

ELECTRONIC, PHONON AND SUPERCONDUCTING PROPERTIES OF MGHG AND CAHG COMPOUNDS: A DFT APPROACH

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Abstract- The structural, electronic, Phonon and superconducting properties of MgHg and CaHg compounds in B2 phase, have been investigated using first-principle calculations based on density functional theory. Calculated ground state property such as lattice constants are in good agreement with pervious reported results. The electronic band structures display the metallic nature of these compounds and the bonding features have been analyzed from charge density difference plots. Positive frequencies of phonon dispersion curves confirm the dynamical stability of both the compounds in cubic CsCl (B2) structure at ambient pressure. The calculated superconducting transition temperatures (TC) are 1.40 K for MgHg and 1.61 K for CaHg compounds which are in good agreement with previous data.

Keywords – Electronic properties, Fermi Surfaces, Phonon Dispersion Curve, Superconductivity

1. INTRODUCTION

The AB type binary mercury-alkaline earth metal (MgHg and CaHg) compounds are great interest due to their high melting and boiling point [1], superconducting properties and light structural materials [2]. These types of compounds are used in automotive, aircraft and aerospace industries applications as well as global energy composition system. They also exhibit desirable chemical properties because of varieties of phenomena related to their chemical bonding [3-4]. In previous years, several authors have investigated the different properties of MgHg and CaHg compounds using theoretical and experimental techniques. The structural stability of Hg based alkaline earth metal (MgHg, CaHg, SrHg and BaHg) compounds have been done by using first principles pseudopotential method and HT approach [2, 5]. Yosis et al. [1] have been performed the computational experiment on mercury based binary AB type compounds and reported that these compounds exhibit the high values of melting and boiling point. The measurements of superconductivity in MgHg, CaHg and BaHg compounds have been reported by chareson et al. [6] using X-ray diffraction. Superconducting transition temperature for MgHg compound has been found 1.4 K by chaelse et al.[7].

In the present work, we study the structural, electronic, phonon and superconducting properties of MgHg and CaHg compounds using density functional theory. We are interested in theoretical calculation of the phonon properties and superconducting transition temperature which are reported first time for these compounds and are compared with their corresponding experimental values [6,7].

2. COMPUTATIONAL METHOD

The MgHg and CaHg compounds crystallized in cubic CsCl (B2) structure (space group Pm3m 221). All calculations were performed using first principle calculations based on Density Functional Theory (DFT) within the plane wave pseudopotential as implemented in Quantum Espresso code [8]. For exchange correlation functional we have used Perdew, Burke and Ernzerhof generalized gradient approximation (PBE-GGA) [9] and the norm-conserving pseudopotential of Troullier and Martins type, generated with the FHI (Fritz- Haber-Institut) code for describing the interaction between the valence electron, nuclei and the core electrons. The convergence is achieved with a kinetic energy cut-off 80 Ry. which is sufficient to fully converge all properties. The Brillouin zone (BZ) integration was carried out by Marzari-Vanderbilt smearing [10] technique using $15 \times 15 \times 15$ k-point mesh and degauss value is taken 0.01 Ry. for both the compounds. Phonon dispersion curves and density of states are calculated using density functional perturbation theory (DFPT) [11]. For calculation of electron-phonon coupling we have considered the scheme of Monkhorst pack [12] has been used to generate k-point mesh and the dynamical matrices are computed on a $5 \times 5 \times 5$ q-point mesh. The superconducting transition temperature has calculated by means of Mc-Millan's equation modified by Allen and Dynes [13,14].

3. RESULT AND DISCUSSION

3.1 Structural and electronic properties –

To calculate the ground state properties of MgHg and CaHg compounds in cubic CsCl (B2) structure (Pm3m. 221), we have fitted the values of total energy to the Birch-Murnaghan's equation of state [15]. The structural properties such as the lattice

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constants (a_0) are presented in Table 1, which show good agreement with previous reported results [6, 7, 16]. To understand the electronic properties of MgHg and CaHg compounds we have calculated the electronic band structure, density of states (total and partial), Fermi surface topology and charge density difference plots. The calculated electronic band structures and density of states (total and partial) for both the compounds are shown in Figure 1(a-b). From the analysis of Figure 1(a-b), it can be noticed that there is no energy gap exist at the Fermi level (E_F) which indicates that both the compounds are metallic nature. The lowest lying bands (flat bands) originate from the d-state of Hg atom and the p-state of Hg atom display the major contribution in both (valance and conduction) regions which responsible for the density of states at the Fermi level $N(E_F)$ as reported in Table 1. The p-state of Mg atom shows their small contribution in conduction band region and hybridized with Hg-p state around the energy range 4 eV for both the compounds.

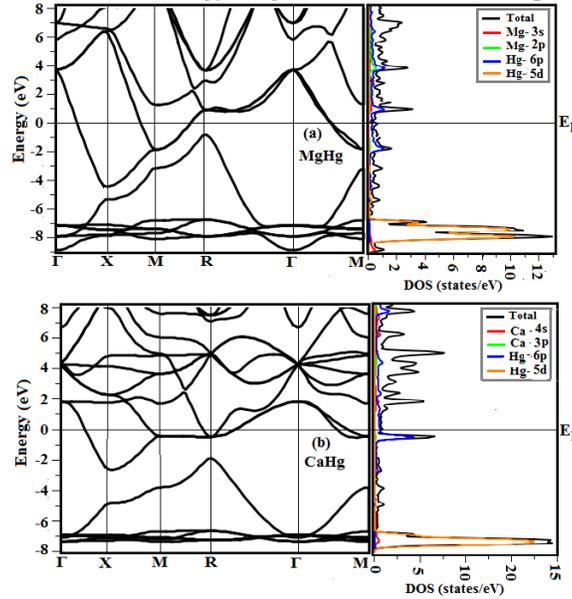


Figure 1. Electronic band structure of (a) MgHg (b) CaHg compounds

The Fermi surfaces (FS) are useful to describe the distribution of energy at different symmetry point in first Brillouin zone. Therefore we have plotted the FS topology for the bands which cross the Fermi level (E_F) and generate Fermi surface sheets as shown in Figure 2 (a-g) for both the compounds. For MgHg compound there are two bands (14 and 15) disperse at the Fermi level and compose two Fermi surface sheets (Figure (a-b)). The first sheet is electron FS sheet creates from 14th band at Γ to R point. The second FS sheet is produced by 15th band which stretched between $X \rightarrow M$ point and the third sheet (Figure (c)) represents the merged FS sheet of these two sheets for MgHg compounds. The energy ranges for the bands 14 and 15 are 0.09-0.067 Ry. and 0.27-0.86 Ry. respectively.

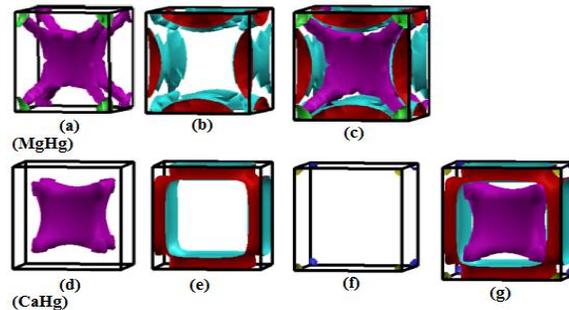


Figure 2. Fermi surface topology for MgHg (a-c) and CaHg (d-g).

Similarly, the Fermi surface topology for CaHg compound is presented in Figure 2 (d-g), there are three bands (namely 14, 15 and 16) crossing the E_F and make three FS sheets. The 14th band composes a closed hole FS sheet centered at Γ point with energy range 0.16-0.48 Ry. The second band constructs electron like FS sheet from the interconnection between M points with energy range 0.31-0.49 Ry. and the 16th band constructs tiny electron pocket at R point between the energy range 0.33-0.66 Ry. The last sheet (Figure 2(g)) is merged FS sheet of these three sheets.

The two dimensional charge density difference plots for MgHg and CaHg compounds in (100) plane are presented in Figure 3(a-b) and the density values are in the range from -0.055 to 0.019 and -0.048 to 0.017 $e/\text{\AA}^3$ for MgHg and CaHg respectively. From the charge density difference plots one can understand the bonding between constitute atoms. From the Figure it can be pointed out that the charges transformation from Hg to Mg and Ca ions due to high electronegativity of Hg

ions [3]. The isolated sphere confirms the ionic bonding between Hg-Mg and Hg-Ca ions. The covalent bonding is present between Mg-Mg and Ca-Ca ions.

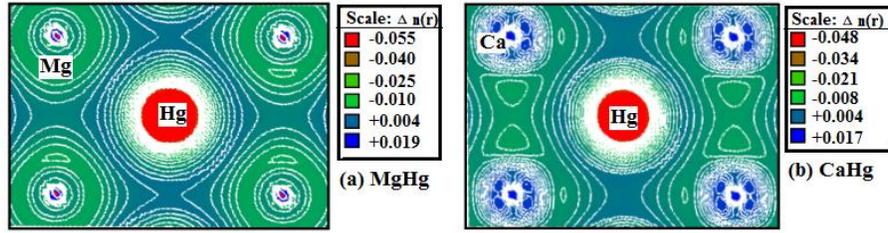


Figure 3. Charge density difference plots in (100) plane for (a) MgHg (b)CaHg compounds

3.2 Phonon properties –

The phonon dispersion curves (PDC) and phonon density of states (PDOS) for B2 phase of MgHg and CaHg compounds along high symmetry direction are calculated at ambient and are presented in Figs. 4(a–b). It can be seen that all the phonon modes are positive throughout the Brillouin zone which confirms the dynamical stability of MgHg and CaHg compounds in CsCl (B2) structure. Phonon dispersion curve contains six phonon modes in both the compounds (Fig. 4 (a-b)), three acoustic and other three are optical branches. The degeneracy of transverse optical (TO) and longitudinal optical (LO) branches around 138cm⁻¹ for MgHg and 127cm⁻¹ for CaHg indicate the metallic nature of both the compounds.

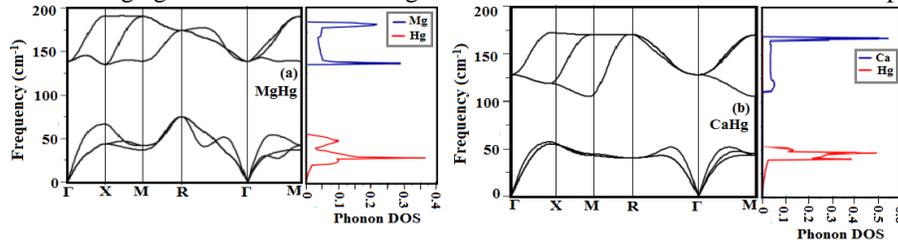


Figure 4. Phonon dispersion curve for MgHg (a) and CaHg (b) compounds in B2 structure (P=0 GPa)

The mass ratio of constituent atoms is responsible for the band gap between acoustic and optical branches which is around 62 cm⁻¹ and 55 cm⁻¹ for MgHg and CaHg compounds respectively. We have also presented the PhDOS along with phonon dispersion curves in which two colors are shown in Figure 4(a-b) where the red color shows the contribution heavy atom (Hg) at low frequency side in acoustic region and blue color shows the contribution of light atom (Mg and Ca) located in optical region.

3.3 Superconducting properties –

The electron-phonon interactions are also studied by employing the linear response method and Migdal-Eliashberg theory [17,18]. The Eliashberg spectral function with the variation of frequency is plotted and shown in Figure.5 (a-b) for MgHg and CaHg respectively. The nature of this function is similar to the phonon density of states and the highest peak is positioned at 1.6 THz for MgHg while it shifts at 1.2 THz in the case of CaHg compound [13,14].

$$T_c = \frac{\omega_{ln}}{1.2} \exp\left(-\frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)}\right)$$

Where ω_{ln} is the logarithmic averaged frequency, λ is the parameter of electron-phonon coupling constant and μ^* is the screen coulomb repulsion constant. The value of μ^* lies between 0.10 and 0.16 [19]. The value of screen coulomb repulsion constant (μ^*) is taken 0.16 (MgHg) and 0.14 (CaHg) for calculating the superconducting transition temperature. The calculated values of superconducting transition temperatures (T_c) is found to be 1.43 K for MgHg and 1.61 K for CaHg, which agree well with experimental value of $T_c = 1.39$ K, 1.4 K (MgHg) and 1.60 K (CaHg) [6,7].

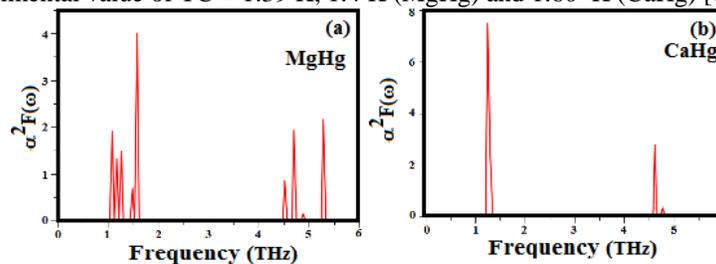


Figure 5. Eliashberg spectral function (a) for MgHg and (b) for CaHg compounds in B2 structure.

Table -1 Calculated Result

Compound	Lattice constant (a0) (Å)	N(EF) States/eV	TC (K)
MgHg (Pre.) Others	3.5138	0.69	1.43
	3.448a	-	1.39a
	3.449c	-	1.4b
CaHg (Pre.) Others	3.8224	1.42	1.61
	3.758a	-	1.6a

Table 1 Calculated lattice parameter (a0), density of states at Fermi level N(EF) and superconducting transition temperature (Tc) for MgHg and CaHg compounds in CsCl structure.

4. CONCLUSION

In conclusion, we have systematically investigate the structural electronic, phonon and superconducting properties of MgHg and CaHg compounds in CsCl phase using the generalized gradient approximation (GGA) scheme within the framework of density functional theory. The calculated lattice constants are in good agreement with the previous data. The electronic band structures and the value of DOS at Fermi level confirm the metallic behavior of these compounds. We have presented the phonon properties for of MgHg and CaHg and satisfied the dynamical stability criteria of of MgHg and CaHg in cubic CsCl phase. Further the superconducting properties of these compounds in CsCl phase are found to be transition temperature TC = 1.43 K and 1.61 K which are in good agreement of reported values of Tc.

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