Short Communication

A temperature-dependent dispersion equation for congruently grown lithium niobate

The refractive indices of lithium niobate have been extensively studied for both stoichiometrically grown and congruently grown crystals. In the former category, Boyd *et al.* [1, 2] measured the wavelength dependence of the refractive indices in the 0.4–4 micron region, while Hobden and Warner [3] examined the temperature dependence and published a temperature-dependent Sellmeier equation covering the visible region of the spectrum. The congruently grown material has been studied by Nelson and Mikulyak [4] who measured the room-temperature refractive indices in the range 0.4–3.1 microns. Subsequently, Smith *et al.* [5] published the results of measurements on the variation with temperature of the refractive index at the two wavelengths 0.632 99 and 3.3922 μm, as well as data on the wavelength dispersion at room temperature. The variation of the refractive indices with the lithium/niobium ratio in the melt has also been studied [6] and was found to have a significant effect on the phase-matching temperature of nonlinear optical processes.

The importance of the congruently grown material lies in the good compositional and optical homogeneity that may be obtained, enabling crystals up to at least 50 mm in length to be used in parametric frequency doubling and mixing experiments. Also, the relatively strong temperature-dependence of the extraordinary refractive index frequently allows the material to be used in the non-critical phase-matching configuration, with consequent advantages in conversion efficiency. For these reasons we have found it desirable to be able to predict the phase-matching temperature of congruently grown lithium niobate for general three beam parametric interactions, and in consequence we have derived from published data a temperature-dependent Sellmeier equation for the refractive indices. The predictions of this equation are compared with some experimental results obtained from difference-frequency mixing of two visible lasers in a lithium niobate crystal to produce infrared radiation.

The form of the equation we used is as described by Hobden and Warner [3]. In a slightly recast form this reads·

$$n^2 = A_1 + \frac{A_2 + B_1 F}{L^2 - (A_3 + B_2 F)^2} + B_3 F - A_4 L^2 \quad (1)$$

where $A_1, A_2, A_3, A_4, B_1, B_2$ and B_3 are constants and L is the vacuum wavelength in microns. F, which contains the temperature dependence, is given by $F = (T - T_0)(T + T_0 + 546)$ where T_0 is a constant and T is the temperature in degrees centigrade. A separate equation is required for each of the two principal refractive indices. When $T = T_0$, the equation reduces to the temperature-independent Sellmeier equation

$$n^2 = A_1 + \frac{A_2}{(L^2 - A_3^2)} - A_4 L^2 \quad (2)$$

The procedure adopted for estimating the parameters in Equation 1 was as follows. First, Equation 2 was fitted by a least-squares method to the wavelength dependent data of Nelson and Mikulyak [4]. This data was obtained at 24.5° C, which gives the value for T_0. For both the ordinary and extraordinary index, the 30 data points of Nelson and Mikulyak were fitted by Equation 2 with a standard deviation of 1.3×10^{-4}, consistent with the quoted experimental uncertainties of $\pm 2 \times 10^{-4}$. Having obtained values for the parameters A_n, the constants B_n were estimated from the temperature data of Smith *et al.* [5]. These authors expressed the temperature dependence of the refractive index for two wavelengths (0.632 99 and 3.3922 microns) in the form of a polynomial:

$$n = n_0(1 + d_1 T + d_2 T^2 + d_3 T^3 + d_4 T^4) \quad (3)$$

where n_0 is the refractive index at 0° C. There are four sets of the coefficients n_0 and d_n, corresponding to the two refractive indices at each of the two wavelengths. Using the published values of these coefficients and the A_n values already obtained, a least-squares fit was performed for

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TABLE I Coefficients of Equation 1 for the ordinary and extraordinary refractive indices

	A_1	A_2	A_3	A_4	B_1	B_2	B_3
Ordinary	4.9048	0.11775	0.21802	0.027153	2.2314×10^{-8}	-2.9671×10^{-8}	2.1429×10^{-8}
Extraordinary	4.5820	0.09921	0.21090	0.021940	5.2716×10^{-8}	-4.9143×10^{-8}	2.2971×10^{-7}

Equation 1 at 20° C intervals in the range 0–500° C. For each refractive index the fit minimized the total deviation from Equation 3 for both wavelengths simultaneously. The standard deviation of the fit over the whole temperature range was 1.8×10^{-4} for both the ordinary and extraordinary indices and the results obtained for the coefficients of Equation 1 are shown in Table I.

As a check on the accuracy of the equation in predicting phase-matching temperatures, we have some data obtained from difference-frequency generation in lithium niobate. Infrared radiation in the range 2.1–3.2 microns was generated by parametric mixing of two visible lasers – a single-mode argon ion laser operating on the 488 nm line and a dye laser tunable over the range 565–640 nm. The mixing was carried out using type 1, non-critical, phase matching in a congruently grown, 50 mm long lithium niobate crystal (grown by Barr and Stroud) held in a stabilized oven. The wavelength of the dye laser could be determined to a precision of about 1 in 10^6 by comparison with a stabilized helium–neon laser in a wavemeter, while the oven temperature was known to within ± 0.05° C in the range 200–400° C. The phase-matching temperatures measured for ten different infrared wavelengths are shown in Table II, together with the temperatures predicted by Equation 1. There is good agreement between the two sets of figures and the systematic difference of about 3° C which is apparent over most of the range is probably not significant given the magnitude of the uncertainties in the input data. ($\partial n_e/\partial T$ is approximately equal to $1 \times 10^{-4} (° C)^{-1}$ at 250° C and so, neglecting the considerably smaller temperature variation of the ordinary index, an error of 1° C in phase-matching temperature is approximately equivalent to an error of 1×10^{-4} in the refractive index at 488 nm.)

In view of the wide range of wavelengths involved in the data of Table II and the good agreement between theory and experiment, we believe that Equation 1 should provide a simple, practical method for predicting phase-matching temperatures with reasonable accuracy in congruently grown lithium niobate over the bulk of the transparency range (0.4–4.5 microns) of the material.

TABLE II The observed and calculated phase-matching temperatures (° C) as a function of infrared wavelength (microns)

Wavelength	T (observed)	T (calculated)
2.159	180.0	176.4
2.249	200.0	200.1
2.337	220.0	221.3
2.432	240.0	242.2
2.541	260.0	264.0
2.643	280.0	282.6
2.771	300.0	303.7
2.901	320.0	323.0
3.049	340.0	342.6
3.235	360.0	364.5

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