

The Grüneisen parameter calculated from the single crystal and the polycrystal TOEC with that calculated from bulk data. Using values for the linear expansion coefficient, $\alpha = 7.02 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$, and specific heat, $c_p = 0.065 \text{ cal g}^{-1} \text{ }^\circ\text{C}^{-1}$ obtained from handbooks and density $\rho = 8.578 \text{ g/cm}^3$, and isothermal bulk modulus $K^T = 1.687 \times 10^{12} \text{ dyn/cm}^2$ obtained in the present work, in the relation

$$\gamma_B = 3\alpha K^T / C_V \rho \quad (4)$$

we get the value for the bulk Grüneisen parameter of $\gamma_B = 1.52$. Using the methods of Ref. 4 for obtaining the Grüneisen parameter from the elastic constants by averaging the contribution of 39 pure-mode phonons that parameter results in values of $\gamma_{SC} = 1.511 \pm 0.026$ and $\gamma_{PC} = 1.546 \pm 0.092$ for the single crystal and the polycrystal, respectively. A useful calculational equation for the polycrystal Grüneisen parameter which can be obtained from the equations of Brugger⁴ by imposing isopy conditions is

$$\gamma = (K^T m_1' - \Delta K) / 6C_{11}^S + (K^T m_2' - \Delta K) / 3C_{44}^S, \quad (5)$$

where m_1' and m_2' are the measured hydrostatic pressure slopes for the polycrystal and $\Delta K = K^S - K^T$ is the difference between the adiabatic and isothermal bulk moduli.

A second check on the values of the TOEC determined here is to compare the measured polycrystal constants with values calculated from the single-crystal constants. Recently, equations permitting this

comparison were derived³⁰ using strain-energy density considerations with the approximation that a uniform state of strain acting on the surface of a homogeneous, quasi-isotropic, polycrystalline body produces a uniform strain throughout the body. This development, analogous to that of Voigt relating the second-order elastic constants,³¹ leads to the following relations between the TOEC:

$$\nu_1 = \frac{1}{35}(C_{111} + 18C_{112} + 16C_{123} - 30C_{144} - 12C_{166} + 16C_{456})$$

$$\nu_2 = \frac{1}{35}(C_{111} + 4C_{112} - 5C_{123} + 19C_{144} + 2C_{166} - 12C_{456})$$

$$\nu_3 = \frac{1}{35}(C_{111} - 3C_{112} + 2C_{123} - 9C_{144} + 9C_{166} + 9C_{456}). \quad (6)$$

Using the values of C_{ijk} for columbium given in Table III in these equations results in $\nu_1 = -4.59 \pm 0.38$, $\nu_2 = -3.80 \pm 0.15$, and $\nu_3 = +0.78 \pm 0.09$, all in units of 10^{12} dyn/cm^2 . These values are seen to be in very good agreement with the values measured for the polycrystal sample B shown in Table V which provides an indirect check on both sets of TOEC.

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³¹ W. Voigt, *Lehrbuch der Kristallphysik* (B. G. Teubner, Leipzig, 1928), p. 962.

Lithium Niobate: Effects of Composition on the Refractive Indices and Optical Second-Harmonic Generation*

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Crystals of lithium niobate have been grown with varying amounts of Li_2O , Nb_2O_5 , and MgO present in the melt. The refractive indices have been measured and the birefringence so obtained correlated with the phase-matching temperature for second-harmonic generation (SHG) and with the composition. The use of optical methods is described for the study of compositional uniformity and a method of correcting the effects of uniform composition gradients on SHG described.

I. INTRODUCTION

An extensive literature already exists on lithium niobate with reference to its use in nonlinear optics for mixing or modulating optical beams. It was first proposed in this connection by Boyd *et al.*,¹ who described

its nonlinear properties and refractive-index data suitable for calculating the details of phase matching. Miller *et al.*² extended this data by pointing out the favorable property of thermally tunable refractive indices present in lithium niobate and showed how this could be used to obtain noncritical phase matching. Hobden and Warner³ have given extensive details of the wavelength and thermal variation of the refractive

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²R. C. Miller, G. D. Boyd, and A. Savage, Appl. Phys. Letters 6, 77 (1965).

³M. V. Hobden and J. Warner, Phys. Letters 22, 243 (1966).