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Supporting Information for

High-Temperature Equation of State of FeH: Implications for Hydrogen in Earth's Inner Core

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Additional Supporting Information

Dataset S1

Magnetic States of fcc FeH at High Pressure

Our first-principles calculations of fcc FeH show the local spin moments (within the muffin-tin sphere of the Fe site) of FM and LMD states as a function of volume at $T = 0$ K (Figure S2a). The local spin moment in the FM state is present down to 76 Bohr^3 (11.26 Å^3) and disappears abruptly below 74 Bohr^3 (10.97 Å^3). On the other hand, that in the LMD state decreases slowly with decreasing volume and is completely quenched at 78 Bohr^3 (11.56 Å^3). The total energy calculations demonstrate that the FM state is stable at ambient pressure (Figure S2b).

Their total energies of FM, LMD, and NM states were fitted to the Vinet EoS;

$$E(V) = \frac{2K_0V_0}{(K_0'-1)^2}\left\{2-\left[5+3\left(\frac{V}{V_0}\right)^{\frac{1}{3}}(K_0'-1)-3K_0'\right]\exp\left[-\frac{3}{2}(K_0'-1)\left[\left(\frac{V}{V_0}\right)^{\frac{1}{3}}-1\right]\right]\right\}+E_0, \qquad \text{(S1)}$$

where E is total energy, and K and K’ are bulk modulus and its pressure derivative, respectively. The subscript 0 denotes the value at zero pressure. For the FM state, fitting was made for the volume range of 76–120 Bohr^3, where the finite local spin moment exists. Similarly, the LMD state was fitted in the range of 80–120 Bohr^3. The obtained fitting parameters sets are V_0 = 13.55 Å^3, K_0 = 186.10 GPa, K_0’ = 4.54 for the FM state, V_0 = 13.22 Å^3, K_0 = 167.08 GPa, K_0’ = 4.04 for the LMD state, and V_0 = 12.51 Å^3, K_0 = 261.71 GPa, K_0’ = 4.08 for the NM state. With these parameters sets, we calculated pressure from the P-V relation of the same form as Eq. 1 in the main text. The FM-NM transition pressure is found to be 47 GPa at 0 K from the comparison of enthalpy ($E + PV$). The FM-PM transition temperature (Curie temperature) is also estimated by comparing the energies of the FM and LMD states as (Sato et al., 2003);

$$T_C = \frac{2}{3k_B}(E_{LMD} - E_{FM}), \qquad \text{(S2)}$$

where T_C is the Curie temperature, k_B is Boltzmann’s constant, and E_{LMD} and E_{FM} are the total energies of the LMD and FM states, respectively (Figure S2c). The obtained Curie temperature is about 1100 K at ambient pressure, which is comparable to the Curie temperature for bcc Fe (1043 K). However, that of fcc FeH rapidly decreases with compression, possibly suggesting changes in the magnetic state at 300 K from FM to PM with local spin moment, which does not contradict previous Mössbauer measurements (Narygina et al., 2011).

Table S1. Thermodynamic parameters for thermal EoSs for FeH and Fe

	fcc FeH	fcc FeH	dhcp FeH	fcc Fe
Ref.	This study	Sakamaki et al. (2009)		Tsujino et al. (2013)
EoS at T_0	Vinet	3rd Birch–Murnaghan	3rd Birch–Murnaghan	3rd Birch–Murnaghan
Magnetic state[a]	NM	FM + PM	FM + PM	FM + PM
T_0 (K)	300	1273	300	1273
$V_{0, T0}$ (Å^3)	13.45(15)	14.15	12.80	12.257
$K_{0, T0}$ (GPa)	183(20)	182	112	111.5
$K'_{0, T0}$	3.84(37)	4	5.4	5.2
n	2	–[b]	2	1
γ_0	0.738(40)	–[b]	1.98	2.28
γ_∞	0.547(83)	–	–	–
q	–	–[b]	-0.6	-0.21
Θ_0 (K)	758	–[b]	340	340
Pressure range	P > 41 GPa	$P < 21$ GPa	$P < 20$ GPa	–

[a]NM, non magnetic state; PM, paramagnetic state with local spin moment

[b]Effect of temperature on the EoS is described by thermal pressure $\Delta P_{th} = P_{T0} + \alpha K_{T0} \times (T - T_0)$, where $\alpha K_{T0} = 1.8 \times 10^{-2}$ GPa/K.

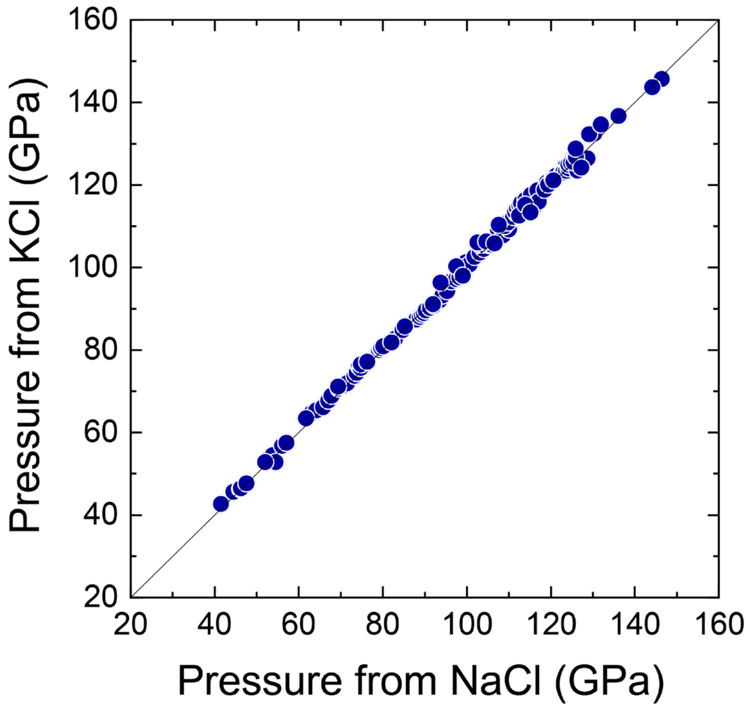

Figure S1. Comparison of pressures determined from an NaCl pressure marker (Dorogokpets & Dewaele, 2007) with those based on KCl (Tateno et al., 2019) in run #3.

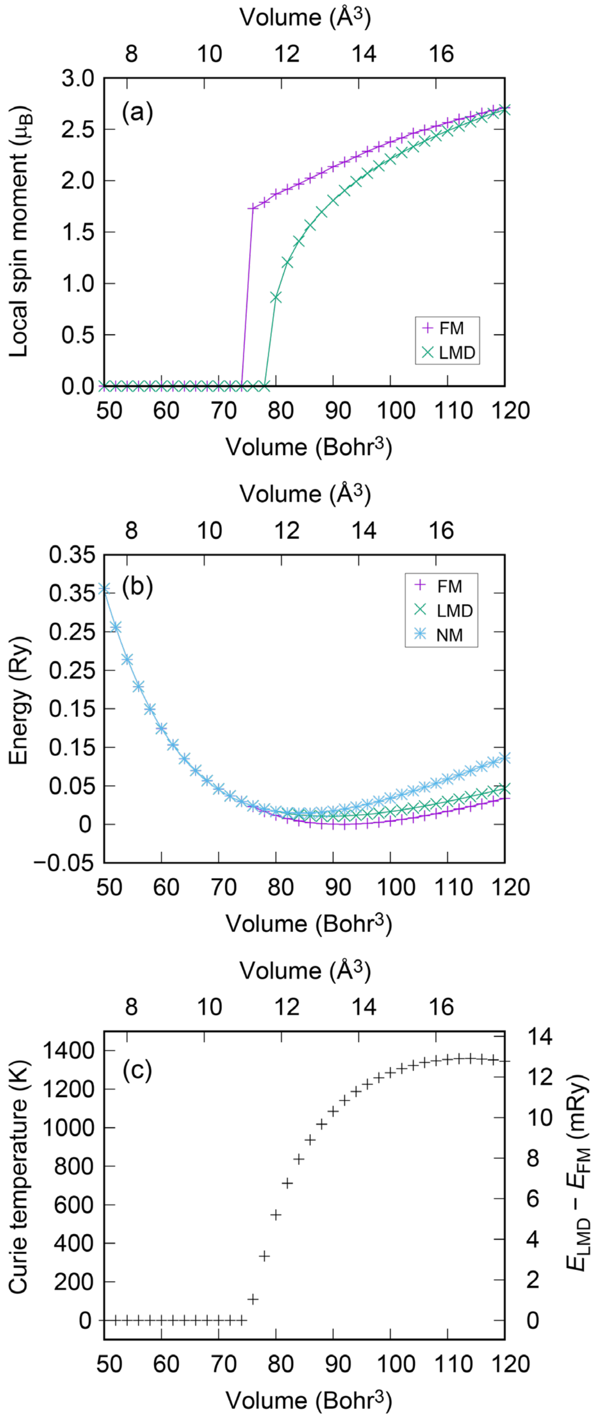

Figure S2. (a) Local spin moment of the FM (purple) and LMD (green) states of fcc FeH as a function of volume. Note that that of the LMD state is quenched at a volume larger than that for the FM state. (b) Total energy of the FM (purple), LMD (green), and NM (blue) states of fcc FeH as a function of volume. Symbols are first-principles results, and lines are fitting curves (see text). (c) Curie temperature of fcc FeH as a function of volume.

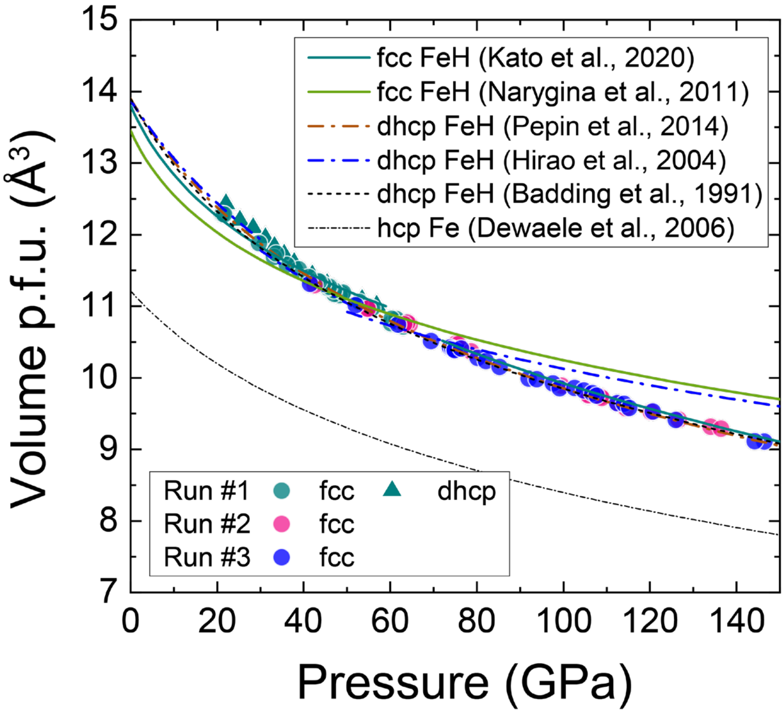

Figure S3. Comparison of the volume per formula unit of FeH obtained at 300 K in this study (circles, fcc; triangles, dhcp) with the compression curves for dhcp and fcc FeH reported by previous studies (Badding et al., 1991; Hirao et al., 2004; Narygina et al., 2011; Pépin et al., 2014; Kato et al., 2020). That of pure Fe is also shown (Dewaele et al., 2006).