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Structural, Elastic, Thermodynamic and Phonon Properties of EuX3 (X=Pd, Sn and In) Intermetallic Compounds

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Abstract. The ab initio computations have been performed to examine the structural, elastic, phonon and thermodynamic properties of the rare-earth intermetallic compounds EuX3 (X=Pd, Sn and In) in AuCu3-type structure. We have performed our calculations within the generalized-gradient approximation (GGA) with ultrasoft pseudopotentials (USPP). The calculated structural parameters, such as the lattice constant, bulk modulus, its pressure derivative, cohesion energy are presented and compared with available data. In order to obtain more information about the mechanical stability we calculated second-order elastic constants, shear modulus, Young's modulus and Poisson's ratio. Phonon dispersion curves have been obtained using the first-principle linear-response approach of the density-functional perturbation theory. We have also performed the thermodynamic properties of EuX3 (X=Pd, Sn and In) intermetallic compounds by using quasi-harmonic approximation. The specific specific heat, internal energy, entropy and F vibration of these three intermetallic compounds are calculated and discussed.

Keywords. Rare-earth intermetallic compounds; Density functional theory; Elastic properties; phonon; ductility; vibrational properties.

INTRODUCTION

Since the discovery of the first intermetallic compound formed by two or more metallic elements, with optionally one or more non-metallic elements (Schulze, 1967) , research stepped up to find new intermetallic compounds. This research revealed several intermetallic compounds (Iandelli and Palenzona, 1974; Maslankiewicz et al., 2008). Intermetallic compounds are the association of metals or Metalloids together by a chemical bond. These compound are of two types, stoichiometric inter metallic compounds and non-Stoichiometric inter metallic compounds. Intermetallic compounds of rare earths have been the subject of

several experimental and theoretical studies and much research continues to attract considerable attention due to their interesting mechanical properties, electric and magnetic, which make them potential candidates for industry (Buschow, 1979). The europium-based intermetallic compounds of the form EuB3 (where B is transition metals, poor metals, etc) have generated considerable interest over many decades and continue to draw considerable attention because of practical applications based on a variety of phenomena because of their superconducting properties and magnetic susceptibility, as functions of their valence electron concentration (Havinga et al., 1970; Toxen et al., 1973; Toxen and Gambino , 1968). Europium-based compounds EuX3 (X=Pd, In and Sn) are binary intermetallic compounds with the cubic AuCu3-type structure space group Pm-3m (No. 221). The Europium (Eu) rare earth is located in 1a (0, 0, 0) position and the elements X (X=In and Sn) occupy 3c (0, 1/2, 1/2) position (Bajorek et al., 2012). The structural (Shafiq et al., 2014; Asadabadi and Akbarzadeh, 2004; Harris et al., 1965), thermo-electrical (Buschow et al., 1969), thermodynamic (Iandelli and Palenzona, 1974), superconductivity (Loewenhaupt and Hufner, 1969; Buschow, 1969; Gorlich et al., 1975; Buschow, 1979), electronic properties (Maslankiewicz et al., 2008) and phase diagrams (Iandelli and Palenzona, 1981) of EuPd3, EuIn3 and EuSn3 have been studied by several groups, employing different theoretical and experimental methods. To the best of our knowledge, no experimental and theoretical studies for vibrational and thermodynamic properties of these compounds in AuCu3-type structure have been appeared in the literature.

In this work, we investigate the structural, elastic, phonon and thermodynamic properties of EuX3 (X=Pd, Sn and In) using a pseudo-potential plane-wave (PPPW) method as implemented in the Quantum ESPRESSO code in the scheme of generalized gradient approximation (GGA). The paper is organized as follows: In Section 2, we present a brief review of the theoretical method. The results and discussions are presented in Section 3. Finally, the conclusions derived from our calculations are summarized in Section 4.

COMPUTATIONAL METHOD

The calculations were performed by employing the self-consistent ultrasoft pseudopotential method based on the density functional theory (Vanderbilt, 1990), within the generalized gradient approximation (GGA), incorporated into the Quantum-Espresso code (QE) (Giannozzi et al., 2009). We employed the Perdew–Burke–Ernzerhof (PBE) for the exchange–correlation potential (Perdew et al., 1996). The electron wave functions were expanded with a plane wave basis set with a kinetic energy of 40 Ry, and an energy cutoff of 500 Ry were included for the charge density. The k-space integration on the Brillouin zone (BZ) for the self-consistent calculations was calculated with a 16×16× 16k-points mesh. The smearing technique with a smearing parameter of 0.02 Ry was used for integration up to the Fermi surface (Methfessel and Paxton, 1989). Lattice dynamic properties were obtained using density functional perturbation theory (DFPT) or linear response (Baroni et al., 1987; Baroni et al., 2001). The phonon dispersive curves, using the phonon code of quantum-Espresso code, have been calculated on a 4×4×4q-point mesh of the first Brillouin zone to obtain eight dynamic matrices. Thermodynamic functions such as specific heat at constant volume, entropy, internal energy and vibrational free energy were calculated using the Quasi Harmonic Approximation (QHA) (Isaev, 2013). We calculate the elastic constants by ElaStic tool which can be interfaced with computer QE package (Golesorkhtabar et al., 2013).

RESULTS AND DISCUSSION

Structural Properties

In order to determine the structural properties of intermetallic compounds EuX3 (X=Pd, In and Sn), we have calculated the total energies as a function of the global volume for both nonmagnetic (NM) and ferromagnetic (FM) states. EuX3 (X=Pd, In and Sn) compounds crystallize in a cubic AuCu3 type crystal structure with four atoms per unit cell. To define the most stable structure among the structure without spin polarized (NM) and the structure with spin polarized (FM), the cohesive energy (Ecoh) was computed from the difference between the total atomic energies, calculated for the ground state configurations of X (X=Pd, In and Sn) and Eu atom, and the minimum energy of bulk compounds calculated at the equilibrium lattice constants. The cohesive energy are estimated by following formulas Ecoh= EEuX3 - (EEu + 3EX), where EEuX3 is the total energy of the intermetallic compounds and EX represent the total energies of the X element. The equilibrium lattice parameters a0, bulk modulus B0 at zero pressure and the cohesive energy for EuX3 (X= Pd, Sn and In) compounds are shown in Table 1. It is noted that all cohesive energy are negative, we conclude that all compounds are thermodynamically stable and the cohesive energy are minimal for non ferromagnetic structure, indicating that the structure without spin polarized is the most energy stable structure. The calculations predicted that the equilibrium lattice constants are in excellent agreement with the theoretical study for EuSn3 and EuIn3 (Asadabadi and Akbarzadeh, 2004) and with the available experimental value for EuSn3 (Villars and Calvert, 1991). In the present study, ordered EuX3 (X= Pd, Sn and In) do not show any magnetic moment.

Table 1. The equilibrium structural parameter a(Å), the bulk modulus B_0 (GPa), Pressure derivative of bulk modulus B' and the cohesive energy (E_{coh}) of EuX3 (X=Pd, In and Sn).

EuX3		Phase	a_0(Å)	B_0(GPa)	B'	E_{coh}(eV)
EuPd3	This work	NM	4.1345	384.3	4.65	-1.3465
	Others work	NM	4.09 (Baroni et al., 2001)			
	This work	FM	4.1234	388.3	4.68	-1.3405
EuSn3	This work	NM	4.6765	192.6	4.69	-1.1768
	Others work	NM	4.6727 (Shafiq et al., 2014)	52.9 (Shafiq et al., 2014)		
	This work	FM	4.6510	168.1	5.23	-1.0931
	Others work	FM	4.6313	73.2400		
	Expt work	FM	4.744 (Sanchez et al., 1976)			
EuIn3	This work	NM	4.6863	172.9	4.43	-0,9315
	Others work	NM	4.6269 (Shafiq et al., 2014)	53.2488 (Shafiq et al., 2014)		
	This work	FM	4.6746	214.5	5.00	-0.9286
	Others work	FM	4.5707 (Shafiq et al., 2014)	63.7229 (Shafiq et al., 2014)		

Mechanical properties

The elastic constants of solids are important parameters due to their close relations with various physical fundamental properties. Thus, it is essential to investigate the elastic constants to understand the mechanical properties of EuX3 (X=Pd, In and Sn) intermetallic compounds. For a cubic crystal, there are three independent elastic constants namely C11, C22 and C44. We have calculated the elastic constants of those intermetallic compounds in AuCu3-type structure without spin polarization (non ferromagnetic case) and with spin polarization (ferromagnetic case) using the Elastic code (Golesorkhtabar et al., 2013). For a cubic structure the Cij matrix should satisfy the Born stability criterion, they are C11 – C12 > 0, and C44 > 0 and C12 < B < C11 (Born and Huang, 1954; Wang and Yip, 1993). In Table 2, the data for second order of elastic constants (Cij), bulk modulus B (GPa) , shear modulus (G) and Poisson's ratio (σ) in Voigt-Reuss-Hill approaches are calculated at the equilibrium parameter. Furthermore, our calculated elastic constants fulfilled the required stability conditions for cubic structures. To our knowledge, there is no experimental work for EuPd3, EuIn3 and EuSn3 intermetallic compounds. The only theoretical available value of elastic constant for EuIn3 and EuSn3 have been compared. The elastic constants for EuIn3 and EuSn3 are in reasonable agreement with those obtained previously (Shafiq et al., 2014). To the best of our knowledge, no experimental values for the elastic constants of these compounds have been appeared in the literature, so our results for AuCu3-type structure can serve as a prediction for future investigations. Our calculated elastic constants indicate that EuX3 (X=Pd, In and Sn) compounds are mechanically stable in the AuCu3-type structure. The brittle behavior of metals and metallic compounds can be measured using Poisson's ratio σ (Frantsevich et al., 1983). For ductile materials, the Poisson's ratio σ is 0.33. For brittle materials, the Poisson's ratio is less than 0.33. From Table 2, it is observed that σ is 0.27 for EuIn3 and EuSn3. This means that the compounds are brittle materials. Whereas EuPd3 with σ equal 0.33 is ductile material.

Vibrational properties and thermodynamics properties

The accurate vibrational and thermodynamic properties can directly provide the valuable information for understanding the dynamical response of materials under different conditions. We study the vibrational properties using the density functional perturbation theory (DFPT) and thermodynamic properties of EuPd3, EuSn3 and EuIn3 by using the quasi-harmonic approximation (QHA) package. The calculated phonon dispersion together with the phonon density of states (PHDOS) of intermetallic compounds EuPd3, EuRn3 and EuSn3, along the principal symmetry direction of the Brillouin zone (BZ) are displayed in figures 1 and 3. It shows that the shape of phonon spectrum curves is similar to each other. The unit cell of EuX3 contains 4 atoms, which gives rise to 12 phonon branches, which contains 9 optical modes and 3 acoustic modes. It clearly depicts that the considered structure of those compounds is dynamically stable as none of the phonon modes have imaginary frequency. The maximum values of these phonon curves are higher for EuPd3 than that for EuSn3 and EuIn3. The phenomenon results from the lattice parameters of EuPd3 (4.1345 Å) being smaller than that for EuSn3 (4.6765 Å) and EuIn3 (4.6863 Å). The shorter bond and smaller lattice parameters can leads to the larger force constants, resulting in higher phonon frequencies (Deligoz et al., 2013). For these compounds, the optical branches and acoustic branches of phonon curves overlap each other and are not separated by a gap. As a result, EuX3 (X=Pd, In and Sn) compounds exhibit a metallic behavior. At gamma point, the calculated frequencies of the optical modes of EuX3 are found to be 109.189, 132.458 and 168.8007 cm-1 for EuPd3, 88.074, 95.120 and 143.041 cm-1 for EuIn3, 81.588, 111.764 and 152.299 cm-1 for EuSn3, respectively. Unfortunately the experimental or theoretical phonon frequencies of EuX3 (X=Pd, In and Sn) compounds are not available in the literature for further comparison. The phonon density of states (PHDOS) associated with phonon

dispersion curve is presented in figures 1 and 3 for both the EuX3 compounds. The PHDOS curves of EuPd3 and EuIn3 and EuSn3 are similar. The PHDOS presented for all three compounds reflect all important features of phonon dispersion. Further, the PHDOS which is an essential quantity to calculate the thermodynamical properties. Figures 1 and 3 show that acoustic and optical branches overlap each other are not separated by a gap. Figures 4 and 7 presents the thermodynamic functions such as specific heat at constant volume, entropy, internal energy and vibrational free energy with temperature for three intermetallic compounds in AuCu3-type structure.

Table 2. Calculated elastic constants (C_{11} , C_{12} et C_{44}) (GPa) with spin polarization (FM) and without spin polarization (NM), the calculated value of Voigt bulk modulus (B_V), Reuss bulk modulus (B_R) , Hill bulk modulus (B_H), Voigt's shear modulus (G_V), Reuss's shear modulus (G_R), Hill's shear modulus (G_H) and Poisson's ratio (σ_V, σ_R, σ_H). Our results are compared with other theoretical calculations (in parenthesis).

	EuPd3	EuSn3	EuIn3
C_{11}	198.0	101.5 (118.316) (Harris et al., 1965)	93.5 (113.716) (Iandelli and Palenzona, 1974)
	412.1	93.8 (99.429) (Iandelli and Palenzona, 1974)	90.2 (100.413) (Iandelli and Palenzona, 1974)
C_{12}	126.6	70.0 (61.175) (Iandelli and Palenzona, 1974)	43.5 (56.308) (Iandelli and Palenzona, 1974)
	158.5	66.3 (65.844) (Iandelli and Palenzona, 1974)	42.3 (67.402) (Iandelli and Palenzona, 1974)
C_{44}	79.3	45.8 (51.377) (Iandelli and Palenzona, 1974)	39.5 (28.450) (Iandelli and Palenzona, 1974)
	190.1	41.6 (28.788) (Iandelli and Palenzona, 1974)	37.9 (22.588) (Iandelli and Palenzona, 1974)
B_V, B_R B_H	150.39	80.49	60.18
G_V	61.86	33.80 (42.254) (Iandelli and Palenzona, 1974)	28.552 (33.70) (Iandelli and Palenzona, 1974)
G_R	53.30	26.01	32.05
		38.943	28.551
G_H	57.58	29.90 (40.598) (Iandelli and Palenzona, 1974)	28.551 (32.87) (Iandelli and Palenzona, 1974)
σ_V	0.32	0.26	0.26
σ_R,	0.34	0.27	0.27
σ_H	0.33	0.27 (0.276) (Iandelli and Palenzona, 1974)	0.27 (0.332) (Iandelli and Palenzona, 1974)

Figure 1 shows the calculated temperature dependence of specific heat at constant volume which clearly reveals that the specific heat behaviour approaches the Dulong and Petit limit (CV~12.0 J.mol-1.K-1) at high temperature. The optic and acoustic modes have large effects on the heat capacity. The variation of the entropies (S) as a function of temperature for EuX3 (X=Pd, In and Sn) are shown in figure 5. As seen in this figure, we note that the entropy increase with increasing temperature for this all intermetallic compounds. The computed temperature variation of phonon contribution to entropy reveals that the entropy increases with temperature. Figure 6 and 7 show behaviour of internal energy and vibrational free energy, respectively, with temperature for all three EuX3 (X=Pd, In and Sn) compounds. It is

clear from figure 6 that the internal energy is highest for EuSn3 than EuIn3 and EuPd3 and it increases continuously with temperature. Figure 7 represents the vibrational free energy at different temperatures which reveals that the free energy decreases with temperature in all cases.

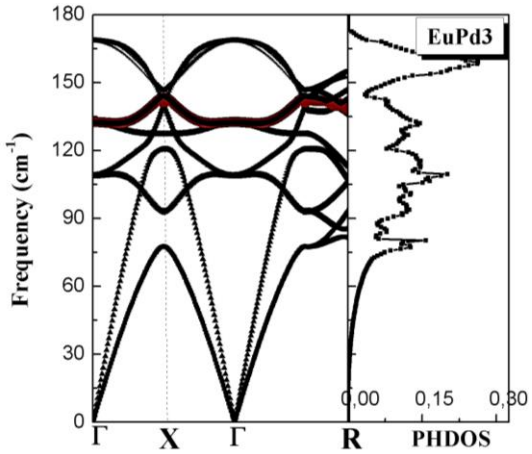

Fig. 1. Calculated phonon dispersion curves and phonon density of states (PHDOS) of EuPd3.

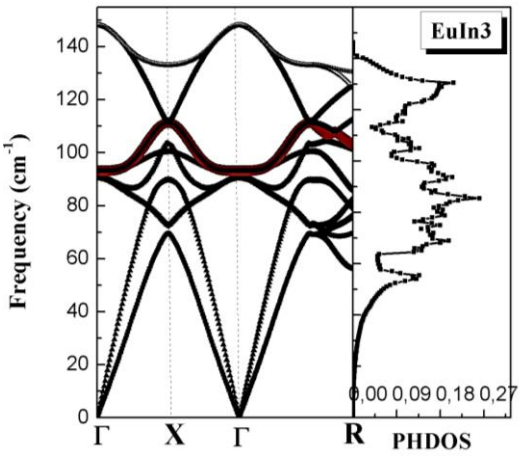

Fig. 2. Calculated phonon dispersion curves and phonon density of states (PHDOS) of EuIn3.

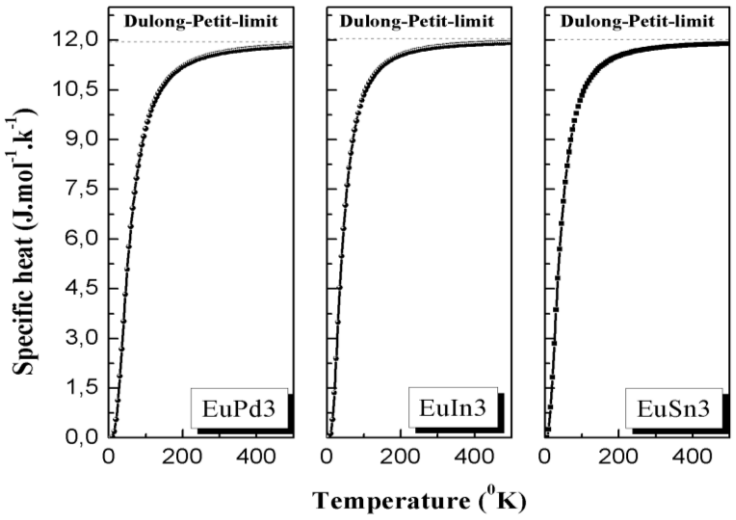

Fig. 4. Variation of specific heat with temperature of EuPd3, EuIn3 and EuSn3 AuCu3-type structure.

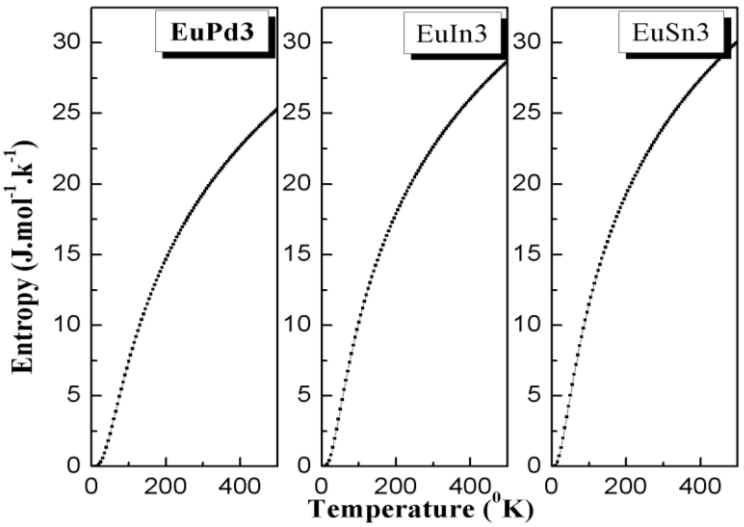

Fig. 5. Variation of entropy with temperature of EuPd3, EuIn3 and EuSn3.

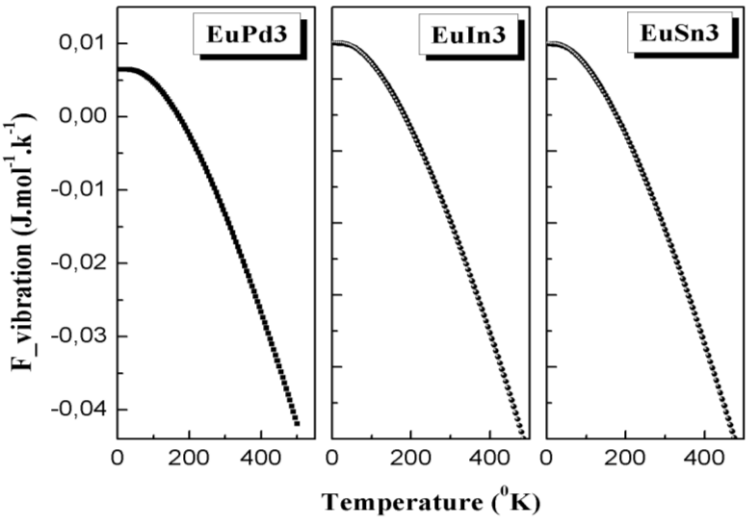

Fig. 7. Variation of vibrational free energy with temperature of EuPd3, EuIn3 and EuSn3.

CONCLUSION

First principle calculations have been performed to investigate theoretically the structural, elastic, electronic properties, vibrational and thermodynamic properties of EuX3 (X=Pd, In and Sn) intermetallic compounds using DFT. The calculated cohesive energy indicate that all compounds are thermodynamically stable and the cohesive energy are minimal for non ferromagnetic structure, indicating that the structure without spin polarized is the most energy stable structure. The ground state lattice parameters are in better agreement with the experiment and theory. The calculated phonon dispersion show no imaginary frequency in entire BZ confirming the dynamical stability of these intermetallic compounds. From phonon dispersion curves, it is found that these compounds are metallic. The acoustic and optical branches overlap each other are not separated by a gap. We have calculated the thermodynamic functions such as specific heat at constant volume, entropy, internal energy and vibrational free energy. We have found that the heat capacity approaches the Dulong–Petit limit at high temperatures. According to Poisson's ratio σ, which implies that all intermetallic compounds are ductile in nature. These parameters are useful to predict new intermetallic materials. The experimental data cannot be found for comparison, and our calculated results provide the reference for future experimental studies.

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