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Abstract. The lifetimes at higher temperatures on lanthanum-modified lead titanate (PLT) are mainly due to the anharmonic decay of optical phonons into low-energy phonons. The temperature-independent contributions from inherent crystal defects and from boundary scattering become comparable to the phonon scattering contribution at lower temperatures. The thermal interaction is large at higher temperatures which decreases the phonon mean free path, and so the decay lifetime decreases as the temperature of the system is increased. This leads to the increased line width at higher temperatures. We made an estimate of the lifetimes for different concentrations and temperatures in PLT.

Keywords. Lifetime, PLT, ceramic, anharmonic, temperature

INTRODUCTION

The phonon decay processes are related with the lattice anharmonic effects. If anharmonic terms are taken into account in the lattice Hamiltonian, the phonons are no longer eigenstates of the system, and they become finite-lived quasiparticles described by a complex self-energy, which in turn determines the lineshape of the associated Raman peak [1,2]. The study is focused on Raman mode lineshape analysis; in an ideal harmonic crystal, the lineshape is expected to be infinitesimally narrow, but experimental peaks of real mate-rials exhibit an intrinsic width. On the one hand, the presence of various decay channels shortens the phonon lifetimes by anharmonic processes involving multiple phonon recombinations conserving both energy and crystal momentum. On the other hand, impurities and defects disturb the translation symmetry of the harmonic crystal. Therefore they do modify the Raman linewidth by elastic scattering processes that also contribute to the phonon-lifetime shortening scenario [3].

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$Pb_{1-x}La_xTiO_3$ is a perovskite type ferroelectric ceramic which has high pyroelectric coefficient at room temperature. It has been investigated for electrical applications such as piezoelectrics or electrooptics as well as pyroelectrics because its electrical properties can be improved easily by making compositional change or doping with other additives, such as lanthanum [6-13]. The dielectric properties of PbTiO3 were reported, in which the dielectric constant of PbTiO3 increases with increasing temperature in tetragonal phase to reach maximum value at curie temperature (486 °C) ,then decreases with increasing temperature in cubic phase [7].The broadening of the phonon lines is due to anharmonicity of the lattice vibrations. The presence of anharmonic forces in a crystal leads to interactions between the harmonic normal modes. These interactions produce temperature-dependent lifetimes for the normal modes [14]. The analysis of the experimental data showed that the temperature dependencies of the phonon frequencies and linewidths were well described by considering the contributions from thermal expansion and lattice anharmonicity. The anharmonic contribution (phonon-phonon coupling) was found to be due to three-phonon processes [1-2]. Recently experiments have found that negative thermal expansion is a common phenomenon in PbTiO3-based materials, and their negative thermal expansion is affected by various substitutions [9].

Now, the calculate Grüneisen parameters or the isothermal Grüneisen parameter and the isobaric Gruüneisen parameter are defined by the following equations:

$$\gamma_{iT} = \frac{B_0}{\omega_i}\left(\frac{\partial \omega_i}{\partial P}\right)_T \quad \text{and} \quad \gamma_{iP} = -\frac{1}{\alpha \omega_i}\left(\frac{\partial \omega_i}{\partial T}\right)_P \tag{1}$$

depends on B_0, the bulk modulus; ω_i, the Raman wave frequency of the ith vibration mode; and on $\left(\frac{\partial \omega_i}{\partial P}\right)_T$, the variation of the ith frequency of vibration with the pressure and at constant temperature. In the isobaric Grüneisen parameter depends on the thermal coefficient, α; and on $\left(\frac{\partial \omega_i}{\partial T}\right)_P$, the variation of the ith frequency of vibration with the temperature and at constant pressure. When γ_{iT} and γ_{iP} are equal, we say that the phonon is harmonic. This anharmonic part also gives an idea of the phonon lifetime in the material.

The thermal interaction is large at higher temperatures, which decreases the phonon mean free path, and so the decay lifetime decreases as the temperature of the system is increased. This leads to the increased line width at higher temperatures. Assuming the true phonon line shape to be Lorentzian, we can use the simple relation to estimate the relaxation time for the decay process.

$$\tau = \frac{1}{\pi c \Gamma} \tag{2}$$

where c is the velocity of light and Γ is the line width. The estimated values of lifetimes for the decay processes are given in fig.1b. These are just approximate data as the equation is used for a semiconductor (ZnSe) [15] not exactly for ceramics.

The aim of the present study is to measure the frequency and the linewidth (full-width at half-maximum ,FWHM). In this letter, we have used Raman spectroscopy to investigate the temperature dependence of the E1 phonon lifetimes from 100K to 400K. Experimental analysis provides strong evidence of a symmetric decay of E_1 phonons in PLT material.

CONCLUSION AND DISCUSSIONS

The agreement between the measured Raman spectra of these PLT ferroelectrics is remarkable for both the frequency position and the intensity of Raman lines and FWHM. The Raman responses on one of the most complex systems, ferroelectrics, and constitutes a step forward in the reliable prediction of their electro-optical responses. In the stability field of tetragonal PbTiO3 we observed pressure-induced reversals between thermal contraction and expansion. This behavior leads to a mathematically infinite number of crossover points in the pressure-volume-temperature space [9,10]. Modern inelastic scattering techniques with neutrons or photons are ideal for sorting out the anharmonic contribution. Here, we analyze the experimental data of the Raman spectra. Understanding of them

then relies on the development of the fundamental theories of the lifetime of the phonons, considering the peak width Raman, with this; we obtained the energy shifts and lifetime broadening of the interacting phonons.

In the figure 1,a) shows the behavior of the FWHM of the Raman E1 mode, this is a low-frequency mode for PLT material. This behavior can be seen for different concentrations of lanthanum keeping the temperature constant. The behavior of the FWHM with lanthanum concentration was analyzed for constant temperatures of 100K, 300K and 400K. As seen in Figure 1, a), the line width is increased with increasing temperature and concentration.

The full width at half maximum of both peaks increases with increasing temperature. The behavior of the change in Raman peak position with temperature varies for different materials, even for a given material the change in phonon frequency with temperature may differ for different phonon modes. The variation in the Raman peak position of the normal modes with temperature is mainly due to the contribution from thermal expansion or volume contribution and from temperature contribution which results from anharmonicity. The phonon frequency can be expressed as a function of volume and temperature as equation (1). Shifts and the broadening of the Raman modes, with increasing temperature, can change both position and shape of the peak. This broadening is due to the anharmonicity of the lattice vibrations.

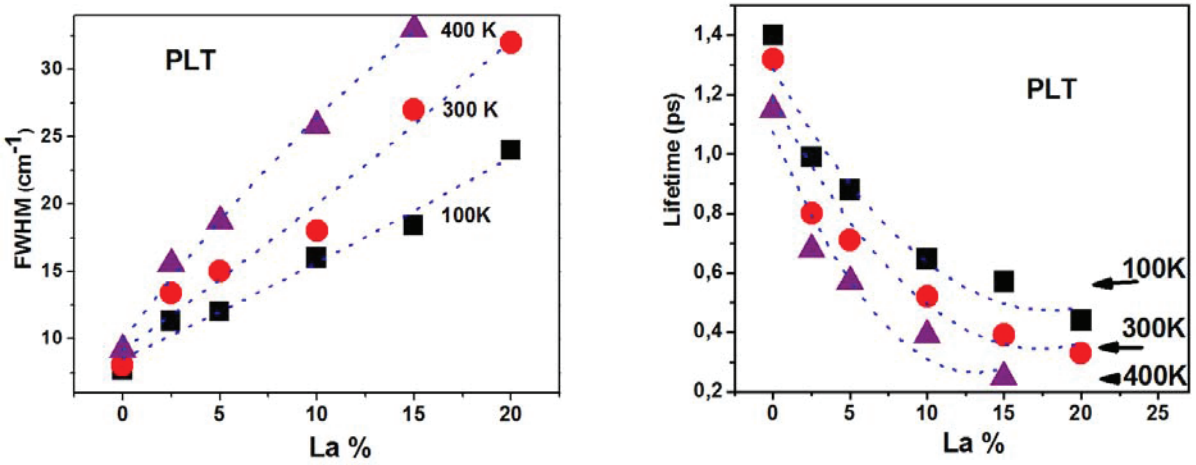

FIGURE 1. (a) The full width at half maximum dependence of the E_1 peak with the concentration (La) for the PT ceramic, for different temperature. Squares for a temperature of 100K and different concentrations of lanthanum, circles for 300K the temperature, and,400K triangles. (b) The phonon lifetime for mode E_1, concentration-dependent and temperature. Squares for a temperature of 100K and different concentrations of lanthanum, circles for 300K the temperature, and,400K triangles.

In Figure 1, b), observe the behavior of the phonon lifetime of E1, these are given for different concentrations keeping the temperature constant in each case. The phonon lifetime is given in picoseconds (ps). By means of a combined model of anharmonic coupling and phonon confinement, the Eg(1,a) Raman spectra were fitted and calculated. The results show that, the temperature-dependent lattice vibration fundamentals are essentially the same for the three samples. The blue shift of Raman frequencies mainly comes from the contribution of three-phonon processes. In order to get good fittings, both the three- and four-phonon processes need to be considered. With increasing temperature, the four-phonon process becomes more important and counteracts the effect of the three-phonon process. The anharmonic-decay-related phonon lifetime increases as concentration decreases, and the smaller concentration of lanthanum have slower anharmonic decay.

The simplest anharmonic approximation, known as the symmetrical three-phonon coupling model, takes into account the optical phonon decay into two phonons with equal energies and opposite momenta. While it provides a fairly accurate description of the phonon temperature dependence in diamond,more general models have been suggested for other materials accounting for anharmonic contributions due to the thermal expansion and/or asymmetric decay into two or more different phonons [16]. We uncover a characteristic temperature dependence of the phonon peak position and linewidth and interpret it in the context of thermal expansion and three-phonon anharmonic decay.

We find that the thermal expansion contribution term accounts for of the total phonon frequency change with temperature. Detailed analysis further reveals that the observed temperature dependence can be well described within a symmetrical three-phonon coupling approximation, where the optical phonon frequency and linewidth Raman are shown. The lifetime of (E1) phonons determines high-temperatures effects; i.e., a sufficiently long E1 phonon lifetime, indicates a lower concentration and lower temperature in the PLT compound.The fundamental mechanism that affects the phonon lifetime is anharmonic decay of a phonon into two or more Brillouin zone phonons subject to energy and wave vector conservation. Finally, the unique anharmonic properties of this mode yield a very long lifetime ultimately limited disorder at low temperature. Then, since the phonon decay is a temperature-dependent process, the study of the temperature dependence of the Raman peak gives insight into the anharmonic interactions and decay channels of the corresponding phonon. We will pursue this approach in a future paper.

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