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Electronic, phononic and superconducting properties of hydrides of yttrium and sulfur at high pressure

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Abstract. The electronic, phononic, and superconducting properties of double hydrides YH_6 and H_3S are analyzed and compared with that for triple hydride YS_4H_4 . A common physical and numerical approach was used to determine both the stability range and the critical superconducting temperature. The characteristics of the high-pressure phase of the triple hydride YS_4H_4 are obtained.

1. Introduction

At present, there is an urgent task to search for new high-temperature superconducting (HTSC) materials with high hydrogen content, in which the transition to the superconducting state occurs under close to normal conditions (ambient pressure and room temperature). In the review paper [1] the properties of 33 hydrides of the main groups of elements at high pressure are discussed and optimum values of pressure corresponding to maximum critical temperature T_c were found. For binary hydrides, superconductivity with T_c of about 100 K was predicted on the base of numerical simulation. Values of T_c above 200 K were predicted both in the covalent hydride H_3S and in the several polyhydrides having the YH_6 type structure.

Along with theoretical estimates of the superconducting properties of various hydrides, experimental results have also been obtained which have confirmed high values of superconducting temperature $T_c \sim 200$ K in sulfur hydrides under high pressure [2]. Thus, high temperature of the superconducting transition T_c is achieved in hydrides under high pressure. One of the main tasks is to search for compounds stable at lower pressures.

Using the USPEX program [3], we have found several structures for triple hydrides based on yttrium and sulfur. Yttrium sulphide (YS , Y_2S_3) exists under normal conditions. For the double hydrides at high pressures YH_6 and SH_3 the calculated values of T_c are sufficiently large. In this paper we present the results obtained for the high-pressure phases of YS_4H_4 .

2. Method of calculation

The calculation of the properties of hydrides $\text{Y}_n\text{X}_m\text{H}_k$ was carried out within the framework of the density functional theory (DFT) in the plane waves basis, using the norm-preserving pseudopotential. The following sequence of calculations was used for each pressure value: 1) structural parameters, enthalpy of the equilibrium structure; 2) electronic Bloch spectra of the equilibrium structure; 3) phononic spectra for the equilibrium structure; 4) transition to the Wannier-functions basis; 5) calculation of electronic spectra, obtaining Eliashberg spectral function and the electron-phonon interaction con-



stant; 6) estimate of the superconducting transition temperature T_c , starting from the series of approximations of the solutions of the Eliashberg equations; 7) calculation of the normal state Wannier-functions of the system, taking into account the effect of the electron-phonon interaction on the electronic spectrum; 8) calculation of the superconducting properties taking into account an influence of the electron-phonon interaction on both T_c and the superconducting gap width.

QUANTUM ESPRESSO software package [4], proprietary software packages based on the results of work [5] and visualization algorithms were used calculations.

3. Results and discussion

A characteristic feature of the structure of the double hydrides YH_6 and H_3S is the high symmetry and the presence of a sublattice of homogeneous hydrogen in the H-H distances in the first coordination spheres. These structures are well studied and both calculated [6-8] and experimental data [2] were obtained for them. Variation of the basic elements of these hydrides leads to different magnitudes of the "chemical" pressure as well as to different ranges of their pressure stability. We recalculated these double hydrides and obtained results, in general, coincide with the results of previous studies [6-8].

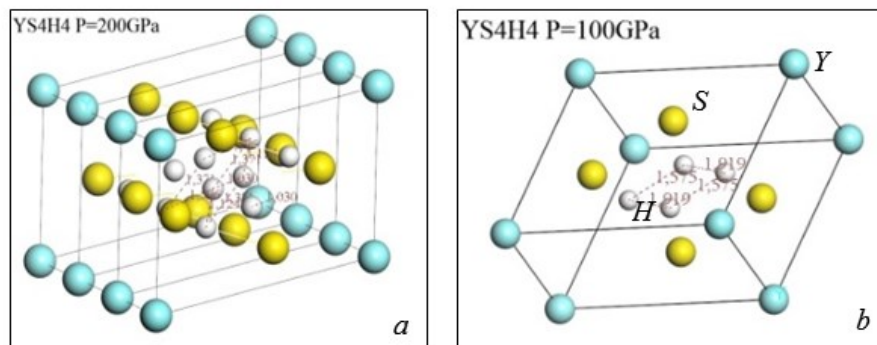


Figure 1 a. Cells YS_4H_4 with symmetry $C2/M$; b. primitive cell of the same structure.

The optimal structure of the triple hydride YS_4H_4 , established by us, is shown in Fig 1. The primitive cell of this structure contains many-electron atoms Y and S. Their interaction keeps required structure symmetry at a lower pressure than the pressures needed to stabilize compounds YH_6 and H_3S . The hydrogen sublattice has the form of an "accordion" with the predominant quasi-chain spatial arrangement of atoms. The H-H interatomic distances have three different values depending on the pressure. Atoms of heavy elements form chains.

Oscillations in the interval 100-200 meV are associated with vibrations of the hydrogen sublattice of these hydrides. Almost the entire frequency range of oscillations is filled only for YH_6 and H_3S , while for YS_4H_4 empty frequency intervals are clearly pronounced. Continuity of the spectrum indicates connection of the sublattices in YH_6 and H_3S . Gaps in the YS_4H_4 spectrum are caused by different environments of different hydrogen atoms. We note that the density of the phonon modes number in the low-frequency region (100 meV for H atoms) is much larger than that for YH_6 .

The phonon densities of the number of states for the YH_6 , H_3S and YS_4H_4 compounds are shown in Fig.2. For YH_6 and H_3S the imaginary frequencies are absent in the spectrum for pressures larger than $P \approx 150$ and 175 GPa, respectively. The maximum phonon oscillation frequencies are about of 200 meV for YH_6 and H_3S , while for YS_4H_4 they are equal to 280 meV.

In these compounds, the electronic states density at the Fermi level is created by electrons of different types (s, p, d). The table shows the values of partial density of states PDOS at the Fermi level for a pressure $P = 200$ GPa for all three types of hydrides.

Table 1 Value of PDOS (E_F) $P = 200$ GPa

substance	s	p	d	Σ
H ₃ S	0.587	0.47	-	1.07
YH ₆	0.294	0,039	0.424	0.762
YS ₄ H ₄	0.133	0.108	0.566	0.807

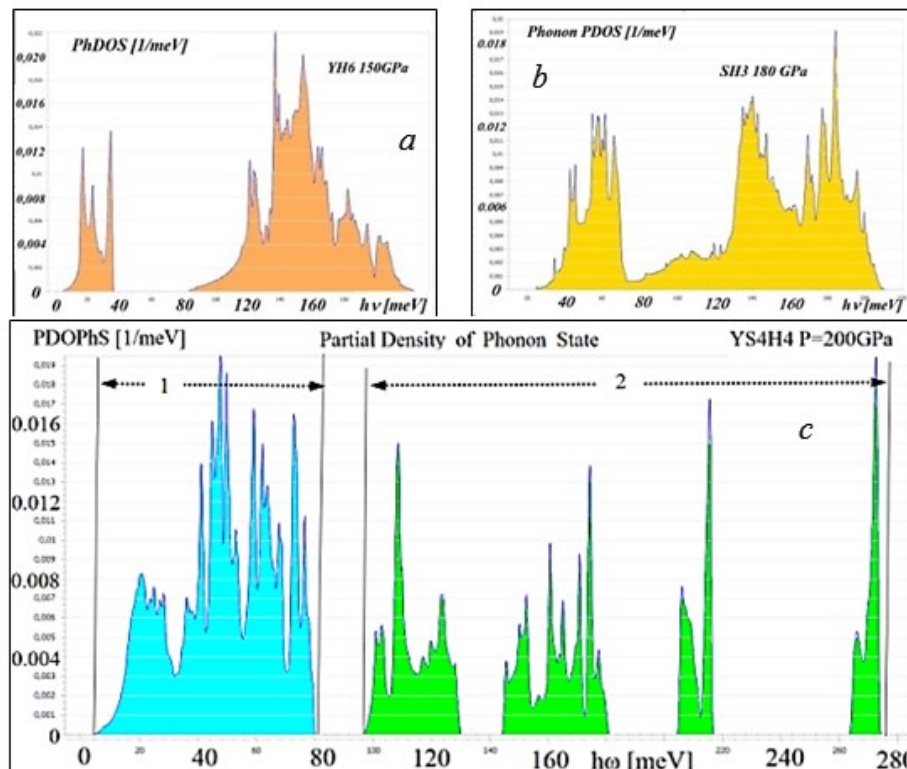


Figure 2. Partial densities of phononic oscillations of PhDOS for YH₆, SH₃ at characteristic pressures $P = 150, 180$ and 200 GPa, respectively. In Figure 2c for YS₄H₄, the vibration frequency band of hydrogen atoms is denoted as range 2, the energy range of the vibration modes of the heavy atoms Y and S is denoted by 1.

Variation of stoichiometry in triple hydrides allows to change the ratio between the partial contribution of different elements to the PDOS at the Fermi level. Such a variation leads also to changing the amplitude (s, p, d) of PDOS at the Fermi level and, therefore, the matrix elements of electron-phonon interaction.

The Eliashberg spectral functions preserve the main features of the phonon partial densities of the number of states PDOS. Calculations of the values of both the electron-phonon interaction constant λ and the critical temperature of the superconducting transition gave the following results: (SH₃ - $P=180$ GPa - $\lambda \approx 2.3$ - $T_c \approx 200$ K); (YH₆ - $P=125$ GPa - $\lambda \approx 3$ - $T_c \approx 165$ K); (YS₄H₄ - $P=200$ GPa - $\lambda \approx 1$ - $T_c \approx 20$ K).

Figure 3 shows the dependence of energy per atom on pressure: $\varepsilon = (E_{cell} - (mE_Y + nE_S + kE_H)) / N_{cell}$, here E_{cell} and N_{cell} - total energy and number of atoms in the calculated cells of the systems; E_Y, E_S, E_H - calculated energies of solitary atoms.

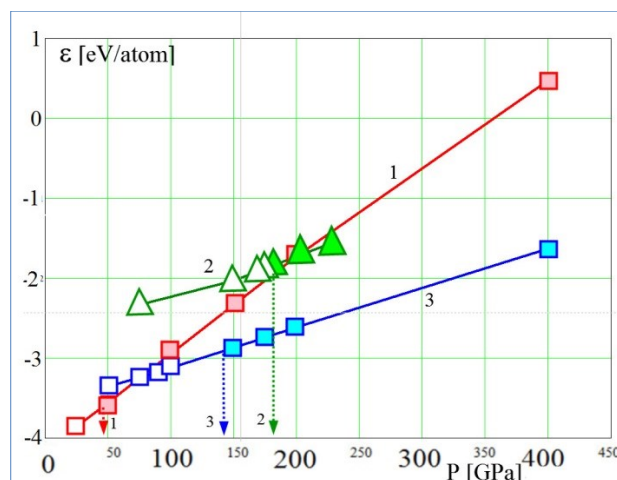


Figure 3. Dependences of the reduced specific energy per atom for the systems YH_6 (3), SH_3 (2), YS_4H_4 (1). The closed symbols show the minimum pressures at which the system is stable. Arrows indicate critical pressures

4. Conclusions

The obtained results indicate the complex character of the interrelationships in the structure, electronic and phononic spectra, types of internal bonds, and the charge state of the atoms of the compounds studied, especially taking into account changes in these properties with a change in pressure. The triple hydrides demonstrate a possibility to stabilize the structure at low pressures due to the "chemical" pressure of the parent matrix atoms.

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