Electronic transition in liquid Fe at P, T conditions: Implication for double-diffusive convection in Mercury's dynamo.

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Article

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1 **Electronic transition in liquid Fe at P , T conditions: Implication for double-**
2 **diffusive convection in Mercury's dynamo.**

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7 **Abstract**

8 A double-diffusive model demonstrated that Mercury's dynamo is driven by thermochemical
9 convection with thermal energy source deep in its core. Although radioactive elements
10 concentrated in the core has been suggested as the source of the energy, arguments were
11 made that there is no geochemical or geophysical validation for this. We report the
12 temperature-dependent resistivity of solid and liquid Fe measured up to 21 GPa in multi anvil.
13 With increasing pressure, we observe resistivity transition with a value change of about 35
14 $\mu\Omega\text{-cm}$ at the melting boundary on the liquid side at ~ 18 GPa. This change in resistivity
15 corresponds to a change in thermal conductivity of about $30 \text{ Wm}^{-1}\text{K}^{-1}$ that would generate
16 ~ 0.94 TW change in heat flux. This indicates that transition in Fe properties is responsible
17 for thermal buoyancy in Mercury's dynamo and not radioactive materials. From the entropy
18 balance, we estimate a thermal dynamo power of about 0.1 TW.

Introduction

The physical and chemical properties of planetary materials impact the mechanism of the planetary heat engine. Since Fe is the major element in the core of rocky planetary bodies, the changes in its transport properties at pressure (P) and temperature (T) conditions should be taken into account in understanding the power to a planetary dynamo. Mercury has a global magnetic field and a dynamo operating in its liquid outer core just like the geodynamo is likely its source (Christensen, 2006; Takahashi et al, 2019). However, Mercury has a very low field intensity, with a surface field strength of about ~1% the strength of the Earth's field (Aharonson et al., 2004; Christensen, 2006) and a morphology unlike any other planetary body (Anderson, 2011; Johnson et al 2012; Takahashi et al., 2019). Application of common assumptions made on Earth to Mercury's dynamo yields a field strength thirty times stronger than the observed (Christensen, 2006). This suggests that Mercury's dynamo operation could be different from the geodynamo and assumptions made in understanding Earth's dynamo may not apply to Mercury. Stanley et al. (2005) suggested that the generation of Mercury's magnetic field may be restricted to a thin fluid shell that generates a strong (hidden) toroidal field, but a weak (observable) poloidal field. An investigation by Christensen (2006) attributed the surface observed Mercury's field to a strong field powered by thermochemical convection deep in the core which diffuses and attenuates by skin effect through a stable stratified conducting part of the outer core. Although the above-mentioned models have successfully explained the weak global field observed on the surface of Mercury, they were based on the concept of co-density, which assumes the same diffusivity for the compositional and thermal buoyancies. On the contrary, compositional and thermal

diffusivities differ by a large factor (Manglik et al., 2010; Bouffard et al., 2019). Using double-diffusive convection model, Manglik et al. (2010) simulated a dynamo driven by thermochemical convection where thermal power is generated from the bottom of the liquid outer core mainly by the heat flux at the inner core boundary (*ICB*) due to the latent heat of solidification from a very large inner core size. In comparison with the observed surface field of Mercury, their model resulted in a stronger multipolar field. Very recently, Takahashi et al. (2019) presented a Mercury's dynamo model where core convection is driven by thermo-compositional double-diffusive convection surrounded by a thermally stable stratified layer at the top of the core and thermally powered by internal heating deep in the liquid core, with a zero latent heat flux at the *ICB*. This model was successful in producing a magnetic field similar in morphology and strength to the observed Mercury's field. However, the origin of the internal heat source has not been constrained well (Takahashi et al., 2019). Although radioactive elements concentrated within the liquid outer core have been suggested as the heat source, arguments were made that there is no obvious geophysical or geochemical justification (Takahashi et al., 2019). Hence, the knowledge of the electrical and thermal transport properties of liquid Fe at the P and T conditions applicable to the deep core interior region of Mercury is needed in understanding the thermal power source of Mercury's dynamo.

Systematic investigations on the electrical resistivities of Fe have been made at relatively lower P conditions (Secco and Schloessin, 1989; Deng et al., 2013; Silber et al., 2018, Yong et al., 2019; Ezenwa and Yoshino, 2020a) reaching the top part of Mercury's core-mantle boundary (*CMB*) at ~ 5 GPa. Although measurements up to P and T conditions

applicable to the deep part of Mercury's core have been made by Yong et al (2019), sample contamination by electrode materials especially in the liquid state may have disguised changes in Fe transport properties. Ezenwa and Yoshino (2020a) recently developed a technique and a cell design capable of investigating the electrical resistivity of transition metals devoid of contamination under high P and T conditions in multi-anvil press. This provided an opportunity for more accurate measurement of liquid Fe electrical resistivity in a wide P and T range. Although measurements can be made at much higher P in laser-heating diamond anvil cell (LH-DAC) (Ohta et al., 2016; Youjun, et al., 2020) relative to multi-anvil large volume press, it is difficult to make fine T control and preserve liquid sample geometry in LH-DAC. Hence, accessible P up to the deep core region of Mercury and preservation of liquid sample geometry in large volume press is highly advantageous for a better understanding of Mercury's core dynamics.

The structure of Mercury's interior has not been well constrained (Rivoldini et al., 2009; Tosi et al., 2013), with its inner core center P at ~ 39 GPa (Chen and Hauck, 2008; Hauck et al., 2013), the dominant phase of Fe at Mercury's core conditions is fcc (γ) Fe. With increasing P and T , materials not only undergo structural changes but also changes in their electronic properties such as spin states. At ambient condition, Fe with bcc structure (α phase) and high spin state is ferromagnetic and changes to paramagnetic above its Curie T . Above Curie T , although unpaired spin states exist, the net-sum of Fe magnetic moments adds up to zero due to spin-randomization. On the other hand, with increasing P , the magnetic moment of Fe changes from ferromagnetic to paramagnetic and hcp structure ($\mathcal{E}$ -phase) with a low spin state at ~ 18 GPa (Manthou et al., 2004; Iota et al., 2007; Mankovsky et al., 2013). One

would expect that at high P above 18 GPa and T conditions, this transformation would not only affect Fe magnetic properties but also its physical transport properties since they are interconnected and governed by their electronic structure (Landrum and Dronskowski, 2000; Ezenwa and Secco, 2019; Ezenwa and Yoshino, 2020b). Thus, we investigate the T -dependent electrical and thermal transport properties of solid and liquid Fe across its magnetic transition (ferromagnetic $\rightarrow$ paramagnetic) with increasing P and discuss its fundamental implications to Mercury's deep dynamo power. A detailed description of the adopted technique has been described by Ezenwa and Yoshino, (2020a), however, slight modifications were made in this present study as detailed in the supplementary material.

Main

Fig. 1 shows the results of our measured electrical resistivity of solid and liquid Fe up to 21 GPa as a function of T in comparison with previous studies at 1 atm and at high P (Chu and Chi, 1981; Ezenwa and Yoshino, 2020a). The electrical resistivity increases with T in both solid and liquid states. From the results of our previous measurements (Ezenwa and Yoshino, 2020a), we observed the appearance of the high P phase of Fe ($\mathcal{E}$) at around 750 K with increasing T at fixed P of 8 GPa. The drop in resistivity at $\mathcal{E}$ - γ transition which is distinguishable at around 12 GPa (Fig. 1B) indicates that $\mathcal{E}$ -Fe has a slightly higher resistivity than γ -Fe. At room temperature, the complete disappearance of the α phase was confirmed at $\sim$ 18 GPa with the eradication of T^2 dependent resistivity with increasing T . The near-linear T -dependent resistivity in both $\mathcal{E}$ and γ phases trails the reduction/termination of magnon contribution to Fe resistivity. In general, the phase boundaries observed in this study are

consistent with Fe phase stability studies using synchrotron X-ray diffraction in a Kawai-type multi anvil press (Uchida et al., 2001; Yamazaki et al., 2012). The sudden rise in the resistivity at higher T indicates the onset of melting while the gradual slope change above this transition marks the completion of melting between two electrodes.

With increasing P , resistivity decreases in the pure phases of α - (Ezenwa and Yoshino, 2020a), γ , and ϵ . However, at around (12-18) GPa where α and ϵ phases could have possibly co-existed, resistivity increases with increasing P in agreement with results of room T measurement study (Gomi et al., 2013). Above 18 GPa, we observed a significant decrease in the overall T -dependent resistivity at high T in the solid-state and well into the liquid as shown in Fig. 1A. Recently Ezenwa and Yoshino (2020b) suggested that the T -dependent electrical resistivity of Pt and other Pt-group metals could compare with the T -dependent resistivity of ϵ -Fe since both are paramagnetic and exhibits close-packed crystal structure at relevant P and T conditions. Interestingly, Youjun et al. (2020) resistivity investigation of ϵ -Fe up to 170 GPa and 3000 K confirmed this prediction and their reported values at the melting point on the solid side are also in a reasonable agreement with a recent direct thermal conductivity investigation by Saha et al., (2020).

Focusing on the resistivity along the melting boundary of Fe, the resistivity of liquid Fe decreases up to 5 GPa (Ezenwa and Yoshino 2020a), then remains constant with a value of $\sim 100 \mu\Omega\text{-cm}$ up to 18 GPa and drops to a value of $\sim 65 \mu\Omega\text{-cm}$ up to the maximum P in our measurement range. At 1 atm, Güntherodt et al. (1975) indicated that spin-disorder contribution to the resistivity of Fe, Co, and Ni could still be present even in their liquid state

but not simply additive. As P broadens and populates d -band states (McMahan and Albers, 1982; McMahan, 1989; Ross et al., 2007), which in turn destabilizes magnetism and reduces/terminates magnon contribution to the T -dependent resistivity of Fe and other similar metals (Ezenwa and Secco 2017; Ezenwa and Secco, 2019) through s - d electron hybridization, one would expect non or negligible magnetic contribution in liquid Fe resistivity at above 18 GPa well into ε and $-\gamma$ phases. The collapse of ordered magnetism across 18 GPa in Fe has been observed in the X-ray magnetic circular dichroism and the X-ray absorption measurements at room T (Mathon et al., 2004; Iota et al., 2007). Since magnetism and electrical charge transports are interconnected with electronic structure, such change could translate to a fundamental change in Fe electronic structure. Remarkably, a recent theoretical study by Korell et al. (2019) demonstrated that paramagnetic state in liquid Fe is stable at 1 atm and it persists with increasing P and becomes diamagnetic beyond 20–50 GPa. Since thermal conductivity and electrical resistivity are fundamentally linked to the dynamics of electrons in metals, it is expected that a change in Fe electrical resistivity would imply a change in thermal conductivity.

The electronic component of thermal conductivity (K_e) of Fe was determined from our measured electrical resistivity at various P and T conditions by plugging resistivity data into Wiedemann-Franz’s relation with Sommerfeld value ($L_o = 2.445 \times 10^{-8} W\Omega/K^2$) of Lorenz function (Secco, 2017) (Fig. 2). With increasing T , K_e of Fe decreases and increases for ε and $-\gamma$ Fe phases, respectively. On melting, K_e decreases and increases slightly with increasing T above the melting point. Along the melting point on the liquid side, as shown in

Fig. 3, a sudden jump of thermal conductivity occurs at $\sim (18-20)$ GPa. A change in thermal conductivity was estimated to be about $30 \text{ Wm}^{-1}\text{K}^{-1}$.

The heat transport in a material by conduction is inherent in its thermal conductivity property at P and T conditions. The percentage of heat transported by convection is more than that which cannot be conducted in planetary cores (Gubbins, 1976). Hence, a reliable determination of the conducted heat flux is essential in calculating the heat transport by convection which translates into available power for the dynamo through thermal buoyancy. A change in Fe electrical and thermal properties as observed in our study would have implications for the core dynamics of planetary bodies such as Mercury whose deep liquid outer core is within the P and T range of this transition. The observed transition would contribute to the available thermal buoyancy power in Mercury's core and dictates the generation and the nature of its magnetic field. To calculate the corresponding change in adiabatic heat flux ($Q_{\text{Cond Difference}}$) across this transition, we use the equation ($Q_{\text{Cond Difference}} = \kappa_e \frac{\alpha g T}{C_p}$), where (α) is thermal expansion, (g) is gravitation acceleration, (C_p) is heat capacity and T is the associated average melting T of Fe determined at ~ 19 GPa in our study (See. Stevenson, 1984). Using parameter values as tabulated in Table 1 and substituting for the change in the κ_e value, we calculated a value of 27 mWm^{-2} for the heat flux difference across this transition. We note that our calculated heat flux is the equivalent of that which cannot be transported by conduction due to the decrease in the thermal conductivity of Fe at the core side of the conducting layer-outer core boundary and as such thermal convection will be the heat transport carrier. The entire radius of Mercury's

core is about 2000 km (Rivoldini and Van Hoolst, 2013; Knibbe and van Westrenen, 2015) with an inner core radius of ~1325 km (Dumberry and Rivoldini 2015), we consider it reasonable to take the mean value of the entire core and the inner core radius value as the radius of the core at this conductivity transition boundary which gives a value of 1662 km. Given that the P at the top of its *CMB* is at ~5GPa (Rivoldini et al., 2009; Tosi et al., 2013) and ~35 GPa at its core center (Chen and Hauck, 2008; Hauck et al., 2013), using simple scaling, we estimate the P at the depth of 1662 km to be around 20-24 GPa. With this estimated radius value, a value of 0.94 TW was found as available heat energy for thermal convection across this region.

The consistent results among thermal evolution studies demonstrated that the heat flux at the *CMB* of Mercury becomes sub-adiabatic in the first billion years (Gyr) of its evolution (Hauck II et al., 2004; Grott et al., 2011; Tosi et al., 2013), thermally stratifying an outer core layer which supports the deep core dynamo powered by thermochemical convection. When the two agents of buoyancy have large different diffusivities, this has been termed double-diffusive convection (Manglik et al., 2010). An investigation has shown that double-diffusive convection where thermal buoyancy deep inside the liquid core powered by internal heating but not by latent heat release from core solidification processes can generate Mercury's like field features (Takahashi et al., 2019). Because of this, slow cooling of the core, retarded inner core growth, and relatively small inner core size has been suggested (Tosi et al 2013; Hauck et al., 2013). Our observed transition in Fe electrical and heat transport properties indicates that thermal convection originating from a change in liquid Fe transport properties is the source of the thermal dynamo power in its double-diffusive thermochemical

convection and may not be a concentration of radioactive elements specifically in this region. Fig. 4 shows an illustration of the core structure of Mercury with the conductivity transition region at its liquid outer core.

The global energy budget determines the core evolution by balancing the heat transported out of the core against the sum of the heat sources within the core. The total heat budget (Q_{Total}) in Mercury's core would include secular cooling(Q_s), latent heat (Q_L), a possible radioactive heat source (Q_{rad}), gravitational energy (Q_g) and energy associated with our observed electronic transition (Q_{Trans}) as follows:

$$Q_{Total} = Q_s + Q_L + Q_{rad} + Q_g + Q_{Trans}.$$

The deeper part of Mercury's core dynamics is largely driven by the extraction of heat by the stratified conducting layer on top of its liquid outer core, possibly Fe-S in composition. By assuming a steady-state system, the amount of heat supplied from the bottom of the core is equal to the amount of heat taking away from the top of the liquid outer core as illustrated in Fig. 4A. Based on entropy balance (Buffett, 2002), the entropy at the boundary between the solid FeS layer and the top of the liquid outer core is balanced by the core fluid motion which powers the dynamo as shown in the equation below and illustrated in Fig. 4B. The available dynamo power contribution from thermal buoyancy ($\phi_{thermal}$) is given by the expression below (Buffett, 2002).

$$\phi_{thermal} \approx \left[\frac{\bar{T}}{T_{out}} - \frac{\bar{T}}{T_{in}} \right] Q \quad (1)$$

$$\phi_{thermal} = \epsilon_T (Q_{FeS-core} - Q_{adiabat}) \quad (2)$$

We note that there would be other contributions to the dynamo power such as compositional buoyancy originating from the release of light alloying elements as the inner core solidifies and grow in size. In this study, we focus on the estimate of the thermal buoyancy component originating from our observed thermal conductivity transition. We estimate a value of 2012 K for the T_{out} , by taking the average of the melting T of FeS at ~18 GPa of value 1900 K (Boehler, 1996) and that of pure Fe at ~18 GPa of value 2125 K as measured in our study. The $\bar{T}$ is the T at the well-mixed convecting region. We take $\bar{T}$ to be the melting T of Fe at 19 GPa of value 2134 K while T_{in} is our measured melting T of pure Fe of value 2142 K at 21 GPa, as determined in this study. The dimensionless Carnot efficiency (ϵ_T) for the thermal component of Mercury's dynamo power was calculated by plugging the values of the T 's in equation (1) and was estimated to be a value of ~0.1. ($Q_{FeS-core} - Q_{adiabat}$) is our calculated heat flux of value 0.94 TW transported by convection due to Fe thermal conductivity difference of value 30 Wm⁻¹K⁻¹. With this, we estimate the available thermal power ($\Phi_{thermal}$) for Mercury's thermochemical double-diffusive convective dynamo to be about 0.1 TW.

The two most likely light elements contained in Mercury's core are sulfur and/or silicon based on geochemical and geophysical arguments (Nittler et al., 2011; Knibbe, and van Westrenen, 2018). These light elements influence the absolute value of the electrical resistivity and thermal conductivity of Fe at P and T conditions as well as the concavity of its T dependent electrical and thermal properties (Berrada et al., 2020). This implies that the amount and nature of light elements alloying with Fe could influence the observed conductivity transition which could lead to a delay or a faster transition with increasing P .

At ambient conditions, magnetic transition in Fe with increasing Si concentration has been observed to occur at ~ (6-13) wt% (Secco, 2017). The effect of a light element on this transition will be investigated in future studies.

Supplementary Materials

See the supplementary materials for a description of the experimental method, and P , T profile of Mercury's core

Declaration of competing interest

No conflict of interest

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Fig.1. T -dependent electrical resistivity of solid and liquid Fe at various fixed P .

Fig.2. T -dependent electronic component of thermal conductivity (K_e) determined using Wiedemann-Franz relation with Sommerfeld of Lorentz number.

Fig.3. Electronic component of thermal conductivity (K_e) determined on melting on the liquid side at various fixed P conditions. The sharp rise in value indicates the conductivity transition.

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459 **Fig.4.** An illustration of the internal structure of Mercury's core including our newly
 460 identified conductivity transition region.

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Parameters	Values	Reference
Change in K_e	$30 \text{ Wm}^{-1}\text{K}^{-1}$	This Study
Thermal expansion (α)	$8.9 \times 10^{-5} \text{ K}^{-1}$	Secco, (2017)
Melting T FeS eutectic at 18GPa	1900 K	Boehler, (1996)
Melting T of Fe at 18GPa	2125 K	This Study
T_{out}	2012 K	Estimated (This Study)
$\bar{T}$	2134 K	Estimated (This Study)
T_{in}	2142 K	This Study
Acceleration gravity (g)	4 m/s^2	Rivoldini and Van Hoolst, (2013)
Heat Capacity (C_p)	$835 \text{ J Kg}^{-1} \text{ K}^{-1}$	Desai, (1989)
Resistivity change	$35 \mu\Omega\text{-cm}$	This Study
Outer core radius	2000 km	(Rivoldini and Van Hoolst, 2013; Knibbe and van Westrenen, 2015)
Inner core radius	1325 km	Rivoldini and Van Hoolst, 2013
Conductivity transition radius	1662 km	This study (estimated value)
Change in heat flux (conductivity transition)	27 mW/m^2	This Study
Total change in heat flux (conductivity transition)	0.94 TW	This Study
ϵ_T (Carnot efficiency)	0.1	This Study
$\emptyset_{\text{thermal}}$	0.1 TW	This Study

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465 **Table. 1.** Parameter values used for the calculated heat flow at the conductivity transition in
466 Mercury's deep liquid core.

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468

Figures

Figure 1

T-dependent electrical resistivity of solid and liquid Fe at various fixed P.

Figure 2

T-dependent electronic component of thermal conductivity (K_e) determined using 443 Wiedemann-Franz relation with Sommerfeld of Lorentz number.

Figure 3

Electronic component of thermal conductivity (K_e) determined on melting on the 453 liquid side at various fixed P conditions. The sharp rise in value indicates the conductivity 454 transition.

Figure 4

An illustration of the internal structure of Mercury's core including our newly 460 identified conductivity transition region.

Supplementary Files

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- [SupplementaryMaterialsMercuryMs.pdf](#)