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A FP-LAPW Study of Structural, Electronic, Elastic and Mechanical Properties of $CePd_3$ Intermetallic Compound

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Abstract

A theoretical study of structural, electronic, elastic and mechanical properties of $CePd_3$ intermetallic compound has been investigated systematically using first principles density functional theory. The calculations are carried out within LSDA for the exchange correlation potential. The ground state properties such as lattice parameter (a_0), bulk modulus (B) and its pressure derivative (B′) are calculated and obtained lattice parameter of this compound shows well agreement with the experimental results. We have calculated the elastic constants (C_{11}, C_{12} and C_{44}), which has not been calculated and measured yet. From energy dispersion curves, it is found that the studied compound is metallic in nature. Ductility of this compound is analyzed using Pugh's criteria and Cauchy's pressure (C_{11}- C_{12}). The mechanical properties such as Young's modulus, shear modulus, anisotropic ratio, Poison's ratio have been calculated for the first time using the Voigt–Reuss–Hill (VRH) averaging scheme. The average sound velocities (v_m), density (ρ) and Debye temperature (θ_D) of this compound are also estimated from the elastic constants.

Keywords: Pugh'scriteria; Density functional theory; Elastic constants; Ductility.

1. Introduction

Studies of Ce-based heavy fermion compounds are of fundamental importance for understanding their complex magnetic and transport properties. A number of recent discoveries have led to the resurgence of interest in the properties of the binary compounds which crystallize in the well known cubic Cu_3Au-type structure [1-4]. Among these, $CeIn_3$ and $CePd_3$ exhibit interesting properties. $CeIn_3$ shows antiferromagnetic ordering with TN = 10 K at ambient pressure, and becomes superconducting with TC=0.175 K at a pressure of 2.55 GPa [1]. On the other hand $CePd_3$ is a canonical intermediate valence compound which has been extensively investigated using many experimental techniques [5-8]. Despite of a low carrier density, 0.3 electrons per formula unit, the low temperature properties of $CePd_3$ are consistent with a simple Fermi-liquid ground state [9]. The commonality between $CeIn_3$ and $CePd_3$ is the presence of a broad peak in the resistivity, at 70 K and 150 K respectively, indicating the onset of a coherent Kondo lattice ground state. A consequence of the IV electron in $CePd_3$ is that the cerium atoms fluctuate between the magnetically active Ce^{3+} and non-magnetic Ce^{4+} configurations at a frequency on the order of 1013 Hz [10]. This fluctuation between magnetic and non-magnetic states results in a random distribution of magnetic moments throughout the lattice that rapidly appear and disappear. Since the average valence of the cerium atoms is approximately +3.5 at room temperature [11, 10], roughly half of the unit cells contain a trivalent cerium atom at any given time. *Ab-initio* electronic structure calculations, within the Kohn–Sham scheme of the density functional theory, are often considered reliable and a powerful tool to provide ground state information on intermetallic compounds. Low-temperature heat capacity data are presented for series of $AuCu_3$ type RPd_3 compounds (R = Ce, La, Lu, Y and Sc). Gardner *et al*. [12] have made measurements of the magnetic susceptibility in a large range of temperature for a complete series of RPd_3 compounds (*R*=rare earths). Some band structure calculations exist on $LaPd_3$ and $CePd_3$. Self-consistent band structure calculations for $LaPd_3$, $CePd_3$, $PrPd_3$, and $NdPd_3$ they found that magnetic ordering only in $PrPd_3$ and $NdPd_3$ and hence are consistent with the experimental findings. The value of lattice parameter for RPd_3 and RPd_3B are presented by Malik *et.al* [13]. Neither $LaPd_3B$ nor $LaPd_3$ are non-magnetic compounds [14].The lattice parameter for $ScPd_3$ and YPd_3 are: 3.9595Å and 4.0727Å [15] respectively. $CePd_3$ is one of the most investigated compounds with intermetallic valence (IV) [16].

2. Calculation method

The electronic, structural and mechanical properties of $CePd_3$ compound are investigated using first principle full potential-linearized augmented plane wave method within density functional theory. We used Perdew and Wang-generalized gradient approximation, which is based on exchange correlation energy [17].The wave vector cut-off

for the plane wave expansion of the wave function in the interstitial region was chosen, $R_{MT}*K_{max}= 7$ where R_{MT} is the smallest muffin-tin radius in the unit cell and K_{max} is the maximum of reciprocal lattice vector. A dense mesh of 10×10×10 k points is used and tetrahedral method [18] has been used for the Brillouin Zone integration. The calculations are iterated until the total energies are converged below 10^{-4} Ry. The total energies are calculated as a function of volume and fitted to Birch-Murnagan equation of state [19] to obtain the ground state properties like zero-pressure equilibrium volume. Information on the influences of pressure and temperature on the elastic moduli and related aggregate properties of single crystals plays an essential role in predicting and understanding the interatomic interactions, strength, mechanical stability, phase transition mechanisms and dynamical response of materials. For a cubic crystal, the three elastic moduli C_{11}, C_{12} and C_{44} fully describe its elastic behavior. C_{11} and C_{12} can be determined from the bulk modulus B and shear constant C_s.

3. Result and discussion

3.1Structural properties

The *Ab-initio* study of ground state properties for $CePd_3$ have been carried out using the Wien2k code based on the full potential linearized augmented plane wave (FP-LAPW) method. The total energies are calculated for this compound for different volumes around the equilibrium cell volume V_0. The calculated total energies are fitted to the Birch-Murnagan equation of state to determine the ground state properties like lattice constant (a_0),bulk modulus (B) and its pressure derivative (B′) at minimum equilibrium volume V_0.The calculated value of B is found to be 158.33(GPa) for $CePd_3$. The value of pressure derivative of bulk modulus (B′) is found to be 4.08(GPa). It is seen from Table 1 that our calculated values of a_0 are in good agreement with the experimental results and other theoretical work.

3.2 Electronic properties

In order to study the electronic structure of the $CePd_3$ compounds, spin-up and spin-down electronic band structures (BS) and density of states (DOS) have been calculated using LSDA approximation, where the Fermi level is considered at zero. The band profile for this compound is shown in figure1. The narrow peak lying above the Fermi level around 3ev is mainly due to *"d"* like states of Ce and Pd atoms. The *"d"* like states of Ce along with the little combination of "d" like states of Pd cross the Fermi level and spread in conduction region. The lowest lying bands below the Fermi level in the energy range -1eV to -6eV are mainly due to *"d"* like states of Pd hybridize with "d" and "p" like states of Ce atom. At Fermi level there is a hybridization of *"d"* and *"p"* like states of Ce and "d" like states of Pd due to which this compound exhibit metallic character. The density of states (DOS) plot provides an even more comprehensive picture of the elemental contributions to the electronic structure of $CePd_3$ compounds. The total and partial densities of states for $CePd_3$ compounds at ambient pressure are presented in figure2. We have calculated the DOS at Fermi level for these compounds. The metallic character of this compound is clearly seen from the finite DOS at Fermi level. The narrow and high peak lying below the Fermi level around at -18eV are mainly due to the *"p"* like states of Ce atoms.

3.3. Elastic properties

The elastic properties play an important role in providing valuable information about the binding characteristic between adjacent atomic planes. Anisotropic characters of binding and structural stability are usually defined by the elastic constants C_{ij}. These constants have been often related to the shear modulus and Young's modulus, which are frequently measured for polycrystalline materials when investigate their hardness. The elastic moduli require knowledge of the derivative of the energy as a function of the lattice strain. We have calculated the elastic constants of the $CePd_3$ compounds in $AuCu_3$ structure using LSDA as exchange correlation at ambient pressure using the method developed by Thomas Charpin and integrated it in the WIEN2k package [22]. The calculated values of elastic constants are given in Table 2. In the absence of any available measured data in the literature, these elastic constants could not be compared. It can be noted that our calculated elastic constants satisfy the stability criterions: $C_{11}- C_{12}> 0$, $C_{44}> 0$, $C_{11} + 2C_{12}> 0$, $C_{12}< B < C_{11}$, which clearly indicate the stability of these compounds in $AuCu_3$ structure.

3.4 Mechanical properties

The theoretical methods are completely independent of experiment and solve the quantum mechanical equations of DFT. Among these studies, the calculation of elastic constants based on the first principle calculation within

the highly accurate all-electron full-potential (linearized) augmented plane wave plus local orbital is very attractive because many other mechanical properties such as bulk modulus, shear modulus, Young's modulus, etc., can be derived from elastic constants. The elastic constants determine the response of the crystal to external forces, as characterized by bulk modulus, Young's modulus, shear modulus and Poisson's ratio which play an important role in determining the strength of the materials. Elastic properties play an important role in providing valuable information about the binding characteristics between adjacent atomic planes, the anisotropic character of binding and structural stability. The elastic constants of solids provide a link between the mechanical and dynamical behaviors of crystals and give important information concerning the nature of the forces operating in solids. These constants can be also predicting the structural stability of materials.

The bulk and shear modulus, defined as

$$B = \frac{1}{3}(C_{11} + 2C_{12}) \text{ and } G_H = \frac{\frac{C_{11} - C_{12} + 3C_{44}}{5} + \frac{5C_{44}(C_{11} - C_{12})}{4C_{44} + 3(C_{11} - C_{12})}}{2}$$

Another important parameter is the elastic anisotropic factor A, which gives a measure of the anisotropy of the elastic wave velocity in a crystal and it is given as:

$$A = \frac{2C_{44}}{C_{11} - C_{12}}$$

which is unity for an isotropic material. It also tells more about the structural stability and it is correlated with the possibility of inducing micro cracks in the materials.

The calculated elastic anisotropic factor for $CePd_3$ compounds is greater than 1, which indicates that these compounds are not elastically isotropic. As suggested by Pugh [23], if B/G_H>1.75; a material behaves in a ductile manner. From Table 2, it can be seen that the of B/G_H is 3.46 for $CePd_3$ Ganeshan *et.al.* [24] have established a correlation between the binding properties and ductility. The bond character of cubic compounds is explained with respect to their Cauchy pressure (C_{12}–C_{44}). The $CePd_3$ has a positive Cauchy pressure; resulting metallic bonding (ductility) in it. Young's modulus is defined as the ratio of stress and strain, and is used to provide a measure of the stiffness of the solid, *i.e.,* the larger value of *E*, the stiffer is the material. The highest value E occurs for Young's modulus (E) is given by

$$E = \frac{9BG_H}{3B + G_H}$$

The Poisson's ratio (σ) is given by

$$\sigma = \frac{(3B - E)}{6B}$$

The value of Poisson's ratio, found to be 0.37 for $CePd_3$ compound.

3.5 Thermal properties

Once we have calculated the Young's modulus E, Bulk modulus B and shear modulus G_H we may obtain the Debye's temperature is by using the average sound velocity v_m. At low temperature the vibrational excitations arise solely from acoustics vibrations. Hence at low temperature the Debye temperature calculated from elastic constants. We have calculated the average sound velocities and Debye temperatures as well as densities for $CePd_3$ compounds by using the calculated elastic constants. In the absence of any measured data in the literature, it could not be compared. Hence, our result can be considered as a prediction for the properties of $CePd_3$ compounds and it will testify future experimental work.

Conclusion

In conclusion, the structural, electronic, thermal, mechanical and elastic properties of cubic magnetic $CePd_3$ compounds which crystallize in the $AuCu_3$ structure have been studied using *ab-initio* full potential linearized augmented plane wave (FP-LAPW) method. The ground states properties such as lattice parameter (a_o), Bulk modulus (B) and its pressure derivative (B') have been obtained using optimization method. The value of lattice parameter for this compound is in good agreement with the experimental data. The electronic band structure of $CePd_3$ exhibits a metallic character. In the present study we found

$B/G_H > 1.75$ and $C_{12} - C_{44} > 0$; which implies that this compound is ductile in nature. We also report the mechanical and thermal properties for this compound and it will be tested in the future experimentally and theoretically.

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Table 1. Calculated lattice parameter a_o (Å), Bulk modulus B(GPa), its pressure derivative B'(GPa) of $CePd_3$ in $AuCu_3$ structure

Solid	Work	a_o (Å)	B (GPa)	B'(GPa)	C_{11} (GPa)	C_{12} (GPa)	C_{44} (GPa)
$CePd_3$	LSDA	4.0558	158.33	4.08	294.46	104.16	29.76
	Exp.	4.132	-	-	-	-	-
	Others	4.128	-	-	-	-	-

Table 2. Calculated Young's modulus (E), shear modulus (G_H), anisotropic factor (A), Poisson's ratio (σ), B/G_H ratio, Cauchy's pressure (C_{12} - C_{44}) , density (ρ), longitudinal (v_l), transverse (v_t), average elastic wave velocities(v_m) and Debye temperature (θ_D) of $CePd_3$ compound.

Solid	*E (GPa)*	*G_H (GPa)*	*A*	*σ*	*B/G_H*	*C_{12}-C_{44} (GPa)*	*ρ x 10^3 (kg/m^3)*	*v_l (m/s)*	*v_t (m/s)*	*v_m (m/s)*	*ϑ_D (K)*
$CePd_3$	132.64	48.48	0.31	0.37	3.46	74.40	8.679	5282	2538	2851	231.3

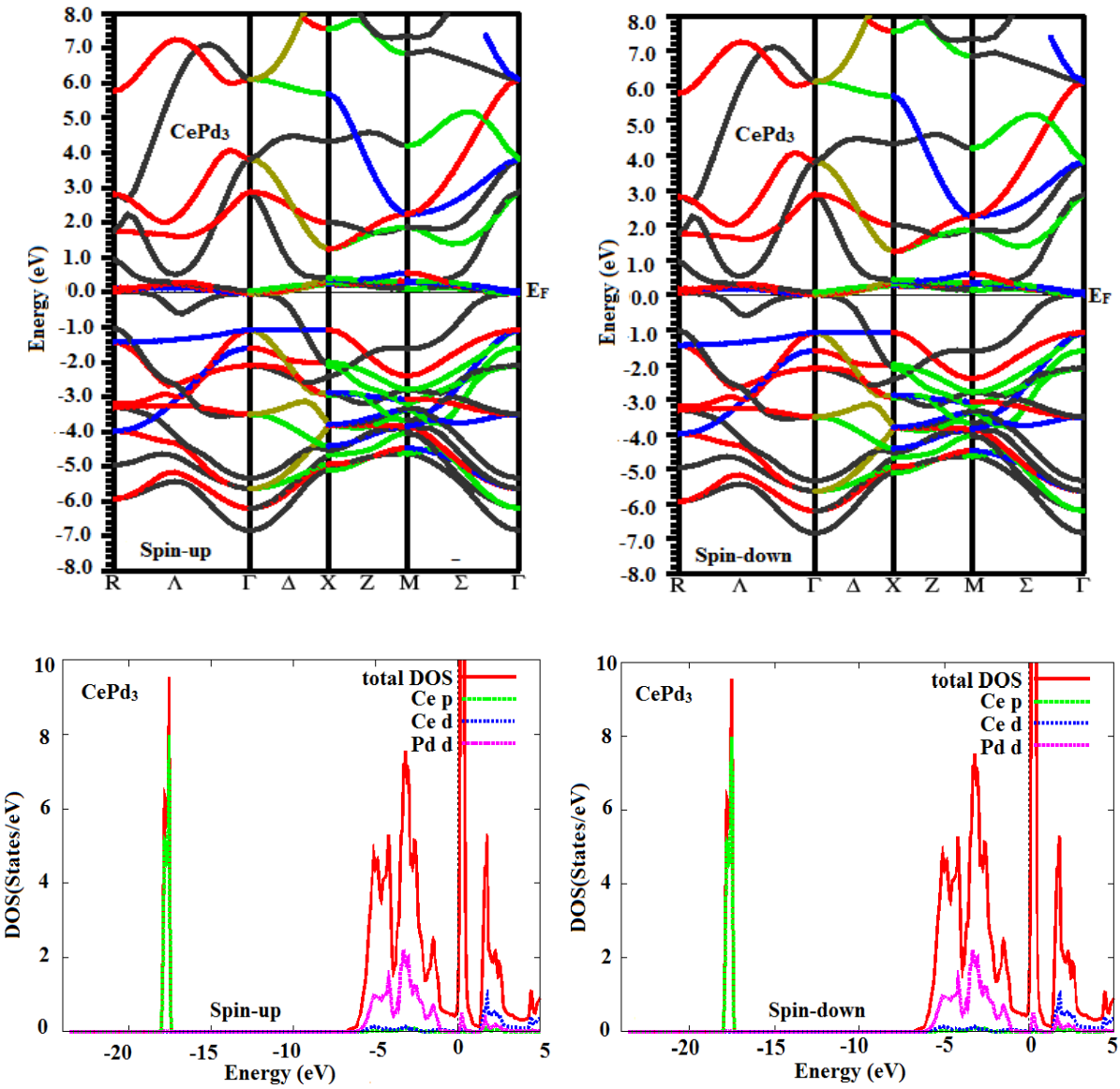

Figure 2. Spin-up and Spin-down Density of States of $CePd_3$

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