Sound Velocities of Iron-Nickel ($Fe_{90}Ni_{10}$) Alloy up to 8 GPa and 773 K: The Effect of Nickel on the Elastic Properties of bcc-Iron at High P-T

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Abstract

Sound velocities of iron and iron-based alloys at high pressure and high temperature are crucial for understanding the composition and structure of Earth's and other telluric planetary cores. In this study, we performed ultrasonic interferometric measurements of both compressional (V_P) and shear (V_S) velocities on a polycrystalline body-centered-cubic (bcc)-$Fe_{90}Ni_{10}$ up to 8 GPa and 773 K. The elastic moduli and their pressure and temperature derivatives are derived from least square fits to third-order finite strain equations, yielding K_{S0} = 154.2(8) GPa, G_0 = 73.2(2) GPa, K_{S0}' = 4.6(2), G_0' = 1.5(1), $\partial K_S/\partial T$ = -0.028(1) GPa/K, and $\partial G/\partial T$ = -0.023(1) GPa/K. A comparison with literature data on bcc-Fe suggests the nickel content not only decreases both P and S wave velocities but also weakens the temperature effects on the elastic moduli of Fe-Ni alloys.

Key words: Fe-Ni alloy; sound velocity; high pressure and high temperature; ultrasonic interferometry

1. Introduction

Understanding the nature of Earth's core, which is the least accessible region of the Earth, is one of the most challenging tasks in geophysical research. Seismic waves can travel inside the Earth and serve as a powerful tool to probe the physical properties of Earth's interior, such as the density, compressional and shear wave velocities depth profiles; see for example the Preliminary Reference Earth Model [*Dziewonski and Anderson*, 1981]. Comparing seismic results with lab-based mineral physics investigations, as well as other evidence from geochemical, and cosmochemical studies, it has been widely accepted that Earth's core is composed of iron alloyed with approximately 10 wt.% nickel and several percent of light elements (such as Si, O, H, S, C, etc.) [e.g., *Birch*, 1952; 1964; *J Li and Fei*, 2003; *Mcdonough and Sun*, 1995]. However, direct studies on the behavior and elasticity properties of iron alloys at high pressure and high temperature (*HPHT*) are still scarce.

Iron-nickel alloys can exist in several crystallographic structures: body-centered-cubic (bcc) structure (α phase), face-centered cubic (fcc) structure (γ phase), and hexagonal close-packed (hcp) structure (ε phase), etc., depending on the pressure (*P*) and temperature (*T*) conditions and the nickel concentration (*Figure 1*). At ambient conditions, iron crystallizes in bcc structure while nickel prefers the fcc structure; when the nickel concentration exceeds ~20 wt.%, Fe-Ni alloys will gradually transform from bcc to fcc structure. However, there is no conclusive consensus on the phase diagram of Fe-Ni systems at high pressure and high temperature. Although most experimental evidences show that Fe-Ni alloy crystallize in the hcp structure at Earth's core conditions [e.g., *H K Mao et al.*, 1990; *Sakai et al.*, 2014; *Tateno et al.*, 2010; *Tateno et al.*, 2012], there are also experimental and theoretic predictions arguing that the bcc structure could be a more stable phase [e.g., *Dubrovinsky et al.*, 2007; *Vocadlo et al.*, 2003].

With the complexity of alloying with light elements, the phase diagram is even more controversial. Thus, more information about the physical and chemical properties (such as, density, sound velocity, bulk modulus, shear modulus, anisotropy, etc.) of the different phases of the Fe-Ni alloys are needed to further constrain the composition and structure of the Earth's core.

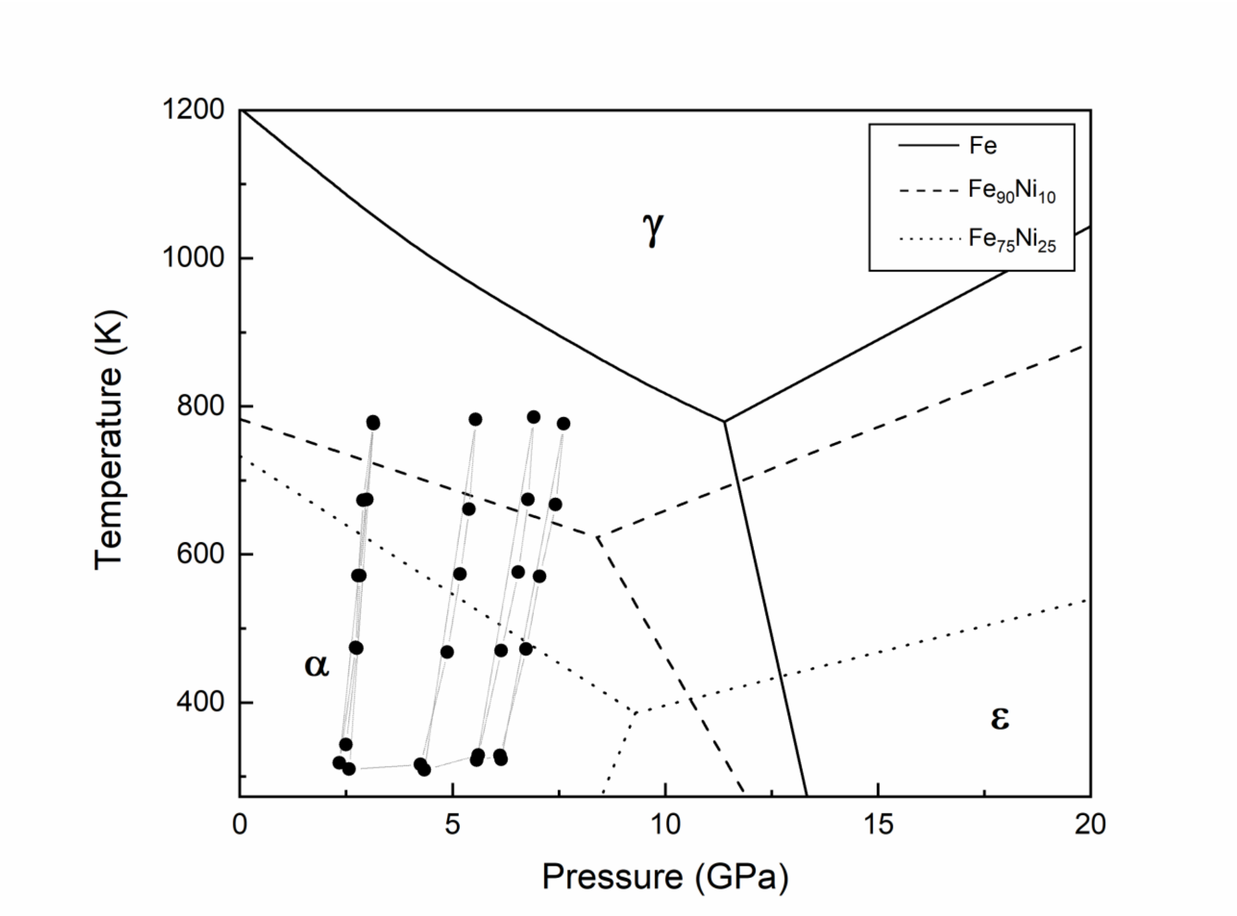

Figure 1 Phase diagram of Fe-Ni alloy system at high P-T based on the data of Huang et al., 1988; Huang et al., 1992. Solid dots indicate the experimental P-T conditions where the ultrasonic data were acquired in this study.

Sound velocities of pure iron (Fe) have been experimentally accessed by ultrasonic interferometry (UI), inelastic X-ray scattering (IXS), nuclear resonant inelastic X-ray scattering (NRIXS), and laser pulses (LP), etc.] at room temperature [e.g., *Chigarev et al.*, 2008; *Decremps et al.*, 2014; *Fiquet et al.*, 2001; *Gleason et al.*, 2013; *H K Mao et al.*, 1998; *Murphy et al.*, 2013] and high temperature [e.g., *Antonangeli et al.*, 2012; *Lin et al.*, 2005; *Liu et al.*, 2014; *Z Mao et al.*, 2012; *Ohtani et al.*, 2013; *Shibazaki et al.*, 2016]. In contrast, experimental studies on the sound velocity of iron-nickel (Fe-Ni) alloys are still limited [*Kantor et al.*, 2007; *Lin et al.*, 2003; *Morrison et al.*, 2019; *Wakamatsu et al.*, 2018], especially for the shear properties under simultaneous high pressure and high temperature conditions.

In the present study, we have carried out ultrasonic interferometric (UI) measurements on a polycrystalline bcc-$Fe_{90}Ni_{10}$ sample at simultaneous pressure and temperature conditions. Compared to other sound velocity measurement techniques, UI in large volume apparatus embeds the advantages of stable and uniform heating of sample and direct measurement of both P and S wave velocities simultaneously. We applied a third-order finite strain approach for data analysis; the resultant compressional and shear velocities as well as the bulk and shear moduli for bcc-$Fe_{90}Ni_{10}$ are compared with those for pure iron to evaluate the effects of nickel content on the elastic properties of Fe-Ni alloys.

2. Experimental methods

The polycrystalline sample of $Fe_{90}Ni_{10}$ (10 wt.% Nickel) was a cylindrical disk cut from a rod purchased from Princeton Scientific Cooperation. Before the ultrasonic measurements, the sample was annealed at 3 GPa, 773 K. Scanning electron microscope (SEM) analysis was conducted on the recovered sample and the results (Figure 2) indicated that the sample was homogenous with an average grain size less than 1 μm. There were no detectable oxygen observed in the Energy-dispersive X-ray spectroscopy (EDS), suggesting that no oxidation reactions occurred during the high temperature annealing process.

To optimize the acoustic signals in the ultrasonic measurement, both sides of the sample were polished using diamond lapping film to 1 μm. The final dimensions of the polished sample were 0.930(2) mm in length and 2.010(2) mm in diameter, with a bulk density of 7.95(3) g/cm^3, as obtained by the Archimedes' method.

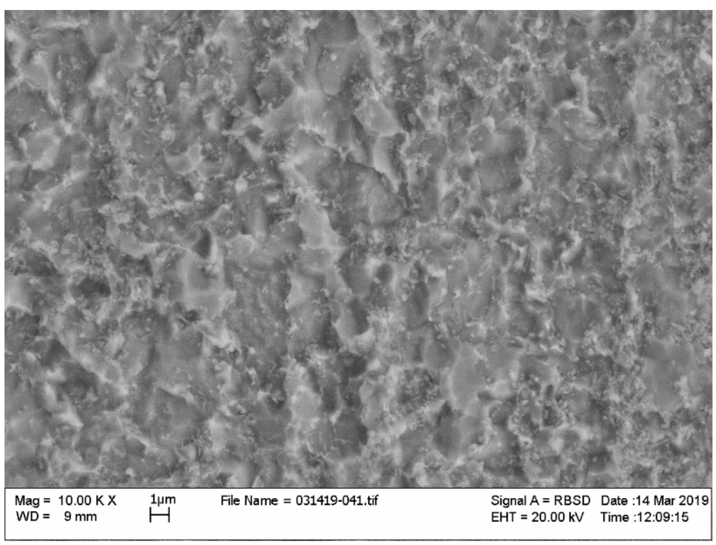

Figure 2 Scanning electron microscope image of sample after high temperature annealing.

High pressure and high temperature ultrasonic measurements were performed to about 8 GPa, 773 K in a 2000-ton uniaxial split-cylinder apparatus (USCA-2000) in the High-Pressure Lab at Stony Brook University. A sketch of the 14/8 cell assembly used in this study is shown in Figure 3. A dual mode $LiNbO_3$ transducer (10º Y-cut) was used to generate and receive both the compressional (P) wave and shear (S) wave simultaneously (50 MHz resonant frequency for P waves and 30 MHz for S waves). A dense alumina rod was placed on the top of the sample and served as the acoustic buffer rod. Due to the low yield strength of NaCl at high temperatures, a disk of NaCl was placed at the back of the sample to provide a pseudo-hydrostatic environment during the experiment. The high temperature environment was generated by a graphite heater and monitored by W/Re3%-W/Re26% type-C thermocouples, with the junction immediately next to the sample near the center of the cell. The temperature measurement uncertainty in current experiment is approximately ±10 K. P and S wave travel times were acquired using the transfer function technique and analyzed using the pulse echo overlap (PEO) method by overlapping the buffer rod and sample echoes (Fig. 4). Details about the transfer function technique for data acquisition and processing have been discussed elsewhere [*B Li et al.*, 2004; *B Li et al.*, 2002]. Cell pressures were calculated from the shear wave travel times of the alumina buffer rod using the pressure scale at high temperature by equation:

$$\mathrm{P} = 242.5(9) \times (1 - t_S/t_{S0}) + 0.01099(5) \times (T - T_0)\#(1.)$$

where P is cell pressure, t_S is the S wave travel time of the buffer rod, and t_{S0} is the S wave travel time at ambient conditions [for further details of the use of alumina as a pressure marker, see *Wang et al.*, 2015]. The pressure uncertainty is estimated to be around ±0.2 GPa in current study.

The experimental P-T path is shown in the Figure 1, superimposed with the previously determined phase diagram from the diamond anvil cell [*Huang et al.*, 1988; *Huang et al.*, 1992]. The sample was first compressed at room temperature to a maximum pressure of ~8 GPa, followed by heating to a peak temperature of 773 K to release the deviatoric stress in the cell, then the ultrasonic data were collected at 100 K intervals along cooling paths to room temperature while the sample was under nearly hydrostatic environment [*B Li et al.*, 2001]. Multiple heating and cooling cycles were performed during decompression to provide a dense coverage of experimental data in P-T space. The P and S wave travel times were obtained at 35 and 27 MHz, respectively, in this study, to maximize the signal-to-noise ratio.

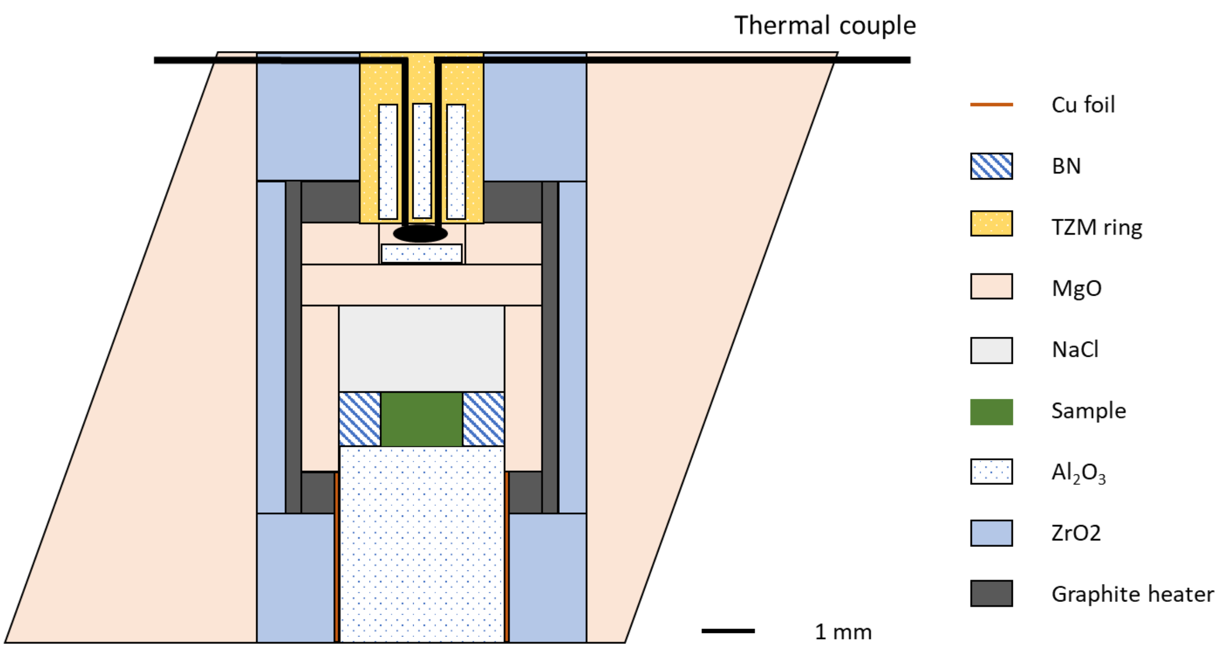

Figure 3 Sketch of the cell assembly used in current study for ultrasonic measurement.

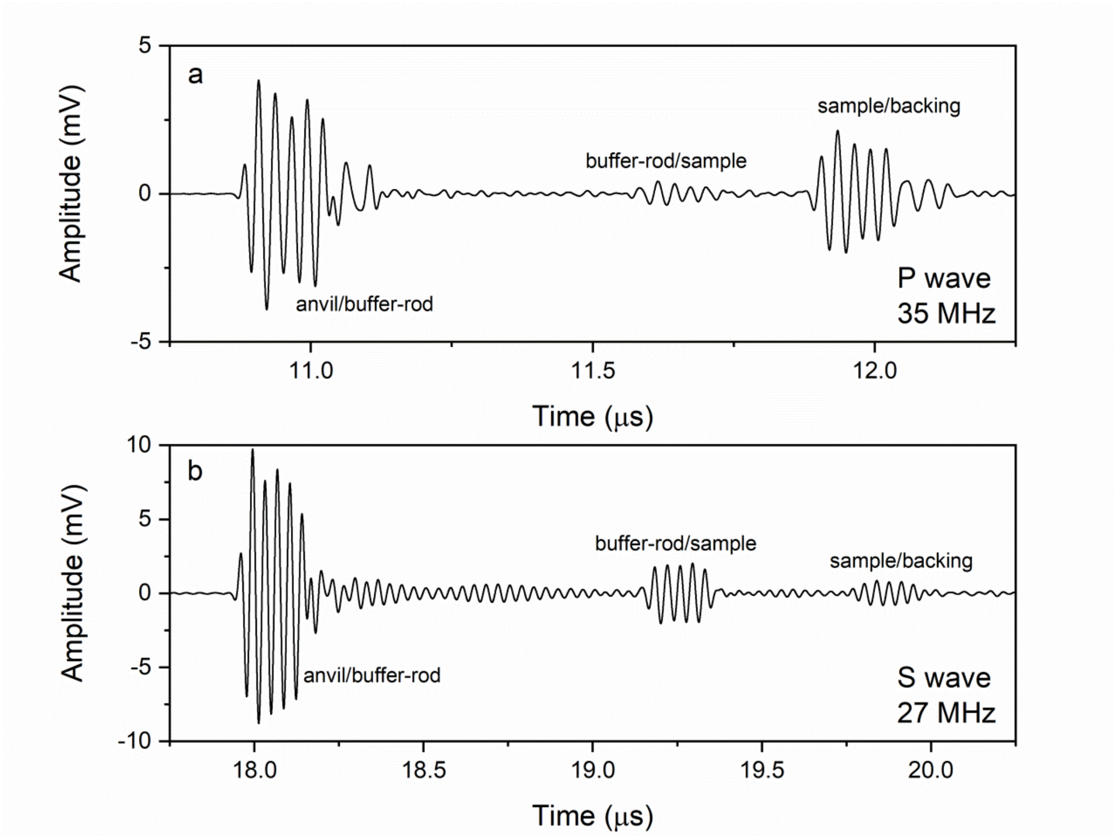

Figure 4 Signal example of (a) P wave and (b) S wave obtained at 7.6 GPa and 776 K.

3. Data analyses

After the experiment, the sample length and diameter were 0.926(2) mm and 2.010(5) mm respectively, indicating that, within 0.4% uncertainty, the sample can be considered to have undergone elastic compression under pseudo-hydrostatic conditions during the entire course of the experiment. As indicated by the P and S wave signals obtained at 7.6 GPa and 776 K in Figure 4, the reflections from the front (buffer rod/sample) and rear (sample/backing) surfaces are highly distinguishable from the background, providing a reliable measurements of travel times with 0.1-0.6% in precision.

The travel time results at all experimental conditions from this study are summarized in **Table 1**. As shown in previous studies, velocities (V_P and V_S), and elastic moduli (K_S and G) as well as their pressure and temperature derivatives (K_S, G, K_S', G', $\partial K_S/\partial T$, and $\partial G/\partial T$) can be obtained by a third-order finite strain approach [*Davies and Dziewonski*, 1975; *B Li and Zhang*, 2005].

First, because the sample has undergone nearly hydrostatic deformation during cooling along decompression the experiment, it is reasonable to assume that density (ρ), volume (V), and length (*l*) have the following relationships:

$$\frac{\rho}{\rho_0} = \frac{V_0}{V} = \left(\frac{l_0}{l}\right)^3 \#(2.)$$

The elastic properties at high pressure and temperature can be calculated through the sound velocities $V_{(P,S)} = \frac{2l}{2t_{(P,S)}}$ and densities ρ by the relationships $K_S = \rho(V_P^2 - \frac{4}{3}V_S^2)$ and $G = \rho V_S^2$ for the bulk and shear modulus, respectively. Under adiabatic compression, the finite strain equations are expressed as the following:

$$\rho V_P^2 = (1-2\varepsilon)^{\frac{5}{2}}(L_1 + L_2\varepsilon) \#(3.)$$

$$\rho V_S^2 = (1-2\varepsilon)^{\frac{5}{2}}(M_1 + M_2\varepsilon) \#(4.)$$

$$K_{S(0,T)} = L_1 - \frac{4}{3}M_1 \#(5.)$$

$$G_{(0,T)} = M_1 \#(6.)$$

$$K'_{S(0,T)} = \frac{5L_1 - L_2}{3K_{S(0,T)}} - \frac{4G'_{(0,T)}}{3} \#(7.)$$

$$G'_{(0,T)} = \frac{5M_1 - M_2}{3K_{S(0,T)}} \#(8.)$$

where the subscript (P,T) indicates values at the pressure P and adiabatic foot temperature T, and the Eulerian strain $\varepsilon = \frac{1}{2}\left[1 - \left(\frac{\rho_{(0,T)}}{\rho_{(0,T_0)}}\right)^{2/3}\right]$. All temperatures reached in the entire experiment are assumed to be raised along separate adiabats from different foot temperatures T_0. Thus, the adiabatic foot temperature for each data point as well as the corresponding density and elastic properties can be extracted through the following equations:

$$\left(\frac{\partial T}{\partial P}\right)_S = \frac{\gamma T}{K_S} \#(9.)$$

$$\rho_{(0,T)} = \rho_{(0,T_0)} e^{-\int \alpha dT} \#(10.)$$

$$K_{S(0,T)} = K_{S(0,T_0)} + (T - T_0)\left(\frac{\partial K_S}{\partial T}\right)_P \#(11.)$$

$$G_{(0,T)} = G_{(0,T_0)} + (T - T_0)\left(\frac{\partial G}{\partial T}\right)_P \#(12.)$$

$$K'_{SS(0,T)} = K'_{SS(0,T_0)} + (T - T_0)\left(\frac{\partial^2 K_S}{\partial P \partial T}\right)_P + \left(\frac{\partial K_S}{\partial T}\right)_P \frac{\gamma T}{K_S} \#(13.)$$

$$G'_{S(0,T)} = G'_{S(0,T_0)} + (T - T_0)\left(\frac{\partial^2 G}{\partial P \partial T}\right)_P + \left(\frac{\partial G}{\partial T}\right)_P \frac{\gamma T}{K_S} \#(14.)$$

$$P = -(1 - 2\varepsilon)^{5/2}\left(3K_{S(0,T_0)}\varepsilon + \frac{1}{2}(36K_{S(0,T_0)} - 9K_{S(0,T_0)}K'_{SS(0,T_0)})\varepsilon^2\right) \#(15.)$$

The sample lengths, as well as the thermoelastic properties K_S, G, K_S', G', $\partial K_S/\partial T$ $\partial G/\partial T$ at ambient conditions, were refined using a least-square fit by minimizing the difference between the observed compressional and shear velocities ($V_{(P,S)} = \frac{2l}{2t_{(P,S)}}$) and pressures ($P$ from [eq.(1)] with those calculated by finite strain theory [eqs. (3), (4), and (15)]. More details about the data analysis procedures can be found elsewhere [*B Li and Zhang*, 2005]. In the *P*-*T* range of the current experiment, the thermal expansivity α was assumed to be a constant value of 4.67×10^{-5} [*Zhang and Guyot*, 1999]; the Grüneisen parameter γ was constrained by the assumption of $\rho\gamma$ = constant with $\gamma_0 = 1.65$ [*Poirier*, 2000; *Quareni and Mulargia*, 1988]; cross derivatives $(\partial^2 K_S/\partial P\partial T)_P$ and $(\partial^2 G/\partial P\partial T)_P$ were assumed to be zero in the current *P*-*T* range. The minimization usually takes only a few iterations to achieve convergence, and the results for the elastic properties are shown in **Table 2**.

Table 1 *Experimental data of bcc-$Fe_{90}Ni_{10}$*

P (GPa)	T (K)	$2t_P$ (μs)	$2t_S$ (μs)	Length (mm)	ρ (g/cm^3)	V_P (km/s)	V_S (km/s)	K_S (GPa)	G (GPa)
6.2	323	0.3080(2)	0.5816(28)	0.9148	8.246	5.94(6)	3.15(3)	182.2(33)	81.6(11)
7.6	776	0.3188(4)	0.6188(18)	0.9174	8.176	5.76(6)	2.97(3)	175.0(30)	71.9(8)
7.4	667	0.3150(4)	0.6060(4)	0.9165	8.200	5.82(6)	3.02(3)	177.6(30)	75.0(8)
7.1	570	0.3124(4)	0.5968(8)	0.9160	8.212	5.86(6)	3.07(3)	179.2(31)	77.4(8)
6.7	472	0.3104(2)	0.5868(22)	0.9155	8.228	5.90(6)	3.12(3)	179.5(32)	80.1(10)
6.1	328	0.3080(4)	0.5810(28)	0.9149	8.243	5.94(6)	3.15(3)	181.9(34)	81.8(11)
5.6	322	0.3092(4)	0.5828(22)	0.9157	8.220	5.92(6)	3.14(3)	180.2(33)	81.2(10)
6.9	785	0.3210(12)	0.6208(16)	0.9188	8.140	5.72(6)	2.96(3)	171.6(35)	71.3(8)
6.8	674	0.3172(6)	0.6096(8)	0.9177	8.167	5.79(6)	3.01(3)	174.7(31)	74.0(8)
6.6	576	0.3146(8)	0.5990(10)	0.9170	8.186	5.83(6)	3.06(3)	175.9(33)	76.7(8)
6.2	470	0.3120(2)	0.5900(10)	0.9165	8.201	5.87(6)	3.11(3)	177.5(31)	79.1(8)
5.6	329	0.3098(2)	0.5826(28)	0.9158	8.219	5.91(6)	3.14(3)	179.0(33)	81.2(11)
4.3	309	0.3140(4)	0.5864(12)	0.9178	8.165	5.85(6)	3.13(3)	172.4(31)	80.0(9)
5.6	782	0.3256(2)	0.6244(6)	0.9213	8.073	5.66(6)	2.95(3)	164.8(28)	70.3(7)
5.4	661	0.3214(2)	0.6130(4)	0.9201	8.103	5.73(6)	3.00(3)	168.3(29)	73.0(7)
5.2	573	0.3192(2)	0.6050(2)	0.9195	8.121	5.76(6)	3.04(3)	169.5(29)	75.0(8)
4.9	468	0.3172(2)	0.5984(2)	0.9188	8.139	5.79(6)	3.07(3)	170.8(29)	76.7(8)
4.3	316	0.3150(2)	0.5878(10)	0.9181	8.158	5.83(6)	3.12(3)	171.0(30)	79.6(8)
2.6	310	0.3206(4)	0.6010(18)	0.9211	8.077	5.75(6)	3.07(3)	165.5(30)	75.9(9)
3.1	779	0.3336(2)	0.6406(8)	0.9260	7.950	5.55(6)	2.89(3)	156.4(26)	66.4(7)
2.9	673	0.3300(2)	0.6268(20)	0.9251	7.974	5.61(6)	2.95(3)	158.0(28)	69.5(8)
2.8	571	0.3266(4)	0.6188(26)	0.9240	8.002	5.66(6)	2.99(3)	161.1(29)	71.4(9)
2.7	474	0.3244(4)	0.6134(38)	0.9229	8.030	5.69(6)	3.01(3)	163.0(31)	72.7(12)
2.3	318	0.3218(2)	0.6028(14)	0.9217	8.062	5.73(6)	3.06(3)	164.0(29)	75.4(8)
3.2	776	0.3334(2)	0.6352(16)	0.9259	7.952	5.55(6)	2.92(3)	155.2(27)	67.6(8)
3.0	674	0.3302(2)	0.6244(4)	0.9249	7.978	5.60(6)	2.96(3)	157.0(27)	70.0(7)
2.8	571	0.3270(2)	0.6188(22)	0.9239	8.003	5.65(6)	2.99(3)	160.4(28)	71.4(9)
2.8	473	0.3246(2)	0.6114(4)	0.9229	8.031	5.69(6)	3.02(3)	162.1(28)	73.2(7)
2.5	343	0.3226(2)	0.6050(10)	0.9218	8.060	5.71(6)	3.05(3)	163.4(29)	74.8(8)

Notes: The uncertainty of length calculated in this study is approximately ±1%.

Table 2 *Comparison of thermoelastic properties of Fe and Fe-Ni alloys*

Reference			P range	T range	K_{S0}	$\partial K_{S0}/\partial P$	$\partial K_{S0}/\partial T$	G_0	$\partial G_0/\partial P$	$\partial G_0/\partial T$	K_{T0}	$\partial K_{T0}/\partial P$	$\partial K_{T0}/\partial T$	EOS	Notes
			(GPa)	(K)	(GPa)		(GPa/K)	(GPa)		(GPa/K)	(GPa)		(GPa/K)		
This study	bcc	$Fe_{90}Ni_{10}$	~8	~773	154.2(8)	4.6(2)	-0.028(1)	73.2(2)	1.5(1)	-0.023(1)	–	–	–	3rd Finite strain	Ultrasonic, adiabatic
				~673	153.8(7)	4.7(2)	-0.027(2)	72.7(2)	1.6(1)	-0.022(1)	–	–	–		
Shibazaki et al. (2016)	bcc	Fe	~6.3	~800	163.2(15)	6.75(33)	–0.038(3)	81.4(6)	1.66(14)	–0.029(1)	–	–	–	Polynomial	Ultrasonic, adiabatic
Adams et al. (2006)	bcc	Fe	0	3-500	166.2	–	-0.029	81.5	–	-0.025	–	–	–	–	Ultrasonic, adiabatic
Isaak and Masuda (1995)	bcc	Fe	0	~800	165.7	–	-0.046	82	–	-0.034	–	–	–	–	Ultrasonic, adiabatic
Dever (1972)	bcc	Fe	0	~773	167.8	–	-0.035	82	–	-0.029	–	–	–	–	Ultrasonic, adiabatic
Zhang and Guyot (1999)	bcc	Fe	~9	~773	–	–	–	–	–	–	155(2)	5.3*	–0.049(6)	3rd Birch Murnaghan	XRD, isothermal
Huang et al. (1987)	bcc	Fe	~12	~723	–	–	–	–	–	–	171(8)	4*	-0.010(16)	Birch Murnaghan	XRD, isothermal
Takahashi et al. (1968)	bcc	Fe	~15	300	–	–	–	–	–	–	162(5)	5.5(8)	–	Murnaghan	XRD, isothermal
	bcc	$Fe_{95}Ni_5$	~16	300	–	–	–	–	–	–	155(10)	4.2(8)	–		
	bcc	$Fe_{90}Ni_{10}$	~17	300	–	–	–	–	–	–	155(10)	5.7(8)	–		
Morrison et al. (2018)	bcc	$Fe_{91}Ni_{09}$	~15	300	–	–	–	–	–	–	146.8(31)	6.39(64)	–	3rd Birch Murnaghan	XRD, isothermal
Kantor et al. (2007)	fcc	$Fe_{78}Ni_{22}$	~72	300	–	–	–	–	–	–	161(1)	4.97(1)	–	3rd Birch Murnaghan	XRD, isothermal
Huang et al. (1992)	fcc	$Fe_{70}Ni_{30}$	~48	300	–	–	–	–	–	–	160(15)	4*	–	Birch Murnaghan	XRD, isothermal

Notes:
*: fixed value

WHY dK/dT from Zhang and Guyot (1999) and Huang (1987) are not included here?

4. Results and discussion

According to Figure 1, some of our experimental data were collected close to the bcc-fcc boundary or within the stability filed of the fcc phase of Fe90Ni10. A close examination of the recorded waveforms as well as the subsequent analysis of P and S wave travel times did not suggest a phase transition to fcc phase at these conditions. This was further tested by performing a separate fit without the data at 773 K. As indicated by the results shown in **Table 2**, within the uncertainty, the inclusion of the data at 773 K has an insignificant effect on the fitting results, and can be reliably treated as the representative values for the bcc phase.

The compressional and shear wave velocities data obtained in this study are compared in Figure 5 with those from ultrasonic measurements [*Shibazaki et al.*, 2016] and an IXS study on bcc-Fe [*Liu et al.*, 2014], as well as data from NRIXS studies on both bcc-Fe and bcc-$Fe_{91}Ni_{09}$

[*Morrison et al.*, 2019]. At room temperature, the velocities of both P and S waves for bcc-$Fe_{90}Ni_{10}$ are consistently lower than pure Fe from *Shibazaki et al.* [2016] by 5% and 6%, respectively, which is in good agreement with the 5-6% (Check?)velocity depression observed in NRIXS studies on Fe-Ni alloys with 0 and 9 at.% nickel [*Morrison et al.*, 2019]. However, the absolute values for both P and S waves from NRIXS are systematically lower than those from other techniques (UI, IXS), which could be attributed to the fact that NRIXS bases on the Debye model to analyze the data instead of measuring the sound velocity directly. Comparing to those for bcc-Fe from *Shibazaki et al.* [2016], V_P of bcc-$Fe_{90}Ni_{10}$ from current study exhibits a slower rate of increase with pressure [~5.1×10^{-2} km/s/GPa versus(*vs.*) 6.9×10^{-2} km/s/GPa for $Fe_{90}Ni_{10}$ and Fe, respectively] while V_S increases at a relatively similar rate (~2.1×10^{-2} km/s/GPa *vs.* 2.0×10^{-2} km/s/GPa for $Fe_{90}Ni_{10}$ and Fe, respectively).

With increasing temperature, both V_P and V_S decrease within the entire *P-T* range of the current experiment with a larger reduction in V_S than V_P. For example, at ~3 GPa, the depression in V_P and V_S from 300 K to 800 K are 4%, 7% for $Fe_{90}Ni_{10}$ and 6%, 8% for pure Fe, respectively. The compressional velocity (V_P) decrease from 300 to 700 K reported by *Liu et al.* [2014] is about 5%, which is larger than the 4% for bcc-$Fe_{90}Ni_{10}$ observed in current study. In addition, nonlinear elastic anomalies indicative of a magnetic transition at high temperature [e.g., *Dever*, 1972] were not observed in the current study, which could possibly be explained by the low temperature range (300-773 K) relative to the Currie temperature (Tc = ????) for FiNi10% .

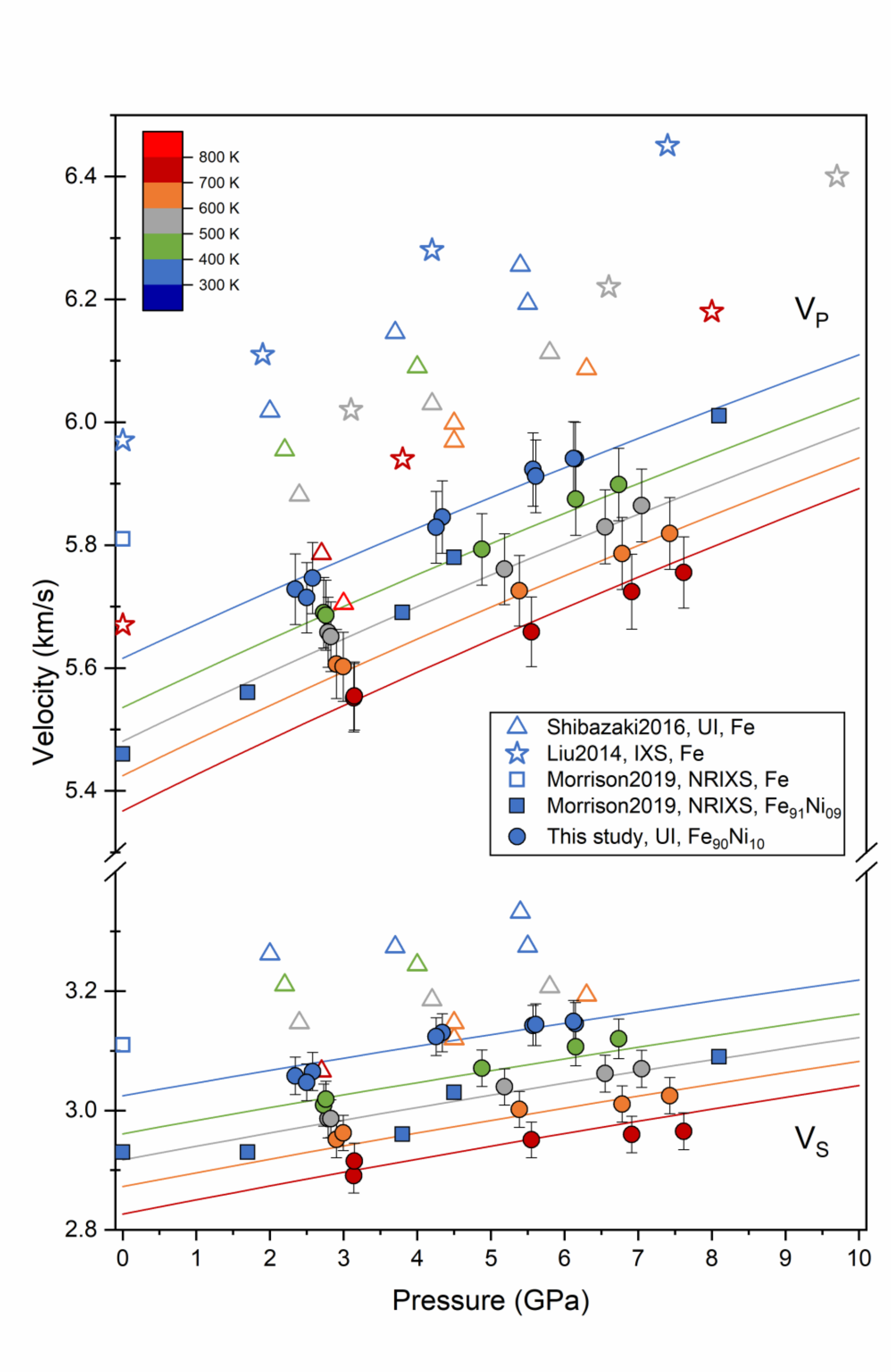

Figure 5 Compressional (V_P) and shear (V_S) wave velocities as a function of pressure and temperature. Solid lines are the finite strain fitted curves from this study. Temperature information are color coded and shown in legend.

The adiabatic bulk modulus (K_S) and shear modulus (G) calculated in current study are plotted in Figure 6 as a function of pressure and temperature. **Table 2** is a comparison of the thermoelastic properties of Fe and Fe-Ni alloys obtained from this study and previous experimental studies. Note in this table that our study reports for the first time the temperature dependences of the elastic bulk and shear moduli ($\partial K_S/\partial T$ and $\partial G/\partial T$) of bcc-$Fe_{90}Ni_{10}$. Comparing with previous ultrasonic studies listed in **Table 2**, the bulk and shear moduli of bcc-$Fe_{90}Ni_{10}$ at ambient conditions [K_{S0} = 154.2(8) GPa and G_0 = 73.2(2) GPa] are lower than those values of bcc-Fe (K_{S0} = 165-168 GPa and G_0 = 81-82 GPa) by approximately 6% and 11%, respectively [*Adams et al.*, 2006; *Dever*, 1972; *Isaak and Masuda*, 1995; *Shibazaki et al.*, 2016]. The effect of nickel content on bulk modulus observed in this study is consistent with previous suggestions based on pressure-volume (*P*-*V*) measurements in diamond anvil cell [*Morrison et al.*, 2018; *Takahashi et al.*, 1968].

It is also worthwhile to note that, comparing with pure bcc-Fe (*Shibazaki et al.* [2016], bcc-$Fe_{90}Ni_{10}$ exhibits a weaker pressure dependence of K_S than bcc-Fe (Fig. 6), which can be quantified by the pressure derivative K_S' [4.6(2) for bcc-$Fe_{90}Ni_{10}$ vs. 6.75(33) for pure Fe]; meanwhile for the shear modulus, bcc-$Fe_{90}Ni_{10}$ and bcc-Fe show close agreement with each other in their pressure derivatives [G_0' = 1.5(1) vs. 1.66(14), respectively]. These comparisons are believed to reveal primarily the intrinsic difference resulted from nickel substitution in the alloy, the different pressure calibration method used in the current experiments (alumina pressure gauge) and those of *Shibazaki et al.* [2016] [equation of state (EOS) of MgO + hBN] may also contribute, but are not considered to be an appreciable effect. Future investigations on Fe-Ni alloys with different Ni contents using either of the pressure calibration method could help further address this issue.

Besides the pressure dependence, we also investigated the effect of 10 wt.% nickel content in our sample on the temperature dependence of both bulk and shear moduli. For pure bcc-Fe, high temperature ultrasonic measurements have been conducted at ambient pressure [e.g., *Adams et al.*, 2006; *Dever*, 1972; *Isaak and Masuda*, 1995] and high pressure [*Shibazaki et al.*, 2016], the reported temperature dependence ranges from -0.029 GPa/K ~ -0.046 GPa/K for the bulk modulus ($\partial K_{S0}/\partial T$) and -0.025 GPa/K ~ -0.034 GPa/K for the shear modulus ($\partial G_0/\partial T$). Our results of $\partial K_{S0}/\partial T$ = -0.028(1) GPa/K and $\partial G_0/\partial T$ = -0.023(1) GPa/K for bcc-$Fe_{90}Ni_{10}$ are only marginally consistent with the lowest values reported for bcc-Fe, indicating that 10 wt% nickel alloying with Fe can noticeably(?) affect the temperature dependence of both bulk and shear moduli.

The current adiabatic value of $\partial K_{S0}/\partial T$ = -0.028(1) GPa/K can be converted to its isothermal counterpart using the differentiated form of the thermodynamic identity $K_T = K_S/(1 + \alpha\gamma T)$, yielding $\partial K_{T0}/\partial T$ = -0.038(1) GPa/K. Two pressure-volume-temperature (*P*-*V*-*T*) investigations have reported $\partial K_{T0}/\partial T$ on bcc-Fe based on X-ray diffraction (XRD) studies and the results are in large discrepancy. While *Huang et al.* [1987] reported a relatively small temperature dependence [$\partial K_{T0}/\partial T$ = -0.010(16) GPa/K], *Zhang and Guyot* [1999] provided a much larger value [$\partial K_{T0}/\partial T$ = -0.049(6) GPa/K], which is more consistent with the current ultrasonic results. To the authors' best knowledge, no *P*-*V*-*T* investigation on bcc-Fe-Ni alloy has yet been reported to provide a direct comparison with the current result.

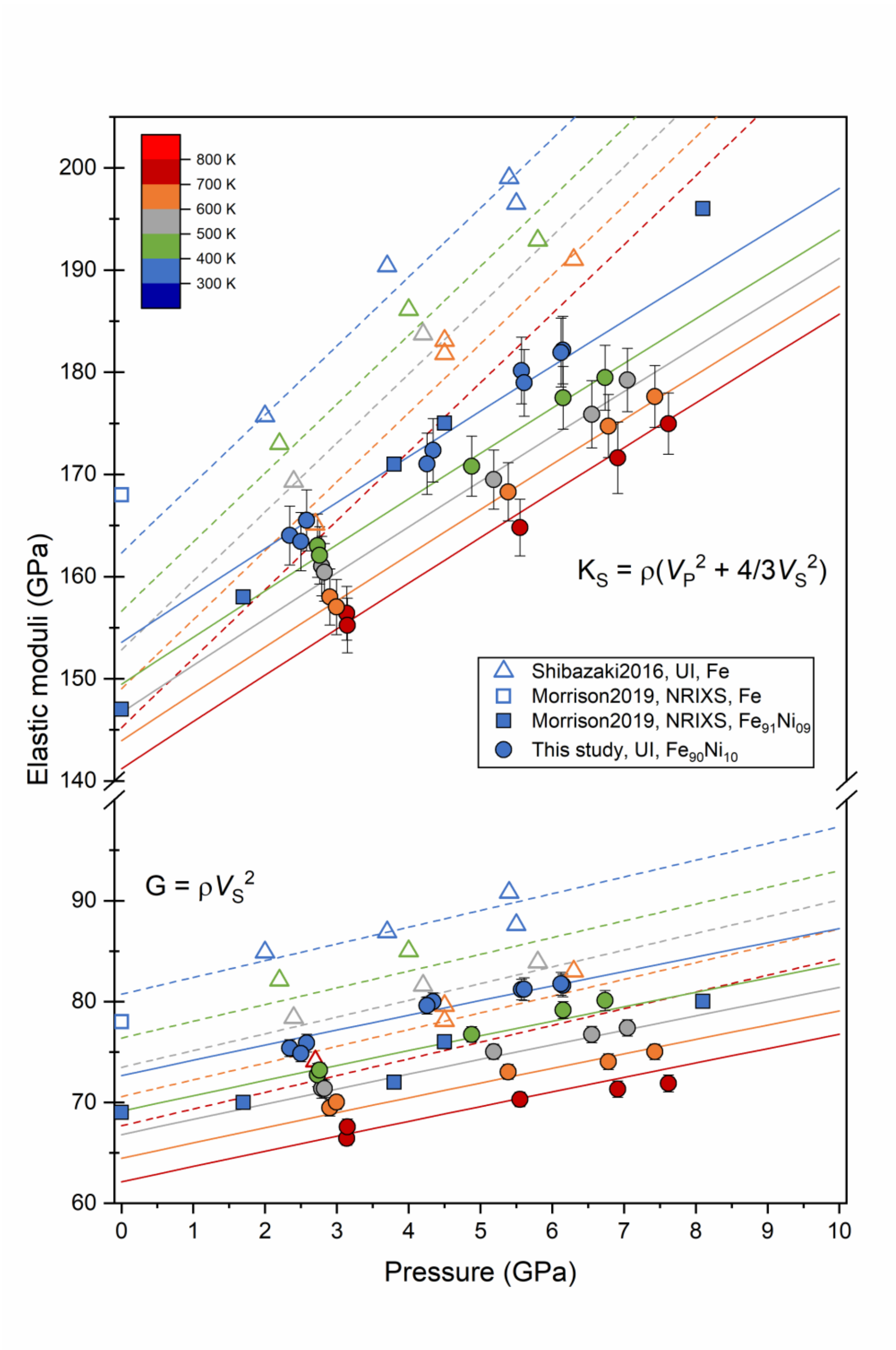

Figure 6 Bulk and shear modulus as a function of pressure and temperature. Solid lines are the finite strain fitted curves from this study. Dashed lines are calculated from Shibazaki et al. [2016]. Temperature information are color coded and shown in legend.

5. Implications

The effect of nickel content on P and S wave velocities observed on bcc phase in this study is roughly consistent with the previously reported results on hcp Fe-Ni alloys [*Lin et al.*, 2003; *Morrison et al.*, 2019; *Wakamatsu et al.*, 2018]. This effect could lead to a velocity decrease as large as ΔV_P = 0.8 km/s at inner core boundary (ICB), as calculated by *Ohtani et al.* [2013], suggesting that nickel content plays as an important factor when we modelling the velocity structure of Earth's core to place constraints on its composition. Moreover, our results suggest that the effect of temperature on the bulk and shear moduli of Fe-Ni alloy with 10 at% Ni is likely to be lower compared to that of Fe. This means the use of temperature derivatives of Fe could produce an upper bound or overestimated values for the Earth's core. In literature, *Lin et al.* [2005] suggested a temperature effect of $\partial V_P/\partial T$ = -0.35 km/(s·10^3K) and $\partial V_S/\partial T$ = -0.46 km/(s·10^3K) at constant density of ~10.25 g/cm^3 from their measurement up to 73 GPa and 1700 K. As it has been generally accepted that the thermal effect could be suppressed by pressure effect at core conditions, thus, we may find *Ohtani et al.* [2013] gave a smaller value of $\partial V_P/\partial T$ = -0.12 km/(s·10^3K) by combing their IXS measurement up to 1100 K with theoretical calculations up to 5500 K. From our measurements, we observed $\partial V_P/\partial$T = -0.18(2) km/(s·10^3K) for bcc-$Fe_{90}Ni_{10}$ at a constant density in current *P*-*T* range (see the velocity-density plot in Figure 7). For bcc-Fe, *Shibazaki et al.* [2016] reported $\partial V_P/\partial$T = -0.33(4) km/(s·10^3K) by UI and *Liu et al.* [2014] reported $\partial V_P/\partial$T = -0.37(3) km/(s·10^3K) by IXS in a similar *P*-*T* range. These values of bcc-Fe are in general agreement with each other but consistently larger than the value for bcc-$Fe_{90}Ni_{10}$, indicating the nickel content can reduce the effect of temperature on the V_P-ρ relationship. If we further analog the weaker temperature effect from nickel alloying to hcp phase, the $\partial V_P/\partial T$ at core conditions could be even smaller. Therefore, the nickel content caused

V_P depression at room temperature could be compensated by the weaker temperature effect at core temperature. Thus, the V_P of hcp-Fe-Ni could be higher than hcp-Fe at core conditions, which may require less V_P increase from alloying with light elements or even V_P decrease from alloying with light elements, depending on the core temperature.

Moreover, we also observed a slightly smaller temperature effect of shear velocity in bcc-$Fe_{90}Ni_{10}$ [$V_S/\partial T$ = -0.28(2) km/(s·10^3K)] than bcc-Fe [$\partial V_S/\partial T$ = -0.39(3) km/(s·10^3K)]. If this effect holds true to hcp phase, the low value of $\partial V_S/\partial T$ would introduce even larger V_S of hcp-Fe-Ni than previous estimations, which requires a more significant pre-melting effects [*Martorell et al.*, 2013] or other mechanism to account for the discrepancy of shear waves with the seismological observations. Thus, the nickel alloying induced decrease in the temperature effect needs to be further evaluated in other structures of Fe-Ni alloys in an expanded *P-T* range. Future acoustic measurements of both V_P and V_S of various Fe-light elements alloys and compounds at simultaneous high *P-T* conditions are also needed to provide a more comprehensive understanding of the composition and thermal structure of Earth's and planetary cores.

Shorten the Implication section? We can have more discussions.

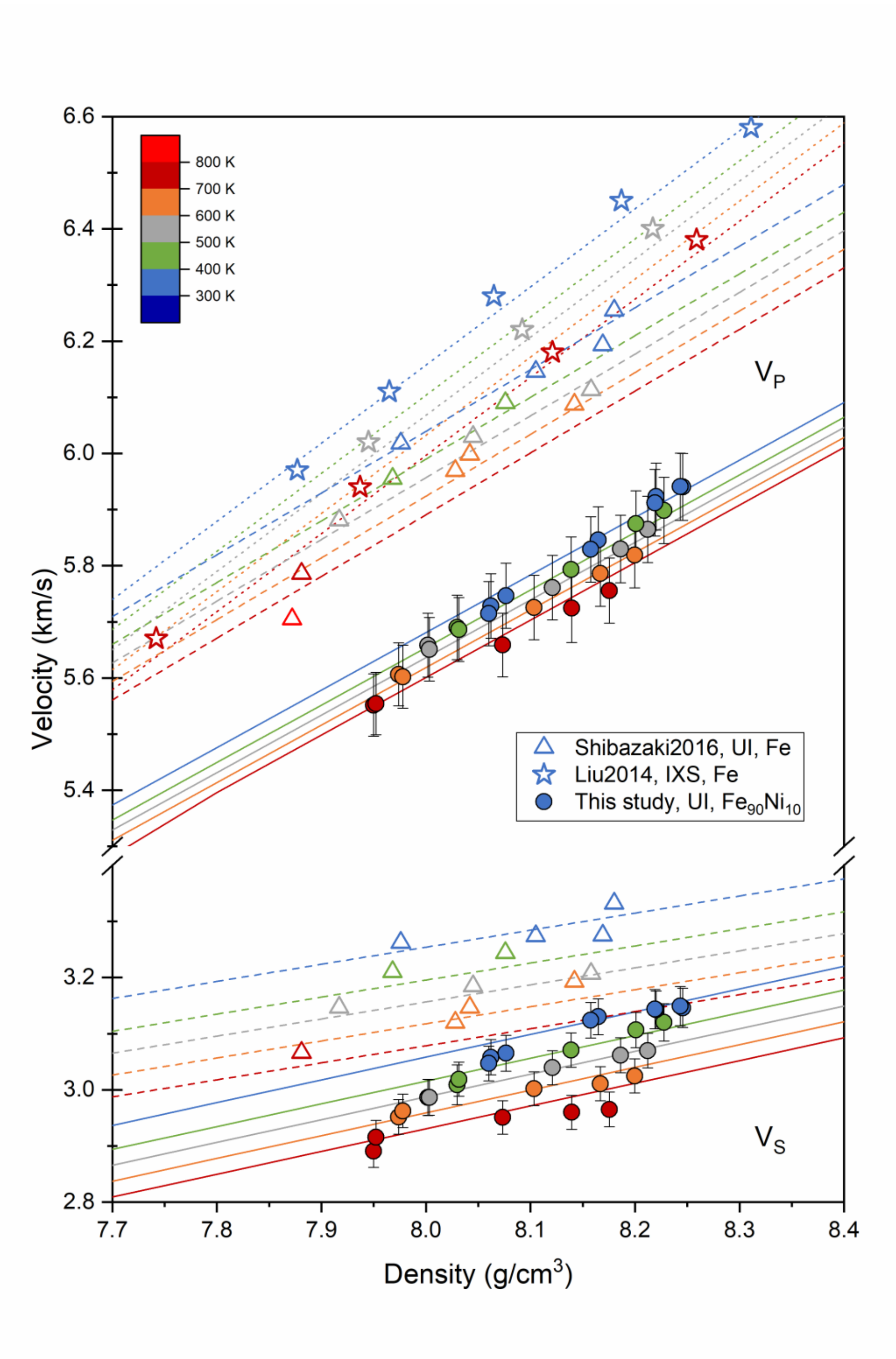

Figure 7 Compressional (V_P) and shear (V_S) wave velocities as a function of density and temperature. Solid lines are the linear curves calculated from this study. Dashed lines are calculated from Shibazaki et al. [2016]. Dot lines are calculated from Liu et al. [2014]. Temperature information are color coded and shown in legend.

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