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Calculation of Low Temperature Thermal Expansion of the High Temperature Superconductor $Bi_2Sr_2CaCu_2O_8$

K. P. Jayachandran and C. S. Menon[1]

School of Pure and Applied Physics, Mahatma Gandhi University, Kottayam-686 560 Kerala, India

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The anisotropy of low-temperature thermal expansion of the high-temperature superconductor $Bi_2Sr_2CaCu_2O_8$ (Bi-2:2:1:2) is analyzed theoretically. The generalized Grüneisen parameters (GPs) of elastic waves propagating in different directions of Bi-2:2:1:2 are calculated using the measured values of second order elastic constants and first order pressure derivatives of second order elastic constants. For this, we have used the measured values of the second order elastic constants of G. A. Saunders *et al*. The 12 third order elastic constants of the Bi-2:2:1:2 system are calculated from the expressions of effective second order elastic constants. All the third order elastic constants are negative except C_{155} and C_{112}. Some values of generalized Grüneisen parameters are negative. The generalized GPs γ_j'' are all positive except γ_2'' (-0.048) at $\theta = 85°$. The Brugger gammas are calculated and the low-temperature limits of the Grüneisen gamma are determined using the procedure of Menon and Ramji Rao. γ_L^- has the value of 4.4 for the Bi-2:2:1:2 system. The low-temperature limit of the Grüneisen gamma is positive. Therefore, we expect volume expansion to be positive down to absolute zero for the Bi-2:2:1:2 system. The anisotropy of thermal expansion along the *c* axis of the compound is very much higher than that along other directions.

INTRODUCTION

The anisotropic thermal expansion of high-temperature superconductors has received a great deal of attention in recent years. The high-T_c cuprate superconductors are textured materials and crystals are typically micaceous (i.e., with a thin plate like morphology) (1). The very small size of the single crystals prohibits the measurement of nonlinear anharmonic properties and therefore the textured samples would have a bright prospect in commercial applications in the foreseeable future (2). Since the discovery of superconductivity in the bismuth–strontium–calcium–copper oxides by Maeda *et al.* (3) in 1988, many authors have determined the nonlinear acoustic properties (4–10). Best results are obtained for stoichiometric starting ratios 2:2:1:2. The Bi-2:2:1:2 phase of bismuth cuprates has the ideal composition $Bi_2Sr_2CaCu_2O_8$ and has a T_c of about 90 K (11). The structure of $Bi_2Sr_2CaCu_2O_8$ is pseudo-tetragonal ($I4/mmm$) (12). The lattice parameters a (=5.4091 Å), b (=5.4209 Å), and c (=30.8445 Å) can be approximated by an orthorhombic cell ($a \approx b$) with the copper oxide layers in the ab plane (13). Hence the symmetry of the Bi-2:2:1:2 is, however, no greater than pseudo-orthorhombic. Bi_2O_2 layers consist of two parallel planar BiO sheets. The bonds between the bismuth oxide layers in the structure are weak and hence the crystallites of these compounds have been shown to cleave readily between these layers (14). The present objective is to study the thermal expansion and the Grüneisen parameters of Bi-2:2:1:2. We have calculated the generalized isothermal Grüneisen parameters γ_j' and γ_j'' for the acoustic modes. For this, the third order elastic constants of $Bi_2Sr_2CaCu_2O_8$ are obtained from second order elastic constants and first order pressure derivatives of second order elastic constants. The expressions for finding out the mode Grüneisen gammas for $Bi_2Sr_2CaCu_2O_8$ are obtained from the effective second order elastic constant expressions. These expressions are derived for the space groups ($4/mmm$, $\bar{4}2m$, 422, $4mm$) and are used to obtain the mode Grüneisen gammas.

THIRD ORDER ELASTIC CONSTANTS OF $Bi_2Sr_2CaCu_2O_8$

Third order elastic constants determine the anharmonic properties of solids such as thermal expansion, temperature and pressure dependence of elastic constants, and interaction of acoustic and thermal phonons. The third order elastic constants can be determined using the finite strain elasticity theory of Murnaghan (15), where elastic stress is nonlinear with elastic strain. If the higher order elastic constants are known, on the basis of continuum approximation, it is possible to calculate the scattering of phonons with the aid of nonlinear elasticity theory. The expressions for the effective second order elastic constants and its pressure derivatives for the strained crystal in te-

[1] To whom correspondence should be addressed.

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TABLE 1
Third Order Elastic Constants of $Bi_2Sr_2CaCu_2O_8$ (in GPa)

$C_{111} = -1048$	$C_{133} = -524$	$C_{344} = -524$
$C_{112} = 110$	$C_{155} = 1174$	$C_{333} = -963$
$C_{113} = -524$	$C_{166} = -458$	$C_{144} = 0$
$C_{123} = -354$	$C_{222} = -1048$	$C_{366} = 0$

tragonal system are given by Ramji Rao and Padmaja (16). The pressure derivatives of the second order elastic constants are in terms of higher order elastic constants up to third order. As the Bi-2:2:1:2 has 6 independent second order elastic constants, we have obtained 6 equations of effective second order elastic constants in terms of strain components ε' and ε''. We have calculated the complete set of 12 independent third order elastic constants and they are presented in Table 1. All of them are negative except C_{155} and C_{112}. The third order elastic constants bear their thermodynamic definitions introduced by Brugger (17).

LOW-TEMPERATURE THERMAL EXPANSION OF $Bi_2Sr_2CaCu_2O_8$

The linear thermal expansion coefficients denoted by $\alpha_\parallel$ and $\alpha_\perp$, parallel and perpendicular to the c axis of Bi-2:2:1:2, are expressed as

$$\begin{aligned} V\alpha_\perp &= [(S_{11} + S_{12})\gamma_\perp^{-1}(-3) + S_{13}\gamma_\parallel^{-1}(-3)]C_v = \gamma_\perp^{Br} C_v \chi \\ V\alpha_\parallel &= [2S_{13}\gamma_\perp^{-1}(-3) + S_{33}\gamma_\parallel^{-1}(-3)]C_v = \gamma_\parallel^{Br} C_v \chi. \end{aligned} \quad [1]$$

Here S_{ij} is the elastic compliance coefficients, V is the molar volume, C_v is the molar specific heat in the T^3 region, and χ is the isothermal compressibility. $\bar{\gamma}_\perp^1(-3)$ and $\bar{\gamma}_\parallel^1(-3)$ are, respectively, the low temperature limits of the effective Grüneisen functions $\bar{\gamma}_\perp^1(T)$ and $\bar{\gamma}_\parallel^1(T)$. $\gamma_\perp^{Br}$ and $\gamma_\parallel^{Br}$ are the average or effective Grüneisen functions in the notation of Brugger and Fritz (18). The effective Grüneisen functions are defined as

$$\bar{\gamma}_\perp^1(T) = \left[\sum_{qj} \gamma''(q,j) C_v(q,j)\right]\left[\sum_{qj} C_v(q,j)\right]^{-1}$$

$$\bar{\gamma}_\parallel^1(T) = \left[\sum_{qj} \gamma'(q,j) C_v(q,j)\right]\left[\sum_{qj} C_v(q,j)\right]^{-1}.$$

These effective Grüneisen functions are weighted averages of the generalized Grüneisen parameters $\gamma(q, j)$. q is the wave vector and j is the polarization index. $C_v(q, j)$ is the contribution of a single normal mode of frequency ω, wave vector q, and polarization index j to the specific heat of the lattice.

$\gamma(q, j)$ are given by

$$\gamma'(q,j) = \frac{-\partial \log \omega(q,j)}{\partial \varepsilon'}$$

$$\gamma''(q,j) = \frac{-\partial \log \omega(q,j)}{\partial \varepsilon''},$$

where ε' is a uniform areal strain in the basal plane perpendicular to the c axis and ε'' is a uniform longitudinal strain along the c axis. Here, instead of choosing arbitrary strain components in defining GPs, we choose such strains that leave the symmetry of the crystal unchanged. Hence the Lagrangian strain parameters η_{ij}, which are used in place of volume change, are selected such that $i = j$ (where $i = j = 1, 2, 3$), in the quasi-harmonic approximation.

At low temperatures, the GPs of the elastic wave frequencies determine the anisotropic thermal expansion of a uniaxial crystal, and the effective Grüneisen functions approach the limits

$$\begin{aligned} \gamma_\perp^{-1}(-3) &= \left[\int \sum_{j=1}^{3} \gamma_j'(\theta,\phi) V_j^{-3}(\theta,\phi))\, d\Omega\right]^{-1} \\ \gamma_\parallel^{-1}(-3) &= \left[\int \sum_{j=1}^{3} \gamma_j''(\theta,\phi) V_j^{-3}(\theta,\phi))\, d\Omega\right]^{-1} \end{aligned} \quad [2]$$

Here $V_j(\theta, \phi)$ is the wave velocity of elastic wave of polarization index j, propagating in the direction (θ, ϕ). γ_j' and γ_j'' are the GPs of this acoustic mode. the Grüneisen parameters for the acoustic modes can be calculated using the second order elastic constants and third order elastic constants and hence, the low temperature limits of GPs can be obtained from the procedure of Menon and Ramji Rao (19, 20). In tetragonal crystals, it is assumed that GPs and the acoustic wave velocities depend only on θ and are independent of the azimuth ϕ.

The average effective Grüneisen functions in the notation of Brugger and Fritz in Eq. [1] are expressed as

$$\begin{aligned} \gamma_\perp^{Br} &= [(S_{11} + S_{12} +)\gamma_\perp^{-1}(-3) + S_{13}\gamma_\parallel^{-1}(-3)]\chi^{-1} \\ \gamma_\parallel^{Br} &= [2S_{13}\gamma_\perp^{-1}(-3) + S_{33}\gamma_\parallel^{-1}(-3)]\chi^{-1}. \end{aligned} \quad [3]$$

THE MODE GRÜNEISEN GAMMAS OF Bi-2:2:1:2 SYSTEM

The general Thurston and Brugger equation for wave propagation is (21)

$$\rho_0\omega^2 U_i = 4\Pi^2 \sum_{kjl} U_j Y_l Y_k \left[C_{ik,jl} + \sum_{mn} \varepsilon_{mn}(C_{ik,jl,mn} + C_{ik,nl}\delta_{mj} + C_{nk,jl}\delta_{mi} + C_{kl,mn}\delta_{ij})\right]. \quad [4]$$

Here Y_i's are the components of the arbitrary direction in which the wave is propagating and U_j's are the components of displacement. ω is the angular frequency and ρ_0 is the density of the crystal in the unstrained state. The C_{ijkl} and C_{ijklmn} values are the second order and third order elastic constants, respectively.

From the elements of the secular determinant formed by the coefficients of U_i's, the expressions for the effective second order elastic constants in terms of the strain components ε'' and ε' are obtained as

$$C'_{11} = C_{11} + [(C_{111} + C_{112} + 3C_{11} + C_{12})(\varepsilon'/2)] + (C_{113} + C_{13})\varepsilon''$$

$$C'_{12} = C_{12} + [(2C_{112} + C_{12} - C_{22})(\varepsilon'/2)] + (C_{123} - C_{13})\varepsilon''$$

$$C'_{13} = C_{13} + [(C_{113} + C_{123} - C_{13} - C_{44})(\varepsilon'/2)] + (C_{133} + C_{13} - C_{33} + C_{44})\varepsilon''$$

$$C'_{33} = C_{33} + [(2C_{133} + 2C_{13})(\varepsilon'/2)] + (C_{333} + 3C_{33})\varepsilon''$$

$$C'_{44} = C_{44} + [(C_{155} + C_{144} + 2C_{44} + 2C_{13})(\varepsilon'/2)] + (C_{344} + C_{33})\varepsilon''$$

$$C'_{66} = C_{66} + [(2C_{166} + 2C_{66} + C_{12} + C_{22})(\varepsilon'/2)] + (C_{366} + C_{13})\varepsilon''.$$

Here the C_{IJ} and C_{IJK} are, respectively, the second order and third order elastic stiffness constants in Voigt notation.

The general expressions for GPs from the determinantal Eq. [4] are

$$\gamma'_j = (-1/2X_j)\left\{\frac{[X_j\partial/\partial\varepsilon'(A+B)] - [\partial/\partial\varepsilon''(AB - C^2)]}{2X_j - (A+B)}\right\}$$

$$\gamma''_j = (-1/2X_j)\left\{\frac{[X_j\partial/\partial\varepsilon''(A+B)] - [\partial/\partial\varepsilon''(AB - C^2)]}{2X_j - (A+B)}\right\},$$

where

$$A = C'_{11}\sin^2\theta + C'_{44}\cos^2\theta,$$

$$B = C'_{44}\sin^2\theta + C'_{33}\cos^2\theta,$$

$$C = \sin\theta\cos\theta(C'_{13} + C'_{44}),$$

and

$$X_j = \rho_0 V_j^2(\theta, \phi).$$

Here we made use of the measured values of C_{IJ} (5) as well as the calculated C_{IJK} (given in Table 1) for the calculation of γ'_j and γ''_j. Table 2 gives the wave velocities and the GPs for the elastic wave propagating at different angles θ to the c axis of $Bi_2Sr_2CaCu_2O_8$. The plots in polar coordinates of the generalized Grüneisen parameters γ'_j and γ''_j for acoustic modes as a function of angle θ, which the direction of propagation makes with the crystal axis, are given in Figs. 1 and 2, respectively. The calculated values of the low-temperature limits of the effective Grüneisen functions $\gamma_\perp^{-1}(-3)$ and $\gamma_\parallel^{-1}(-3)$ from Eq. [2] for the $Bi_2Sr_2CaCu_2O_8$ system are -3.9 and 6.7. Now from Eq. [3] the $\gamma_\perp^{Sr}$ and $\gamma_\parallel^{Br}$ are evaluated as -0.9 and 6.2, respectively. The low-temperature volume lattice thermal expansion (in terms of the Grüneisen parameter) $\overline{\gamma}_L = 2\gamma_\perp^{Br} + \gamma_\parallel^{Br}$ is 4.4.

RESULTS AND DISCUSSION

The theory of low-temperature thermal expansion has been applied to determine the mode gammas and low-temperature limit of the thermal expansion in $Bi_2Sr_2CaCu_2O_8$. The mode Grüneisen parameters of the acoustic modes in different wave propagation directions have been calculated, which show anisotropy in thermal expansion of the compound in all the acoustic wave propagation directions. The low-temperature limit of the Grüneisen parameter of the crystal $\overline{\gamma}_L$ obtained in the present calculation is 4.4. The mean acoustic mode Grüneisen parameter for certain polycrystalline high-T_c superconductors determined by ultrasonic techniques spread over a wide range from 1.5 to 23.7 (22–26). Hence the value 4.4 obtained for $\overline{\gamma}_L$ is a reasonable one. The thermal Grüneisen parameter γ_o^{th}, deduced by G. K. White (27) for Bi-2:2:1:2, is 2.3 and it is related to the volume thermal expansion coefficient β, the specific heat C_p, the volume V, and the bulk modulus B^s by

$$\gamma_o^{th} = \frac{\beta B^s V}{c_p}.$$

The mean acoustic anode Grüneisen parameter γ^{el} deduced (5) for Bi-2:2:1:2 is 2.5, which accounts for the contribution of overall long-wavelength acoustic modes to the thermal expansion. γ^{el} is evaluated using

$$\gamma^{el} = (1/3)(\gamma_l + 2\gamma_s).$$

Here γ_l and γ_s are longitudinal and shear acoustic mode Grüneisen parameters, respectively. It must be noted here that although γ_o^{th} and γ^{el} are related to $\overline{\gamma}_L$, they could not be identified with $\overline{\gamma}_L$. The average Grüneisen function $\gamma_\parallel^{Br}$ parallel to the c axis is 6.2. This is in good agreement with the longitudinal mode Grüneisen parameter γ_L value of 7.9 along the [001] direction of Bi-2:2:1:2 obtained by G. A. Saunders *et al.* (5). The negative value of average

TABLE 2
Generalized GPs for the Elastic Waves Propagating at Different Angles θ to the Crystal Axis in the $Bi_2Sr_2CaCu_2O_8$ System

θ^0	γ_1'	γ_1''	$\rho_0 v_1^2$	γ_2'	γ_2''	$\rho_0 v_2^2$	γ_3'	γ_3''	$\rho_0 v_3^2$
5	15.66	11.93	19.55	−16.76	12.43	19.14	5.732	9.519	44.21
15	12.63	8.157	23.40	−15.22	11.04	20.25	4.818	10.45	44.77
25	9.115	3.911	28.06	−10.57	8.790	22.34	3.669	10.68	48.41
35	4.983	4.446	29.15	−6.435	6.351	25.15	3.032	8.098	58.50
45	0.788	6.297	27.42	−2.727	4.166	28.35	2.399	5.537	72.93
55	−3.781	8.160	24.79	0.2299	2.424	31.55	1.862	3.936	88.27
65	−8.643	9.973	22.21	2.375	1.160	34.36	1.488	2.986	102.0
75	−13.05	11.54	20.22	3.753	0.347	36.45	1.259	2.451	112.3
85	−15.77	12.50	19.14	4.424	−0.048	37.56	1.152	2.209	117.8

Note. ρ_0 is the density of the crystal in the unstrained state.

Grüneisen function $\gamma_{\perp}^{Br}$ (= −0.9) suggests that the thermal expansion along the *ab* plane of Bi-2:2:1:2 could be negative. The transverse acoustic branch γ_1'' assumes a minimum value 3.91 at $\theta = 25°$ to the *c* axis and a maximum value 12.5 at $\theta = 85°$. For γ_2'' and γ_3'' the minimum values are −0.048 and 2.21, respectively (occur at $\theta = 85°$ for both). The acoustic branches of the GPs γ_2' and γ_1' have positive as well as negative values (ranging from 4.42 to −16.76 and 15.66 to −15.77). γ_3' are found to be completely positive with a minimum of 1.15 at $\theta = 85°$ and a maximum of 5.73 at $\theta = 5°$. This anisotropy in the data of mode gammas is mounting evidence for pronounced vibrational anharmonicity in the compound in certain acoustic wave propagation directions. The variation of generalized Grüneisen parameters γ_j' and γ_j'' with angle θ, which the direction of propagation makes with the crystal axis of the Bi-2:2:1:2 system, are shown in Figs. 3 and 4, respectively.

Fanggao *et al.* (7) have clearly indicated that, in the Bi-2:2:1:2 system, weak interlayer forces are responsible for the softening of mean elastic moduli and of the elastic stiffness C_{33}. Thus, the main effect of application of hydrostatic pressure to a layer-like compound is to squeeze the widely spaced weakly bonded layers together. Therefore, the influence of the comparatively small repulsive forces

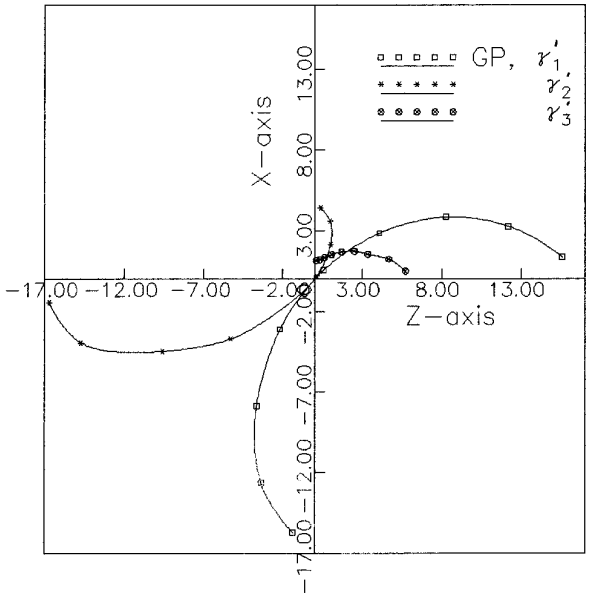

FIG. 1. Polar diagram showing the plot of generalized GPs γ_j' for the acoustic branches as a function of the angle θ, which the direction of propagation makes with the crystal axis of $Bi_2Sr_2CaCu_2O_8$. Here the scale chosen for plotting the polar coordinate *r* (i.e., gamma values) along the *XZ* plane is marked on the *X* axis and the *Z* axis.

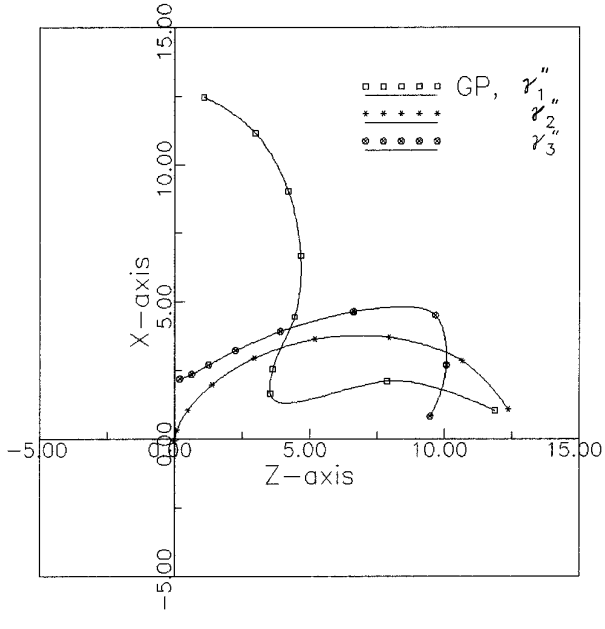

FIG. 2. Polar diagram showing the plot of generalized GPs γ_j'' for the acoustic branches as a function of the angle θ, which the direction of propagation makes with the crystal axis of $Bi_2Sr_2CaCu_2O_8$. Here the scale chosen for plotting the polar coordinate *r* (i.e., the gamma values) along the *XZ* plane is marked on the *X* axis and the *Z* axis.

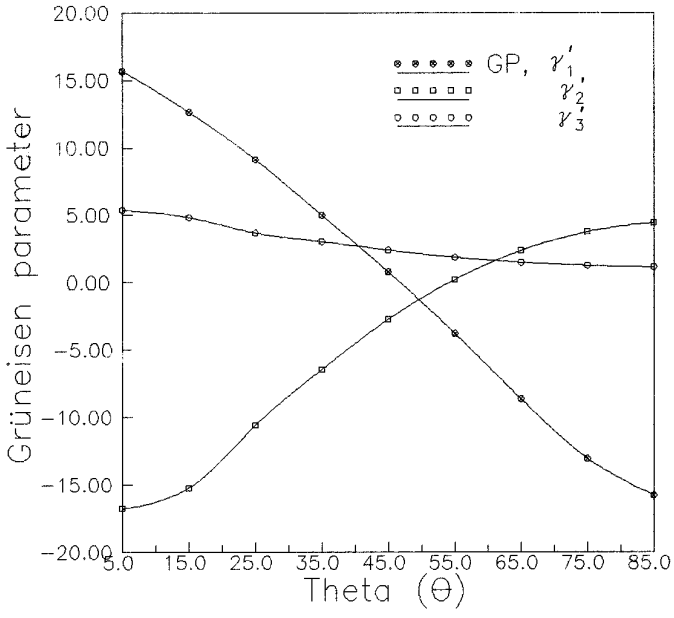

FIG. 3. Variation of generalized GPs γ_j' with θ for different acoustic waves in $Bi_2Sr_2CaCu_2O_8$.

that act between pairs of BiO layers must play an especially important role in the behavior of $Bi_2Sr_2CaCu_2O_8$ under pressure. The energy and velocity of the acoustic mode propagated down the *c* axis should increase substantially when pressure is applied. Therefore, the corresponding

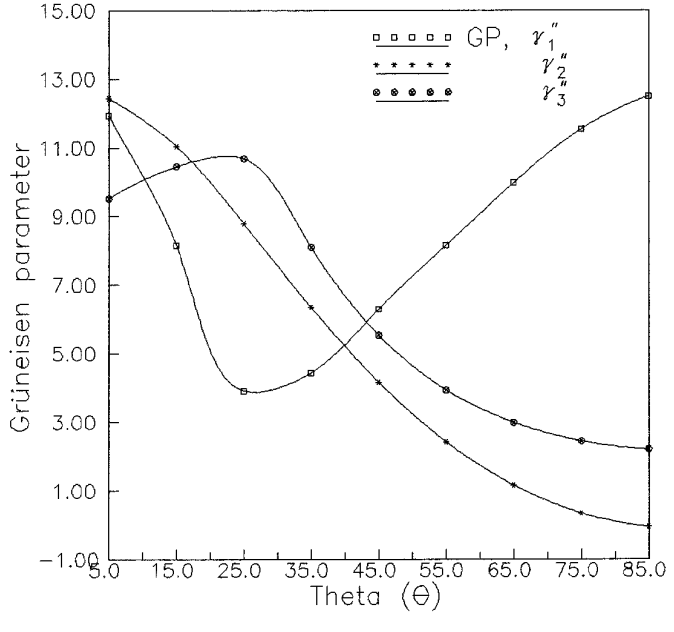

FIG. 4. Variation of generalized GPs γ_j'' with θ for different acoustic waves in $Bi_2Sr_2CaCu_2O_8$.

mode Grüneisen parameters must be positive and much larger than those of other acoustic modes. In the present study, the mode gammas corresponding to the acoustic modes propagating down the *c* axis of Bi-2:2:1:2 (i.e., the gamma values corresponding to $\theta = 5°$ to the *c* axis) are all positive and much higher than the values along other directions, except for the case of γ_2'. This lends more support to the conclusion that the vibrational anharmonicity and hence the anisotropy in thermal expansion along the *c* axis of Bi-2:2:1:2 is possibly due to the weak interlayer forces between BiO sheets.

CONCLUSION

The mode Grüneisen parameters of $Bi_2Sr_2CaCu_2O_8$ for different acoustic wave propagation directions have been calculated. The data give evidence for thermal expansion anisotropy of the material for various acoustic modes. The average Grüneisen functions $\gamma_\perp^{Br}$ and $\gamma_\parallel^{Br}$ are -0.9 and 6.2, respectively. These values suggest that the anisotropy in thermal expansion along the *c* axis is more pronounced than that along the *ab* plane. Since the average Grüneisen gamma along the *ab* plane is a negative value compared to that along the *c* axis, there could be a slight contraction (due to the small mangitude of the value of $\gamma_\perp^{Br}$) of the crystal along this plane at low temperatures. The Grüneisen parameter studies provide concrete evidence to the anisotropy in thermal expansion of $Bi_2Sr_2CaCu_2O_8$. The low-temperature limit of the Grüneisen gamma is positive, so we expect volume expansion to be positive down to O K for $Bi_2Sr_2CaCu_2O_8$. The theory followed here to calculate Grüneisen gammas along various orientations can be applied to other tetragonal systems ($\bar{4}2m$, 422, $4mm$) as well.

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