

Figure S1. Spin configurations of (a) AFM-I and (b) AFM-II FeH_x drawn by the VESTA program (Momma and Izumi, 2011). Brown spheres with arrows indicate Fe atoms. Pink spheres are H atoms located at the octahedral interstitial sites. Red and blue vectors represent up and down spins, respectively.

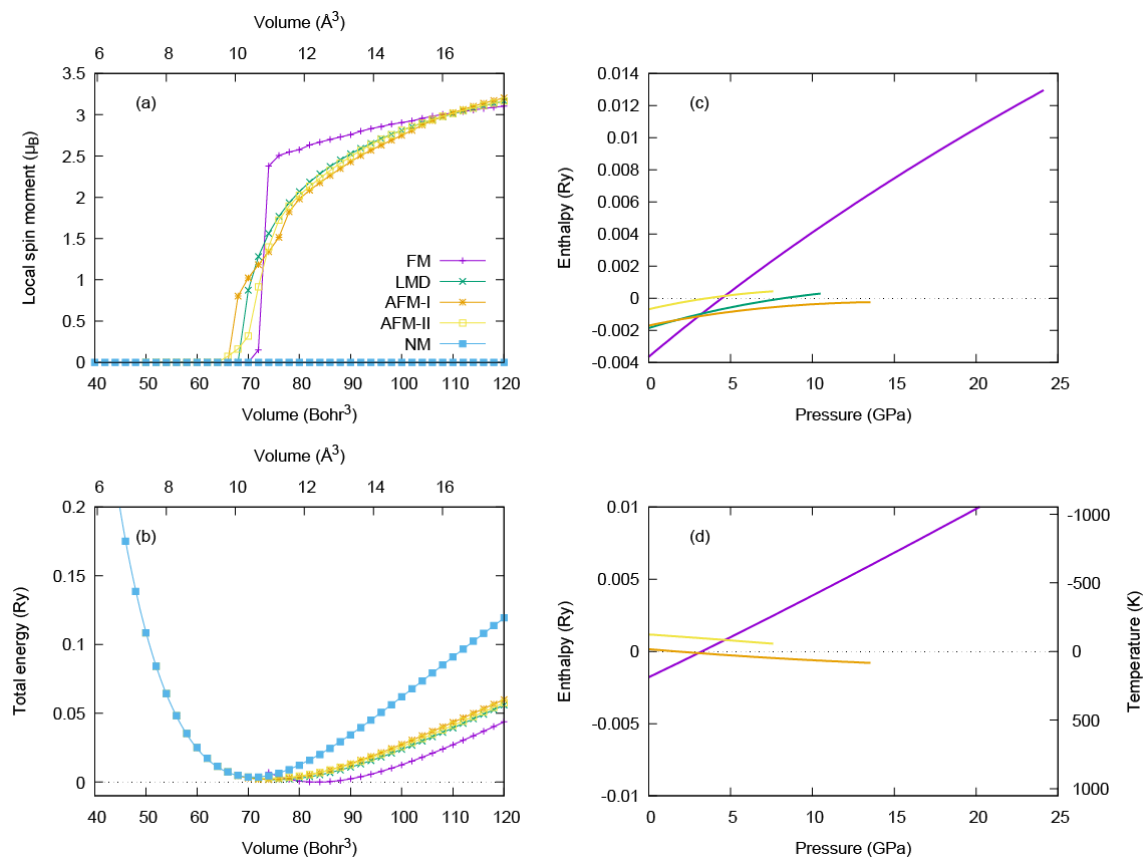


Figure S2. Calculation results of fcc Fe with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

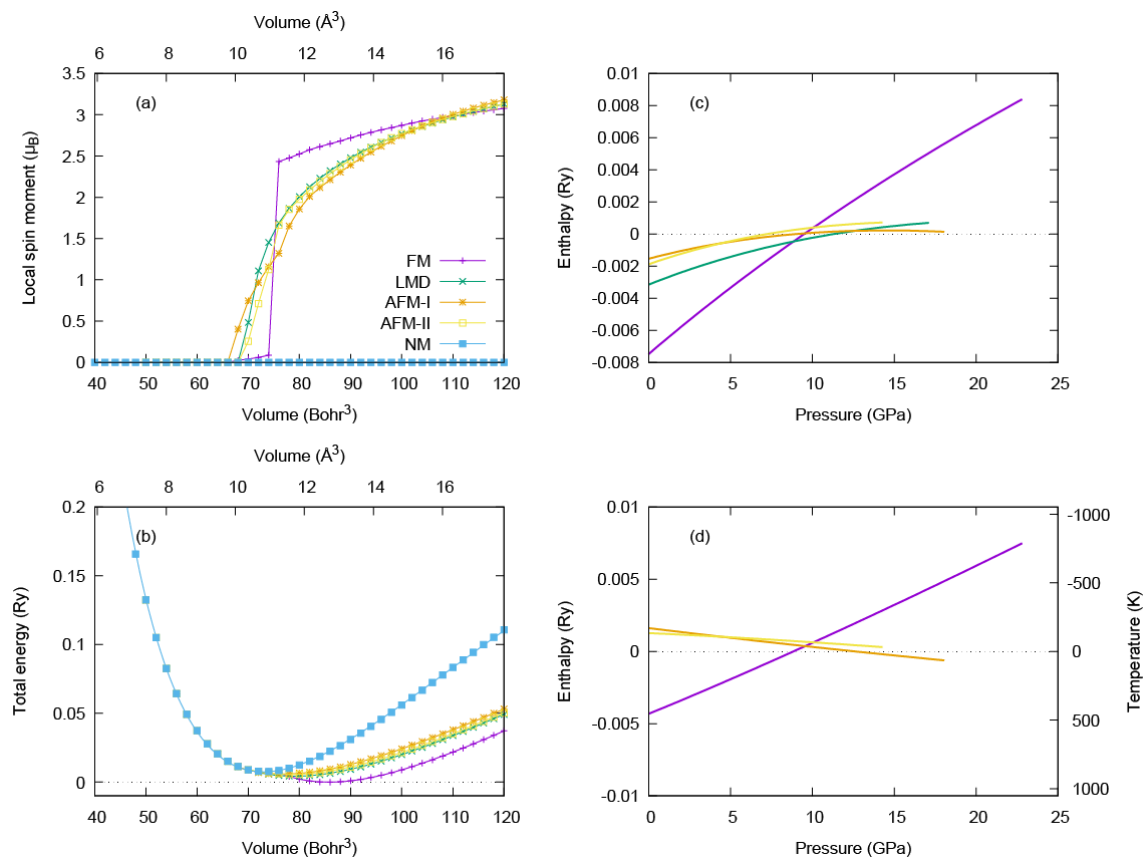


Figure S3. Calculation results of fcc FeH_{0.1} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

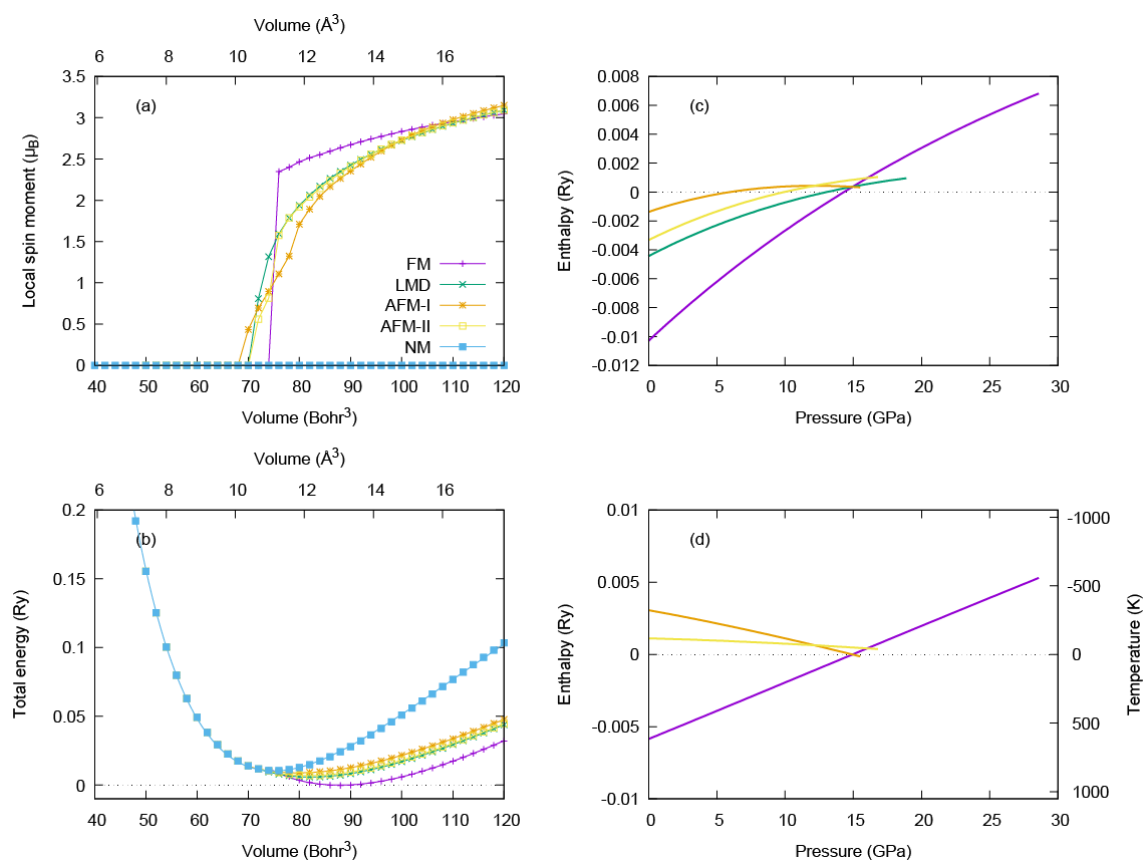


Figure S4. Calculation results of fcc FeH_{0.2} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

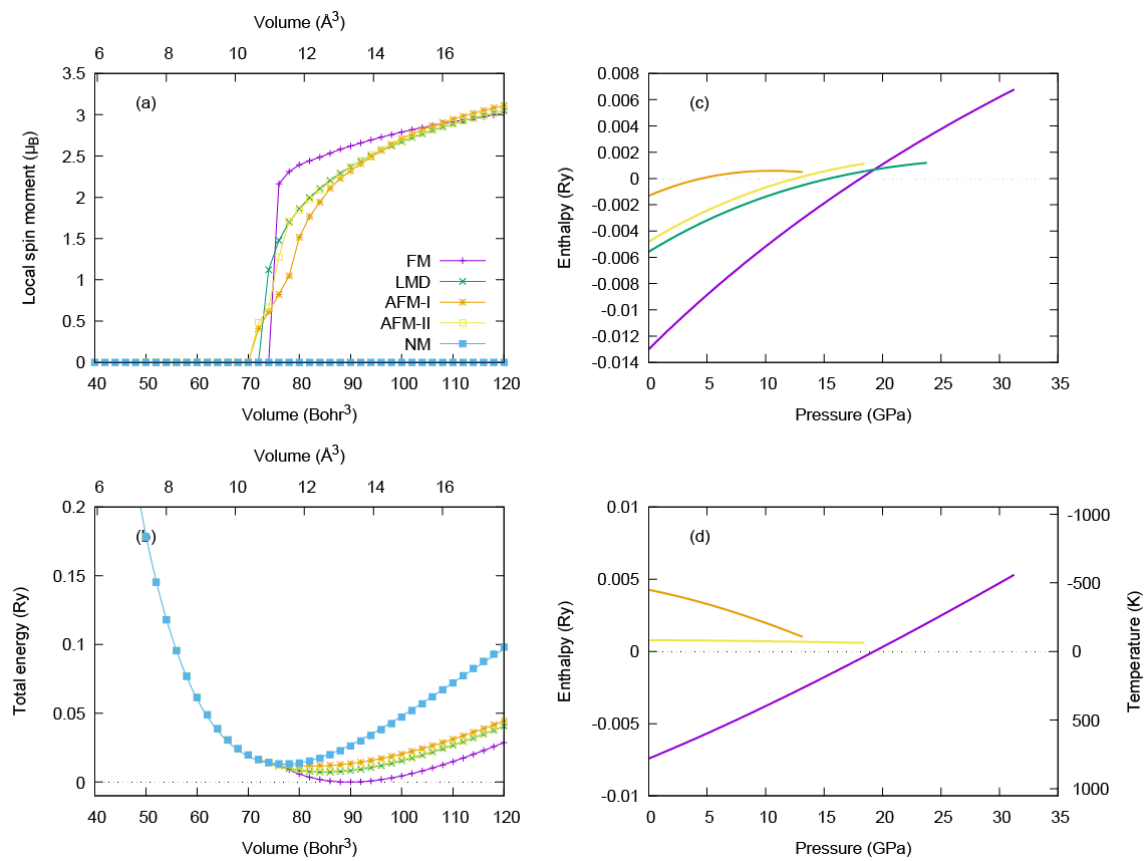


Figure S5. Calculation results of fcc FeH_{0.3} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

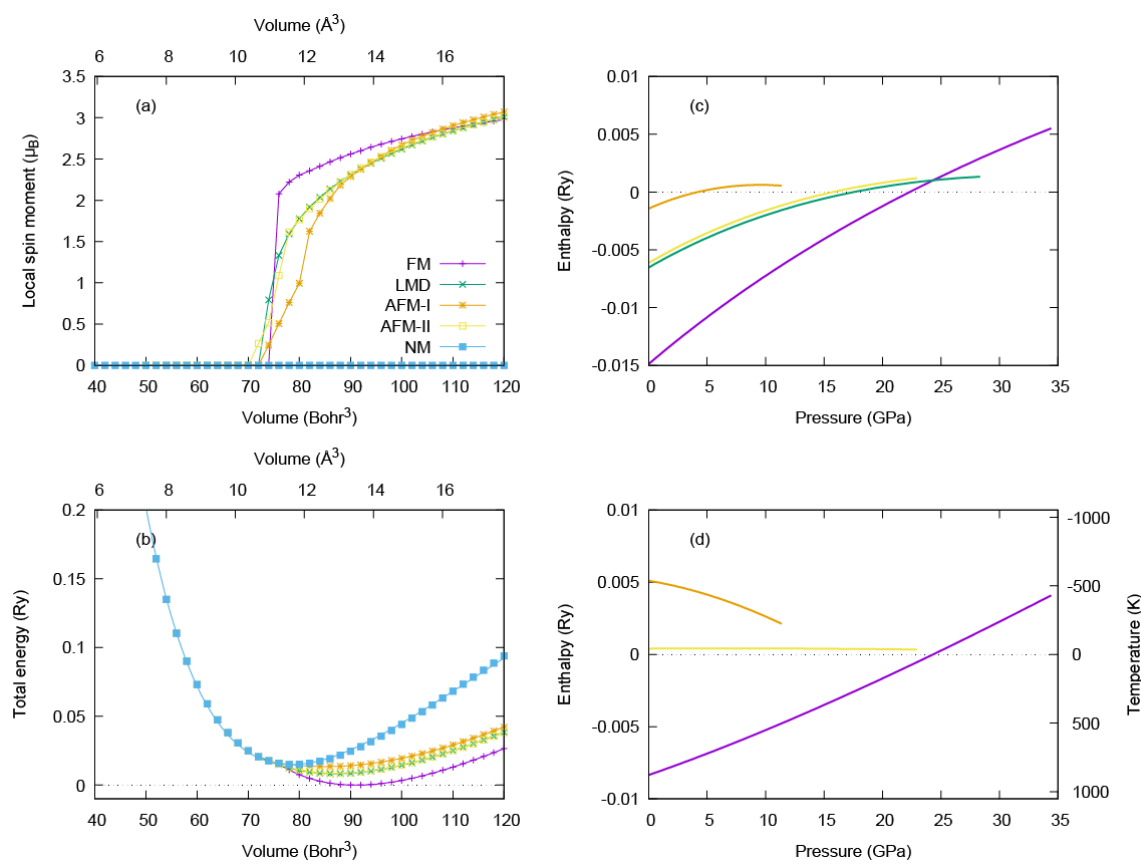


Figure S6. Calculation results of fcc FeH_{0.4} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

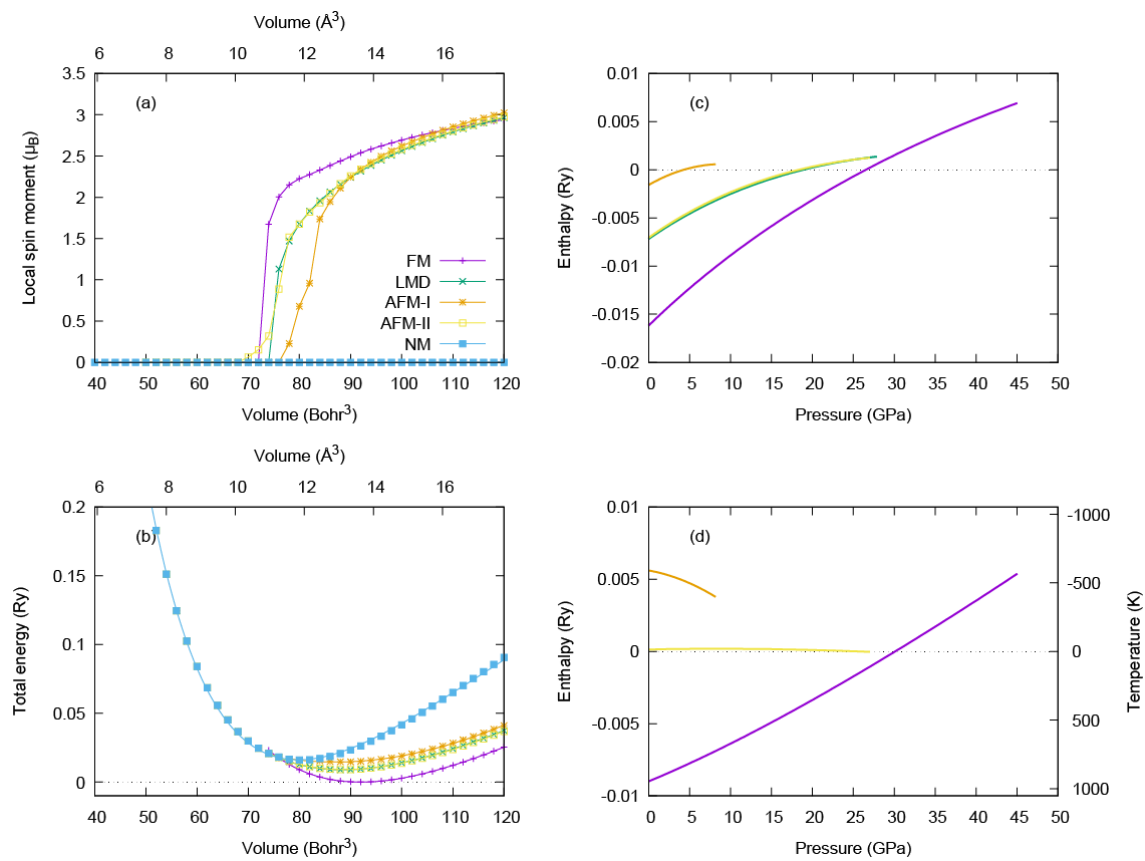


Figure S7. Calculation results of fcc FeH_{0.5} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

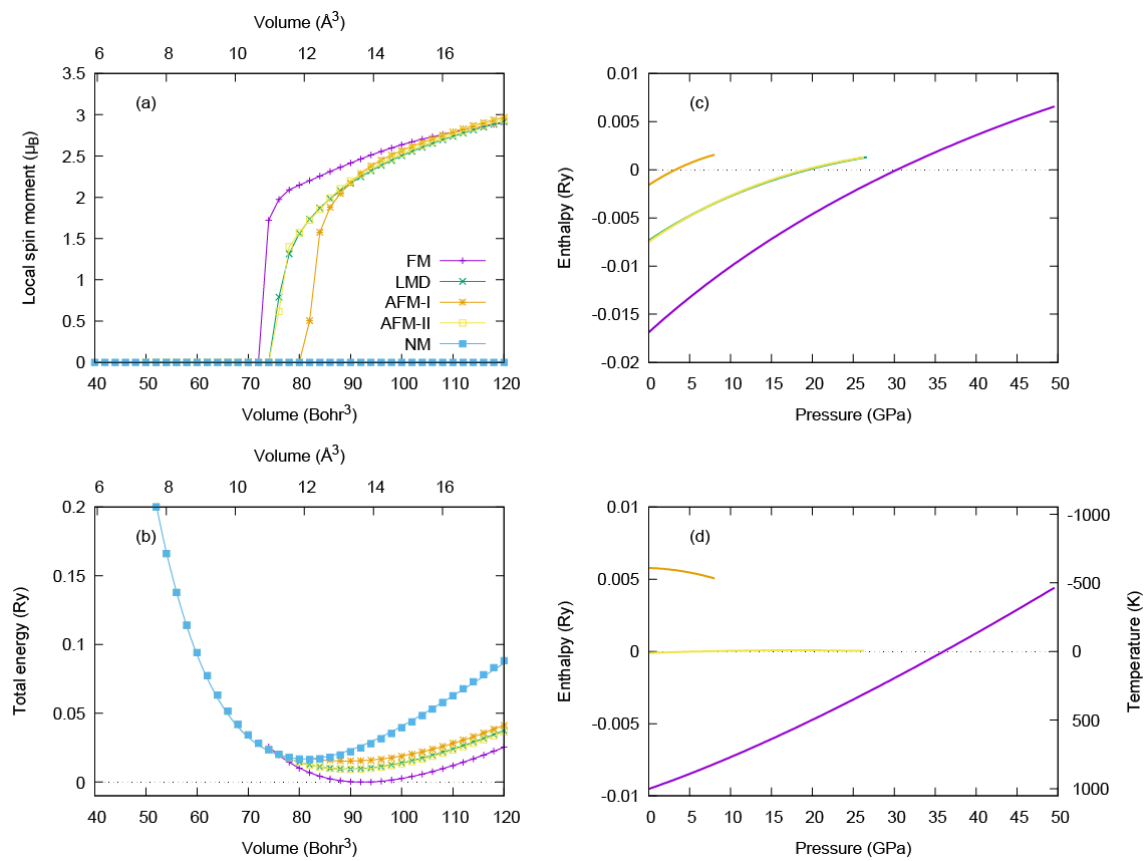


Figure S8. Calculation results of fcc FeH_{0.6} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

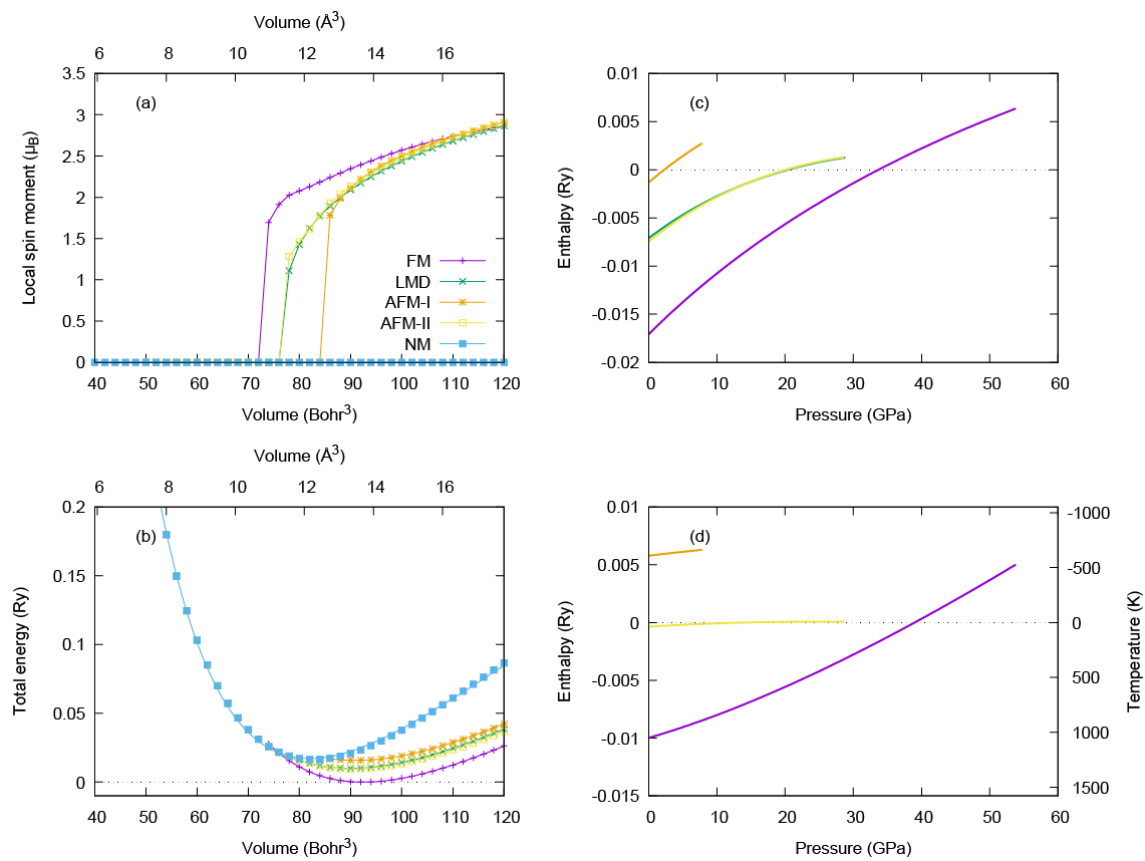


Figure S9. Calculation results of fcc FeH_{0.7} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

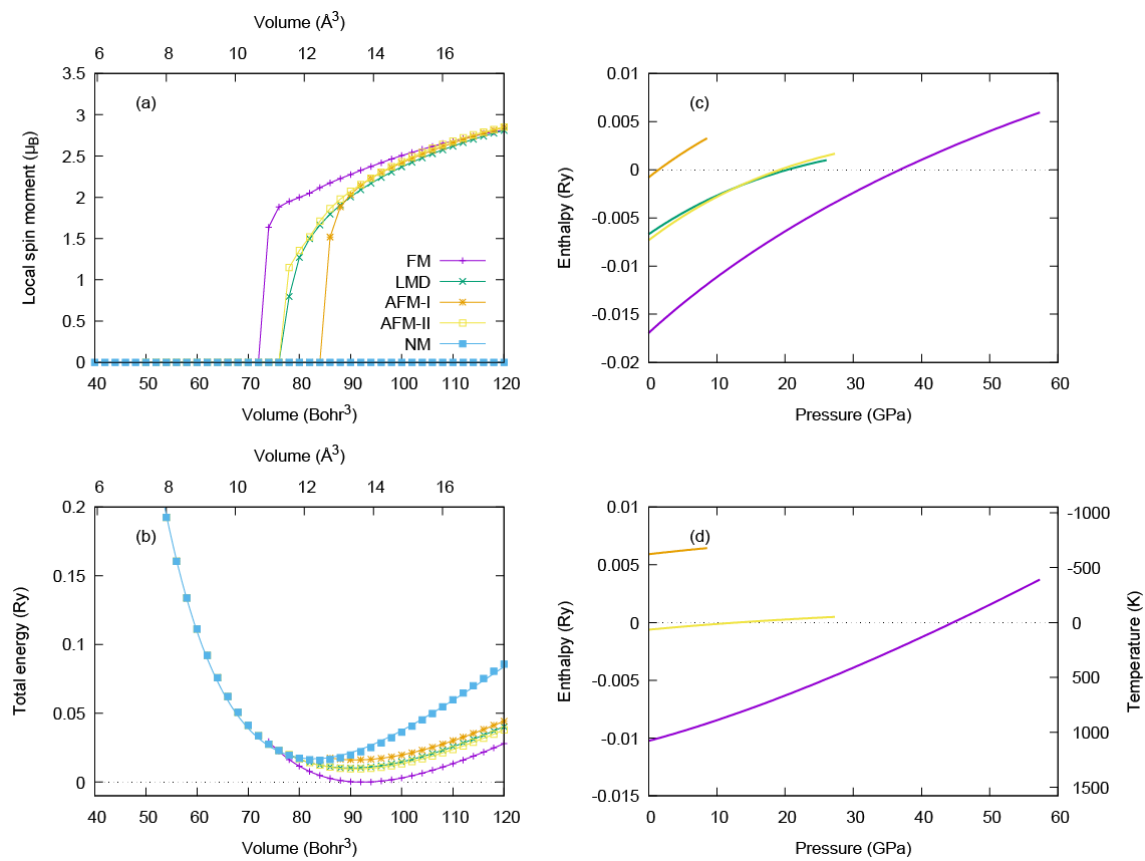


Figure S10. Calculation results of fcc FeH_{0.8} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

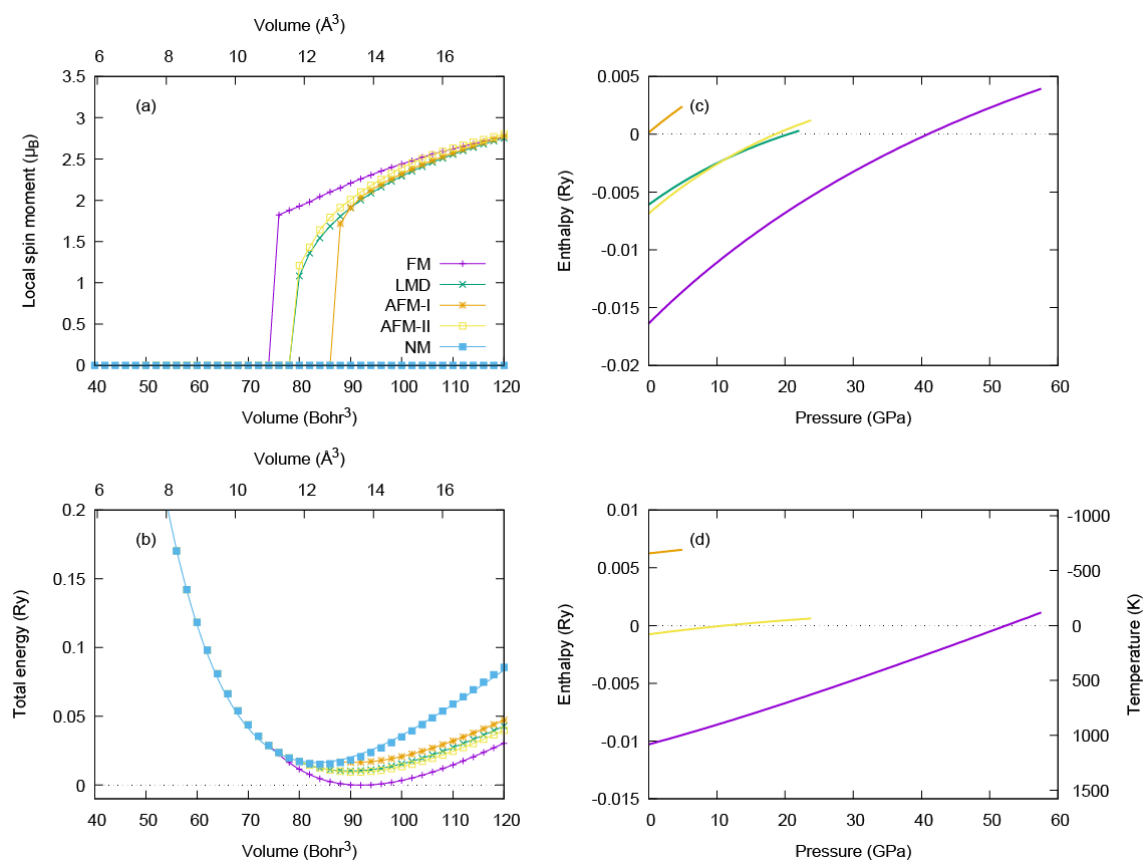


Figure S11. Calculation results of fcc FeH_{0.9} with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

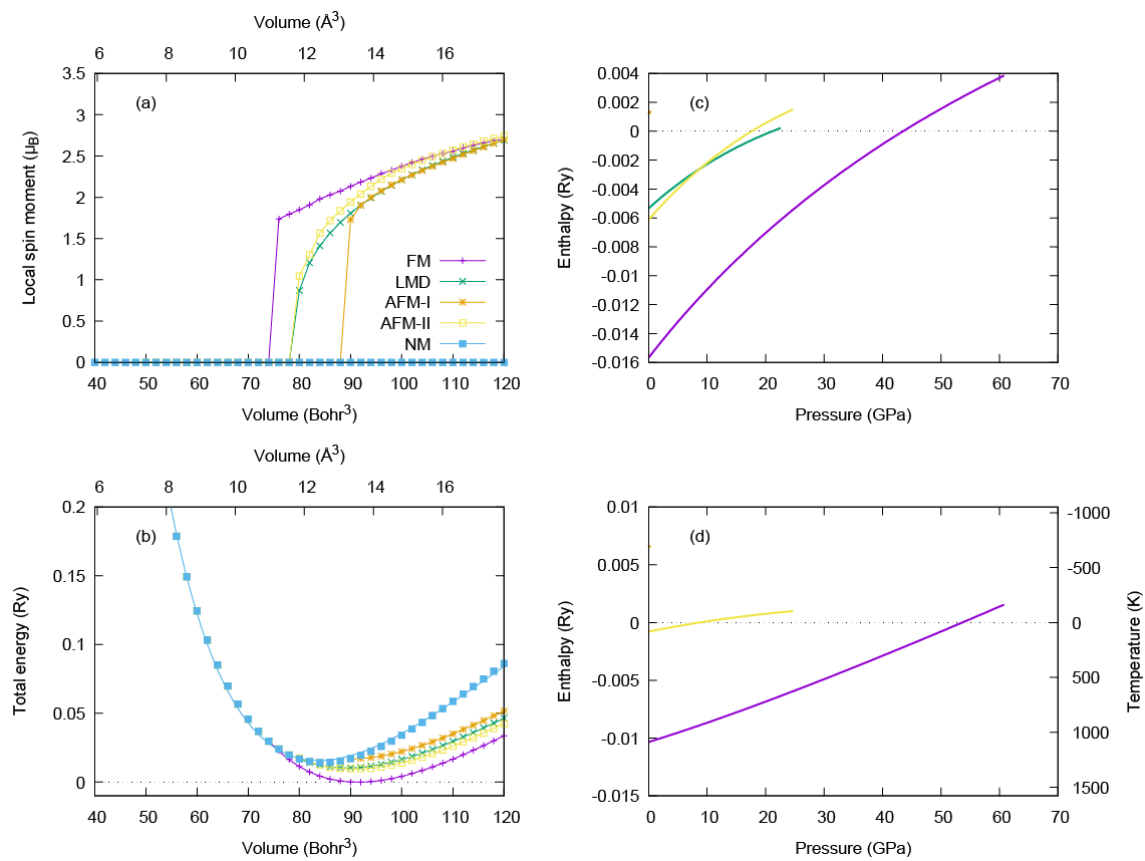


Figure S12. Calculation results of fcc FeH with FM (purple), LMD (green), AFM-I (orange), AFM-II (yellow) and NM (cyan) spin states. (a) Local magnetic moment. (b) Total energy. (c) Relative enthalpy from NM state. (d) Relative enthalpy from LMD state, which can be used to estimate the approximate magnetic transition temperature.

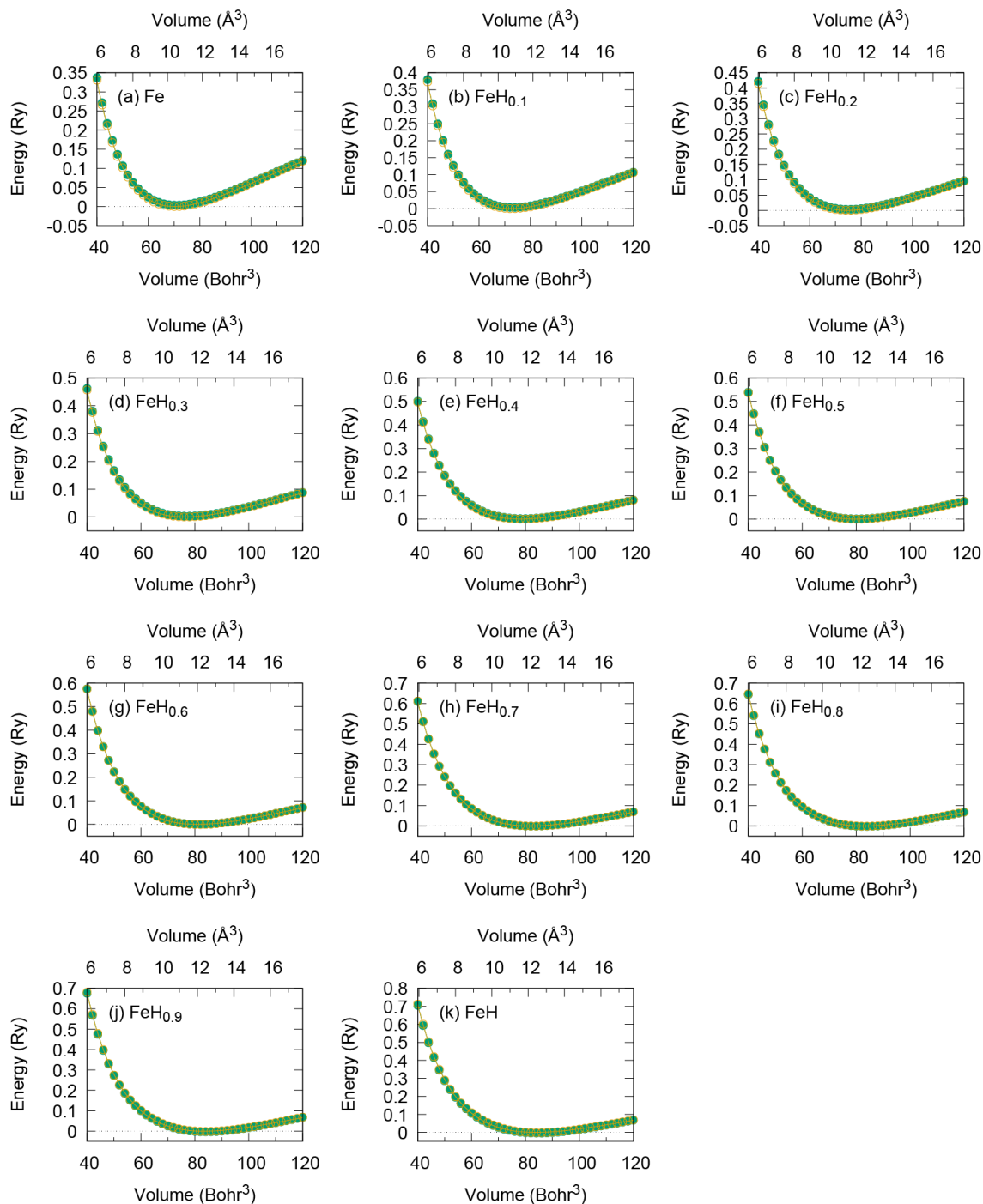


Figure S13. Total energy of NM hcp (orange) and dhcp (green) FeH_x.

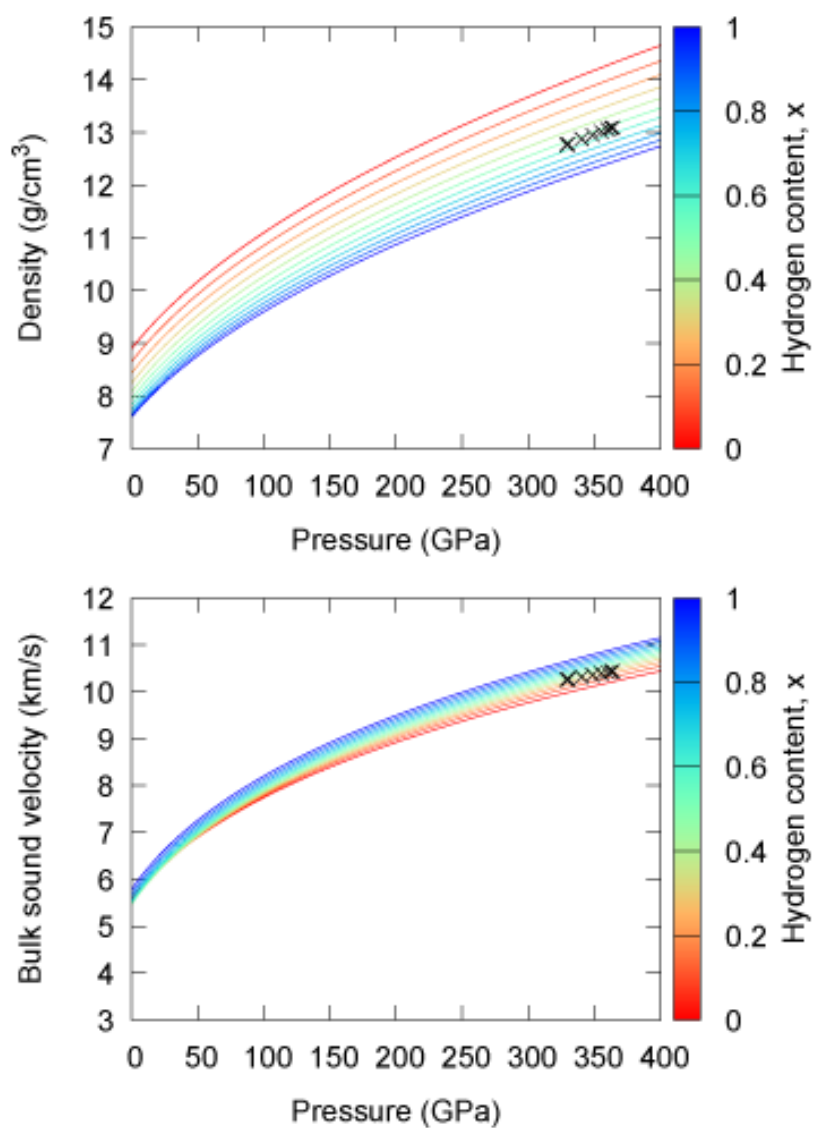


Figure S14. (a) Density and (b) bulk sound velocity of NM hcp FeH_{*x*} ($x = 0.0, 0.1, 0.2, \dots, 1.0$) as functions of pressure. Colors represent the composition from pure Fe (red) to FeH (blue). Cross symbols indicated the PREM at the inner core (Dziewonski and Anderson 1981).

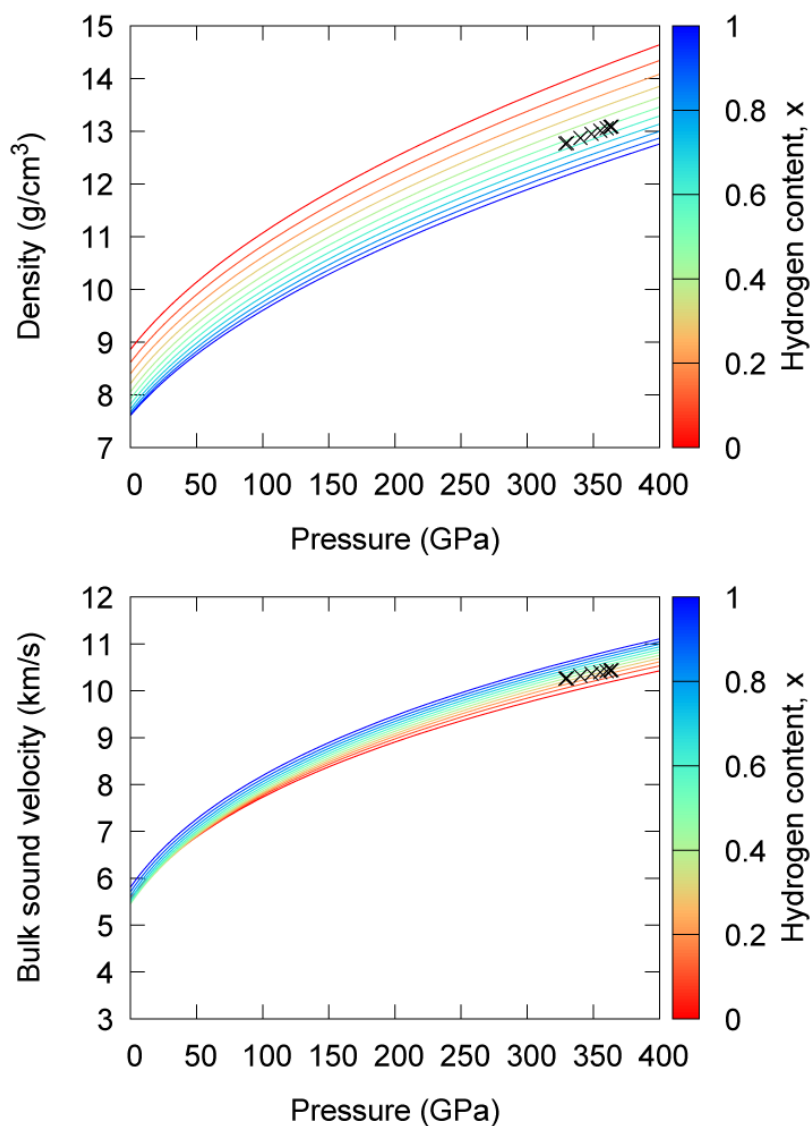


Figure S15. (a) Density and (b) bulk sound velocity of NM dhcp FeH_x ($x = 0.0, 0.1, 0.2, \dots, 1.0$) as functions of pressure. Colors represent the composition from pure Fe (red) to FeH (blue). Cross symbols indicated the PREM at the inner core (Dziewonski and Anderson 1981).