Insight into Thermoelectric Response of LaCoCrGa Quaternary Heusler alloy for Green Energy Devices

Srishti Singh[a], Saleem Yousuf, and Dinesh C. Gupta[b]

Condensed Matter Theory Group, School of Studies in Physics, Jiwaji University, Gwalior
[a)]Corresponding author: srishtisingh0026@gamil.com
b)sosfizix@gmail.com

Abstract. Employing the first-principles simulation packages and post Semi-classical Boltzmann transport theory, the investigation of thermoelectric transport properties of LaCoCrGa Heusler alloy are analyzed. We tried to find the ways of optimizing the thermoelectric response against the chemical potential and its robustness against the pressure variation. The different heat transport coefficients convey the material as *n*-type with nearly independent nature against the pressure. Understanding the variation against the chemical potential convey that the alloy shows higher peaks with increase in temperature, while as the increase of pressure only shifts the peak towards the higher chemical potential. The information of chemical potential could lead to explore the optimized range for application purposes. The overall strength of the all-physical properties conveys its potential stand for the heat recovery devices as green energy source for future technologies.

INTRODUCTION

The immense demand of alternate sources of energy, due to the fast reduction of the fossil fuels have paved new ways in technology developments like that of thermoelectrics [1]. The material candidate for application purposes require the high value of output and durability, that scientific community is engaged in development of new functional materials. Thermoelectric devices require the material candidate that has high value of efficiency with low values of thermal conductance. Experimentalists and theorists are engaged in material simulation with many materials already explored and synthesized. Here, we report on the investigation of the various thermoelectric transport mechanism of the quaternary LaCoCrGa Heusler compound using the density functional theory simulation package under the semi-classical transport theory. It is the exploration of the transport mechanism using the two-current model [2-3]. According to this theory Seebeck coefficient and power factor can be calculated from electronic structures. LaCoCrGa has a narrow band gap in the spin down state, so it is predictable that the minority spin state will give a larger value of S. In this work, we study the thermoelectric properties of LaCoCrGa in the ferromagnetic phase at different pressure range.

COMPUTATIONAL METHOD

We have calculated the ground state properties of rare earth based LaCoCrGa alloy by using the full-potential linearized augmented plane wave (FPLAPW) method, based on the density functional theory framework [4]. The generalized gradient approximation (GGA) in the scheme of Perdew–Burke–Ernzehof (PBE) functional is used for the exchange and correlation interaction by WEIN2k Package [5]. We have placed the precise approximation in muffin tin. There is no charge leakage so we choose the muffin tin radii of 2.54 a.u. for La and 2.50 a.u. for Co and for Cr and Ga as 2.48 a.u. and 2.37 a.u. respectively. Further, for the energy convergence, the cutoff parameter $K_{MAX}R_{MT}$ = 7 Ry is used. The selected energy between the valence and the core states is set to 6 Ry. For Brillouin zone (BZ) integration, we have used a mesh of 14×14×14 k points [6]. The integrated charge difference between two successive iterations as less than 10^{-4} e/a.u.^{3} was obtained and the convergence criterion for energy was set to 10^{-4} Ry.

Result and Discussion

The atomic structure of LaCoCrGa compound is known to crystallize in a cubic of LiMgPdSn-type structure, which has F-43m (216) space group, where La and Co atoms occupy 4c (0.25, 0.25, 0.25) and 4a (0, 0, 0) sites, Cr on 4b (0.5, 0.5, 0.5) and Ga on 4d (0.75, 0.75, 0.75) site, respectively. It has been found that the material stabilizes in the ferromagnetic ground state with lattice parameter of 6.38 Å.

The thermoelectric properties are evaluated using the Boltzmann transport theory under the constant relaxation time approximation [7]. Effect of temperature and pressure on various transport properties i.e., Seebeck coefficient (S) and power factor (PF) with chemical potential has been studied. For selection of the proper doping element for increasing thermoelectric response, the chemical potential behavior with varying temperature and pressure is important. The transport properties are discussed by the Seebeck coefficient (S) conveyed as

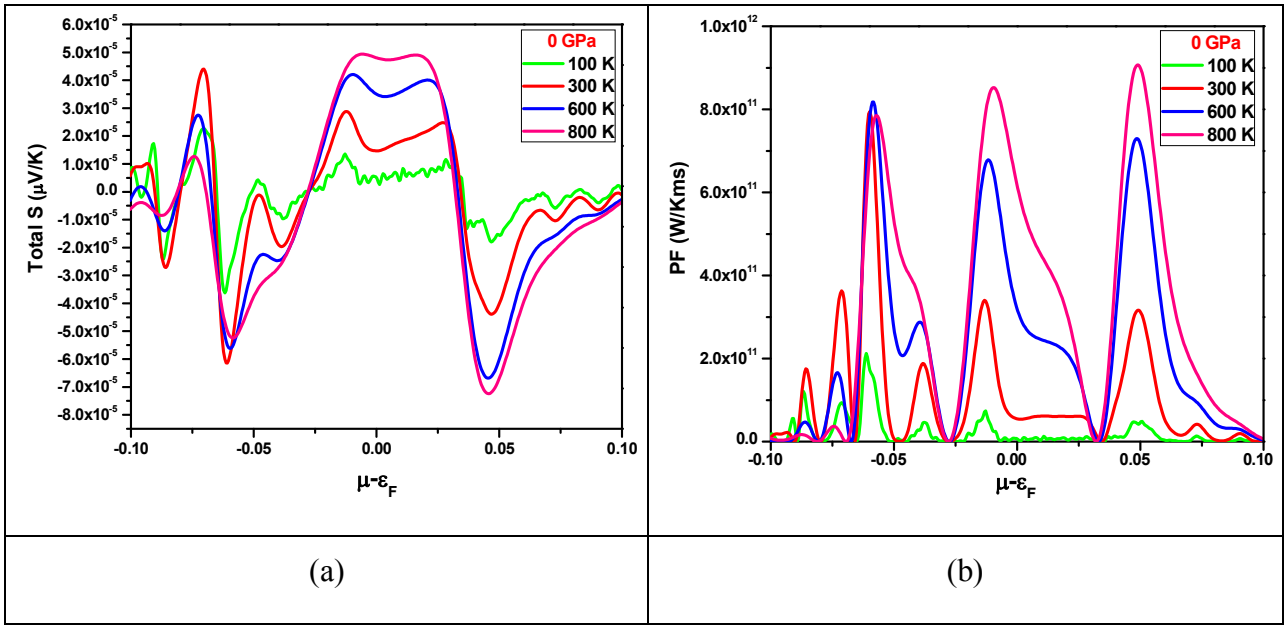
To use this "Old Style Equation" as a "template," highlight the entire line, then use cut and paste to the new location.Note that the equation number will automatically update (increment).

$$S = \frac{8}{3eh^2}\pi^2 k^2{}_B m^*{}_{DOS} T\left(\frac{\pi}{3n}\right)^{2/3} \quad \text{(i)}$$

Here e, h, k_B, m^*_{DOS}, T and n are the electronic charge, Planck constant, Boltzmann constant, density of state (DOS) effective mass, absolute temperature and carrier concentration, respectively.

The variation of the S shows that high temperature results in high values of S and at optimum chemical potential, large value of S, can be obtained. We can also predict that below the Fermi level of compound there is a rapid decrease in S, which designates that there is a drastic decrease in the electronic conductivity and increase in the lattice part of thermal conductivity. Further, we can see that upon increase in pressure, the peak value shifts towards the higher chemical potential values, which designate that there is sharp shift in the electronic properties of the material along the Fermi level.

On seeing the variation of the power factor (PF), we can say that as temperature increases there is an increase in the modulus of PF. While as the variation of the pressure, several peaks occur along the chemical potential. These peaks designate the type of chemical potential behavior of the thermal transport of the carriers along the Fermi level.



(a) (b)

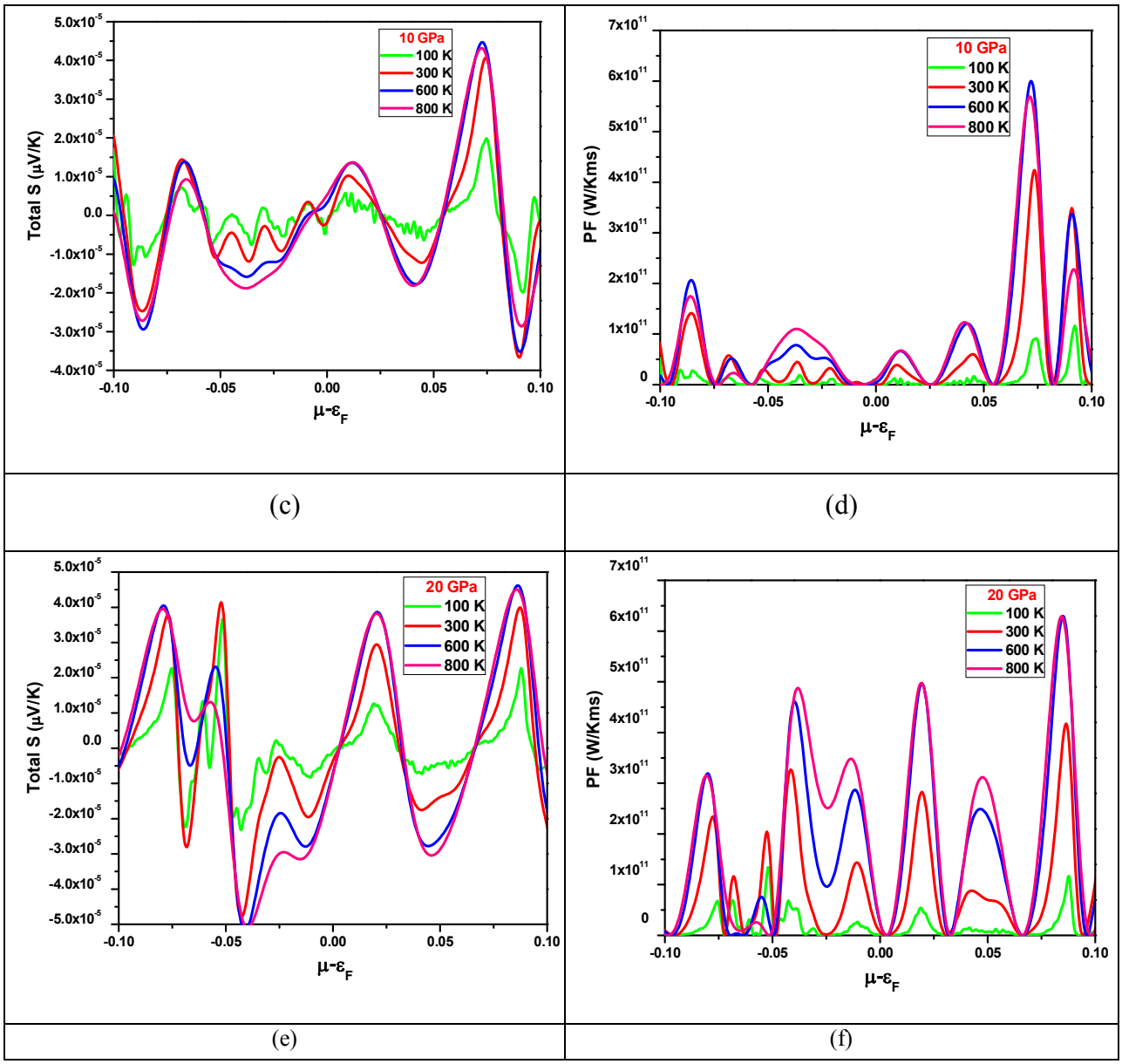


FIGURE 1. Variation of various transport coefficients with temperature, pressure and chemical potential. S defines the Seebeck coefficient (μV/K), PF as Power factor (W/Kms).

CONCLUSION

We have investigated detailed first-principles calculations on LaCoCrGa quaternary Heusler alloys to define the effect of chemical potential with pressure variation on their thermoelectric properties. Understanding the effects of pressure variation deliver a unique tool to develop the best materials for applications perspectives for thermoelectrics and spintronics. Thermoelectric parameters have been considered at different temperature range from 100 to 800 K and pressure range from 0 to 20 GPa. Here we obtain a large Seebeck coefficient along with huge power factor with chemical potential. The material displays the half-metallicnature with descent values of transport coefficients and a way to enhance the thermoelectric response.

ACKNOWLEDGMENTS

The authors, especially Srishti Singh, are thankful to Department of Science and Technology (DST), New Delhi for providing the financial support for this work. (Grant no. SR/WOS-A/PM-82/2017).

REFERENCES

1. S. Datta and B. Das, Appl. Phys. Lett., 56 665 (1990).
2. H.J. Xiang and D.J. Singh, Phys. Rev., B 76 195111 (2007).
3. S. Yousuf and D.C. Gupta, J. Elec. Mater., 47 2468 (2017).
4. K. Schwarz, P. Blaha and G.K.H. Madsen, Comput. Phys. Commun., 147 71 (2002).
5. J.P. Perdew, K. Burke and M. Ernzerhof, Phys. Rev. Lett., 77 3865 (1996).
6. G.K.H. Madsen and D.J. Singh, Comput. Phys. Commun., 175 67 (2006).
7. G. Ding, G. Gao and K. Yao, Sci. Rep., **5** 9567 (2015).