Supporting Information

New high-pressure phases of Fe_7N_3 and Fe_7C_3 stable at Earth's core conditions: Evidence for carbon-nitrogen isomorphism in Fe-compounds

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Text S1. Method

Here we present the details of the calculating method used to consider the temperature effect, the phonon dispersion curves of the predicted phases, and the comparison of crystal structure predictions by USPEX and AIRSS methods.

In calculations using the lattice dynamics method within QHA, the Helmholtz free energy of a system with a volume V at temperature T has the form:

$$F(V,T) = F_0(V) + F_{vib}(V,T) + F_{el}(V,T) \quad (1)$$

where $F_0(V)$ is the energy of the static lattice at 0 K; $F_{vib}(V,T)$ is the energy contribution of vibrations; $F_{el}(V,T)$ is the electronic contribution to the free energy.

The vibrational contribution in the QHA expressed as:

$$F_{vib}(V,T) = \sum_{q,i}\left[\frac{1}{2}\hbar\omega_{q,i} + k_B T\, ln\left(1 - e^{-\frac{\hbar\omega_{q,i}}{k_B T}}\right)\right] \quad (2)$$

where $\hbar$ is the Planck constant, k_B - the Boltzmann constant, $\omega(q,i)$ - the vibrational frequency of the i-th mode at the q-point and T - temperature.

The electronic contribution to the free energy is often believed to be negligible and therefore it can be ignored. If we deal with dielectric materials, this assumption is valid. For metals, the contribution should be taken into account, since it becomes significant with temperature. The free energy of electrons, in this case, can be expressed as:

$$F_e(V,T) = E_e(V,T) + TS_e(V,T) \quad (3)$$

where $E_e(V,T)$ is the internal energy of the electrons, $S_e(V,T)$ - the electronic entrophy.

For electrons, E_e and S_e can be written through the Fermi-Dirac distribution function and the electron density of states (DOS):

$$E_e(V,T) = \int_0^{\infty} n(\varepsilon,V) f(\varepsilon)\varepsilon d\varepsilon + \int_0^{\varepsilon_F} n(\varepsilon,V)\varepsilon d\varepsilon \quad (4)$$

$$S_e(V,T) = \int_0^{\infty} n(\varepsilon,V)[f\ln f + (1 - f)\ln(1 - f)]d\varepsilon \quad (5)$$

where ε is the Kohn-Sham orbital's energy, ε_F - the Fermi level, n(ε,V) - the electron DOS, f(ε) - the Fermi-Dirac distribution function.

$$(\varepsilon,V) = \frac{1}{N_k}\sum_{k,i}\delta\big(\varepsilon(V) - \varepsilon_{k,i}(V)\big)$$

It should be noted that , where k and i are the wave vector and energy eigenvalue index, respectively, does not depend on temperature, and the temperature dependence, in this case, is manifested only through the Fermi-Dirac function.

Considering the dependence of the Helmholtz free energy on the volume at a given temperature, we can write the pressure as: $P = -(\partial F/\partial V)_T$. Thus, knowing the pressure dependence on volume and temperature, one can calculate the Gibbs free energy as: $G = F + PV$.

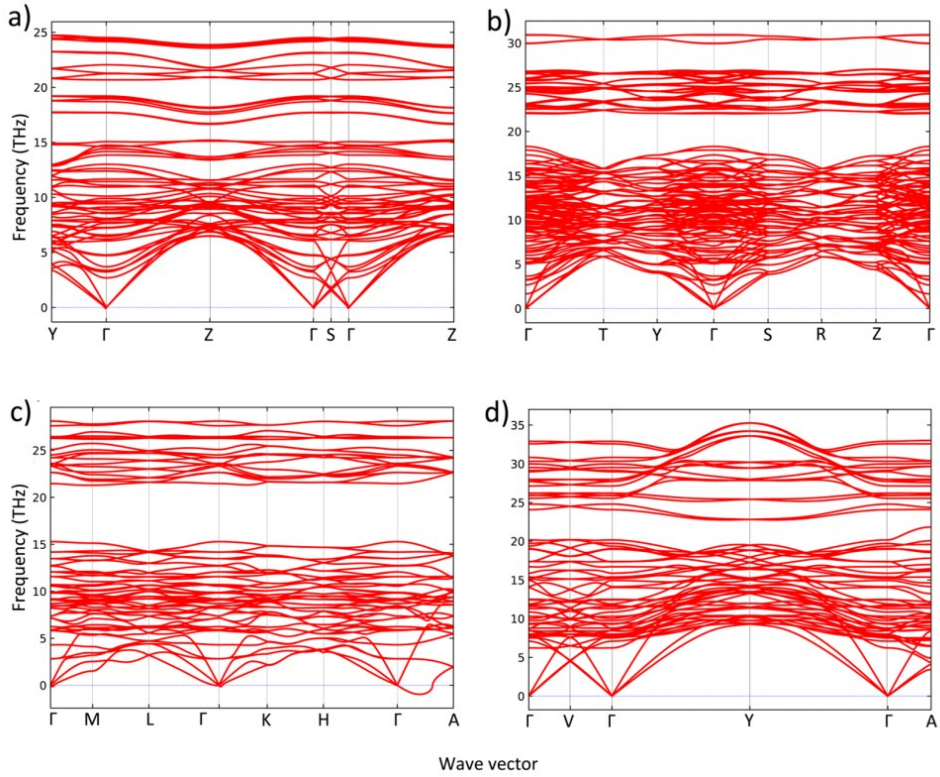

Figure S1. Phonon dispersion curves of *Amm*2-Fe_7N_3 at 60GPa (a), *Cmc*2_1- Fe_7N_3 at 200GPa (b), β-Fe_7N_3 at 150GPa (c), and *C*2/*m*- Fe_7N_3 at 400GPa (d).

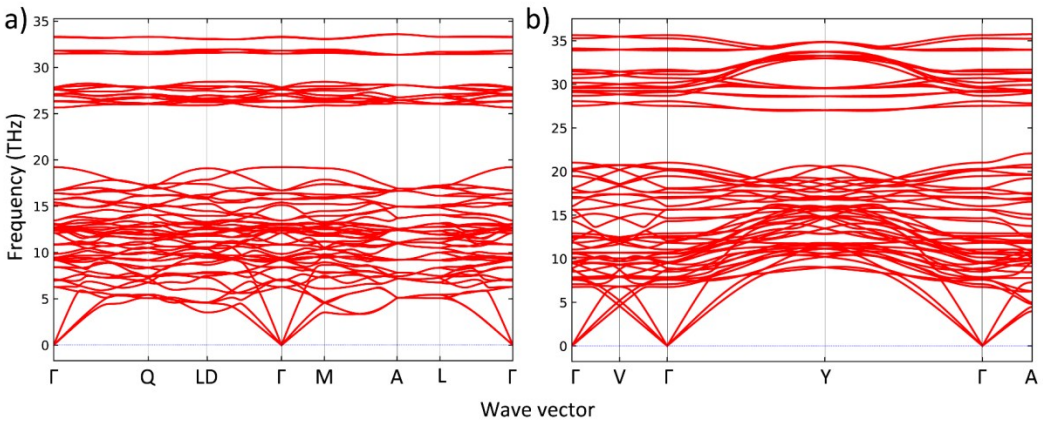

Figure S2. Phonon dispersion curves of h-Fe_7C_3 at 250 GPa (a) and *C*2/*m*-Fe_7C_3 at 400 GPa (b).

Table S1. Structures predicted by USPEX and AIRSS method, Z - the number of formula units in the unit cell.

Pressure (GPa)	USPEX		AIRSS	
	Space group	Z	Space group	Z
150	$P6_3mc$	2	$P6_3mc$	2
200	$Cmc2_1$	4	$P6_3mc$	2
300	$Cmc2_1$	4	$Cmc2_1$	4
400	$C2/m$	2	$Cmc2_1$	4