1 **Electrical resistivity of substitutionally disordered hcp Fe–Si and**
2 **Fe–Ni alloys: Chemically-induced resistivity saturation in the Earth's**
3 **core**

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16 **ABSTRACT**

17 The thermal conductivity of the Earth's core can be estimated from its electrical
18 resistivity via the Wiedemann-Franz law. However, previously reported resistivity
19 values are rather scattered, mainly due to the lack of knowledge with regard to
20 resistivity saturation (violations of the Bloch-Grüneisen law and the Matthiessen's rule).
21 Here we conducted high-pressure experiments and first-principles calculations in order
22 to clarify the relationship between the resistivity saturation and the impurity resistivity
23 of substitutional silicon in hexagonal-close-packed (hcp) iron. We measured the
24 electrical resistivity of Fe–Si alloys (iron with 1, 2, 4, 6.5, and 9 wt.% silicon) using

25 four-terminal method in a diamond-anvil cell up to 90 GPa at 300 K. We also computed
26 the electronic band structure of substitutionally disordered hcp Fe–Si and Fe–Ni alloy
27 systems by means of Korringa-Kohn-Rostoker method with coherent potential
28 approximation (KKR-CPA). The electrical resistivity was then calculated from the
29 Kubo-Greenwood formula. These experimental and theoretical results show excellent
30 agreement with each other, and the first principles results show the saturation behavior
31 at high silicon concentration. We further calculated the resistivity of Fe–Ni–Si ternary
32 alloys and found the violation of the Matthiessen’s rule as a consequence of the
33 resistivity saturation. Such resistivity saturation has important implications for core
34 dynamics. The saturation effect places the upper limit of the resistivity, resulting in that
35 the total resistivity value has almost no temperature dependence. As a consequence, the
36 core thermal conductivity has a lower bound and exhibits a linear temperature
37 dependence. We predict the electrical resistivity at the top of the Earth’s core to be 1.12
38 $\times 10^{-6} \Omega\text{m}$, which corresponds to the thermal conductivity of 87.1 W/m/K. Such high
39 thermal conductivity suggests high isentropic heat flow, leading to young inner core age
40 (< 0.85 Gyr old) and high initial core temperature. It also strongly suppresses thermal
41 convection in the core, which results in no convective motion in inner core and possibly
42 thermally stratified layer in the outer core.

43 *Keywords:* core; electrical resistivity; resistivity saturation; diamond-anvil cell;
44 KKR-CPA; thermal conductivity

45 **1. Introduction**

46 Thermal conductivity strongly constrains the thermal evolution of the Earth's core
47 because it is likely that long-lived geomagnetic field has been sustained by thermal
48 convection at least before the nucleation of the inner core (e.g., Gomi et al., 2013;
49 Labrosse, 2015). Since heat is mainly transported by mobile electrons in metals, the
50 thermal conductivity of the core can be reasonably estimated from the electrical
51 resistivity via the Wiedemann-Franz law, $k = LT/\rho$, where k is thermal conductivity, $L =$
52 $2.45 \times 10^{-8} \text{ W}\Omega/\text{K}^2$ is the Lorenz number, T is temperature, and ρ is electrical resistivity.
53 However, a wide range of outer core resistivity, $0.6\text{--}4.7 \times 10^{-6} \Omega\text{m}$, has been reported
54 (see a review in Gomi and Hirose, 2015). Stacey and Anderson (2001) estimated the
55 resistivity of $\text{Fe}_{0.65}\text{Ni}_{0.1}\text{Si}_{0.25}$ at the core-mantle boundary (CMB) condition to be $2.12 \times$
56 $10^{-6} \Omega\text{m}$ based on the previous shock compression experiments reported by Matassov
57 (1977). The recent experimental work by Seagle et al. (2013) showed $1.5\text{--}2.1 \times 10^{-6} \Omega\text{m}$
58 for Fe-16.43at.%Si at the CMB condition, while Gomi et al. (2013) estimated $1.02 \times$
59 $10^{-6} \Omega\text{m}$ for Fe-22.5at.%Si. Later, Ohta et al. (2016) confirmed the Gomi et al. (2013)'s
60 value from high pressure and temperature resistivity measurements. de Koker et al.
61 (2012) reported $1.0 \times 10^{-6} \Omega\text{m}$ for Fe-25at.%Si at the CMB by first-principles
62 molecular dynamics (FPMD) calculations. Pozzo et al. (2013) also conducted FPMD
63 calculations and reported $0.9 \times 10^{-6} \Omega\text{m}$ for $\text{Fe}_{0.82}\text{Si}_{0.10}\text{O}_{0.08}$ and $\text{Fe}_{0.79}\text{Si}_{0.08}\text{O}_{0.13}$ at the
64 CMB. Konôpková et al. (2016) conducted direct measurement of thermal conductivity
65 at high pressure and temperature, and the corresponding resistivity is $3.7 \times 10^{-6} \Omega\text{m}$,
66 which is the highest resistivity among previous studies, and consistent with the Stacey
67 and Loper (2007)'s estimate of $3.62 \times 10^{-6} \Omega\text{m}$. Indeed, a factor of 2 or more
68 uncertainties in the estimated resistivity has a strong impact on the energy balance of

69 geodynamo and the thermal history of the Earth's core (e.g., Labrosse, 2015; Gomi et al,
70 2013; Stacey and Loper, 2007).

71 Traditionally the core electrical resistivity has been estimated on the basis of the
72 Bloch-Grüneisen law and the Matthiessen's rule (Stacey and Anderson, 2001; Stacey
73 and Loper, 2007; Seagle et al., 2013). The former predicts linear temperature
74 dependence of the resistivity above Debye temperature, and the latter shows that the
75 total resistivity is the sum of each scattering terms. However, these simple rules are
76 expected to be violated at high resistivity because of the resistivity saturation effect,
77 which is widely known in metallurgy (Bohnenkamp et al., 2002; Gunnarsson et al.,
78 2003; Mooij, 1973; Wiesmann et al., 1977). Gomi et al. (2013) pointed out its
79 significance at the Earth's core conditions. The resistivity saturation has been observed
80 in metals with high resistivity induced by thermal and/or chemical disorder (Mooij,
81 1973). Recently, thermally-induced saturation (Ohta et al., 2016; Pozzo and Alfè, 2016),
82 as well as pressure-induced saturation (Kiarasi and Secco, 2015), was confirmed at high
83 pressure.

84 In this study, we focus on the chemically-induced resistivity saturation in hcp iron at
85 high pressure. High-pressure electrical resistivity measurements of hcp Fe-Si alloys
86 were conducted in a diamond-anvil cell (DAC). We also performed first-principles
87 calculations on hcp Fe-Si, Fe-Ni, and Fe-Ni-Si alloys. Based on these experimental
88 and theoretical results, the relationship between the impurity resistivity and the
89 resistivity saturation is demonstrated. We also discuss the implications of the high
90 thermal conductivity of the Earth's core, resulting from the resistivity saturation effect.

91 **2. Methods**

92 *2.1. Diamond-anvil cell experiments*

93 We conducted electrical resistivity measurements at high pressure in a DAC with
94 flat 300- μm or beveled 200- μm culet diamonds up to 90 GPa at room temperature. A
95 rhenium gasket was pre-indented to a thickness of $\sim 30\text{ }\mu\text{m}$. A hole with diameter of
96 $\sim 80\%$ of a culet size was drilled at the center of an indent. Al_2O_3 powder filled the hole
97 and covered the surface of the indent in order to electrically insulate the rhenium gasket
98 from a sample and electrical leads. They were then re-compressed to about 20 GPa to
99 make Al_2O_3 inner gasket and drilled a hole with about 40- μm diameter as a sample
100 chamber. Pressure was determined from the Raman spectrum of a diamond anvil at
101 room temperature (Akahama and Kawamura, 2004). The sample resistance was
102 obtained by the four-terminal method under a constant DC current of 10 mA with a
103 digital multi-meter (ADCMT 6581). Six separate runs were conducted for five Fe–Si
104 alloys with different Si concentrations. The samples were foils of Fe alloyed with 1
105 wt.% (1.97 at.%) Si, 2 wt.% (3.90 at.%) Si, 4 wt.% (7.65 at.%) Si, 6.5 wt.% (12.14
106 at.%) Si, and 9 wt.% (16.43 at.%) Si (*Rare Metallic*, 99.99% purity) with initial
107 thickness of $\sim 10\text{ }\mu\text{m}$. We confirmed that no significant chemical heterogeneity within
108 sample foils with SEM/EDS. Four-terminal-shaped samples were cut by Nd:YAG laser
109 or focused-ion beam (FIB) from a single foil. It was loaded into the hole in the gasket
110 together with Al_2O_3 pressure medium. The sample and Pt electrodes were connected
111 mechanically by applying pressure (Fig. 1).

112 Following Gomi et al. (2013), we collected room-temperature resistance data with
113 decreasing pressure because the shape of the sample changed very little during
114 decompression. Assuming an isotropic deformation, sample resistivity at high pressure
115 was obtained from the ratio of resistance obtained at high pressure (R) and 1 bar (R_0),

116 taking the effect of volume change into account:

$$117 \quad \rho = \rho_0 \left(\frac{V}{V_0} \right)^{\frac{1}{3}} \frac{R}{R_0}, \quad (1)$$

118 where V is volume and R is resistance, and subscript zero indicates ambient condition.
119 The volume of the sample is calculated by using the equation of state of pure iron
120 (Dewaele et al., 2006). The resistivity of Fe–Si alloys at ambient condition is from
121 Schwerer et al. (1969). The uncertainty in measured resistivity is derived from the
122 possibility that the sample deformed anisotropically during decompression. The
123 maximum and minimum resistivities are therefore obtained as: $\rho_{\max} = \rho_0 (R/R_0)$, $\rho_{\min} = \rho_0$
124 $(V/V_0) (R/R_0)$ (Gomi et al., 2013).

125 *2.2. First-principles KKR-CPA calculations*

126 We also performed first-principles calculations by using the AkaiKKR
127 (machikaneyama) package, which employed the Korringa-Kohn-Rostoker (KKR)
128 method with the atomic sphere approximation (ASA) within a framework of the local
129 density approximation (LDA) for exchange-correlation potential (Moruzzi et al., 1978).
130 The relativistic effects are taken into account within the scalar relativistic approximation.
131 The wave functions are calculated up to $l = 3$, where l is the angular momentum
132 quantum number. The coherent potential approximation (CPA) is adopted to treat
133 substitutional disorder effect on an electronic band structure (e.g., Akai, 1989). The
134 electrical resistivity is calculated from the Kubo-Greenwood formula with the vertex
135 correction (Butler, 1985; Oshita et al., 2009). In the conductivity calculation, we used
136 58632 k -points in the irreducible Brillouin zone (Supplementary Fig. S1).

137 The hcp structure is a stable form of iron under the Earth's core

pressure-temperature conditions (e.g., Tateno et al., 2010, 2015). Therefore, we calculated the electrical resistivity of all iron alloys with the hcp structure, although the stability field of the substantially-disordered hcp phase is limited near the iron end member for both iron–silicon and iron–nickel alloys. We carried out non spin polarized calculations on the Fe–Si (1.97, 3.90, 7.65, 12.14, 16.43, 20, 30, 40, 50, 60, 70, 80, 90 at.% Si) and Fe–Ni (4.77, 9.56, 14.38, 20, 30, 40, 50, 60, 70, 80, 90 at.% Ni) binary systems at lattice volumes of 22.43, 19.10, 17.41, and 16.27 Å³. These values are equivalent to experimentally-determined room-temperature volumes of pure hcp iron at 0, 40, 80 and 120 GPa, respectively (Dewaele et al., 2006). The axial ratio was set to the ideal value ($c/a = 1.633$) in all calculations.

3. Results

3.1. Experimental results

Fig. 2a shows the measured electrical resistivity of Fe–Si alloys with different Si contents (1 to 9 wt.% Si) up to 90 GPa at 300 K. A sharp resistivity enhancement was observed at the transition between body-centered cubic (bcc) and hcp phases, consistent with previous studies (Gomi and Hirose, 2015; Gomi et al., 2013; Seagle et al., 2013). The resistivities of Fe–Si alloys are very close to the values of Gomi et al. (2013) (2 wt.% Si) and Seagle et al. (2013) (9 wt.% Si) at around 20 GPa, but show slightly lower values at higher pressures.

The resistivity increases linearly with increasing silicon impurity concentration. Such a linear effect implies that a shunt-resistor model cannot describe the resistivity of an alloy with high concentration of impurity elements. Therefore, we considered the resistivity of alloys at room temperature is the sum of phonon-contributed resistivity and

161 impurity resistivity as predicted by the Matthiessen's rule:

$$162 \quad \rho_{\text{ideal}}(V, 300 \text{ K}) = \rho_{\text{ph}}(V, 300 \text{ K}) + \rho_i \times \chi_i \quad (2)$$

163 where $\rho_{\text{ph}}(V, T)$ is phonon-contributed resistivity of iron, which was reported by Gomi
164 et al. (2013) as follows:

$$165 \quad \rho_{\text{ph}}(V, 300 \text{ K}) = 5.26 \times \left(1.24 - \frac{V}{V_0}\right)^{-3.21} \times 10^{-9} \quad (\Omega\text{m}) \quad (3)$$

166 in which $\rho_i(V)$ and χ_i are impurity resistivity and impurity concentration of element i ,
167 respectively. For the impurity resistivity of silicon, the following equation well
168 represents measured values, as shown in Fig. 2a:

$$169 \quad \rho_{\text{Si}}(V) = F_1 \times \left(F_2 \times \frac{V}{V_0}\right)^{F_3} = 4.34 \times \left(2.56 - \frac{V}{V_0}\right)^{-7.79} \times 10^{-6} \quad (\Omega\text{m/at.}\%). \quad (4)$$

170 In addition, the impurity resistivity of nickel has been examined experimentally
171 under the same conditions by Gomi and Hirose (2015). They obtained the following
172 parameters (Fig. 2b):

$$173 \quad \rho_{\text{Ni}}(V) = 2.40 \times \left(11.6 - \frac{V}{V_0}\right)^{-0.918} \times 10^{-7} \quad (\Omega\text{m/at.}\%). \quad (5)$$

174 These phase relationships are consistent with those reported by Huang et al. (1988).

175 *3.2. First-principles results*

176 We calculated the electronic density of states (DOS), band structure, cross section of
177 the Fermi surface, and electrical resistivity of compressed hcp Fe–Si and Fe–Ni alloys.

178 Figs. 3 and 4 show the DOS and band structure of nonmagnetic hcp Fe–Si (1.97 to
179 30 at.% Si) and Fe–Ni alloys (4.77 to 50 at.% Ni) at about 40 GPa. We calculated the
180 Bloch spectrum function along the path, which connects high symmetry points in the

181 Brillion zone of the hexagonal lattice ($\Gamma \rightarrow M \rightarrow K \rightarrow \Gamma \rightarrow A \rightarrow L \rightarrow H \rightarrow A$).
182 At low impurity limit, these plots should be equivalent to the band structure of pure
183 ordered hcp iron. In such a perfectly ordered system, wave vector should be a good
184 quantum number with energy eigenvalue, conduction electrons have infinite life time,
185 and electron bands should become thin lines. On the other hand, once substitutional
186 disorder is introduced, wave vector is no longer a good quantum number, conduction
187 electrons have finite life time due to impurity scattering, and electron energy bands
188 become broaden with energy uncertainty predicted by uncertainty relationship between
189 energy and time; $\Delta E \Delta t \geq \hbar/2$, where ΔE is the uncertainty in energy, Δt is electron life
190 time, and $\hbar$ is the reduced Planck's constant (the Dirac's constant). These broadening
191 features are also found in the DOS plot for Si-rich Fe–Si alloys, whereas alloys with low
192 Si concentrations show sharp peaks (Fig. 3).

193 Since electrons in the vicinity of the Fermi level are only effective to electron
194 transport properties (Banhart et al., 1989; Glasbrenner et al., 2014), we further analyzed
195 the Bloch spectrum function at the Fermi level. Figs. 5 and 6 show the cross sections of
196 the Bloch spectrum function at the Fermi energy of hcp Fe–Si and Fe–Ni alloys at about
197 40 GPa. At low impurity limit, this cross section should be equivalent to the cross
198 section of the Fermi surface of pure Fe. However, with increasing impurity
199 concentration, the cross section becomes broaden due to the effect of impurity disorder.
200 At low, but finite, impurity concentrations (e.g. Fig 5 a-c), one can find that the cross
201 section still has distinguishable features of the Fermi surface of host metal. This
202 situation is referred as well-defined Fermi surface (Butler and Stocks, 1983).

203 The electrical resistivities of Fe–Si and Fe–Ni alloys were calculated by using the
204 Kubo–Greenwood formula (Butler, 1985; Ohshita et al. 2009) at several volumes.

205 Supplementary Fig. S2 shows the electrical resistivity of Fe–Si alloys calculated both
 206 perpendicular and parallel to the c -axis of the hcp structure as a function of silicon
 207 content at about 40 GPa. The electrical resistivity increases with increasing impurity
 208 concentration up to about 50 at.% Si for both directions, and then decreases to another
 209 end-member. The electrical resistivity shows anisotropy that the resistivity
 210 perpendicular to the c -axis is $\sim 30\%$ larger than the value parallel to the c -axis at around
 211 10 at.% Si (Supplementary Fig. S2). For simplification, we consider that the resistivity
 212 of bulk polycrystalline alloys is given by $(2\rho_{\perp} + \rho_{\parallel})/3$, where $\rho_{\perp}$ and $\rho_{\parallel}$ are the resistivity
 213 calculated perpendicular and parallel to the c -axis, respectively (see supplementary
 214 materials). Fig. 7 illustrates the resistivity of such polycrystalline alloys at about 40 GPa.
 215 Within the compositional range experimentally examined, the first-principles and
 216 experimental results show a good agreement. Because the first-principles calculations
 217 did not include the finite temperature effect (e.g., phonon scattering), experimental
 218 resistivities are slightly higher than the first-principles values. Additionally, static lattice
 219 imperfection and zero point vibration have positive contribution to the resistivity,
 220 although our calculation neglected these effects. The resistivity of Fe–Si alloys
 221 increases almost linearly with increasing silicon concentration, whereas the
 222 first-principles result predicts breakdown of such linearity at ~ 20 at.% Si. This nonlinear
 223 behavior is associated with resistivity saturation, which is discussed in section 4.1. The
 224 results of calculations performed at about 0, 40, 80, and 120 GPa are compared with the
 225 present and previous measurements (Matassov, 1977; Ho et al. 1983; Varga et al. 2002
 226 and references therein) in section 4.2. We also calculated the resistivity of $\text{Fe}_{90-x}\text{Ni}_{10}\text{Si}_x$
 227 ternary alloys at the lattice volume of $16.27 \text{ \AA}^3$ (Fig. 9). At low resistivity, i.e. low
 228 silicon concentration, the difference in the resistivity between these binary and ternary

229 alloys is large, whereas it becomes small at high resistivity. This behavior is indicative
230 to the breakdown of the Matthiessen's rule.

231 **4. Discussion**

232 *4.1. Resistivity saturation and the Fermi surface*

233 In the previous section, we found two characteristic behaviors of resistivity; the
234 deviation from linear impurity effect for Fe–Si binary alloys at around 20 at.% Si (Figs.
235 7 and 8), and the breakdown of the Matthiessen's rule (Fig. 9). These two features are
236 well understood within the resistivity saturation theory, closely related to the collapse of
237 the Fermi surface (Fig. 5) (Butler, 1985; Glasbrenner et al., 2014). It also predicts
238 smaller temperature dependence than the prediction from the Bloch-Grüneisen law.

239 It is widely accepted that dominant sources of the resistivity of metals are 1)
240 phonon-contributed resistivity $\rho_{\text{ph}}(T)$ that is caused by thermal disorder (atomic
241 displacement), 2) impurity resistivity ρ_i that is caused by static chemical disorder, and 3)
242 spin disorder resistivity $\rho_{\text{mag}}(T)$. In addition, Pourovskii et al. (2014) suggested that the
243 resistivity caused by electron–electron scattering $\rho_{\text{ee}}(T)$ is negligible for hcp iron at the
244 core condition. At low resistivity, the phonon-contributed resistivity exhibits linear
245 temperature dependence above the Debye temperature (the Bloch-Grüneisen law), and
246 the total resistivity can be expressed as the sum of each scattering terms (the
247 Matthiessen's rule).

248 However, both the Bloch-Grüneisen law and the Matthiessen's rule are no longer
249 valid at high resistivity. The resistivity dose not readily increase beyond a certain
250 “saturation” value of $\sim 1.5 \times 10^{-6} \Omega\text{m}$, when the resistivity increases with increasing the
251 degree of disorder due to the scattering mechanisms described above. The resistivity

252 saturation behavior was first reported on disordered transition metal alloys with
253 concentrated impurity element (Mooij, 1973). Later, the saturation was found in almost
254 all kinds of metals with high resistivity including iron alloys at 1 bar (Bohnenkamp et
255 al., 2002) and high pressure (Kiarasi and Secco, 2015). Furthermore, resistivity increase
256 upon melting was observed to be small (Deng et al., 2013; Secco and Schloessin, 1989),
257 which is considered to be a consequence of the saturation (Mott, 1972).

258 A physical mechanism of the resistivity saturation is largely investigated by
259 theoretical studies (see Gunnarsson et al., 2003 for a recent review). The basic idea is
260 that the mean free path of conduction electrons hardly becomes shorter than interatomic
261 distance, which is so-called “Ioffe-Regel condition” (Gurvitch, 1981). Wiesmann et al.
262 (1977) formulated the temperature dependence of A-15 super conductor as a
263 shunt-resistor model. This model is also applied to pure metals, ordered metallic
264 compounds, and disordered alloys. Subsequently Gurvitch (1981) theoretically derived
265 the shunt-resistor model assuming a distribution of electron life times with a minimum
266 value. Butler (1985) developed a first-principles calculation method to examine the
267 resistivity of disordered alloys combined with coherent potential approximation (CPA)
268 and the Kubo-Greenwood equation. This method reproduces the resistivity of
269 substantially disordered alloys well (e.g., Banhart et al., 1989; Oshita et al., 2009).
270 Butler (1985) also discussed that the resistivity saturation is the result of progressive
271 smearing toward “completely smeared out” of an energy band by disorder. In principal,
272 this method can treat more than one scattering mechanisms on equal footing
273 simultaneously (Glasbrenner et al., 2014; Ebert et al., 2015). Recently, Glasbrenner et al.
274 (2014) demonstrated the resistivity saturation of bcc Fe and hcp Gd with both thermal
275 and spin disorder. In this study, we computed the electronic band structure and the

276 electrical resistivity of hcp Fe-Si and Fe-Ni alloys based on the KKR-CPA and
277 Kubo-Greenwood equation. Following the arguments presented by Glasbrenner et al.
278 (2014), we analyze the DOS and the cross sections of the Fermi surface in order to
279 understand the origin of the resistivity saturation.

280 As mentioned in Section 3.2, electron life time is relevant to energy via uncertainty
281 relationship. Since the inverse of the mean free path can be obtained from the width of
282 the peak in the cross section of the Fermi surface, the Bloch spectral function gives us
283 information about electron transport (e.g., Butler and Stocks, 1984; Banhart et al., 1989;
284 Glasbrenner et al., 2014). Indeed, we can find that the Bloch spectral function becomes
285 broaden with increasing impurity concentration (Figs. 3 and 4). In Fig. 7, the Fe-Ni
286 resistivity increases with increasing nickel concentration, and the maximum resistivity
287 is found around 50 at.% Ni. At higher nickel concentrations, the resistivity decreases
288 with increasing the nickel content. This parabolic-like behavior could be explained by
289 Nordheim's rule. On the other hand, the resistivity of Fe-Si alloys increases with
290 increasing silicon concentration almost linearly, and then a kink is found ~20 at.% Si
291 (Fig. 7). At the same point, the Bloch spectral function becomes broaden, and the Fermi
292 surface is almost smeared out at ~20 at.% Si (Figs. 3 and 5). The collapse of a large
293 portion of the Fermi surface is related to the resistivity saturation (Butler 1985;
294 Glasbrenner et al. 2014). The Ioffe-Regel condition suggests that the mean free path
295 hardly becomes shorter than interatomic distance. In the wave number space, this
296 semi-classical picture corresponds to the following; the width becomes broaden all over
297 the Brillouin zone, which has the order of the inverse of lattice constants. Butler (1985)
298 estimated the resistivity of alloys with energy bands, which are completely smeared out.
299 The value is approximately $1.5 \times 10^{-6} \Omega\text{m}$, and thus he argued that this limitation may

300 be the origin of the resistivity saturation. In this study, we only considered the impurity
301 resistivity, but the same argument could also be applied to the phonon-contributed and
302 magnetic-disorder resistivity as demonstrated by Glasbrenner et al. (2014).

303 We observed three characteristic behaviors closely related to resistivity saturation:
304 1) bending in resistivity change, 2) coincident with the collapse of a large portion of the
305 Fermi surface (the Ioffe-Regel condition), and 3) the breakdown of the Matthiessen's
306 rule. Although individual phenomenon is rather qualitative, we consider resistivity
307 saturation is robust considering all of these three pieces of evidence.

308 *4.2. Comparison between measurements and calculations*

309 Although the resistivity saturation is widely observed in highly disordered transition
310 metals and alloys, there are a few exceptions that have higher resistivity than the
311 Ioffe-Regel value of $\sim 1.5 \times 10^{-6} \Omega\text{m}$ (Gunnarsson et al., 2003). In this section, we
312 compare our results with previously reported resistivity data of Fe–Si and Fe–Ni
313 binaries, in order to evaluate the applicability of our theory to the Earth's core.

314 Fig. 8a summarizes the electrical resistivity of Fe–Si and Fe–Ni alloys at ambient
315 pressure. The resistivity increases with increasing silicon concentration within the bcc
316 stability field, and then the resistivity decreases because of the formation of Fe₃Si
317 component with DO₃ structure. The local minimum is observed at stoichiometric
318 composition, which reflects a re-appearance of chemical ordering. Varga et al. (2002)
319 obtained single-phase highly disordered alloys with 28–34 at.% Si composition by
320 means of rapid-quenching technique, although a mixture of Fe₃Si and Fe₅Si₃ component
321 is thermodynamically stable. Fe₃Si-based ternary alloys (Fe_{1-x}M_x)₃Si exhibit
322 anomalously high resistivity due to the ferromagnetism (e.g., Nishino et al., 1993).

323 The impurity resistivity of nickel in iron is smaller than that of silicon, which is
324 consistent with the fact that the impurity resistivity of a transition metal impurity in
325 transition metal host is small compared to that of non-transition metal impurity, in
326 general. At 1 bar, the resistivity increases with increasing nickel impurity concentration
327 within the bcc stability field (Fig. 8a). A discontinuous change in the resistivity at $x_{\text{Ni}} \approx$
328 30 at.% is attributed to the phase transition from bcc to face-centered cubic (fcc)
329 structure (Ho et al., 1983). After the phase transition, anomalously high resistivity
330 region is found up to 40 at.% Ni, which may be explained by the weak ferromagnet
331 model (Ho et al. 1983). In this compositional range, very small thermal expansion
332 coefficient has been reported, which is characteristic of Invar alloys. Our calculations
333 did not reproduce such anomaly, because we only considered the non-magnetic hcp
334 phase. At the nickel concentrations higher than 50 at.%, the resistivity decreases with
335 increasing nickel content.

336 Fig. 8b compares experimental results with first-principles calculations on the
337 resistivities of hcp Fe–Si and Fe–Ni alloys at ~40 GPa. The first-principles resistivity of
338 Fe–Si alloys increases almost linearly with increasing silicon content up to ~20 at.% Si,
339 which shows excellent agreement with the present DAC measurements. Furthermore,
340 previous shockwave results on Fe–Si alloys (Matassov, 1977) are also very close to our
341 measurements and calculations, except at high silicon content (cf. 34.2 at.% Si), even
342 though these shock-wave data were obtained along the Hugoniot. This is consistent with
343 the saturation theory that temperature dependence becomes small at high resistivity
344 (Rossiter, 1987). Such a small, or sometimes negative, temperature coefficient of
345 resistivity (TCR) is known as Mooij’s law (Mooij, 1973). Recently, this TCR behavior
346 was confirmed by high-pressure experiments in a cubic anvil press (Kiarasi and Secco,

2015). The modeling of the TCR will be presented in the next section. On the other hand, the shock result on Fe–34.2at.%Si shows abnormally high resistivity, which is also found in results obtained at ambient pressure (Varga et al., 2002 and references therein) (Fig. 8a). Note also that the maximum silicon content in the Earth’s core should be less than 25 at.% Si (e.g., Hirose et al., 2013). Our previous DAC results on Fe–Ni alloys at room temperature are in good agreement with theoretical calculations at 0 K. The shock compression result on Fe–19.2at.%Ni by Matassov (1977) reported larger resistivity, but it could be due to the effects of high temperature and the phase transformation to the fcc phase (Kuwayama et al., 2008).

The comparison is made at 80 and 120 GPa in Figs. 8c and 8d, respectively. The present first-principles predictions are very consistent with the DAC measurements at these higher pressures too. We see the consistency and the difference between previous shock results and theoretical calculations, similar to those found at 40 GPa (Fig. 8b).

4.3. Resistivity modeling considering both thermally- and chemically-induced resistivity saturation

The resistivity saturation strongly limits the total resistivity of the core, because 1) the bulk resistivity rarely exceeds the saturation resistivity and 2) the saturation decreases the temperature coefficient of resistivity. Considering the Wiedemann-Franz law with the Sommerfeld value of the Lorenz number is a good approximation at the Earth’s core conditions (Gomi and Hirose, 2015), the thermal conductivity of the Earth’s core should have a lower limit depending on its temperature and increase linearly with increasing temperature.

Without any specific modeling, we can roughly estimate the resistivity of the core

370 from the saturation value. For instance, assuming the saturation resistivity to be $1.5 \times$
 371 $10^{-6} \Omega\text{m}$ and temperature to be 4000 and 5500 K, the thermal conductivity should be
 372 greater than 65 and 90 W/m/K, respectively. For a total resistivity of $1.0 \times 10^{-6} \Omega\text{m}$, the
 373 calculated thermal conductivity would be 98 W/m/K at 4000 K and 135 W/m/K at 5500
 374 K. These values are indeed very close to recent high values of the thermal conductivity
 375 estimates (de Koker et al., 2012; Gomi and Hirose, 2015; Gomi et al., 2013; Pozzo et al.,
 376 2012, 2013, 2014), and thus we emphasize that the resistivity saturation is the most
 377 important factor to constrain the thermal conductivity of the Earth's core.

378 The present first-principles calculations demonstrate the chemically-induced
 379 resistivity saturation. Now we formulate a core conductivity model as functions of
 380 volume and temperature, in order to demonstrate how the effect of temperature will be
 381 suppressed by the saturation.

382 It is widely accepted that the saturation behavior of pure metals can be well
 383 described by a shunt-resistor model (Gunnarsson et al., 2003). However, several models
 384 are proposed for highly disordered alloys (Rossiter, 1987). In our previous studies
 385 (Gomi et al., 2013; Gomi and Hirose, 2015), we applied the model described by
 386 Bohnenkamp et al. (2002). However, it is only valid for dilute alloys. Instead, we
 387 employ the one reported by Cote and Meisel (1978):

$$388 \quad \rho_{\text{tot}}(V, T) = \left(1 - \frac{\rho_{\text{tot}}(V, T)}{\rho_{\text{sat}}(V)}\right) \rho_{\text{ph,ideal}}(V, T) + \rho_{\text{imp}}(V) \exp(-2W(T)) \quad (6)$$

389 Note that Eq. (6) is equivalent to the shunt-resistor model (Eq. S6), when impurity
 390 resistivity term is neglected. We obtain phonon-contributed resistivity $\rho_{\text{ph,ideal}}(V, T)$ from
 391 the Bloch-Grüneisen formula (Gomi et al., 2013) and impurity resistivity $\rho_{\text{imp}}(V)$ from
 392 the present first-principles calculations. Bohnenkamp et al. (2002) reported the

393 saturation resistivity to be $\rho_{\text{sat}} = 1.68 \times 10^{-6} \Omega\text{m}$ for iron and iron based alloys at 1 bar.
 394 Gomi et al. (2013) assumed that it diminishes at high pressure as a function of
 395 inter-atomic distance. The remaining term in Eq. (6) is the Debye-Waller factor,
 396 $\exp(-2W(T))$, which is negligible in general (Rossiter, 1978; Markowitz, 1976).
 397 Assuming the Debye model, $W(T)$ can be obtained as follows (Markowitz, 1976):

$$398 \quad W(T) = \frac{3\hbar^2 K^2 T^2}{2mk_B \Theta_D^3} \int_0^{\Theta_D/T} \left(\frac{1}{\exp(z) - 1} + \frac{1}{2} \right) z dz \quad (7)$$

399 where $\hbar$ is the reduced Planck's constant (the Dirac's constant), k_B is the Boltzmann
 400 constant, m is atomic mass, and K is electronic wave vector transfer that is roughly
 401 equal to the Fermi wave vector, k_F . With $K = \pi/a = 1.39 \text{ \AA}^{-1}$, $M = 55.85 \text{ g/mol}$, and Θ_D
 402 $= 710 \text{ K}$, the Debye-Waller factor is calculated to be $\exp(-2W(T)) \approx 0.95$ at $V = 16.27 \text{ \AA}^3$
 403 and $T = 5000 \text{ K}$ ($P \sim 169 \text{ GPa}$), which is indeed very close to unity. The total resistivity
 404 of the Fe-Si system is shown in Supplementary Fig. S4. Our model is broadly
 405 consistent with previous shock compression measurements (Matassov, 1977). Applying
 406 the resistivity model described above, the electrical resistivity (thermal conductivity) of
 407 $\text{Fe}_{65}\text{Ni}_{10}\text{Si}_{25}$ (at $V = 16.27 \text{ \AA}^3$) are $1.12 \times 10^{-6} \Omega\text{m}$ (87.1 W/m/K) at 4000 K (156.19
 408 GPa) and $1.16 \times 10^{-6} \Omega\text{m}$ (116 W/m/K) at 5500 K (175.2 GPa).

409 4.4. Implications for the Earth's core

410 Since thermal convection can occur only when the total heat flow is higher than the
 411 conductive heat flow along isentropic temperature gradient,

$$412 \quad Q_s(r, t) = -4\pi r^2 k(r, t) \left(\frac{\partial T(r, t)}{\partial r} \right)_s, \quad (8)$$

413 the high thermal conductivity, arising from the resistivity saturation, strongly suppresses

414 thermal convection (see supplementary material). Here we compute the thermal
 415 evolution of the Earth's core based on the core energy model (Labrosse, 2015) with our
 416 new value of the thermal conductivity and highly concentrated uranium and thorium
 417 (Wohlers and Wood, 2015). We assume that the core resistivity is close to saturation and
 418 hence the thermal conductivity depends only on the temperature (see supplementary
 419 material):

$$420 \quad k(r, t) = k_{\text{CMBp}} \frac{T(r, t)}{T_{\text{CMBp}}} \quad (9)$$

421 where $k_{\text{CMBp}} = 87.1$ W/m/K is the present-day thermal conductivity at the top of the
 422 core.

423 Our calculation without any radioactivity indicates that the inner core age is younger
 424 than 0.71 Ga, which is inconsistent with the Mesoproterozoic (~1.3 Ga) inner core
 425 nucleation proposed by Biggin et al. (2015). Furthermore, incorporation of 8 ppb
 426 uranium and 8 ppb thorium (Wohlers and Wood, 2015) cannot extend the inner core age
 427 older than 0.85 Ga. We also examined the effect of potassium, which did not
 428 significantly alter the conclusion (Fig. 10). Additional energy sources, such as chemical
 429 buoyancy arising from magnesium precipitation (O'Rourke and Stevenson, 2016),
 430 might explain the Mesoproterozoic inner core nucleation. But, we prefer the very young
 431 inner core (< 0.5 Ga) inferred from more recent analysis of palaeomagnetic database by
 432 Smirnov et al. (2016).

433 **5. Summary and conclusion**

434 We conducted high-pressure resistivity measurements on Fe-Si alloys (with 1, 2, 4,
 435 6.5, and 9 wt.% Si) in a diamond-anvil cell up to 90 GPa at 300 K by means of the

436 four-terminal method. We also performed first-principles calculations on
 437 substitutionally disordered hcp Fe-Si, Fe-Ni, and Fe-Ni-Si alloy systems by using the
 438 AkaiKKR package, which employs the KKR-CPA methods and the Kubo-Greenwood
 439 formula (Butler, 1985; Oshita et al., 2009). Experimentally-determined electrical
 440 resistivity increases linearly with increasing silicon content up to 9 wt.% Si (16.43 at.%
 441 Si), which shows good agreement with our calculations. However, such linearity is
 442 violated at higher Si concentrations with a kink around 20 at.% Si coincident with the
 443 collapse of the large portion of the Fermi surface (the Ioffe-Regel condition). We also
 444 calculated the electrical resistivity of $\text{Fe}_{100-x}\text{Ni}_{10}\text{Si}_x$ ($0 \leq x \leq 30$), which clearly shows the
 445 breakdown of the Matthiessen's rule. Furthermore, the resistivity of Fe-Si alloys
 446 obtained by our room-temperature measurements and zero-temperature calculations is
 447 very close to the that found in previous shock-compression experiments under high
 448 temperature (Matassov, 1977), which implies very small temperature coefficient of the
 449 resistivity. These non-linear behaviors are, indeed, frequently found in the resistivity of
 450 transition metals and alloys, and called "resistivity saturation" (Bohnenkamp et al.,
 451 2002; Gunnarsson et al., 2003; Kiarasi and Secco, 2015).

452 Considering the resistivity saturation, the resistivity of the Earth's core is estimated to
 453 be $\sim 1.0 \times 10^{-6} \Omega\text{m}$, which may be insensitive to temperature and composition. This
 454 value is one of the lowest values ever estimated (Gomi and Hirose, 2015). The
 455 resistivity saturation has significant implications for the thermal structure, dynamics,
 456 and evolution of the Earth's core, because the low electrical resistivity means the high
 457 thermal conductivity through the Wiedemann-Franz law. The high thermal conductivity
 458 strongly suppresses the thermal convection. We conducted thermal evolution calculation
 459 with radioactive uranium and thorium (Wohlert and Wood, 2015). The result indicates

460 that the inner core age is younger than 0.85 Ga, which is consistent with very young
461 inner core (< 0.5 Ga) inferred from elaborate paleomagnetic intensity analysis reported
462 by Smirnov et al. (2016).

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467

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593

594 **Figure captions**

595 **Fig. 1.** A sample configuration in a DAC. An Fe–Si alloy foil was cut to the
596 four-terminal shape by using a Nd:YAG laser or an FIB. It was then loaded into an
597 Al_2O_3 insulating gasket. Four platinum foil electrodes were connected with the Fe–Si
598 sample.

599 **Fig. 2.** (a) Electrical resistivity of Fe–Si alloys as a function of pressure. The yellow (1
600 wt.% Si), light green (2 wt.% Si), deep green (4 wt. % Si), blue (6.5 wt.% Si), and
601 purple (9 wt.% Si) circles and diamonds are present measurements. Previous
602 measurements conducted by Gomi et al. (2013) (red and orange circles for pure Fe) and
603 Seagle et al. (2013) (purple triangles for 9 wt.% Si alloy) are also plotted for
604 comparison. (b) Electrical resistivity of Fe–Ni alloys as a function of pressure from
605 Gomi and Hirose (2015); yellow (5 wt.% Ni), green (10 wt.% Ni), and blue circles (15
606 wt.% Ni). The red and orange symbols indicate the resistivity of pure iron (Gomi et al.,
607 2013).

608 **Fig. 3.** Electronic density of states (DOS) and band structure of $\text{Fe}_{98.03}\text{Si}_{1.97}$ (a, b),
609 $\text{Fe}_{92.35}\text{Si}_{7.65}$ (c, d), $\text{Fe}_{80}\text{Si}_{20}$ (e, f) and $\text{Fe}_{70}\text{Si}_{30}$ (g, h) at $19.10 \text{ \AA}^3$ ($\sim 40 \text{ GPa}$). The Fermi
610 energy is set to be 0 Ry.

611 **Fig. 4.** Electronic density of states (DOS) and band structure of $\text{Fe}_{95.23}\text{Ni}_{4.77}$ (a, b),
612 $\text{Fe}_{90.44}\text{Ni}_{9.56}$ (c, d), $\text{Fe}_{85.62}\text{Ni}_{14.38}$ (e, f) and $\text{Fe}_{50}\text{Ni}_{50}$ (g, h) at $19.10 \text{ \AA}^3$ ($\sim 40 \text{ GPa}$). The
613 Fermi energy is set to be 0 Ry.

614 **Fig. 5.** The Bloch spectrum function of iron silicon alloys of $\text{Fe}_{98.03}\text{Si}_{1.97}$ (a, b, c),
615 $\text{Fe}_{92.35}\text{Si}_{7.65}$ (d, e, f), $\text{Fe}_{80}\text{Si}_{20}$ (g, h, i) and $\text{Fe}_{70}\text{Si}_{30}$ (j, k, l) with volume of $19.10 \text{ \AA}^3$ (~40
616 GPa) at the Fermi energy.

617 **Fig. 6.** The Bloch spectrum function of iron nickel alloys of $\text{Fe}_{95.23}\text{Ni}_{4.77}$ (a, b, c),
618 $\text{Fe}_{90.44}\text{Ni}_{9.56}$ (d, e, f), $\text{Fe}_{85.62}\text{Ni}_{14.38}$ (g, h, i) and $\text{Fe}_{50}\text{Ni}_{50}$ (j, k, l) with volume of $19.10 \text{ \AA}^3$
619 (~40 GPa) at the Fermi energy.

620 **Fig. 7.** Electrical resistivity of (a) Fe-Si and (b) Fe-Ni alloys calculated by using the
621 KKR-CPA combined with the Kubo-Greenwood formula (solid circles) compared with
622 DAC experimental results (open circles) at ~40 GPa.

623 **Fig. 8.** Present (circle) and previous (square and diamond) electrical resistivity data for
624 Fe-Si (blue) and Fe-Ni alloys (green) as a function of silicon and nickel concentration.
625 Filled circles, present calculations of hcp alloys; open circles, present measurements of
626 Fe-Si alloys and the previous measurements on Fe-Ni alloys (Gomi and Hirose, 2015);
627 diamonds, previous shock compression measurements by Matassov (1977). Blue open
628 squares show the resistivity of single-phase Fe-Si alloys reported by Varga et al. (2002)
629 and references therein. The literature values for Fe-Ni alloys (Ho et al., 1983) are
630 shown as green broken line. (a) 1 bar, (b) 40 GPa, (c) 80 GPa, and (d) 120 GPa. Blue
631 and green filled circles are present calculations of hcp iron-silicon and iron-nickel
632 alloys.

633 **Fig. 9.** Electrical resistivity of $\text{Fe}_{100-x}\text{Si}_x$ binary (blue) and $\text{Fe}_{90-x}\text{Ni}_{10}\text{Si}_x$ ternary (red)
634 alloys as a function of silicon content at the volume of $16.27 \text{ \AA}^3$ (~120 GPa). Gray

635 broken line shows the sum of the resistivities of $\text{Fe}_{90}\text{Ni}_{10}$ and $\text{Fe}_{100-x}\text{Si}_x$ (the
636 Matthiessen's rule). Note that the Matthiessen's rule is violated at high resistivity.

637 **Fig. 10.** Inner core age as functions of concentration of radioactive elements in the
638 Earth's core. (a) Uranium and thorium. (b) Uranium and potassium. Radioactive
639 elements produce large portion of energy available for the dynamo at early Earth
640 (Wohlers and Wood, 2015), however, ~ 8 ppb uranium and ~ 8 ppb thorium are not
641 sufficient to extend the inner core age of ~ 1.3 Ga, which is proposed by Biggin et al.
642 (2015).

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