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Field Dependent Thermal Conductivity of $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ Ferroelectric Perovskites

Gaurav Kumar Aggarwal[1], Ashok Kumar[2], UC Naithani[3]
[1,2,3]Department of Physics, HNB Garhwal Central University, Pauri Campus, Pauri (Garhwal), Uttarakhand., Pin – 246 001, India

Abstract*—Expressions are obtained for the thermal conductivity of some ABO_3 type displacive ferroelectrics in paraelectric phase, using Kubo formalism and double time thermal Green's function technique. The total thermal conductivity obtained for these perovskites which is sum of two terms contributed by optical and acoustical phonons, is in agreement with the experimental results of Steigmeier. The variation of thermal conductivity with temperature and external electric field is discussed, for the case of $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ in presence of anharmonicity. Thermal conductivity decreases with increase of temperature in the cases of all these three perovskites. Also the total thermal conductivity which is the sum of two terms contributed by optical and acoustical phonons is seen to increase with the applied electric field, which is in agreement with the experimental results of Steigmeir. Around the Curie temperature, the thermal conductivity shows an anomalous behaviour and soft mode is held responsible for it.

***Keywords*—Anharmonicity, ferroelectric crystals, paraelectric phase, perovskites, soft mode, thermal conductivity.**

I. INTRODUCTION

Interest in ferroelectric properties, materials and devices has been considerable over the last 10 years. This interest has been driven by the exciting possibility of using ferroelectric thin films for nonvolatile memory applications and new micro electromechanical systems (MEMS).The main interest is in ceramic ferroelectrics and thin films, which are easier to make and which offer a larger variety of easily achievable compositional modifications**.** Ferroelectric substances are useful in many ways such as in Non-Volatile RAMs (memory), Dynamic RAMs (capacitors), Tunable Microwave Devices, Pyroelectric Detectors/Sensors, Optical Waveguides, and Piezoelectric Sensors/Actuators etc.

Perovskite is a family name of a group of materials and the mineral name of calcium titanate (CaTiO3) having a structure of the type ABO_3. Many piezoelectric (including ferroelectric) ceramics such as Barium Titanate (BaTiO3), Strontium Titanate ($SrTiO_3$), Potassium Tantalate ($KTaO_3$), Lead Titanate (PbTiO3), Lead Zirconate Titanate (PZT), Lead Lanthanum Zirconate Titanate (PLZT), Lead Magnesium Niobate (PMN), Potassium Niobate (KNbO3), Potassium Sodium Niobate (KxNa1-xNbO3), and Potassium Tantalate Niobate (K(TaxNb1-x)O3) have a perovskite type structure. These materials may be envisaged by consideration of a non-polar, cubic basic building block (Fig.1).

Thermal conductivity for phonon scattering by anharmonicity in crystals has been a subject of considerable interest in the past. Considerable review of the work is available in the literature [1]-[5]. These studies mostly use the phonon Boltzmann equation, first derived by Peierls [6], in the relaxation time approximation and involved some assumptions which suffer from the usual shortcoming of the kinetic theories as enumerated by Hardy [7] and Allen and Ford [8]. The generalization of phonon Boltzmann equation using equation of motion method for non-equilibrium Green's functions, is used by many workers to derive an expression for the transport coefficient [9]-[12].

The theory of heat conduction in solids acquires a more rigorous theoretical basis when recent theories on phonon transport express the thermal conductivity in terms of correlation functions of energy flux. Many workers have successfully employed this theory to calculate the lattice thermal conductivity of solid using various techniques. Semwal and Sharma [13] and Sharma and Bahadur [14] have theoretically investigated the thermal conductivity of a harmonic Bravais crystal. Although they have calculated both diagonal and non-diagonal contributions of K yet they have not shown the variation of K with temperature and frequency.

Naithani & Semwal [15] and Naithani et. al. [16] have also obtained general expressions for thermal conductivity in an anharmonic crystal but did not choose any specific case. The present study differs from them in view that we have calculated the thermal conductivity here taking the cases of $SrTiO_3$, $BaTiO_3$ and $KTaO_3$.

The properties of ferroelectric crystals are investigated both theoretically and experimentally by many workers. These properties reveal many interesting applications in the ceramic industries, optoelectric device for use in optical communication, memory display, coherent optical processing, modulators, beam reflectors and holographic storage media. There is also some published work on the thermal conductivity of pure and mixed crystals [17]-[22].

The aim of the present work is to study theoretically the variation of thermal conductivity with temperature and external electric field by using the method of double time temperature dependent Green's function in anharmonic ferroelectric crystals such as $SrTiO_3$, $BaTiO_3$ and $KTaO_3$, using a transformed model Hamiltonian for ferroelectric crystal, augmented with anharmonicity up to fourth order and electric dipole moment terms. Present calculated results are compared with the results of other workers.

II. GENERAL FORMULATION

Kubo formula expresses the thermal conductivity as

$$K = \lim_{\varepsilon\to 0}(k_B\beta/3V)\int_0^\infty dt\, e^{-\varepsilon t}\int_0^\beta d\lambda\, < Q(0)\,;Q(t+i\hbar\lambda) > \qquad \dots (1)$$

where k_B, V, $\hbar$, T and Q(t) are the Boltzmann Constant, volume of the crystal, Plank constant divided by 2π, absolute temperature and the heat current operators of lattice, in the Heisenberg representation and the angular brackets <--> indicate the thermal average over the canonical ensemble described by the Hamiltonian

$$<O> = Tr.\,(e^{-\beta H}, O)/\,Tr.\,(e^{-\beta H}) \qquad \dots (2)$$

where Tr. denotes the trace of the expression and H be Hamiltonian of the system.

We write the diagonal part of the flux operator Q as a sum of two operators contributed by optical and acoustical phonons separately, i.e.,

$$Q(t) = \sum_k \hbar\,\omega^a_k v^a_k N^a_k(t) + \sum_k{}' \hbar\,\omega^o_k v^o_k N^o_k(t)$$
$$= \sum_{k,\lambda} \hbar\,\omega^\lambda_k v^\lambda_k N^\lambda_k(t), \qquad \dots (3)$$

where o and a are used for optical and acoustical modes respectively. Single index k represents K_s, denoting polarization and v^λ_k represents the velocity of sound for optic (k = o) and acoustical (k = a). The diagonal contribution to the thermal conductivity from equation (3), becomes

$K = K^a + K^o = \Sigma K^\lambda$; λ=o, a;

where,

$$K^\lambda = \lim_{\varepsilon\to 0}(\hbar^2 k_B\beta^2/3V)\sum \omega^\lambda_k\,\omega^\lambda_{k'}\,v^\lambda_{kv}{}^\lambda_k\int_0^\infty dt\, e^{-\varepsilon t}\int_0^\beta d\,\lambda' <N^\lambda_k(0);\, N^\lambda_{k'}(t+i\hbar\lambda)>, \qquad \dots (4.1)$$

$$<N^\lambda_k(0);\, N^\lambda_{k'}(t')> = <a^{\lambda\dagger}_k(0)\, a^\lambda_k(t)> <a^\lambda_k(0)\, a^{\lambda\dagger}_k(t)>. \qquad \dots (4.2)$$

Equation (4.2) is obtained after the use of a decoupling scheme discussed in a previous study [23].

III. GREEN'S FUNCTION AND HAMILTONIAN

In order to calculate the correlation function (4.2), we define the Green's functions for system as

$$G^\lambda_{k,k'}(t-t') = <<A^\lambda_k(t):A^\lambda_{k'}(t')>>$$
$$= i\theta(t\,t') <[A^\lambda_k(t):A^\lambda_{k'}(t')]>, \qquad \dots (5)$$

where θ (t) is the Heaviside step function and $A^\lambda_k = a^\lambda_k + a^{\lambda\dagger}_{-k} = A^{\lambda\dagger}_{-k}$.

The modified transformed Hamiltonian which induced the fourth order anharmonicity due to interaction of the soft mode coordinates, resonant interaction and the scattering terms in presence of external electric field is exactly the same as given by eq. (3.15) of our previous study [16].The notation used are the same and in the same sense.

Writing the equation of motion for the acoustical Green's function $G^a_{k,k'}$ (t- t') with the help of modified Hamiltonian, Fourier transforming and writing it in the Dyson's equation form, one obtain it value

$$G^a_{k,k'}(\omega+i\varepsilon) = \omega^a_k\delta_{kk'}/\Pi[\omega^2-(\bar{\varpi}^a_k)^2(\omega)+2i\omega^a_k\Gamma^a_k(\omega)], \qquad \dots (6)$$

In a similar manner we obtain following values for optical Green's function,

$$G^o_{k,k'}(\omega+i\varepsilon) = \omega^o_k\delta_{kk'}/\Pi[\omega^2-(\bar{\varpi}^o_k)^2(\omega)+2i\omega^o_k\Gamma^o_k(\omega)], \qquad \dots (7)$$

We can combine equation (6) and (7) as,

$$G^\lambda_{k,k'}(\omega+i\varepsilon) = \omega^\lambda_k\delta_{k,k'}/\Pi[\omega^2-(\bar{\varpi}^\lambda_k)^2(\omega)+2i\omega^\lambda_k\Gamma^\lambda_k(\omega)], \qquad \dots (8)$$

and

$$(\bar{\varpi}^\lambda_k)^2(\omega) = (\omega^\lambda_k)^2+2\omega^\lambda_k\,\Delta^\lambda_k(\omega), \qquad \dots (9)$$

Eqn. (8) can be rewritten as,

$$G^\lambda_{k,k'}(\omega+i\varepsilon) = \{\omega^\lambda_k\delta_{k,k'}/\Pi\}\times[\omega^2-(\bar{\varpi}^\lambda_k)^2(\omega)-2i\omega^\lambda_k\Gamma^\lambda_k(\omega)]/[\{\omega^2-(\bar{\varpi}^\lambda_k)^2(\omega)\}^2+4(\omega^\lambda_k)^2(\Gamma^\lambda_k(\omega))^2], \qquad \dots (10)$$

The spectral density function $J^\lambda_k(\omega)$ is given by,

$$J^\lambda_k(\omega) = \lim_{\varepsilon\to 0}\{\,i/(e^{\beta\hbar\omega}-1)\}[\,G^\lambda_{k,k'}(\omega+i\varepsilon) - G^\lambda_{k,k'}(\omega-i\varepsilon)]. \qquad \dots (11.1)$$

We are also able to calculate the correlation function (4.2), using the following relations:

$$<a^\lambda_k{}^\dagger(t'):\, a^\lambda_k(t)> = (1/4)\int_{-\infty}^{\infty}(1+\omega/\,\bar{\varpi}^\lambda_k)^2\, J^\lambda_k(\omega)e^{-i\omega(t-t')}\,d\omega, \qquad \dots (11.2)$$

Substituting the value of $G^\lambda_{k,k'}$ into equation (11) and then in equation (11.2), we get

$$<a^\lambda_k{}^\dagger(t'):a^\lambda_{k'}(t)> = (\delta_{k,k'}/\Pi)\int_{-\infty}^{\infty} d\omega[\{(\omega+\bar{\varpi}^\lambda_k)^2\,\Gamma^\lambda_k(\omega)e^{-i\omega(t-t')}\}/(e^{\beta\hbar\omega}-1)\{(\omega^2-(\bar{\varpi}^\lambda_k)^2)^2+4(\bar{\varpi}^\lambda_k)^2(\Gamma^\lambda_k(\omega))^2\}]. \qquad \dots (12)$$

IV. THERMAL CONDUCTIVITY

Now substituting the value of the correlation function in equation (4.1) and then we get the diagonal contribution to the thermal conductivity is given by

$$K=\sum_{\lambda} K^{\lambda} = (\hbar^2 k_B \beta^2/3\Pi V) \sum_{k,\lambda} (\omega^{\lambda}_k)^2 (v^{\lambda}_k)^2 \int_{-\infty}^{\infty} d\omega \, \{e^{\beta\hbar\omega}/(e^{\beta\hbar\omega}-1)^2\} \times [(\omega+\bar{\varpi}^{\lambda}_k)^4 (\Gamma^{\lambda}_k(\omega))^2/\{(\omega^2-(\bar{\varpi}^{\lambda}_k)^2)^2+4(\bar{\varpi}^{\lambda}_k)^2(\Gamma^{\lambda}_k(\omega))^2\}^2]. \quad \text{... (13)}$$

For small values of $\Gamma^{\lambda}_k(\omega)$, the integrand of the equation (13) is peaked around $\omega \approx \varpi^{\lambda}_k$ and integration gives thermal conductivity as,

$$K=(\hbar^2 k_B \beta^2/3V) \sum_{k,\lambda} (\omega^{\lambda}_k)^2 (v^{\lambda}_k)^2 [\exp(\beta\hbar\bar{\varpi}^{\lambda}_k)/\{\exp(\beta\hbar\bar{\varpi}^{\lambda}_k)-1\}^2] \times [1/\Gamma^{\lambda}_k(\omega)] \times [(\omega^{\lambda}_k+\bar{\varpi}^{\lambda}_k)/2\omega^{\lambda}_k]^4. \quad \text{... (14)}$$

In the pure crystal case the higher order anharmonic terms may be neglected, then equation (14) for thermal conductivity is reduced to give the familiar expression discussed by Carruthers [4] and Klemens [2] based on the Boltzmann transport equation,

$$K = \left(\frac{\hbar^2 k_B \beta^2}{3V}\right) \sum_{k,\lambda} (\omega^{\lambda}_k)^2 (v^{\lambda}_k)^2 \; \frac{\exp(\beta\hbar\varpi^{\lambda}_k)}{\{\exp(\beta\hbar\varpi^{\lambda}_k)-1\}^2} \cdot \frac{1}{\Gamma^{\lambda}_k(\omega)} \quad \text{... (15)}$$

4.1. TEMPERATURE AND ELECTRIC FIELD DEPENDENCE OF THERMAL CONDUCTIVITY

It is now clear from equation (15) that the temperature dependence of thermal conductivity is a clear consequence of the temperature dependence of $\Gamma^{\lambda}_k(\omega)$, so before discussing the former it is necessary to discuss latter. In high temperature limit, temperature dependence of $\Gamma^{\lambda}_k(\omega)$ can be expressed as,

$$\Gamma^{\lambda}_k(\omega) = A + BT + CT^2 \quad , \quad \text{... (16)}$$

where the coefficient A is independent of temperature and depends upon impurity only and is zero for a pure crystal and the coefficients B and C are the coefficients of T and T^2 respectively. They depend upon third and fourth order anharmonic coefficients in the potential energy expression. Thus for pure crystal equation (16) can be reduce to

$$\Gamma^{\lambda}_k(\omega) = BT + CT^2 + DE^2T, \quad \text{... (17)}$$

At very high temperature limit i.e. well above the Curie temperature the coefficient C becomes negligible small, so the temperature dependence of $\Gamma^{\lambda}_k(\omega)$ can be expressed as:

$$\Gamma^{\lambda}_k(\omega) = BT + DE^2T = (B+DE^2)T \quad \text{... (18)}$$

The temperature and electric field dependence of the soft mode frequency is given by:

$$(\omega^{\lambda}_k) = \Omega \approx \Omega_{k,E,T} \approx \Omega_{k,T}(E^2+1)^{1/2}(T-T'_c)^{1/2}/\,(T-T_c)^{1/2} \quad \text{... (19)}$$

Where T'_c is the changed Curie temperature in presence of electric field.

It is clear from equations (15), (18) and (19) that at very high temperature thermal conductivity of a pure crystal varies inversely with temperature and electric field, and can be expressed as,

$$K = \frac{G'(E^2+1)(T-T'_C)}{T(1+G''E^2)(T-T_C)} \quad \text{... (20)}$$

Where $G'' = D/B$, and $G' = D$, also D is a constant. The values of D for $SrTiO_3$ (3132 N/m), for $BaTiO_3$ (1200 N/m) and $KTaO_3$ (6000 N/m) have been calculated by reference [25] by best fit of data. With the help of equation (20), we have calculated the thermal conductivity of pure anharmonic $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ crystals in their Paraelectric phase. These calculated values are plotted as given in figures 2, 3 and 4 respectively. Figures 2, 3 and 4 show the variation of thermal conductivity with temperature in presence of applied electric field strengths for $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ crystals in their paraelectric phase respectively.

V. RESULT, DISCUSSION AND CONCLUSION

In the present study we have discussed the temperature and electric field dependence of thermal conductivity of ABO_3 type displacive ferroelectrics (such as $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ etc.) in the paraelectric phase using double time temperature dependent thermal Green's function technique.

The technique and approach used to obtain the expressions for thermal conductivity in these perovskites is different from other workers. It has involved numerical estimation of the thermal conductivity in $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ to show the variation in thermal conductivity with temperature and frequency. It is clear from equation (15) that temperature and electric field dependence of thermal conductivity is a clear consequence of temperature and electric field dependence of $\Gamma^{\lambda}_k(\omega)$ and ω^{λ}_k. Now in the higher temperature case $\Gamma^{\lambda}_k(\omega)$ and ω^{λ}_k varies as temperature and electric field. Figures 2, 3 and 4 show the temperature and electric field dependence of thermal conductivity in paraelectric phase in $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ respectively.

It is evident from these figures that thermal conductivity decreases with increase of temperature in $SrTiO_3$, $BaTiO_3$ and $KTaO_3$. This decrement is large in low temperature case but small in high temperature case. These results are in good agreement with the results of other workers [2], [4], [14]-[16], [26]-[27]. N^o_o become anomalously large. This anomalous increase in the value of N^o_o will decrease phonon half with vanishingly and hence thermal conductivity will be anomalously large near the Curie temperature. Soft mode is held responsible for this variation of thermal conductivity.

The effect of an increasing electric field is to increase the total thermal conductivity. This agrees with the experimental results of Steigmeir. The increment in thermal conductivity is appreciable in the vicinity of Curie temperature for all three cases. This increment in thermal conductivity becomes very small in higher temperature range. In the vicinity of Curie temperature T_C the thermal conductivity increases anomalously in agreement with the maximum in conductivity vs. temperature curve for $SrTiO_3$ near 32 K (T_C) observed by Sievers and Pohl. The soft mode is held responsible for this effect.

Figures 2, 3 and 4 also show the electric field dependence of thermal conductivity in paraelectric phase in $SrTiO_3$, $BaTiO_3$ and $KTaO_3$ respectively.

This variation is similar to the result of others [1], [15]-[16], [25]-[26].

Recently we have applied double time thermal Green's function technique in obtaining expression for electric field dependent inelastic scattering cross section of neutrons [28] and specific heat [29] in $BaTiO_3$, $SrTiO_3$ and $KTaO_3$ displacive ferroelectric perovskites.

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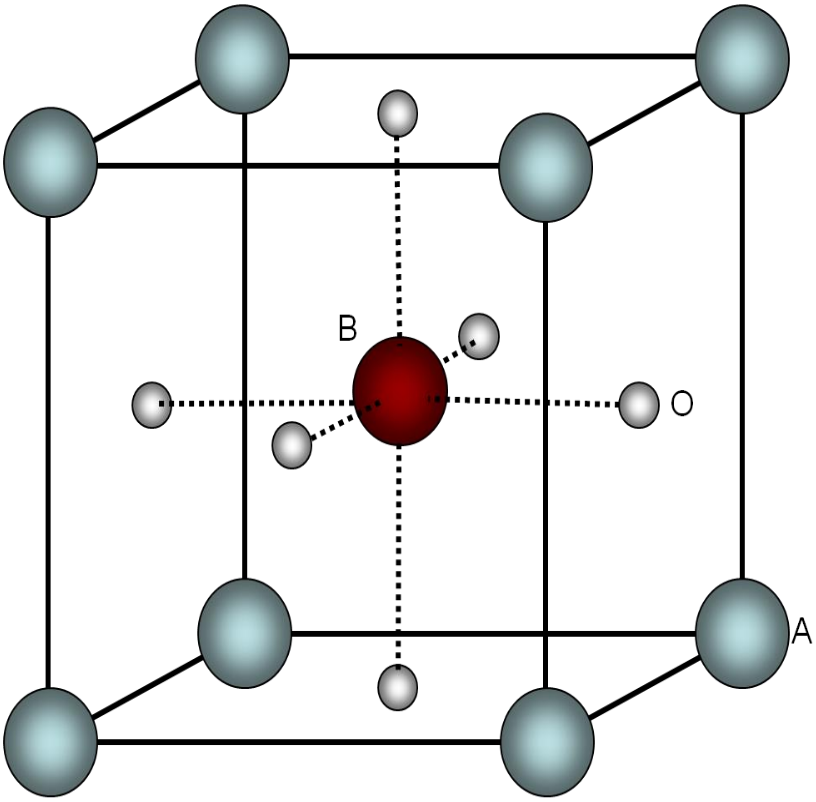

Fig. 1 - ABO_3 type Ferroelectric perovskites' structure

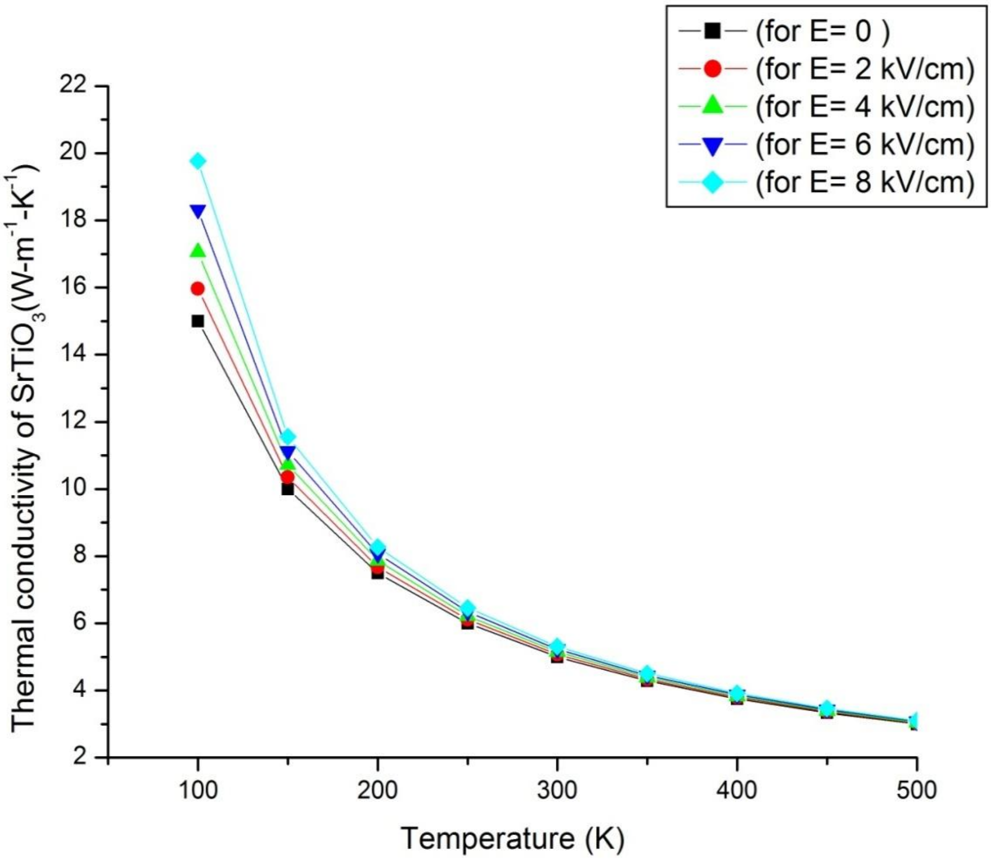

Fig. 2 Thermal Conductivity versus temperature (in K) for $SrTiO_3$ at different Electric Fields

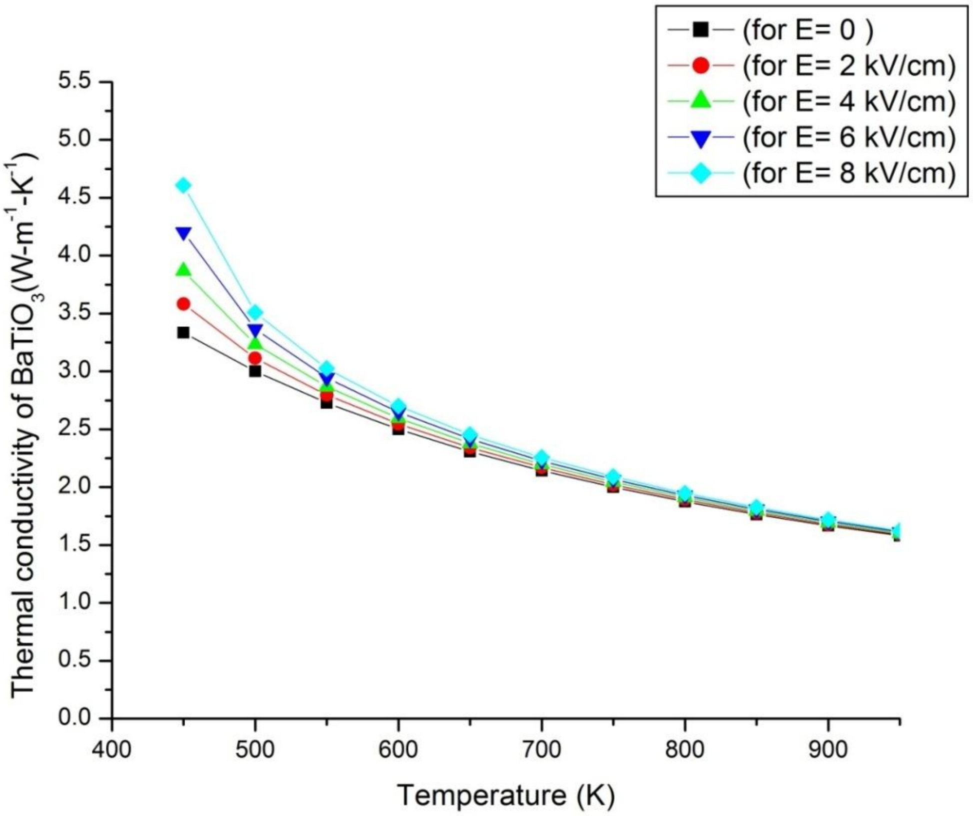

Fig. 3 Thermal Conductivity versus temperature (in K) for BaTiO$_3$ at different Electric Fields

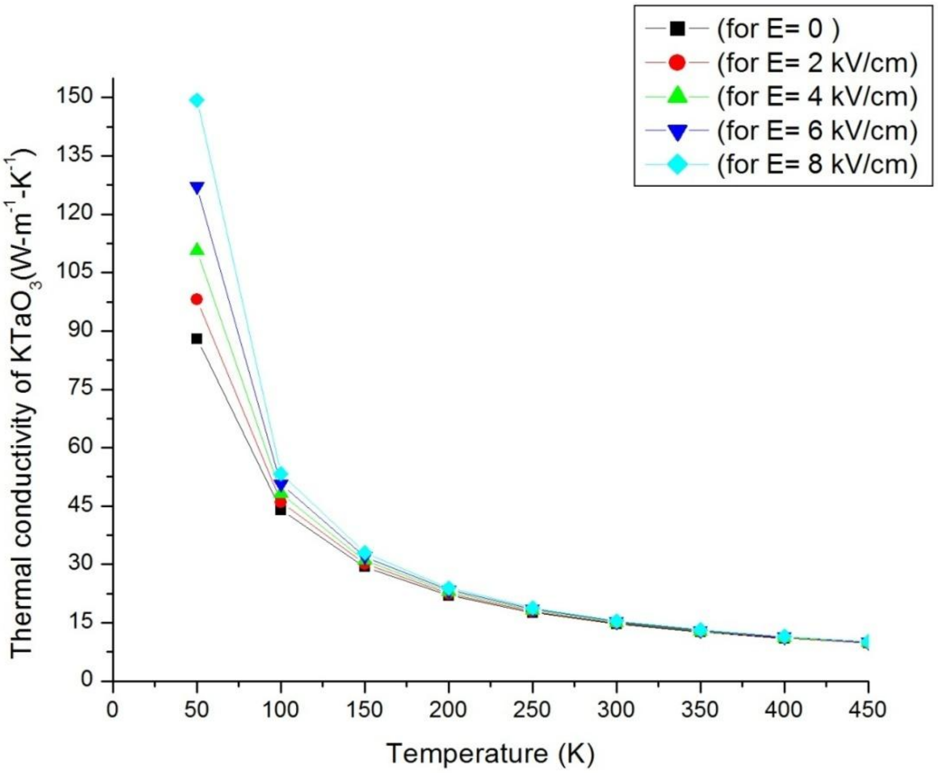

Fig.4 Thermal Conductivity versus temperature (in K) for KTaO$_3$ at different Electric Fields

Authors

Mr. Gaurav Kumar Aggarwal is pursuing D.Phil in Physics from H.N.B. Garhwal University, Srinagar, Uttarakhand, India. He received his M.Sc. degree in Physics from H.N.B. Garhwal University. His area of interest is Condensed Matter Physics and Electronics. He is actively involved in teaching and research. His field of research is ferroelectric materials particularly displacive ferroelectric perovskites.

Dr. Ashok Kumar obtained his PhD in Physics from H.N.B. Garhwal University, Srinagar, Uttarakhand, India. His area of interest is Condensed Matter Physics. He is actively involved in theoretical research in the field of ferroelectrics. He has published more than 30 research papers in reputed journals. His recent research is focussed on designing and development of ferroelectric perovskites.

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Prof. U.C. Naithani presently holds the chair of Professor and Head, Deptt. of Physics, H.N.B. Garhwal University, Pauri Campus, Pauri, Uttarakhand, India. He did his PhD in Condensed Matter Physics. He is actively engaged in teaching and research since last 37 years. His major field of research is in the area of dielectric materials particularly ferroelectric perovskites. He has produced a dozen of PhD scholars and more than 85 research papers are to his credit in this field.