase space were considered and learned. More details can be found in Ref. (82).

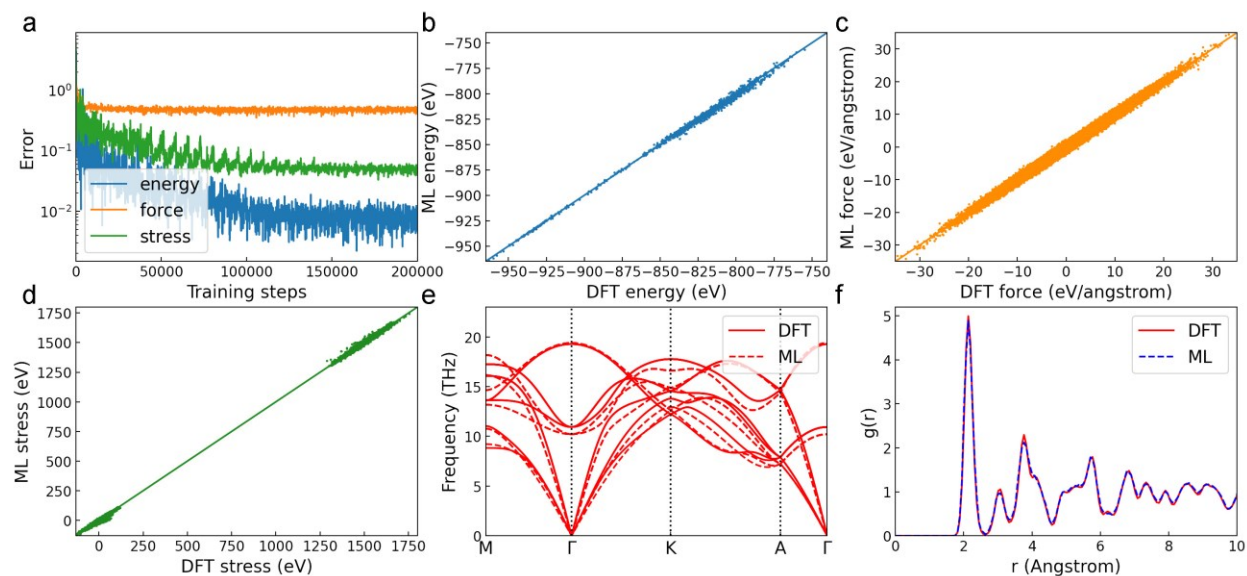


Fig. S6. Training errors of the machine learning (ML) and comparisons between DFT results and our machine learning model. (a) Convergence of training errors (root mean square error) for energy, force, and stress during the machine learning (ML) training. (b-d) Comparisons of energy, force, and stress (shear and normal) between our ML model and DFT calculations. The comparisons show remarkable consistency among these predicted quantities. The solid diagonal lines serve as visual aids to facilitate interpretation. (e-f) Comparisons of lattice dynamics (phonon spectrum) and radial distribution functions $g(r)$ of hcp-Fe at 330 GPa and 0 K between DFT results and our ML model.

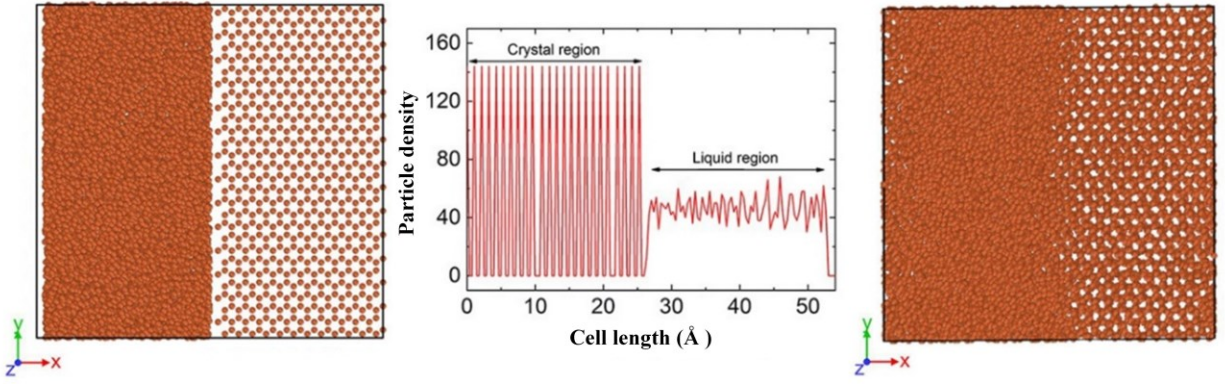


Fig. S7. Initial configuration of iron in two-phase method (left) and its particle density along the x-axis (middle), as well as the final configuration after 50 ps (right). The configuration contains 20160 iron atoms. The NPT simulations were conducted at the melting point of 5350 K at the pressure of 230 GPa and at the melting point of 6200 K at the pressure of 330 GPa, respectively. The initial interface between solid and liquid was divided by a 1 Å vacuum layer to prevent unreasonable structures. The coexistence of solid and liquid in the right figure locates the melting.

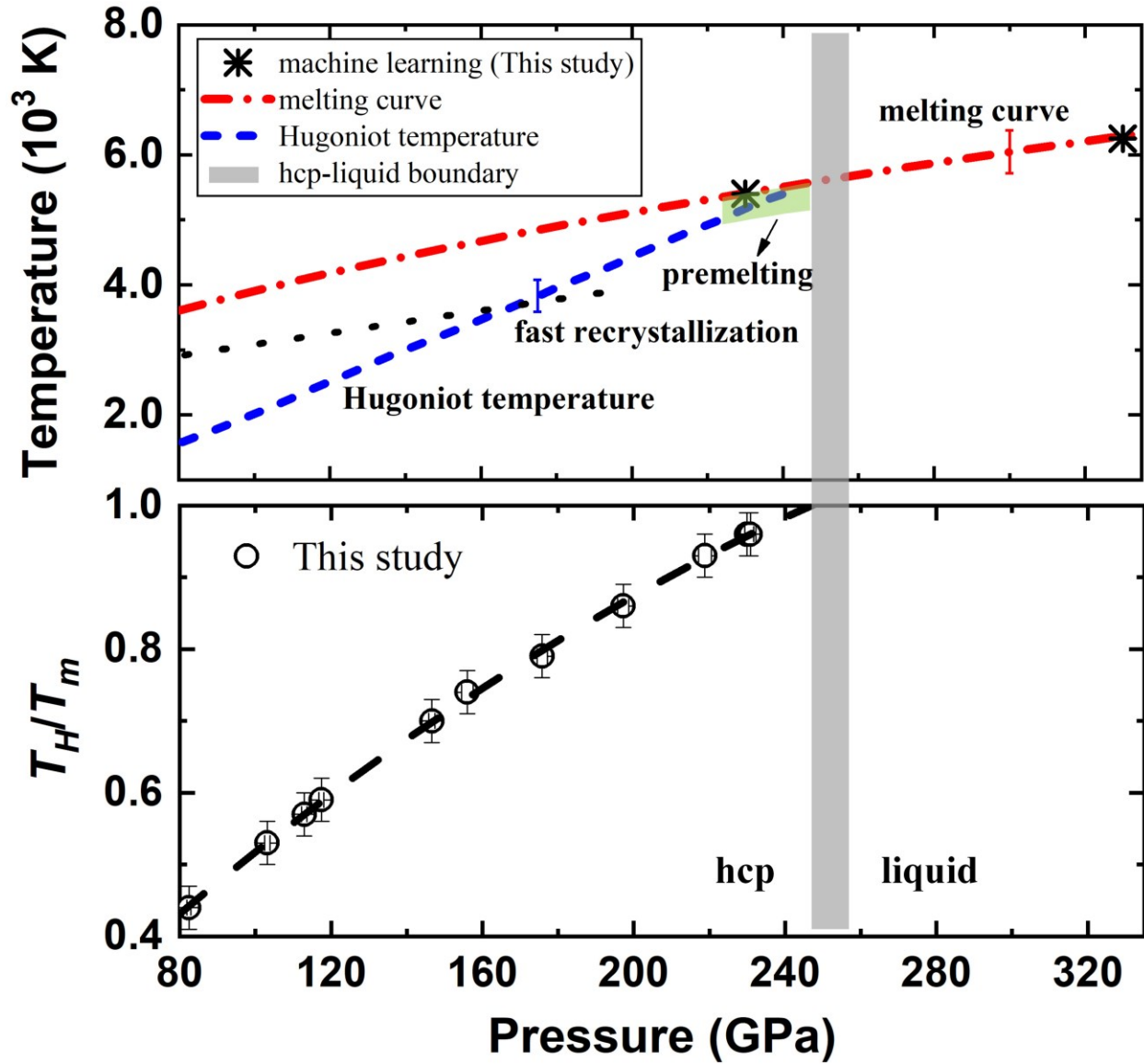


Fig. S8. Melting temperature of iron at high pressure and the ratio of shock Hugoniot temperature to melting point. **Top**, Pressure vs. temperature relation in shock-compressed iron and the melting curve of iron at high pressure. The * represents the present calculated melting points of hcp-Fe at 230 GPa and 330 GPa by the two-phase method using machine learning MD. The red dash-dot line represents the melting curve of iron at high pressure from both experiments (17-19) and theoretical computations (22, 23). The blue short-dash line represents the measured Hugoniot temperature in solid iron from literature (17, 19, 23, 52). The light-green region represents the ratio of shock temperature to the melting point of iron above 0.96, where collective atomic motion occurs in iron under shock compression. The temperature threshold for collective motion in solid iron is significantly higher the temperature boundary at which fast recrystallization occurs in polycrystalline iron (black dotted line). **Bottom**, The ratio of Hugoniot temperature of iron to its melting point (T_H/T_m) as a function of shock pressure. Open circles represent the T_H/T_m conditions in the present sound velocity measurements under shock compression. The hcp-liquid boundary of shocked-iron was determined by *in situ* XRD (19) and sound velocity measurements (48) under shock compression, which happens at 247-260 GPa.

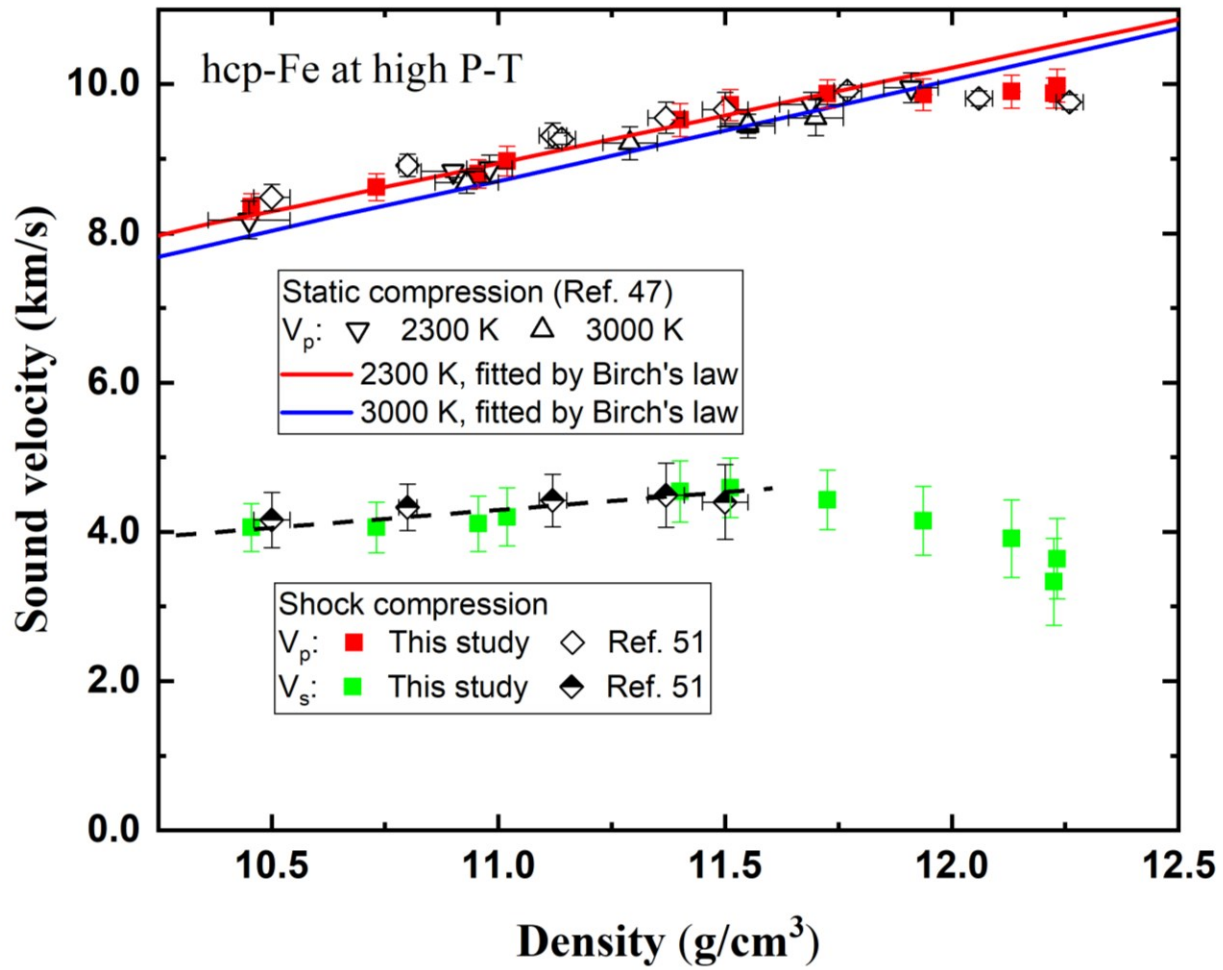


Fig. S9. Sound velocity of hcp-Fe as a function of density under high pressure and temperature conditions by static and shock compression. The V_p of hcp-Fe up to 163 GPa and 3000 K was measured using inelastic x-ray scattering from a laser-heated sample in a diamond anvil cell (49). The V_p and V_s was determined up to 231 GPa and 5200 K under shock compression in this study. At relatively low temperatures (*e.g.*, up to 3000 K), the density-velocity relation of hcp-Fe could be reasonably described by a quasi-linear Birch's law. However, as it approached the premelting region, the sound wave velocity is significantly softened so that the Birch's law is broken down. The quasi-linear dashed-line serves as a visual guide, representing a quasi-linear relationship between the V_s and density of iron at the relatively low P-T conditions.

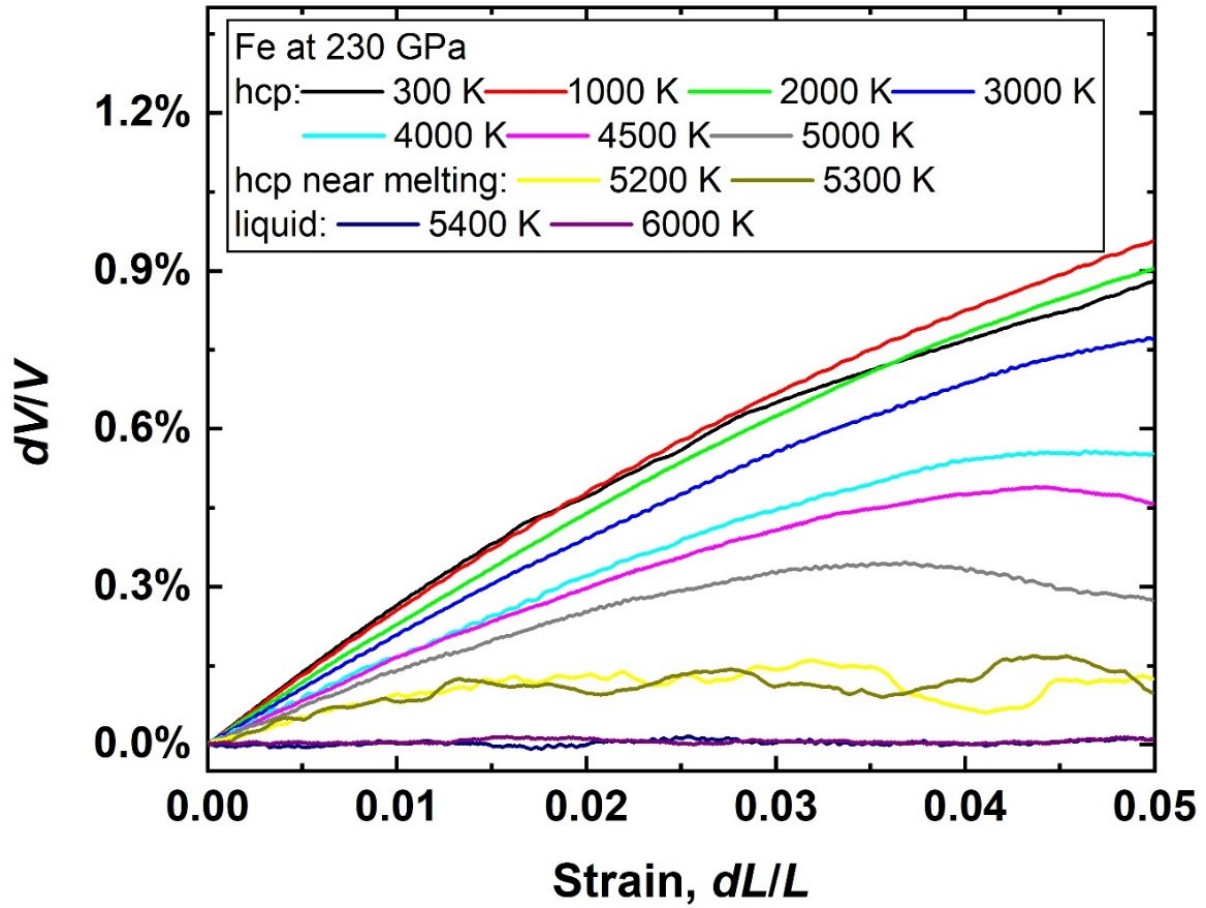


Fig. S10. Volume change vs. gradual deformation under axial strain in hcp-Fe at 230 GPa and different temperatures. The Poisson's effect could be simulated in this way. The volume change indicates different responses of hcp-Fe. For solid (< 5100 K), the system shows a strong elasticity with a linear increase under a small deformation. For hcp-Fe at near melting point (> 5200 K), collective atomic motion induces the softening and the slope of the volume response curve decreases rapidly. For liquid (> 5400 K), the volume would not change with the deformation due to its incompressibility.

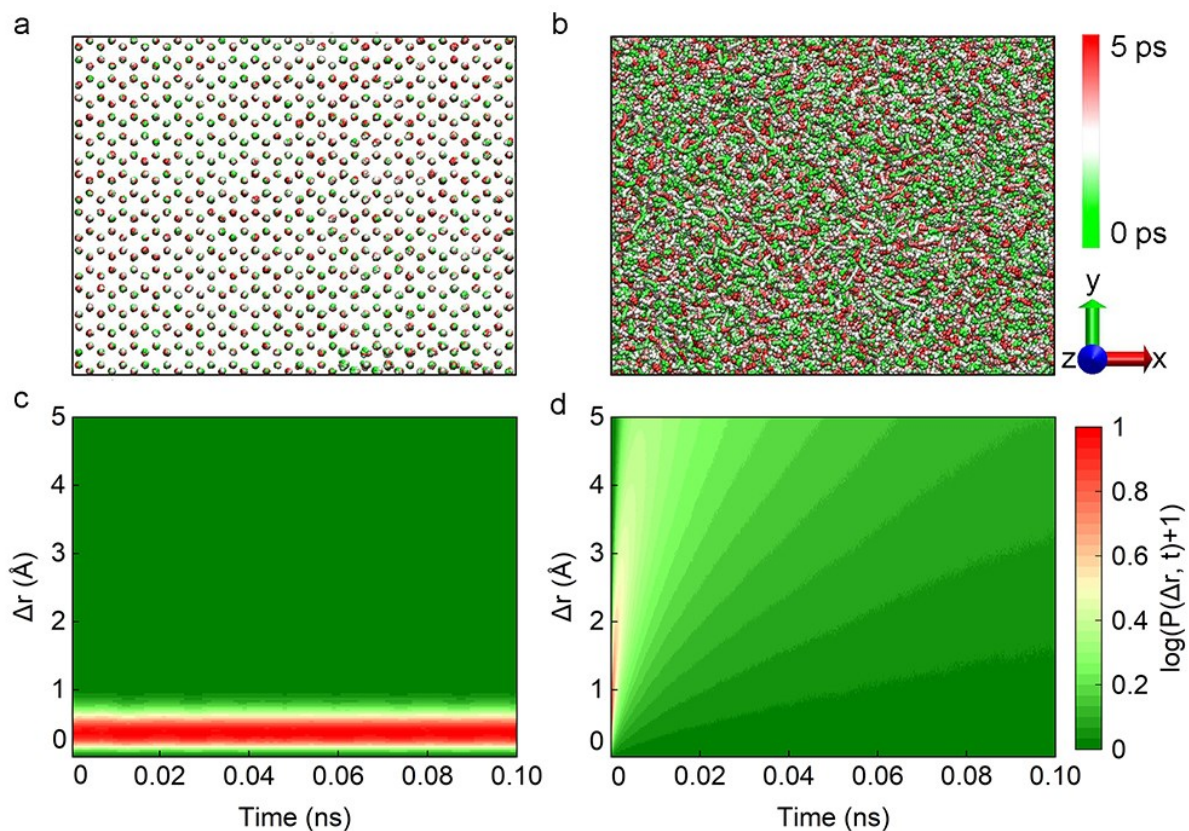


Fig. S11. Relaxed structures' snapshots of two-phase method simulations at 230 GPa and different temperatures. (a) 5000 K in ideal hcp-Fe and (b) 5500 K in full liquid iron. Self van Hove correlation functions corresponding to (c) 5000 K in ideal hcp structure and (d) 5500 K in full liquid iron. Stable Gaussian distribution in (c) indicates a typical solid behavior where atoms vibrate at equilibrium positions. Self van Hove correlation function diverges with time in (d) shows atoms diffuse disorderly in liquid iron.

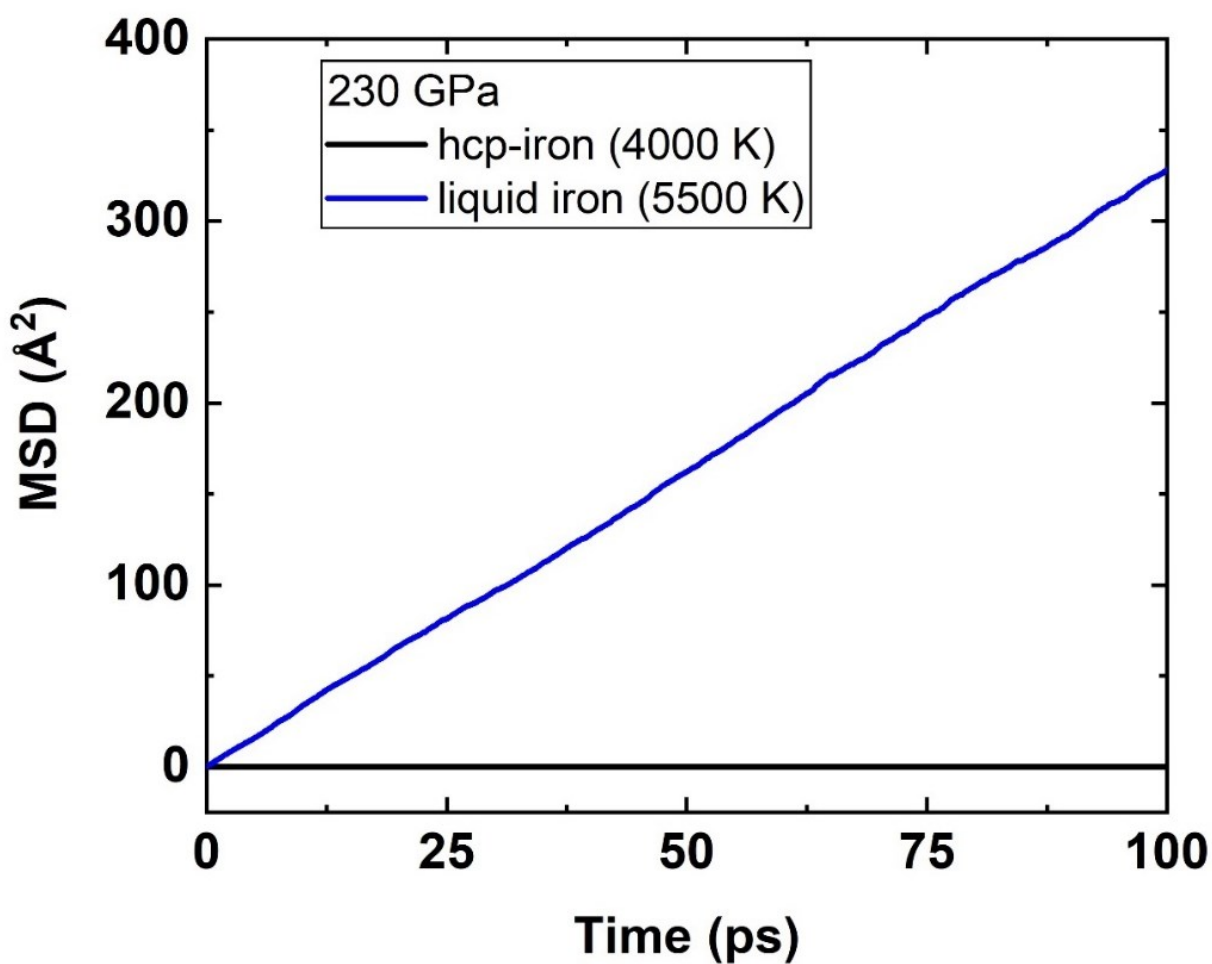


Fig. S12. Calculated mean squared displacement (MSD) of atoms in hcp and liquid iron at 230 GPa. In liquid iron, the MSD ascends steeply throughout the whole simulation time.

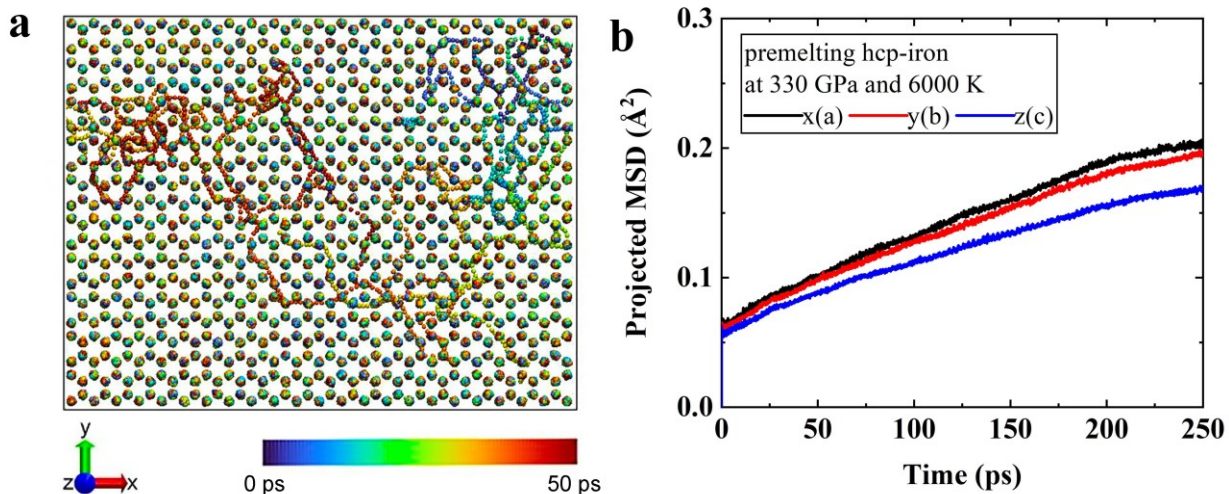


Fig. S13. Collective atomic motion of premelting hcp-Fe at the inner core conditions (~330 GPa and 6000 K). (a) Dynamical snapshots of premelting hcp-Fe at ~330 GPa and 6000 K ($T/T_m \sim 0.97$) relevant to the inner core P-T conditions. The collective motion emerges in hcp-Fe at the relevant inner-core boundary conditions. Iron atoms are colored with time progression during 50 ps. (b) Projected MSD of hcp-Fe atoms at ~330 GPa and near the melting point of 6000 K indicating the diffusivity. The diffusion is anisotropic and mostly appears in a-b (x-y) planes.

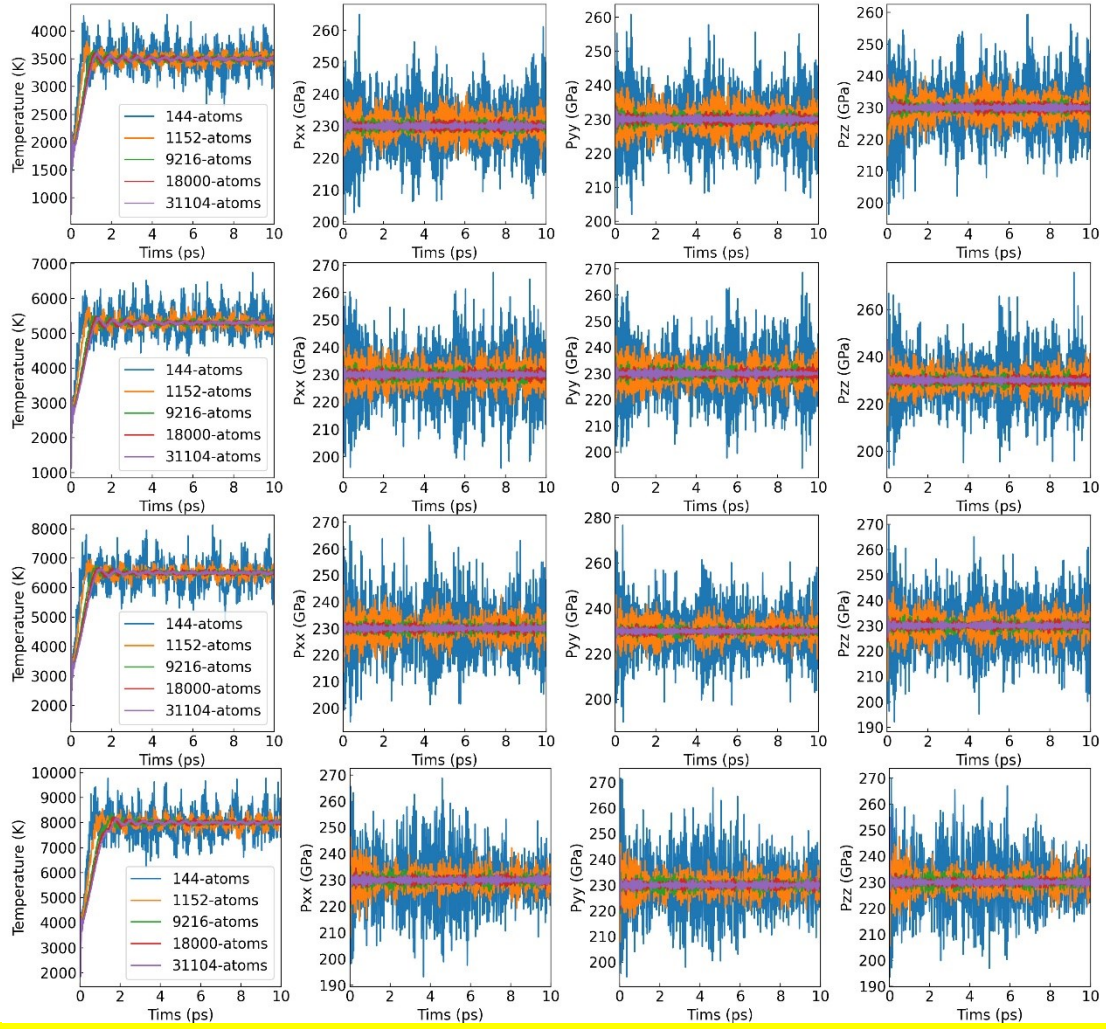


Fig. S14. Fluctuations in temperature and diagonal stress tensors of iron at 230 GPa and various temperatures using machine learning simulations with different supercell sizes. The utilized supercell configurations contained 144, 1152, 9216, 18000, and 31104 atoms, respectively. The diagonal stress tensors along the xx, yy, and zz directions were denoted as Pxx, Pyy, and Pzz, respectively. In the leftmost row, a specific temperature is represented, while the subsequent three rows indicate the three respective diagonal stress tensors (pressure) at the corresponding temperatures.

Movie S1. Diffusions of atomic collective motion of hcp-Fe at high pressure-temperature (230 GPa and $\sim 0.98 T/T_m$). The collective motion emerges near melting in both two-phase simulations without superheating and one-phase simulations with superheating. Diffusing iron atoms are marked by red.

Table S1. Conditions of the reverse-impact experiments and the sound velocities of hcp-Fe at high pressure-temperature under shock compression.

Shot No.	W (km/s)	Thickness (δ , mm)	Window materials	U_s (km/s)	u_p (km/s)	P_H (GPa)	ρ (g cm ⁻³)	T_H (K)	T_H/T_M at P_H	V_p (km/s)	V_b (km/s)	V_s (km/s)	V_b/V_p	v
1	4.866(14)	1.466(2)	LiF	6.485(14)	1.616(9)	82.4(5)	10.455(8)	1600(100)	0.44(3)	8.36(17)	6.92(14)	4.06(32)	0.83(2)	0.347(21)
2	5.680(17)	1.477(2)	LiF	6.925(15)	1.895(11)	103.1(5)	10.731(9)	2090(120)	0.53(3)	8.62(18)	7.23(14)	4.06(34)	0.84(2)	0.358(20)
3	6.036(18)	1.480(2)	LiF	7.119(16)	2.018(12)	112.9(5)	10.956(10)	2330(150)	0.57(3)	8.80(19)	7.41(15)	4.11(37)	0.84(2)	0.358(21)
4	6.195(18)	1.456(2)	LiF	7.205(16)	2.072(11)	117.4(5)	11.019(10)	2450(150)	0.59(3)	8.97(20)	7.62(15)	4.10(39)	0.85(2)	0.368(20)
5	5.541(16)	0.661(1)	Al ₂ O ₃	7.738(14)	2.410(10)	146.6(6)	11.401(10)	3150(200)	0.70(3)	9.52(22)	7.95(16)	4.54(41)	0.84(2)	0.358(21)
6	7.472(22)	1.682(2)	LiF	7.901(20)	2.513(15)	156.0(8)	11.511(14)	3400(200)	0.74(3)	9.72(21)	8.15(16)	4.59(40)	0.84(2)	0.358(20)
7	6.332(19)	0.734(1)	Al ₂ O ₃	8.226(16)	2.719(12)	175.8(8)	11.726(12)	3840(200)	0.79(3)	9.87(19)	8.44(17)	4.43(40)	0.86(2)	0.379(18)
8	6.881(21)	0.662(1)	Al ₂ O ₃	8.560(18)	2.931(13)	197.2(9)	11.937(13)	4380(250)	0.86(3)	9.86(21)	8.62(17)	4.15(46)	0.87(2)	0.389(19)
9	7.411(22)	0.662(1)	Al ₂ O ₃	8.882(19)	3.135(14)	218.8(10)	12.132(14)	4920(250)	0.93(3)	9.90(22)	8.81(18)	3.91(52)	0.89(3)	0.408(19)
10*	7.673(24)	0.736(1)	Al ₂ O ₃	9.040(20)	3.235(15)	229.9(10)	12.225(15)	5180(250)	0.96(3)	9.88(20)	9.10(18)	3.33(58)	0.92(3)	0.435(20)
11	7.695(23)	0.664(1)	Al ₂ O ₃	9.052(19)	3.243(14)	230.8(10)	12.232(15)	5200(250)	0.96(3)	9.98(20)	9.05(18)	3.64(54)	0.91(3)	0.426(19)

Note: Eleven shots were conducted, where the pressure in iron ranges from ~82 to 231 GPa. W is the impact velocity, P_H and T_H is the shock Hugoniot pressure and temperature of iron, respectively, ρ is the density at Hugoniot pressure, U_s and u_p are the shock wave velocity and particle velocity of iron, respectively, calculated from W and the known Hugoniot relations of iron and windows, V_p , V_b , and V_s is the longitudinal, bulk, and shear velocity of iron under shock compression, respectively. The Hugoniot relation ($U_s = C_0 + \lambda u_p$) used in this study are U_s (km/s) = 3.935 + 1.578 u_p with the initial density of $\rho_0 = 7.850(2)$ g cm⁻³ for iron (73), U_s (km/s) = 5.215 + 1.351 u_p with the initial density of $\rho_0 = 2.640(2)$ g cm⁻³ for LiF (89), U_s (km/s) = 8.740(44) + 0.957(10) u_p with the initial density of 3.988(2) g cm⁻³ for sapphire Al₂O₃. The parameters in parentheses are standard deviations. The used LiF and Al₂O₃ crystals are <100> and <0001> orientation, respectively. The sapphire is denser than that of LiF crystal. Therefore, iron impacting onto sapphire can generate much higher pressures than that by LiF. *In shot No. 10, only the longitudinal sound velocity was measured, the bulk and shear sound velocity were estimated from the thermodynamic calculations.

