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Comparison Of Optical Properties Of PLZT(96/4/65/35) And PZT(65/35) Ceramics In Rhombohedral Phase

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ABSTRACT

In this paper optical properties of PZT(65/35) composition and PLZT(96/4/65/35) at rhombohedral phase have been studied and compared. Optical properties such as, Dielectrics function, Refraction index and Optical gap and Density of states, have been calculated. The calculations were performed in the frame of density functional theory (DFT), using the full potentional linearized augmented plane wave (FP-LAPW) method with the generalized gradient approximation (GGA). The optical constants were obtained by using the Kramers-Kronig transformations in imagery dielectric tensor. we show in this paper that density of states in PLZT(96/4/65/35) have been more than PZT(65/35) compound above Fermi level and this system able to using as laser active matter due to existence of La-4f level that more localized to Ti-3d.

Keywords – PLZT, PZT, Optical properties, DFT, GGA

I. INTRODUCTION

Lanthanum-doped lead zirconium titanate ferroelectric ceramics $Pb_{1-y}La_y(Zr_xTi_{1-x})O_3$ (PLZT) are very interesting due to their high optical transparency in optical applications. PLZT ceramics are desirable candidates for much device application as light shutters, modulators, color filters, memories and image storage devices [1-3]. It exhibits a much larger electro-optical effect than $LiNbO_3$, which is commonly used in commercial waveguide devices [4].

The incorporation of lanthanum into the lattice enhanced the densification rates of the PZT ceramic, leading to pore-free homogeneous microstructures [5]. This compound can be made in a broad range of composition stoichiometry with a variety and outstanding electro-optical properties. According to the content of La, these compounds are crystallized in different crystal structure phases [6]. The room temperature phase diagram of PLZT system is shown in Fig. 1. The different phases in the diagram are tetragonal ferroelectric, rhombohedral ferroelectric, cubic relaxor ferroelectric, orthorhombic antiferroelectric and cubic paraelectric phases which have been shown as FE_{Tet}, FE_{Rh}, SFE, AFE and PE_{Cubic} respectively [1]. PLZT has perovskite structure with ABO_3 formula. Because the atomic size of La is comparable to Pb, it usually substitutes in Pb position in PZT compound.

II. METHOD OF CALCULATIONS

The calculations were performed in the framework of Density Functional Theory (DFT) [7], using Full Potentional Linearized Augmented Plane Wave (FP-LAPW) method. The Generalized Gradient Approximation (GGA) of Perdew et al. [8] was selected for the exchange-correlation energy in our calculations.

The wave functions were expanded into spherical harmonics within atomic Muffin-Tin (MT) spheres and in the form of plane wave in the remaining space. The MT spheres are supposed not overlap with each other. The cut off energy, which defines the separation of the valence and core states, was chosen about -6 Ry. $R_{MT}\times k_{max}$ and number of k-points in the first Brillouin zone has been chosen to be equal to 6 and 2000, respectively that R_{MT} and k_{Max} are the smallest radius of atoms and the maximum value of the reciprocal lattice vector respectively.

The converge criterion of calculations have been considered when the integrate charge difference between input and output charge is less than 0.0001. Keramers-Kronig transformations have been used for analyzing data [9].This transformation allows us to determine real part of dielectric function by having its imagery part. Real part of dielectric function has been extracted from imaginary part of dielectric function by the Keramers-Kronig's transformations. As know, the imaginary and real part of dielectric function can be written as:

$$Im\,\varepsilon_{ij}(\omega)=\frac{4\pi e^2}{m^2\omega^2}\sum_{c,v}\int dk < C_k|p_i|V_k >< V_k|p_j|C_k > \times\delta(\varepsilon_{ck}-\varepsilon_{vk}-\omega)$$

$$Re\,\varepsilon_{ij}(\omega)=\sigma_{ij}+\frac{2}{\pi}p\int_0^{\infty}\frac{\omega' Im\,\varepsilon_{ij}(\omega')}{\omega'^2-\omega^2}d\omega'$$

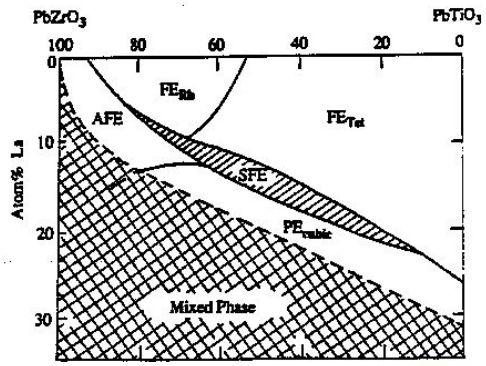

Fig 1. Room temperature phase diagram of PLZT system [1].

III. RESULTS AND DISCUSSIONS

The $Pb(Zr_{1-x}Ti_x)O_3$ compound with x=0.65 is crystalline in Rhombohedral phase with R3m space group. This space group has 6 symmetry operations. When La atoms are doped in PZT compound because of radii of La atom is close to Pb atom, the La atoms are substituted at Pb positions or vacancies of Pb atoms. Due to existence of more localized La-4f level, density of states increase above the Fermi level. These levels are more localized than Ti-3d. The figures 2 and 3 show comparison between total density of states for PZT and PLZT in the rhombohedral phase.

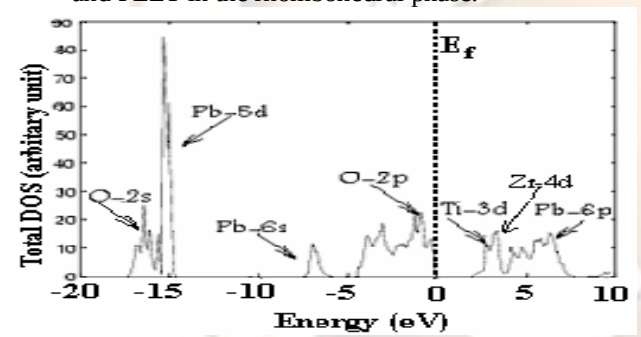

Fig 2. Total density of states PZT(65/35) [10]

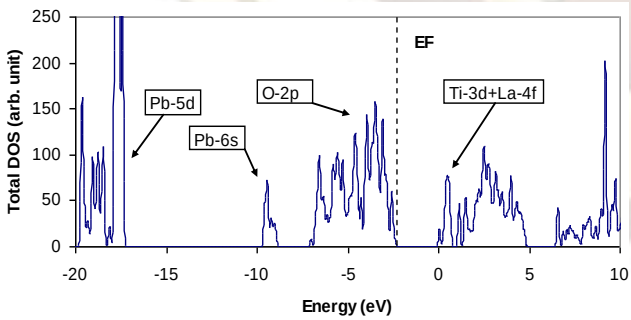

Fig 3. Total density of states PLZT(96/4/63/35)

The increment of density of states above the Fermi level, increase the response probability of electrons in valance band to external electromagnetic fields. Maximum of La-4f is laid about 0.6 eV higher than Ti-3d. Because of La-4f is more localized than Ti-3d so, electrons which are exciting to f state can descend to d state and then transit to the ground state p at the more time.

So, due to these two states are close and also there are O-2p states, this 3 level system able to using as a Laser active matter. The dielectric function is studying for optical responses. Figures 4, 5 show real part of dielectric function for PZT and PLZT. The roots of dielectric function indicate plasma energies. Number of roots for PZT compound are 6 roots and for PLZT, increase to 12 roots. Dielectric function at both cases is not so different at x- and z-direction. PLZT' roots are close between 10 eV to 12 eV. So, plasma oscillations are more.

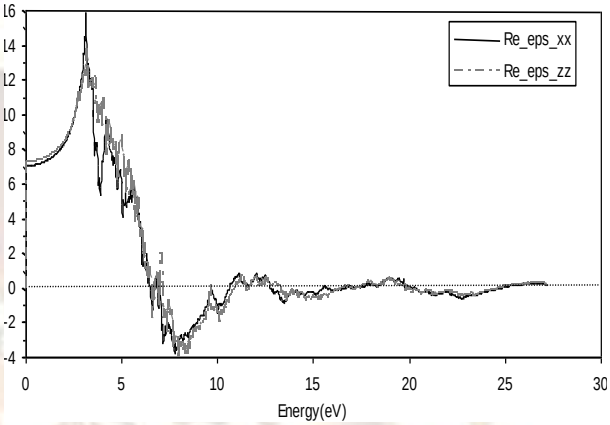

Fig 4. Real part of dielectric function of PZT(65/35)[10]

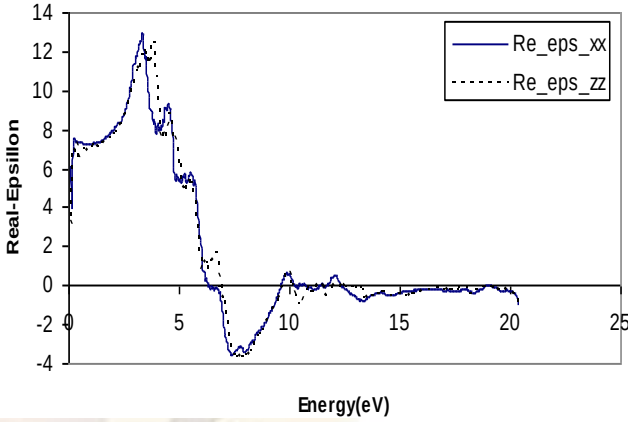

Fig 5. Real part of dielectric function of PLZT(96/4/65/35)

Real part of dielectric function' roots are necessary condition for plasma oscillations, but we need to consider the energy loss function as enough condition. Calculated roots for PLZT are 6.30, 6.47, 9.63, 10.22, 10.42, 10.47, 11.42, 11.53, 11.70, 12.32, 18.88, 19 electron Volt.

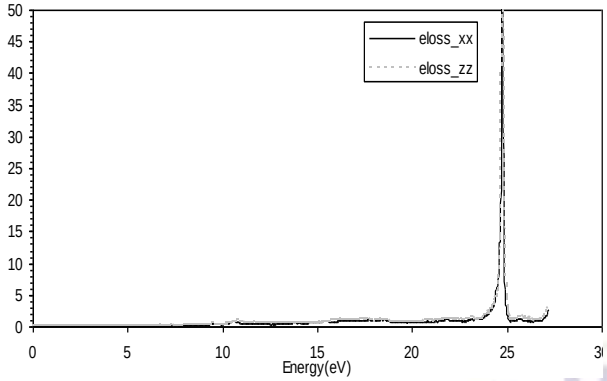

Fig 6. Energy loss function of PZT(65/35)[10]

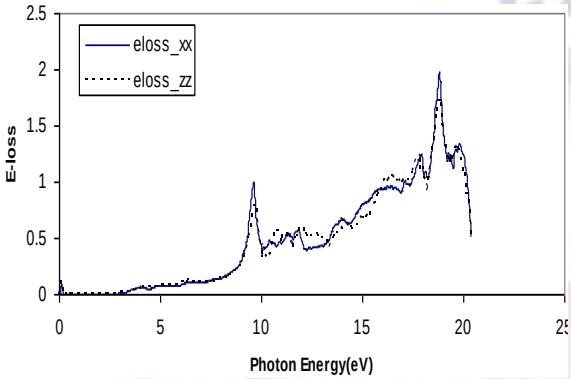

Fig 7. Energy loss function of PLZT(96/4/65/35)

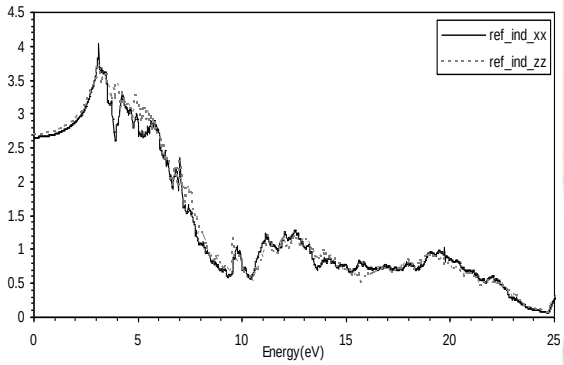

Fig 8. Refractive index of PZT(65/35) to incident photon energy[10]

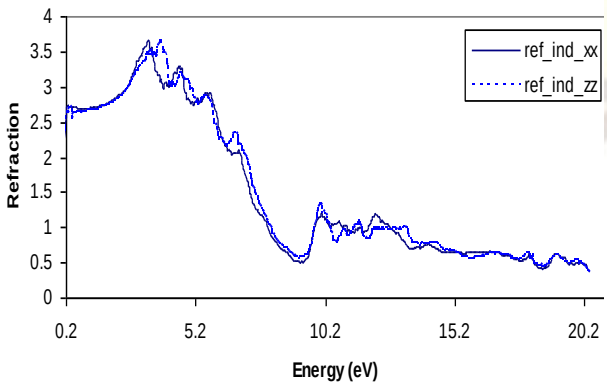

Fig 9. Refractive index of PLZT(96/4/65/35) to incident photon energy

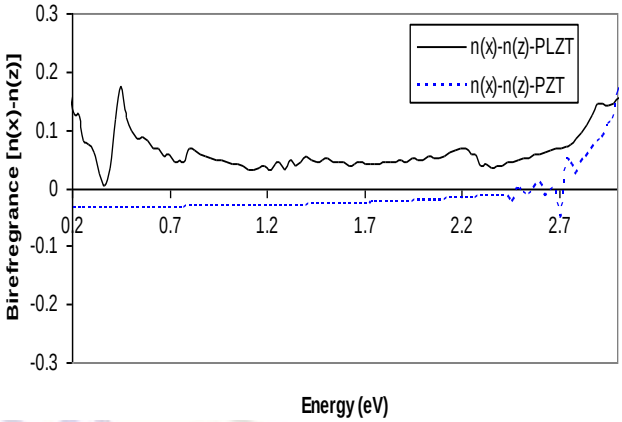

Fig 10. Birefringence of PLZT and PZT

Figures 6, 7 show energy loss function for PZT and PLZT compounds. According to figures 6, 7 we found that existence of La atoms in PZT are caused plasma oscillations occurs at lower energy. Figure 8, 9 show refractive index for PZT and PLZT compounds. The static refractive index and refractive index at 665 nm are summarized in table 1.

Table 1. Comparison static and refractive index at 665 nm

Compound	n_{0x}	n_{0z}	n_x at 665 nm	n_z at 665 nm
PZT	2.69[10]	2.69[10]	2.86[10]	2.88[10]
PLZT	2.59	2.88	2.79	2.75

According table 1 we can found La atom in PZT compound decrease refractive index and birefringence has been increased. Figure 10 show birefringence changes to incident photon energy. Also, we see that fluctuations of refractive index in x- and z-directions less than PZT case however birefringence of PLZT is more than PZT.

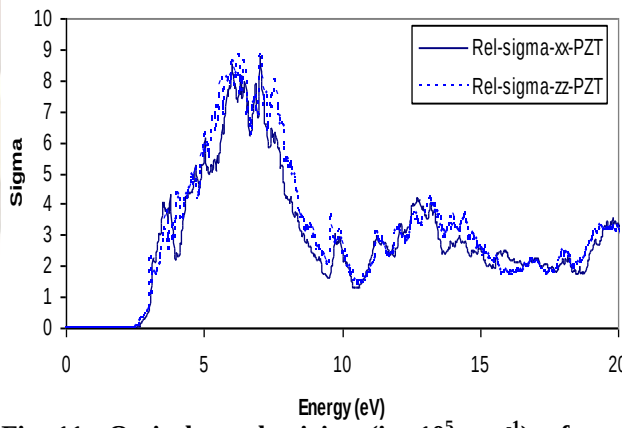

Fig 11. Optical conductivity (in 10^5 cm^{-1}) of PZT(65/35) to incident photon energy[10]

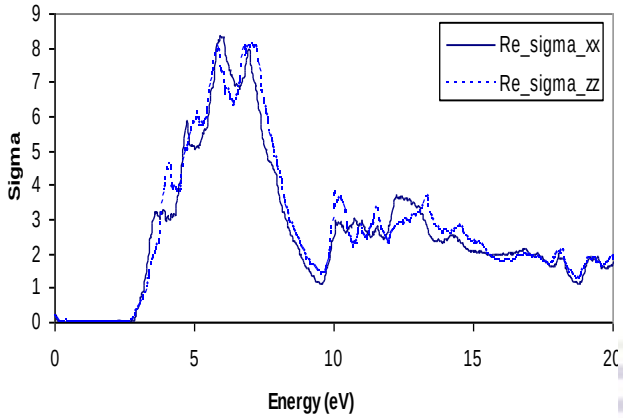

Fig 12. Optical conductivity (in 10^5 cm^{-1}) of PLZT(96/4/65/35) to incident photon energy

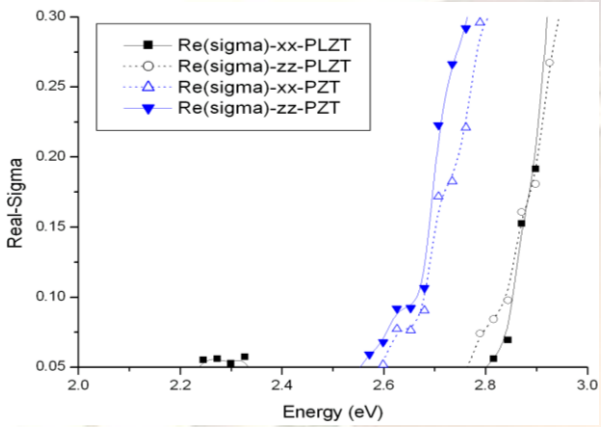

Fig 13. Threshold of optical conductivity for PLZT and PZT

The occupied states electrons are exciting to non occupied states above Fermi level by photon absorption. Figure 11, 12 shows the optical conductivity for PZT and PLZT compounds. Threshold of optical conductivity for PZT is 2.43 eV and 2.6 eV in x- and z-directions respectively. By doping La atom in PZT, threshold of optical conductivity are laid at 2.70 eV and 2.80 eV in x- and z-directions.

IV. CONCLUSION

we show in this paper that density of states in PLZT(96/4/65/35) have been more than PZT(65/35) compound above Fermi level and this system able to using as laser active matter due to existence of La-4f level that more localized to Ti-3d. We see that fluctuations of refractive index in x- and z-directions less than PZT case however birefringence of PLZT is more than PZT. Also, optical gap of PLZT is more than PZT so photons are absorbed at higher energy to PZT compound, however the rhombohedral phase of PLZT is more stable than tetragonal phase but in comparison with tetragonal phase, optical properties has been decreased[11].

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