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Phonon Frequency Spectrum through Lattice Dynamics and Normal Coordinate Analysis of HTSC Tl₂Ca₃Ba₂Cu₄O₁₂

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Abstract

The lattice dynamics of the high temperature superconductors $Tl_2Ca_3Ba_2Cu_4O_{12}$ has been investigated on the basis of the three body force shell model (TSM). The various interactions between ions are treated on a general way without making them numerically equal. The phonon frequency at the zone centre of Brillouin zone are presented and the vibrational assignments are discussed. Further the normal coordinate calculation has also been employed to study the vibrational analysis of this compound. The normal coordinate analysis of the superconductor Brillouin zone has been calculated by using Wilson's F-G matrix method which is useful for the confirmation of our present investigation. The vibrational frequencies and the potential energy distribution (PED) of the optically active phonon modes are also presented.

Keywords: Lattice dynamics, Normal Coordinate, Brillouin zone, Three shell Force shell Model, Tl2Ca3Ba2Cu4O12, Zero Wave Vector, high-temperature superconductors, Raman, infrared. Phonon Frequency, etc.

1. Introduction

The study of the lattice dynamics of the high-temperature superconductors is of importance not only for the overall physical characterization of these compounds but also for an assessment of the role played by the phonons i.e., the superconducting phenomenon. A lattice dynamical study requires the knowledge of the crystal structure and the particle interactions. Usually the crystal structure is determined using X-ray diffraction. For particle interactions one has to use models, which represent the characteristic of the electronic structure and its effect on ionic interaction in a relevant manner. In lattice dynamics the ionic interactions are expressed interms of force constants. Cox et.al. (1988) have refined the structure of high-temperature superconductors $Tl_2Ca_3Ba_2Cu_4O_{12}$ using neutron and powder diffraction data. Raman and infrared active modes of $Tl_2Ca_3Ba_2Cu_4O_{12}$ have been calculated by Kulkarni et al.(1989) in the frame work of shell models. Belosiudov et al. (1991) have calculated vibrational spectrum of $Tl_2Ca_3Ba_2Cu_4O_{12}$ using interactions. A high resolution neutron diffraction study on $Tl_2Ca_3Ba_2Cu_4O_{12}$ using interactions.

In the present work we start with a more general approach in the framework of the three body-force shell model (TSM) with R # S # T to calculate the lattice dynamics frequencies. The values of the phonon frequencies calculated in this present work at zone center by the three body force shell model is in good agreement with the available Raman and infrared values. Further a normal coordinate analysis has also been attempted for the superconductor $Tl_2Ca_3Ba_2Cu_4O_{12}$ using Wilson's F-G matrix method for the confirmation of our present investigations. The vibrational frequencies and the potential energy distribution (PED) of the optically active modes are also reported.

2. Theoretical Consideration

2.1 Lattice dynamics of $Tl_2Ca_3Ba_2Cu_4O_{12}$ based on the shell model

The calculation of lattice dynamical vibration frequencies of $Tl_2Ca_3Ba_2Cu_4O_{12}$ system is performed by three-body force shell model (TSM) calculations. In the shell model calculation the equations of the motion for the code coordinate U and the shell coordinate W are expressed as follows:

$$-M\varpi^{2}U = (R + ZC'Z) U + (T + ZC'Y) W$$

$$0 = (T' - YC'Z) U (S + K + YC'Y) W$$
 (1)

With ZC'Z = Z[Z + 2f(a)] C + V where M, Z and Y are diagonal matrices representing the mass ionic charge on the shell. R, S and T are matrices specifying short–range core-core, shell-shell and core-shell interactions, respectively. V is the matrix describing the three-body overlap interactions and

f(a) is related to overlap integrals of electron wave function. U and W are the vectors describing the ionic displacements and deformations respectively.

In the earlier approaches the R, S and T elements were considered to be equal to one another. In the present investigation, we have started with an approach such that R # S # T (Moha., et al. 1986). That is the various interactions between the ions are treated in a more general way without making them numerically equal. The dynamical matrix of the model consists of long-range Coulomb and three-body interactions and the short-range overlap repulsions. The off-diagonal elements of this matrix along the symmetry directions chain a completely new term having a significant contribution for unequal R, S and T.

The lattice dynamical calculation of high-temperature superconductors is explained using an interionic potential consisting of long-range Coulomb part and a short-range Potential of Born-Mayer from (Onari,S., et al. 1988)

(2)

$$a_{ij} = a_{ij} \exp(-b_{ij}r)$$

Where i, j label the ions and r is their separation. The parameters a_{ij} and b_{ij} are the pair potentials and the parameters Y and K determine the electronic polarizabilities. The parameters used in the present calculations are given in Table 1. Phonon frequencies are calculated using the force constants derived from the interionic potential. Following (Leaner et al. 1982) interionic pair potentials for short-rang interactions can be transferred from one structure to another in similar environments. The force constants evaluated by this method are in good agreement with the evaluated values (Onari,S., et al 1990; Thomsen, C., et al.1987).

2.2 Normal Coordinate Analysis of the Zero Wave Vector Vibrations of Tl₂Ca₃Ba₂Cu₄O₁₂

The high Tc superconductor $Tl_2Ca_3Ba_2Cu_4O_{12}$ System crystallizes in the simple tetragonal (st) system, which belongs to the space group $P4/_{mmm}(D'_{4h})$ The simple tetragonal (st) unit cell of $Tl_2Ca_3Ba_2Cu_4O_{12}$ and the numbering of the atoms are shown in Fig.1. The 21 atoms of the unit cell yield a total of 42 optical vibrational modes. All the above calculations are made at q = 0. Once of A_{2u} and E_u modes corresponds to acoustic vibrations with frequency $\omega = 0$. These normal modes are distributed as follows.

$A_{2u} + E_u$	\Box from the motion of a TI atoms
$A_{lg} + E_g + A_{2u} + E_u$	\Box from the motion of 2 Ca atoms
A2u + Eu	\Box from the motion of a Ca atoms
$A_{lg} + E_g + A_{2u} + E_u$	\Box from the motion of 2 Ba atoms
$2A_{lg} + 2E_g + 2A_{2u} + 2E_u$	\Box from the motion of 4 Cu atoms
$2A_{1g} + 2B_{1g} + 2E_g + 2A_{2u} + 2B_{2u} + 3E_u$	\Box from the motion of six O (1) atoms along c-axis.
$A_{1g}+2 E_g+A_{2u}+B_{2u}+2E_u$	\Box from the motion of four O(2) atoms along b-axis
Eg + A2u + B2u + Eu	\Box from the motion O (4) atoms along a-axis

Subtracting the translation modes A2u + B2u + Eu the q = 0 optical modes involved in an irreducible representation are as follows.

$$\Gamma \text{opt} = 7\text{A1g} + 2\text{B1g} + 9\text{Eg} + 10\text{A2u} + 2\text{B2u} + 12\text{Eu}$$
(3)

The species belonging to A1g and Eg Raman active modes whereas A2u Bu are infrared active modes. The A2u and A_{1g} modes involve displacement along crystallographic c-axis, the B_{2u} and E_g modes along the b-axis and E_u modes along the a-axis. The normal coordinate calculation was performed using the programs GMAT and FPERT given by (Fuhrer et al.1976). The general agreement between the evaluated and observed normal frequencies of $Tl_2Ca_3Ba_2Cu_4O_{12}$ is good. The calculated force constants using the above programs are given in Table 2. It is interesting to note that the evaluated frequencies given in Table 3. agree favourably with the experiment values.

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To check whether the chosen set of vibrational frequencies makes the maximum contribution to the potential energy associated with the normal coordinate frequencies of the superconducting material, the potential energy distribution was calculated using the equation.

(4)

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$$PED = (F_{ii} \quad L^{2}_{ik}) / \lambda_{k}$$

Where Potential Energy Distribution (PED) is the combination of the i-th symmetry coordinate to the potential energy of the vibration whose frequency is v_k , F_{ij} are potential constants, L_{ik} , are L matrix elements and $\lambda_k = 4 \pi^2 C^2 v_{k}^2$.

3. Results and Discussion:

3.1 Lattice Dynamical Calculation using Shell Model(Tl₂Ca₃Ba₂Cu₄O₁₂)

The Lattice Dynamical calculations based on modified TSM reproduce the observed frequencies of Raman and infrared active modes reasonable which are given in table 3. The calculated frequencies are in good agreement with the available experimental values. The lowest calculated Raman active A_{1g} mode frequency at 101 cm^{-1} is due to the vibration of Ba atoms and this agrees very well with the experimental frequency at 104 cm^{-1} . Similarly the calculated Raman frequency in A_{1g} symmetry at 126 cm⁻¹ and 449 cm⁻¹ are due to the vibration of T1 and O(1) atoms respectively and the observed frequencies at 127 and 459 cm⁻¹ agrees very well with the calculated frequency. The highest calculated Raman frequency 528 cm⁻¹ in A_{1g} symmetry is due to the vibration of O(2) atoms which also agrees very well with the observed frequency at 528 cm⁻¹.

The Raman Phonon frequency at 206 and 298cm⁻¹ in B_{1g} symmetry are due to the vibration of O(1) and O(4) atoms respectively.

The calculated Raman Phonon frequency in E_g symmetry at 68 cm⁻¹ 244cm⁻¹, 399cm⁻¹, 545cm⁻¹ are due to the vibration of Ba₁, Cu₁, O(2) and O(3) atoms respectively and it agrees very well with the observed frequencies at 65 cm⁻¹, 245cm⁻¹, 400cm⁻¹ and 545cm⁻¹ respectively.

The phonon frequencies at 107cm^{-1} is due to the vibration of Ba, Ca and Cu. The Ca and Cu vibrates at 180^{0} out of phase to Ba atom. Similarly the calculated phonon frequency at 365cm^{-1} is due to the vibration of O(4) atom , which is out of phase to O(1) atom and its observed frequency at 365cm^{-1} agrees very well with the calculated phonon frequencies. The calculated Raman phonon frequency at 398 cm^{-1} is due the vibration due to the O(1) atom and it performs bending bond vibration in the O(4) atom and Ca atom is out of phase to O(1) and O(4) atom. The observed frequencies at 398 cm^{-1} agrees very well with the calculated phonon frequencies. The highest phonon frequencies in symmetry E_{g} is due to the vibration of O(4) atom and performing stretching bond vibration.

The Calculated IR frequency in A2u symmetry at 79cm-1 and 143cm-1 are due to the vibrations of Cu(1) and Cu(2). The frequencies at 139cm⁻¹ is due to the vibration of Tl and Ba atom. The frequency at 167cm⁻¹ is due to the vibration of Ca, O(1) and O(4) atoms and its observed frequency at 157cm⁻¹ is agrees very well with calculated frequencies. Similarly the frequency at 295cm⁻¹ is due to vibration of O(1) and O(2) and Ca atoms and Ca atom vibrates at 180[°] out of phase to O(1) and O(2) atoms. Its observed frequency 456 cm⁻¹ is due to the vibration of Ca, O(1) and O(2) atom where as O(1) atom vibrates at 180[°] out of phase to Cu and O(2). The highest frequency in A_{2u} symmetry is 570cm⁻¹ is due to vibration of O(3) atom and its observed frequency is 574cm⁻¹ agrees very well with the calculated frequency.

The calculated Infrared frequency in B2u symmetry at 126cm-1 and 284cm-1 are due to vibration of O(1) and O(4) atoms respectively.

The calculated infrared frequencies in Eu symmetry at 68cm-1 is due to vibration of Tl atom and its observed frequency 68cm-1 agrees very well with the calculated frequency. The calculated frequency at 247cm-1, 375cm-1, 380cm-1 are due to the bending bond vibration and it agrees with observed frequencies at 247cm-1, 372cm-1 and 380 cm -1 agrees very well with the calculated frequencies. To calculate frequencies at 559cm-1 and 568cm-1 are due to vibration of O(1) and O(4) atoms respectively performing stretching bond vibration and its observed frequency 559 cm-1 and 568cm-1 agrees very well with the calculated phonon frequency. The observed frequencies at 565cm-1 agrees very well with the phonon frequency.

3.2 Normal co-ordinate analysis of $Tl_2Ca_3Ba_2Cu_4O_{12}$

The evaluated frequencies using the normal coordinate analysis method listed in table 3. agrees favourably with the calculated lattice dynamical frequencies and observed experimental frequencies.

The calculated Raman phonon frequency in A1g symmetry at 107cm-1 is due to the vibration of Ba atom which is the lowest frequency in this mode due to the stretched vibration of Ba-O(2). The calculated frequency at 124cm-1 is due to the vibration of Tl atom which is due to the stretched vibration of Cu-O(1). The frequency at 147cm-1 is due to the vibration of O(1)-Cu-O(1). Similarly the highest frequency in this symmetry 530cm-1 is due to the vibrations of O(2) atom and it is due to the bending vibration of O(1)-Ca-O(2) which agrees very well

with the observed frequency at 528cm-1, which is confirmed potential energy distribution calculation.

The frequency at 60cm-1 in Eg symmetry is due the vibration of Ba atom which is due to stretched vibration of Ba-O(2). The calculated frequency at 390 cm-1 is due to the vibration of O(1) atom and it is due to bending vibration of Tl-O(2)-Ba. The highest frequency in this symmetry is 560cm-1 is due to the vibration O(4) atom which is due to the stretched vibration. This frequency agrees very well with the observed frequency at 567cm⁻¹ which is confirmed by potential energy distribution calculation.

The lowest frequency in the A2u symmetry is 80cm-1 is due to the vibration of Cu(1) atom which is due to the bending vibration Tl-O(3)-Tl. The calculated frequency at 300cm-1 is due to the vibration of O(2) atom which is due to the bending vibration O(1)-Cu-O(1). The highest calculated frequency in this symmetry is 588 cm-1, which is due to the vibration of O(3) atom which performs stretched vibration of Ca-O(1). The calculated frequencies is confirmed by potential energy distribution calculation.

The calculated frequency in Eu symmetry at 70 cm-1 is due to the vibration of Cu(2) atom and which is due to bending vibration of O(2)-Tl-O(3). The frequency at 365 cm-1 is due to the vibration of O(1) atom and which is due to the bending vibration. The highest calculated Infrared Raman frequency 570 cm-1 is due to the vibration of O(4) atom which is performed by stretched vibration of Tl-O(4).

The G-matrix elements have been calculated from the equilibrium geometry. The initial force constants were taken from the related molecules. The final set of potential constants provide the stability of the crystal in relation to all vibrational modes. The vibrational frequencies and potential energy distribution values are presented in this work. The potential energy distribution indicates the contribution of an individual force constant to the vibrational energy of normal modes. It clearly indicates that there is mixing of the internal displacement coordinates.

Vibrational modes on the region 400-500 cm-1 are attributed to Ca-O(2) stretching. The present potential energy distribution confirms our conclusion. The lower frequency modes involve the small displacement of Ca-O(2) and Ba-O and the angular displacement of O-Ca-O. The evaluated frequencies using the normal coordinate analysis method listed in table 3 agree favourably with the calculated lattice dynamical frequencies and observed experimental frequencies.

4.Conclusion

The theoretical phonon frequencies obtained by the lattice dynamics and the normal coordinate analysis method agrees very well will the available Raman and infrared frequencies. This calculation attributes not only the phonon frequency in center of the Brilioun zone but also supports the strong electron-phonon interaction in the high-temperature superconductor Tl2Ca3Ba2Cu4O12.

methods viz, lattice dynamical study and normal coordinate analysis yielded the same phonon frequency for Tl2Ca3Ba2Cu4O12 high temperature superconductor. It is also seen from the tables, that the agreement between the calculated and observed frequencies wherever available are very good for the system under consideration. This fact supports It is not uncommon in physics that two different methods yield the same result. In the present work, two different that the present vibrational assignments made for the infrared and Raman Spectra are adequate. Therefore, it is concluded that normal coordinate analysis of the optically active lattice vibrations is very useful for the theoretical interpretation of the vibrational spectra at the center of the Brillouin zone in cuprate oxides. Further it is also a good method to characterize the high temperature superconductors.

It is also shown that it is possible to obtain is a fairly reasonable representation of the lattice vibration in Tl2Ca3Ba2Cu4O12. Using the three-body force shell model, with the assumption of unequal short-range interactions, the present approach leads to yields a complete phonon frequency of high Tc superconductor under consideration in the present work. Summarizing, the theoretical phonon frequencies obtained by the lattice dynamics and the normal co-ordinate analysis agree very well with the available Raman and infrared frequencies.

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Fig. 1 Unit Cell of $Tl_2Ca_3Ba_2Cu_4O_{12}$

Table :1

Parameters of the model: a, b are Born- Mayer constants: Z, Y, K, ionic charge, shell charge and on-site core-shell force constant of the ion, va is the volume of the unit cell

Interaction		a (eV)	b (Å ⁻¹)	
T1-O (Same pla	ane)	3000	2.80	
T1-O (adj plane	e)	3000	3.55	
Ba-O		3220	2.90	
Ca-O		2510	3.10	
Cu-O		1260	3.35	
0-0		1000	3.00	
ion	Z(e)	Y(e)	$k(e^2 v_a)$	
T1	2.70	2.00	1000	
Ba	2.00	2.32	207	
Ca	2.02	-0.50	1350	
Cu	2.00	3.22	1248	
O(Cu-O) plane	-1.90) -2.70	310	
O(T1-O) plane	-1.93	-2.70	210	
O(Ba-O) plane	-1.93	-2.70	310 (K)	
			2100(K⊥)

Table : 2

Force constants for $Tl_2Ca_3Ba_2Cu_4O_{12}$ (in units of 10^2 Nm⁻¹ (stretching) and 10^{-18} Nm rad⁻² (bending))

Potential	bond type		distance (Å)	initial value
Constant				
\mathbf{f}_{a}	Ca-O(1)	2.467	1.06	
\mathbf{f}_{b}	Ba-O(1)	2.798	0.75	
$\mathbf{f}_{\mathbf{c}}$	Ba-O(2)	2.819	1.10	
\mathbf{f}_{d}	Ba-O(3)	2.851	0.81	
f_e	Tl-O(1)	2.003	0.30	
\mathbf{f}_{g}	Tl-O(2)	2.097	0.30	
$\mathbf{f}_{\mathbf{h}}$	Tl-O(3)	3.108	0.61	
$\mathbf{f}_{\mathbf{k}}$	Tl-O(3)	2.402	0.48	
\mathbf{f}_{l}	Cu-O(1)	1.932	0.145	
$\mathbf{f}_{\mathbf{m}}$	Cu-O(2)	2.648	1.65	
$\mathbf{f}_{\mathbf{n}}$	Tl-O(3) –Tl		0.31	
$\mathbf{f}_{\mathbf{p}}$	O(1)-Cu-O(1)		0.25	
f_{α}	Tl-O(2)-Ba		0.46	
f_{β}	O(2)-Tl-O(3)		0.80	

Symmetry species	Frequency(cm ⁻¹) Using Lattice Dynamics	Using Normal Coordinate analysis	PotentialEnergy Distribution(%)
A _{1g} (Raman)	101 (104)	107	$f_{c}(58) f_{d}(24) f_{a}(11)$
	126 (127)	124	f _c (55) f _l (30)
	148 (142)	147	$f_l(71) f_d(12) f_p(11)$
	256 (256)	256	$f_{a}(51) f_{a}(20)$
	370 (360)	377	$f_a (68) f_u (20) f_c (10)$
	449 (439)	438	$f_{a}(61) f_{c}(20) f_{m}(10)$
	528 (528)	530	f_a (59) f_e (29) f_m (11)
	625(628)	630	$F_1(69) f_e(19) f_m(17)$
B _{1g}	206	210	f_a (56) f_k (14) f_a (10)
	298	296	$f_a(52) f_k(20) f_{\beta}(6)$
Eg	68(65)	60	$f_{c}(60) f_{e}(14) f_{\beta}(6)$
	107(107)	110	$f_{\rm m}$ (70) $f_{\rm e}(18)$ $f_{\beta}(10)$
	125 (125)	125	$f_{m}(70) f_{l}(21)$
	244 (245)	240	$f_e(52) f_m(16) f_c(12)$
	3656 (365)	370	$f_{l}(45) f_{n}(31) f_{c}(20)$
	378 (400)	390	$f_a(42) f_g(25) f_n(15)$
	399 (410)	405	$f_{\beta}(56) f_k(17) f_g(16)$
	545 (54)	550	$f_n(62) f_h(22) f_m(14)$
	555 (567)	560	$f_{\alpha}(68) f_n(21)$
A2u	79 (79)	80	$f_n(70) f_a(24)$
	139 (137)	136	$f_m(65) f_u(20) f_d(11)$
	143 (143)	150	$f_{\rm m}(80) f_{\rm l}(14)$
	160 (167)	165	$f_{a}(81) f_{e}(16)$
	295(313)	300	$f_p(54)f_n(30)$
	422 (432)	430	$f_p(62) f_{\beta}(18) f_n(10)$
	432 (437)	440	$f_a(40) f_g(30) f_n(10)$
	456 (450)	450	$f_{\beta}(41) f_{p}(21) f_{m}(26)$
	570 (574)	580	$f_a(60) f_e(21) f_m(16)$

Table:3. The Phonor	n frequency	Spectrum	of Tl ₂ Ca ₃	Ba ₂ Cu ₄ O _{12.}
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B2u	126	130	$f_p(54) f_n(26) f_e(15)$
	284	283	$f_{\beta}(28) f_{p}(21) fu(30) f_{e}(19)$
Eu	68(68)	70	$f_{\beta}(70) f_{c}(20)$
	107 (107)	110	$f_{a}(71) f_{e}(20)$
	132 (133)	134	$f_a(46) f_d(18) f_b(10)$
	247 (247)	247	$f_n(52) f_a(17) f_e(24)$
	255 (255)	250	$f_a(55) f_d(18) f_c(10)$
	355 (36)	365	$f_c(61) f_e(16) f_u(14)$
	375 (372)	378	$f_a(55) f_e(14) f_u(20)$
	380 (380)	381	$f_{p}(64) f_{c}(16)$
	455 (455)	460	$f_{\beta}(66) f_{p}(25)$
	559 (559)	562	$f_n(65) f_\alpha(21)$
	568 (568)	570	$f_n(68) f_a(25)$

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