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Thermoacoustic generation in anisotropic media

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The generation of acoustic waves by a modulated thermal source is examined for anisotropic materials. A wave equation is developed to include the effect of a thermoacoustic source for the anisotropic case. The dependence of the thermoacoustic source term upon the material elastic constants is identified for a thermal source varying in one dimension. This dependence is examined for several classes of crystalline anisotropy and it is found that thermoacoustic generation varies with crystallographic orientation. The directions of maximum and minimum thermoacoustic generation are not consistent for a given crystal class and are dependent upon the specific values of the elastic constants for the material.

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INTRODUCTION

It has long been known that a modulated thermal source may be used to create acoustic waves. The effect was discovered in the 1880s by Alexander Graham Bell, who studied the audible sound generated by illuminating matter with a periodically interrupted light. The basic principle of thermoacoustic generation within a solid involves the coupling of energy from thermal expansion and contraction into an acoustic wave.2 The problem of thermoacoustic generation at the surface of an isotropic solid was studied by White,³ who related the production of elastic waves to thermal and elastic properties of the solid. More recently, thermoacoustic generation at the surface of a solid has been used as a means of spectroscopic characterization⁴ and as a source for imaging. 5 Thermoacoustic generation is a particularly useful technique to study the thermal and elastic properties of thin layers of material because thermoacoustic waves can be generated within a layer which is thinner than the generated acoustic wavelength. In contrast, a layer several acoustic wavelengths thick is required to interact effectively with externally generated acoustic waves.

This paper assesses the effect of elastic anisotropy on the generation of acoustic waves by a modulated thermal source. It will be shown that the solid elastic constants enter into the efficiency of thermoacoustic generation and this results in an orientation dependence for thermoacoustic generation within anisotropic solids.

I. THERMOACOUSTIC WAVE EQUATION

The first step in this analysis is to develop a wave equation for thermoacoustic generation in anisotropic materials. The presence of a locally nonuniform temperature distribution within the material produces an additional strain without a corresponding change in stress. The thermal strain S_{Θ} is proportional to the temperature increase Θ and to the linear thermal expansion coefficient α . This strain is purely dilatational and can be written in abbreviated subscript notation as

$$S_{\Theta} = \alpha \Theta \begin{bmatrix} 1 \\ 1 \\ 1 \\ 0 \\ 0 \end{bmatrix}, \tag{1}$$

where α and Θ are taken to be scalar quantities. The strain due to acoustic waves in the material S_a can be obtained from the difference between the total strain S and the temperature associated strain S_{Θ} ,

$$S_a = S - S_{\Theta}, \tag{2}$$

giving the strain displacement relation,

$$S_a = \nabla_s u - S_{\Theta}, \tag{3}$$

where u is the displacement and ∇_s is the symmetric part of the displacement gradient. Recall the acoustic equation of motion

$$\nabla \cdot T = \rho \frac{\partial^2 u}{\partial t^2} \tag{4}$$

and the constitutive equation

$$T = c: S_a, \tag{5}$$

where T is the stress, ρ is the density, c is the stiffness matrix, and the double dot product represents matrix multiplication. Substituting S_a into the constitutive equation gives

$$T = c : S - \alpha c : \Theta \begin{bmatrix} 1 \\ 1 \\ 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}. \tag{6}$$

This is the stress-strain relation for thermoacoustic generation and, if it is simplified to the case of isotropic media, it becomes

$$T = c : S - 3B\alpha\Theta \begin{bmatrix} 1\\1\\1\\0\\0\\0 \end{bmatrix}, \tag{7}$$

which is called the Duhamel-Neumann law, 8 where B is the bulk modulus of elasticity.

Combining Eqs. (3)-(5) gives the thermoacoustic wave equation9

$$\nabla \cdot (c : \nabla_s u) - \rho \frac{\partial^2 u}{\partial t^2} = \nabla \cdot (c : S_{\Theta}). \tag{8}$$

II. THERMOACOUSTIC SOURCE TERM

The right-hand side of Eq. (8) can be viewed as a source term due to the nonuniform temperature distribution. Expanding this term gives

$$\nabla \cdot (c : S_{\Theta}) = \alpha \nabla \cdot c : \Theta \begin{bmatrix} 1 \\ 1 \\ 1 \\ 0 \\ 0 \\ 0 \end{bmatrix}. \tag{9}$$

For the most general anisotropic case, there are 21 independent constants in the c matrix (e.g., triclinic crystal class) and the thermoacoustic source term can be written as follows:

$$\nabla \cdot (c:S_{\Theta}) = \left(\frac{\partial \Theta}{\partial x} \left(c_{11} + c_{12} + c_{13}\right) + \frac{\partial \Theta}{\partial y} \left(c_{16} + c_{26} + c_{36}\right) + \frac{\partial \Theta}{\partial z} \left(c_{15} + c_{25} + c_{35}\right)\right) \alpha \hat{x}$$

$$+ \left(\frac{\partial \Theta}{\partial x} \left(c_{16} + c_{26} + c_{36}\right) + \frac{\partial \Theta}{\partial y} \left(c_{12} + c_{22} + c_{23}\right) + \frac{\partial \Theta}{\partial z} \left(c_{14} + c_{24} + c_{34}\right)\right) \alpha \hat{y}$$

$$+ \left(\frac{\partial \Theta}{\partial x} \left(c_{15} + c_{25} + c_{35}\right) + \frac{\partial \Theta}{\partial y} \left(c_{14} + c_{24} + c_{34}\right) + \frac{\partial \Theta}{\partial z} \left(c_{13} + c_{23} + c_{33}\right)\right) \alpha \hat{z}. \tag{10}$$

Examining the components of the thermoacoustic source term indicates that thermoacoustic generation is enhanced for large thermal gradients, large coefficients of thermal expansion, and large values for the elastic constants. In addition, anisotropic materials allow for generation of acoustic waves in directions perpendicular to the thermal gradients. Under the assumption of propagation in an isotropic medium, Eq. (10) becomes

$$\nabla \cdot (c : S_{\Theta}) = 3B\alpha \nabla \Theta. \tag{11}$$

With the additional assumption of purely dilatational displacements, the thermoacoustic wave equation (8) becomes

$$c_{11}\nabla^2 u - \rho \frac{\partial^2 u}{\partial t^2} = 3B\alpha\nabla\Theta, \tag{12}$$

which can also be directly obtained from the Dunhamel-Neumann law.

In order to predict the efficiency of thermoacoustic generation in anisotropic materials, the thermoacoustic wave equation (8) will be examined for the simple case of a thermal gradient in one direction. In particular, assume that harmonic thermal energy is absorbed at the boundary between two semi-infinite media with a resulting thermal distribution which varies in the \hat{x} direction only. The thermoacoustic wave equation in the \hat{x} direction is then

$$\frac{\partial^2 u_x}{\partial x^2} - \frac{1}{v^2} \frac{\partial^2 u_x}{\partial t^2} = \alpha \frac{(c_{11} + c_{12} + c_{13})}{c_{11}} \frac{\partial \Theta}{\partial x}, \quad (13)$$

where $v = \sqrt{c_{11}/\rho}$ is the acoustic velocity. The effect of the elastic constants on the thermoacoustic source term can now be isolated to the factor

$$\beta_x = (c_{11} + c_{12} + c_{13})/c_{11}. \tag{14}$$

Likewise, for thermoacoustic generation due to thermal gra-

dients confined to the \hat{y} or \hat{z} directions, the elastic portion of the source terms becomes

$$\beta_{\nu} = (c_{12} + c_{22} + c_{23})/c_{22}, \tag{15}$$

$$\beta_z = (c_{13} + c_{23} + c_{33})/c_{33}. \tag{16}$$

III. THERMOACOUSTIC GENERATION IN CRYSTALS

The thermoacoustic generation factor β is given in Tables I-V for various examples from the orthorhombic, tetragonal, trigonal, hexagonal, and cubic crystal classes. The values of the elastic constants used to calculate β were obtained from Auld. 10 The orthorhombic crystals (see Table I) have generation factors β_x , β_y , and β_z which are independent because of the independence of the elastic constants

$$c_{11} \neq c_{22} \neq c_{33}$$

and

$$c_{12} \neq c_{13} \neq c_{23}$$

For the three orthorhombic crystals in Table I, no simple pattern is observed for the thermoacoustic generation factors β_x , β_y , and β_z . The relative magnitude of β in the three calculated directions is different in each case: For barium sodium niobate, $\beta_z > \beta_x > \beta_y$; for iodic acid, $\beta_x > \beta_z > \beta_y$;

TABLE I. Thermoacoustic generation factor for orthorhombic crystals.

| Material | $\boldsymbol{\beta}_{x}$ | $\boldsymbol{\beta}_{y}$ | $\boldsymbol{\beta}_z$ |
|------------------------------------|--------------------------|--------------------------|------------------------|
| Barium sodium niobate ^a | 1.64 | 1.63 | 1.76 |
| Iodic acid ^a | 1.78 | 1.32 | 1.39 |
| Rochelle salt* | 2.16 | 1.90 | 1.88 |

^a Piezoelectric crystal, values are calculated using c^E .

TABLE II. Thermoacoustic generation factor for tetragonal crystals.

| Material | $oldsymbol{eta}_{	ext{trans}}$ | β |
|---------------------------------------|--------------------------------|------|
| Ammonium dihydrogen phosphate (ADP) | 1.39 | 2.18 |
| Barium titanate | 2.20 | 2.83 |
| Calcium molybdate | 1.77 | 1.62 |
| Indium | 2.80 | 2.84 |
| Lead molybdate | 2.11 | 1.97 |
| Potassium dihydrogen phosphate (KDP)* | 1.90 | 2.01 |
| Rutile | 2.16 | 1.58 |
| Tellurium dioxide | 2.31 | 1.41 |

^{*}Piezoelectric crystal, values are calculated using c^E .

and for Rochelle salt, $\beta_x > \beta_y > \beta_z$. Likewise, the range of variation of β with direction is different in each case: for Barium sodium niobate, 7%; for iodic acid, 26%; and for Rochelle salt, 13%. All three of the orthorhombic crystals in Table I are piezoelectric and the stiffening due to this effect has been ignored in the calculation of β by using c^E , the elastic stiffness matrix. The inclusion of piezoelectic effects in β could be accomplished by use of c^D , the elastic stiffness at zero electric displacement.

For tetragonal, trigonal, and hexagonal crystals, the thermoacoustic generation factors are equal in the \hat{x} and \hat{y} directions ($\beta_{\text{trans}} \equiv \beta_x = \beta_y$) due to the equivalence of the elastic constants

$$c_{11} = c_{22}$$

and

$$c_{23}=c_{13}$$

For the tetragonal crystals in Table II, no clear pattern emerges for the relative magnitudes of β_{trans} and β_z . The examples shown are equally divided between cases where $\beta_{\text{trans}} > \beta_z$ and $\beta_z > \beta_{\text{trans}}$. A substantial difference between the relative magnitude of β_{trans} and β_z is observed for tellurium dioxide (39%), ADP (36%), and rutile (27%), indicating that for these crystals the magnitude of thermoacoustic generation will vary significantly with orientation. Also note the large absolute values of β_z for barium titanate (2.83) and indium (2.84).

For the trigonal crystals in Table III, there is no consistent pattern for the relative magnitudes of β_{trans} and β_z . The examples show that larger thermoacoustic generation may be observed in either direction. However, note the consistently small values of β for the trigonal crystals and in particular the small β for quartz (1.22).

For the hexagonal crystals in Table IV, in each case $\beta_{\text{trans}} > \beta_z$, indicating that thermoacoustic generation is

TABLE III. Thermoacoustic generation factor for trigonal crystals.

| Material | $oldsymbol{eta}_{	ext{trans}}$ | β, | |
|--------------------|--------------------------------|------|--|
| Lithium niobate* | 1.63 | 1.73 | |
| Lithium tantalate* | 1.55 | 1.69 | |
| Quartz* | 1.22 | 1.22 | |
| Sapphire | 1.55 | 1.46 | |

^{*} Piezoelectric crystal, values are calculated using c^E .

TABLE IV. Thermoacoustic generation factor for hexagonal crystals.

| Material | $oldsymbol{eta}_{	ext{trans}}$ | β, | |
|------------------------------|--------------------------------|------|--|
| Beryllium oxide ^a | 1.47 | 1.36 | |
| Cadmium selenide | 2.14 | 1.94 | |
| Cadmium sulfide ^a | 2.20 | 2.09 | |
| Titanium, crystal | 1.99 | 1.76 | |
| Zinc oxide | 2.08 | 2.00 | |
| Zinc sulfide | 2.09 | 1.97 | |

^{*}Piezoelectric crystal, values are calculated using c^{E} .

consistently greater in the transverse directions relative to the \hat{z} direction. However, note that the list in Table IV is only a sampling of hexagonal crystals and others may deviate from this pattern. Note that in the x-y plane of a hexagonal crystal, elastic wave propagation is isotropic resulting in thermoacoustic generation which is independent of direction.

For cubic crystals, the thermoacoustic generation factor β is equal in the \hat{x} , \hat{y} , and \hat{z} directions and is designated β_{100} . The factor β can be calculated for other directions by application of a coordinate transformation to the c matrix. ¹² Using a 45° rotation about the \hat{z} axis, the effective elastic constants for propagation in the [110] direction of a cubic crystal are given as follows:

$$c'_{11} = (c_{11} + c_{12} + 2c_{44})/2, \quad c'_{12} = (c_{11} + c_{12} - 2c_{44})/2,$$
(17)

 $c_{13}'=c_{12},$

and

$$\beta_{110} = (c'_{11} + c'_{12} + c'_{13})/c'_{11}. \tag{18}$$

TABLE V. Thermoacoustic generation factor for cubic crystals.

| Material | $oldsymbol{eta_{100}}$ | $oldsymbol{eta_{110}}$ | β_{111} |
|-------------------------------|------------------------|------------------------|---------------|
| Aluminum, crystal | 2.14 | 2.04 | 2.01 |
| Barium fluoride | 1.91 | 1.90 | 1.89 |
| Bismuth germanate* | 1.47 | 1.48 | 1.48 |
| Bismuth germanium oxide | 1.48 | 1.80 | 1.95 |
| Diamond | 1.49 | 1.35 | 1.31 |
| Europium iron garnet | 1.85 | 1.82 | 1.81 |
| Gallium arsenide* | 1.91 | 1.55 | 1.46 |
| Gallium phosphide | 1.89 | 1.55 | 1.46 |
| Germanium | 1.75 | 1.45 | 1.37 |
| Gold, crystal | 2.69 | 2.34 | 2.25 |
| Indium antimonidea | 2.09 | 1.71 | 1.61 |
| Indium arsenide* | 2.09 | 1.67 | 1.57 |
| Indium phosphide* | 2.13 | 1.73 | 1.62 |
| Iron, crystal | 2.19 | 1.70 | 1.58 |
| Lithium fluoride | 1.76 | 1.40 | 1.31 |
| Magnesium oxide | 1.61 | 1.38 | 1.31 |
| Nickel, crystal | 2.28 | 1.76 | 1.64 |
| Silicon | 1.77 | 1.51 | 1.44 |
| Silver, crystal | 2.50 | 2.01 | 1.89 |
| Sodium fluoride | 1.50 | 1.64 | 1.69 |
| Strontium titanate | 1.64 | 1.56 | 1.54 |
| Tungsten, crystal | 1.79 | 1.79 | 1.79 |
| Yttrium aluminum garnet (YAG) | 1.66 | 1.65 | 1.64 |
| Yttrium gallium garnet | 1.81 | 1.75 | 1.74 |
| Yttrium iron garnet (YIG) | 1.83 | 1.84 | 1.84 |

Piezoelectric crystal, values are calculated using c^E .

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By application of an additional rotation about the \hat{y}' axis, the elastic constants for propagation in the [111] direction are found to be

$$c_{11}'' = (c_{11} + 2c_{12} + 4c_{44})/2,$$

$$c_{12}'' = (c_{11} + 2c_{12} - 2c_{44})/2,$$

$$c_{13}'' = c_{12}'',$$
(19)

and β_{111} can be calculated from the c'' values as above. Using these expressions, the values of β_{100} , β_{110} , and β_{111} are given in Table V for various cubic crystals. For most of the examples in Table V, the relative magnitude of the generation factor is $\beta_{100} > \beta_{110} > \beta_{111}$, indicating that thermoacoustic generation is greatest along an axis [100] of the cubic crystals. However, in four cases (bismuth germanate, bismuth germanium oxide, sodium fluoride, and yttrium iron garnet), the preferred direction for thermoacoustic generation is [111].

Experimental investigation of the thermoacoustic effect in crystalline materials has been reported by Tam and Leung¹³ and by Vladimirtsev et al. ¹⁴ Tam and Leung used photoacoustically generated sound to measure the ultrasonic velocity as a function of orientation in a sample of polycrystalline aluminum. Although their paper does not mention variation in the amplitude of photoacoustic generation with crystal orientation, their experimental apparatus is well suited to measurements of this kind. Vladimirtsev et al. produced photoacoustic generation in piezoelectric crystals and observed the excitation of normal modes. They observed that surface optical defects increased the intensity of photoacoustic generation; however, they do not mention variation in photoacoustic generation with crystal orientation.

IV. SUMMARY

A wave equation has been derived which includes the generation of elastic waves by a modulated thermal source. The dependence of the thermoacoustic generation on material elastic constants has been explored for anisotropic materials. Although substantial variation in thermoacoustic generation is predicted for changes in orientation, the directions of maximum and minimum generation are not consistent for

a single crystal class and instead depend upon the specific values of the elastic constants.

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