

# Improved Sellmeier equation for congruently grown lithium tantalate

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**Abstract** We present a Sellmeier equation that describes the refractive index dispersion of congruently grown LiTaO<sub>3</sub>, accurate from 0.3 to 5 μm and from 23 to above 200°C. Data were acquired by two different ways: directly by the minimal deviation technique in the visible and near-infrared range and indirectly by quasi-phase-matched optical parametric generation in PPLT in the mid-infrared range. The data was fit to a Sellmeier equation based on two oscillators in the UV and an infrared correction. The resulting equation accurately predicts the tuning curves for optical parametric generation in the infrared, as well as the correct quasi-phase-matching conditions for frequency conversion into the ultraviolet by second-harmonic generation.

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## 1 Introduction

Lithium tantalate (LiTaO<sub>3</sub>) is a ferroelectric which has been used extensively to obtain frequency conversion processes, such as second-harmonic generation [1], optical parametric generation [2], and sum-difference frequency generation [3]. Due to its large transparency range (0.28–5.5 μm) and its large nonlinear coefficient  $d_{33}$  it has been used to obtain radiation in the ultraviolet, visible, and mid infrared.

The wavelength and temperature dependencies of the refractive index play a very important role in determining the wavelengths that will be generated in any given quasi-phase-matched frequency conversion process. In general, the refractive index is calculated using an empirical Sellmeier equation, which in turn is obtained by fitting refractive index data to an equation derived from the Lorentz model of the refractive index,

$$n^2(\omega) = 1 + \sum_j \frac{f_j}{\omega_{0,j}^2 - \omega^2}. \quad (1)$$

Here  $n$  is the refractive index,  $\omega$  is the optical frequency,  $\omega_{0,j}$  is the resonant frequency of the  $j$ th oscillator of the medium, and  $f_j$  is the “oscillator strength” of the  $j$ th oscillator. In general, the fitting parameters  $\omega_{0,j}$  and  $f_j$  are temperature dependent. DiDomenico and Wemple [4] proposed a model for ferroelectrics with oxygen-octahedra, such as lithium tantalate, where the main contributions to the dispersion in the visible and near-infrared stem from two inter-band transitions with resonance frequencies inside the ultraviolet region, one at 9 eV (~120 nm) and another at 5 eV (~240 nm); this model predicts the general optical properties of several oxygen-octahedra ferroelectrics quite well. However, in order to perform predictions for quasi-phase-matching it is necessary to know the refractive index with an accuracy of at least one part in  $10^3$ , preferably one part in  $10^4$  or better, for all the waves involved in the nonlinear process; in this case a more accurate Sellmeier equation is required.

There are several publications that report different Sellmeier equations for the refractive index of congruently grown and stoichiometric lithium tantalate. Bond [5] measured the index of congruently grown lithium tantalate from 0.4 to 4 μm at room temperature; Iwasaki et al. [6] reported

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the refractive index between 0.46 and 0.644  $\mu\text{m}$  from 23 to 630°C; they described the temperature dependence of the extraordinary index  $n_e$  in terms of the temperature dependence of the spontaneous polarization. Abedin and Ito [7] proposed a temperature-dependent Sellmeier equation based on the measurements given in Refs. [5] and [6], valid from 0.4 to 4.0  $\mu\text{m}$  in a temperature range of 25 to 300°C. Meyn and Fejer [8] proposed an equation that accurately predicts the dispersion of the refractive index of congruently-grown lithium tantalate for wavelengths between 0.325 to 0.532  $\mu\text{m}$  and temperatures between 20 to 260°C. Bruner et al. [9] found their own temperature-dependent equation for both congruently grown and stoichiometric lithium tantalate, based on three resonances, valid from 0.39 to 4.1  $\mu\text{m}$  and from 30 to 200°C. Nakamura et al. [10] presented temperature-independent Sellmeier equations for the ordinary and extraordinary refractive indices of stoichiometric lithium tantalate (undoped and MgO doped), valid from 0.440 to 1.050  $\mu\text{m}$ , and Kolev et al. [11] proposed a correction of the temperature-dependent Sellmeier equation of Ref. [9] to extend its validity up to the 6  $\mu\text{m}$  absorption edge of stoichiometric lithium tantalate. Juvalta et al. [12] have reported new equations for 0.25 to 0.5  $\mu\text{m}$  at room temperature for congruently grown and stoichiometric lithium tantalate. Although their precision is lower than that of the previously mentioned equations, they can be used to predict the refractive index below 300 nm.

The refractive indices of the congruently grown lithium tantalate predicted by Refs. [8] and [9] are similar but different enough to predict quasi-phase-matching tuning curves that are significantly different. In this work we present another temperature-dependent Sellmeier equation for congruently grown lithium tantalate, valid from below 0.3 to above 5  $\mu\text{m}$ . The refractive index between 0.458 and 1.06  $\mu\text{m}$  was measured directly using the minimum deviation angle technique and the data from 1.3 to 5  $\mu\text{m}$  was obtained indirectly by measuring the spectra obtained from quasi-phase-matched optical parametric generation. This equation accurately predicts the quasi-phase-matched optical parametric generation from 1.3 to over 5 microns obtained by pumping a periodically poled crystal with a 1.064  $\mu\text{m}$  beam and, in addition, predicts virtually the same indices of refraction in the ultraviolet reported by Juvalta et al. [12] and Meyn et al. [8].

## 2 Experiment

We measured the refractive index in the visible and near infrared directly using the technique of minimum deviation [5]. The measurements were performed on prisms obtained by cutting and polishing congruently grown z-cut single-domain lithium tantalate wafers (thickness of 0.51 mm; supplier CASTECH). The prisms were mounted inside an oven

capable of rising the temperature up to 230°C; the temperature of the prism was monitored by a 10 k $\Omega$  thermistor placed as close to the prism as possible ( $\sim 1$  mm); the apex angle of prism at room temperature was of 33.78°. The uncertainty with which the temperature was known was estimated to be  $\pm 0.2^\circ\text{C}$  for values close to room temperature; this uncertainty increased to about  $\pm 0.5^\circ\text{C}$  at temperatures around 200°C. The setup was mounted on a computer-controlled rotation stage that had a precision of  $1 \times 10^{-2}$  degrees. As light sources we used a tunable Ar<sup>+</sup> laser (457.9, 476.5, 488.0, 496.5, and 514.5 nm), a HeNe laser (632.8 nm), and a Nd:YVO<sub>4</sub> laser (1064.2 nm). The refractive indices were obtained from

$$n_e = \frac{\sin(\frac{\theta+\alpha}{2})}{\sin(\frac{\alpha}{2})}, \quad (2)$$

where  $n_e$  is the extraordinary refractive index,  $\theta$  is the angle of minimum deviation, and  $\alpha$  is the apex angle of the prism. The main source of error of these measurements was the uncertainty of  $\theta$ . The maximum uncertainty was  $\delta\theta \approx 5 \times 10^{-2}$  degrees, which introduced a relative uncertainty of the refractive index  $\delta n/n \approx 5 \times 10^{-4}$ .

The refractive index data at mid IR were obtained indirectly by measuring the wavelength of the signal beam produced by quasi-phase-matched optical parametric generation, similar to the technique used by Jundt [13] to determine the dispersion of lithium niobate. Conservation of energy and momentum lead to the well-known relationships

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i}, \quad (3)$$

and

$$\frac{n_p}{\lambda_p} = \frac{n_s}{\lambda_s} + \frac{n_i}{\lambda_i} \pm \frac{1}{\Lambda}, \quad (4)$$

where  $\lambda_p, \lambda_s, \lambda_i$  are the pump, signal and idler wavelengths,  $n_p, n_s,$  and  $n_i$  are the pump, signal and idler refractive indices and  $\Lambda$  is the poling periodicity. From these equations we find that the refractive index at the idler wavelength is given by

$$n_i = \frac{\lambda_p \lambda_s}{\lambda_s - \lambda_p} \left[ \frac{n_p}{\lambda_p} - \frac{n_s}{\lambda_s} \mp \frac{1}{\Lambda} \right]. \quad (5)$$

From (5), we see that if  $\lambda_s$  is measured and  $n_p$  and  $n_s$  are assumed to be known, we can determine  $n_i$ .

Using standard electrical poling techniques we made PPLT (periodically polarized lithium tantalate) structures with periodicities of 26.0, 27.0, 28.0, 29.0, 30.0, and 31.0  $\mu\text{m}$  to obtain optical parametric generation from 1.3 to 5  $\mu\text{m}$ . The mask-fabrication technique that we used [14] limited the accuracy of the periodicities to  $\approx \pm 50$  nm. Due to experimental constraints we did not measure the signal

wavelength directly. Instead, we measured the wavelength of the beam resulting from the sum-frequency mixing of the signal and pump beams, which always appears to some extent in periodically poled crystals. Using energy conservation we find that the signal wavelength is given by

$$\lambda_s = \frac{\lambda_p \lambda_{s+p}}{\lambda_p - \lambda_{s+p}}, \tag{6}$$

where  $\lambda_{s+p}$  is the measured sum-frequency wavelength, which typically is in the orange-red region of the spectrum. These wavelengths were measured with a CCD-based Ocean Optics spectrometer with  $\sim 0.3$  nm of resolution.

### 3 Sellmeier equation

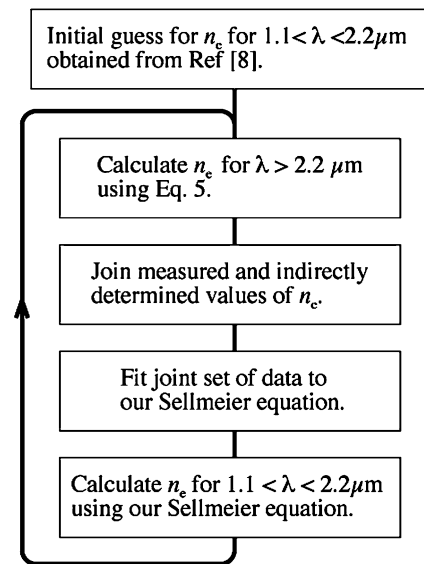
We fit the data to a Sellmeier equation with two oscillators with resonant frequencies located in the ultraviolet and a third oscillator in the mid infrared. From (1) we get

$$n_e^2(\lambda) = 1 + \frac{A\lambda^2}{\lambda^2 - \lambda_A^2} + \frac{B\lambda^2}{\lambda^2 - \lambda_B^2} + C\lambda^2, \tag{7}$$

where  $A$  and  $B$  are constants (for a given temperature) proportional to the oscillator strengths of the resonances located in the UV,  $\lambda_A$  and  $\lambda_B$  are the resonant wavelengths in the UV. The last term represents the contribution from the oscillator in the mid infrared, where the approximation  $D\lambda^2/(\lambda^2 - \lambda_B^2) \approx -D\lambda^2/\lambda_B^2 \equiv C\lambda^2$  has been made assuming wavelengths much smaller than the mid-infrared resonant wavelength.

To obtain the fitting parameters  $A$ ,  $B$ ,  $\lambda_A$ ,  $\lambda_B$ , and  $C$  for a given temperature, we use the following procedure. Assuming we have a reasonable estimate for the refractive indices of the pump and signal beams, obtained from the Sellmeier equation given in Ref. [8], we obtain our first estimate of the refractive index at the idler wavelengths by using the data obtained from optical parametric generation and (5). These inferred indices, together with the indices measured directly with the minimum deviation angle technique, are then fit to (7) using the Levenberg–Marquardt algorithm implemented in a commercial software program. This equation is then used to estimate the indices of the pump and signal beams, the indices of the idler wavelengths are once again calculated as before and the new set of data is fitted once again, and so on. This procedure, which is depicted graphically in Fig. 1, is repeated until the fitting parameters do not change significantly.

In order to make the curve fit converge it was necessary to fix one of the fitting parameters. Based on the work of Wemple and DiDomenico [4] we initially assumed that the UV resonances occur at approximately 9 and 5 eV, which correspond to 138 and 248 nm, respectively. Since all of our



**Fig. 1** Flow chart of the procedure employed to obtain the Sellmeier equation coefficients

**Table 1** Sellmeier equation coefficients for  $T = 23^\circ\text{C}$

$A$	$2.97584 \pm 0.00038$
$\lambda_A$	$0.138 \mu\text{m}$ (fixed arbitrarily)
$B$	$0.54622 \pm 0.0007$
$\lambda_B$	$0.24028 \pm 0.00065 \mu\text{m}$
$C$	$-0.023497 \pm 5.9 \times 10^{-5} \mu\text{m}^{-2}$

**Table 2** Temperature dependent Sellmeier equation coefficients

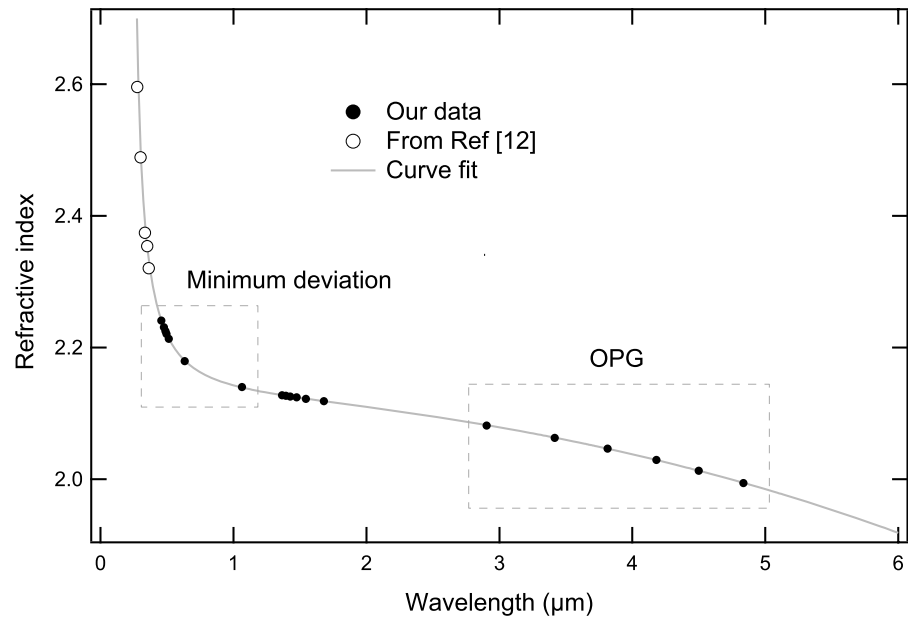
$A$	2.976
$\lambda_A$	$(0.13798 + 3.2 \times 10^{-8} T^2) \mu\text{m}$
$B$	$0.5387 + 3.6 \times 10^{-4} T - 1.2 \times 10^{-7} T^2$
$\lambda_B$	$0.24028 \mu\text{m}$
$C$	$(-0.02337 - 4.3 \times 10^{-6} T + 3.5 \times 10^{-9} T^2) \mu\text{m}^{-2}$

data were taken at wavelengths above 458 nm, the dispersion is not sensitive to the exact value of the first resonance, since  $A\lambda^2/(\lambda^2 - \lambda_A^2) \approx A$ . We arbitrarily fixed the value of  $\lambda_A$  at 138 nm in order to make the curve fit converge.

Figure 2 shows the data and curve fit obtained at room temperature ( $T = 23^\circ\text{C}$ ). As can be seen, the theoretical fit follows the data quite faithfully. We have included in this graph the data reported by Juvalta et al. [12]. Although the data were not included in our fitting routine, we see that within experimental error the theoretical fit predicts even the refractive index in the UV accurately. The coefficients that we obtained for the data at room temperature are given in Table 1.

The same process was repeated at different temperatures (23, 50, 80, 120, 180, and  $230^\circ\text{C}$ ). We took into account the

**Fig. 2** Dispersion of the refractive index at  $T = 23^\circ\text{C}$



thermal expansion of the crystal by assuming that periodicity at any temperature is given by

$$\Lambda(T) = \Lambda_{25^\circ\text{C}}(1 + a(T - 25) + b(T - 25)^2), \quad (8)$$

where  $\Lambda_{25^\circ\text{C}}$  is the periodicity at room temperature,  $a = 1.61 \times 10^{-5}$ ,  $b = 7 \times 10^{-9}$  and  $T$  is the temperature in  $^\circ\text{C}$  [15]. We found empirically that the temperature dependence of the refractive index could be predicted well if the temperature dependence was ascribed only to  $\lambda_A$ ,  $B$  and  $C$ , and letting  $A$  and  $\lambda_B$  constant. The final values for the temperature-dependent Sellmeier equation are given in Table 2.

In order to validate the usefulness of this equation, we compared its predictions with several experimental results. Figure 3 shows the signal wavelengths obtained by optical parametric generation using several periodicities (26.0, 27.0, 28.0, 29.0, and 30.0  $\mu\text{m}$ ) at two different temperatures (23 and  $180^\circ\text{C}$ ) and the theoretical predictions obtained using our formula. For comparison we include the prediction using the formula given in Ref. [8]. As can be seen, at  $23^\circ\text{C}$  both formulas agree quite well among themselves and with the experimental data. However, at  $180^\circ$  the difference between the predictions of the two equations is more noticeable: our equation tends to predict slightly longer wavelengths than what are observed experimentally while the equation from Ref. [8] predicts slightly lower wavelengths. From this data it is difficult to state which equation predicts the optical parametric generation tuning curve more accurately.

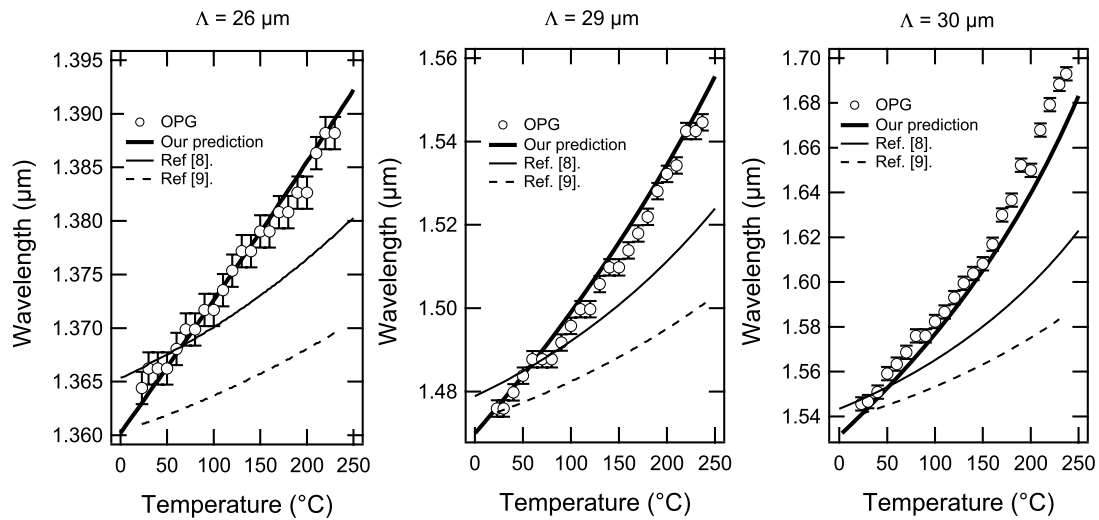
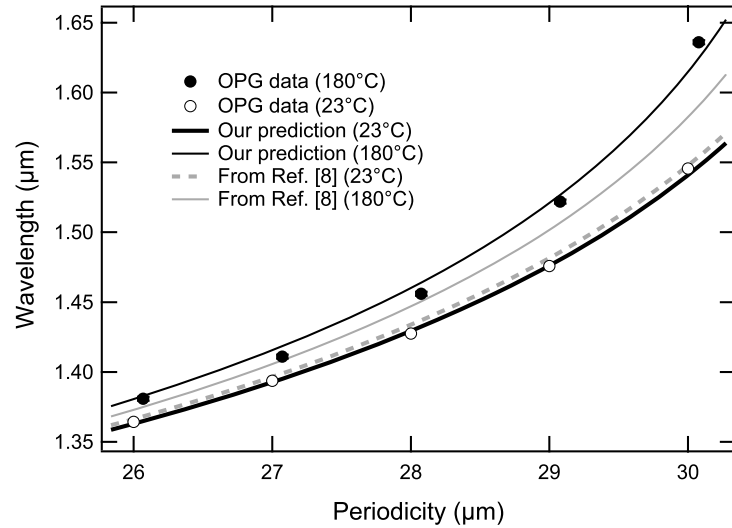
We made additional experiments to verify the accuracy of our equation in the infrared. We measured the signal wavelengths obtained by optical parametric generation approximately every  $10^\circ\text{C}$ , starting at 23 and ending at  $230^\circ\text{C}$ . Fig-

ure 4 shows the results from the measurements at 3 different periodicities (26.0, 29.0, and 30.0  $\mu\text{m}$ ), along with the theoretical predictions obtained from the equations given in Refs. [8, 9] and our equation. The idler wavelengths corresponding to the data shown in Fig. 4a are between 4.5 and 4.8  $\mu\text{m}$ . In all the cases the data agreed better with the predictions from the equation presented in this work. This is not surprising, since the temperature dependence of the equation of Ref. [8] was inferred solely from the temperature dependence of the second-harmonic generation of visible and near infrared to ultraviolet radiation; the data in the mid infrared was taken from a previous publication [5] which did not include temperature dependence. In order to accurately predict the curves shown in Fig. 4, the Sellmeier must be accurate at both the signal and idler wavelengths.

We also checked if our equation accurately predicted the data obtained by other researchers. To check its validity in the ultraviolet, we compared the experimental second-harmonic generation results presented in Ref. [8] with our predictions. Figure 5 shows the wavelength at which the peak of the second-harmonic process occurs as a function of the temperature for several periodicities. The circles correspond to data extracted from Fig. 2 of Ref. [8] and the solid lines are the predictions obtained using our equation. Thermal expansion of the crystal was taken into consideration in the theoretical predictions. As can be seen, the agreement between this data and the theoretical predictions is very good.

We also compared our predictions with data given in Ref. [16] by Luo et al.; this reference reports simultaneous second- and third-harmonic generation using the same poling periodicity. Using the Sellmeier equation of Ref. [8], they calculated that with a poling periodicity of 14.778  $\mu\text{m}$

**Fig. 3** Signal wavelength obtained by optical parametric generation at 23 and 180°C



**Fig. 4** Signal wavelength vs. temperature for three different poling periodicities (26, 29, and 30  $\mu\text{m}$ )

and at temperature of 74.1 $^{\circ}\text{C}$  they could obtain the second-harmonic of 1.342 nm radiation by first-order quasi-phase-matching, and at the same time the third-harmonic by third-order quasi-phase-matched sum-frequency mixing of the fundamental and the second harmonic. Although they did observe simultaneous second and third-harmonic generation, the temperatures at which each process reached its maximum efficiency were different, 92.5 $^{\circ}\text{C}$  for second-harmonic generation and 85.6 $^{\circ}\text{C}$  for third-harmonic generation, instead of the predicted value of 74.1 $^{\circ}\text{C}$ . Our equation predicts that at 85.6 $^{\circ}\text{C}$  the required periodicity to obtain third-harmonic generation is 14.78  $\mu\text{m}$ , which is identical to the nominal poling periodicity of the crystal.

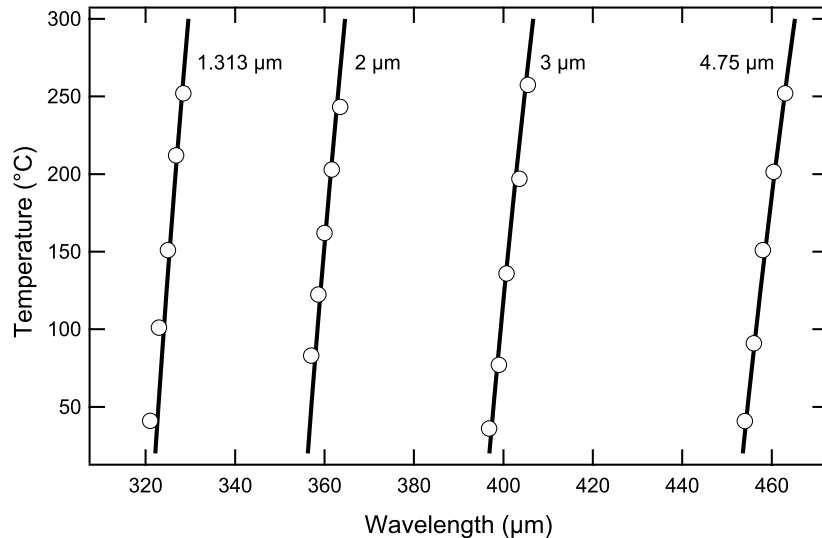
Finally, some of the discrepancies between the results presented in this work and the results of other researchers may be due to slightly different stoichiometric compositions

of the samples, which is typically quantified by the parameter  $x$  given by

$$x = \frac{[\text{Li}]}{[\text{Li}] + [\text{Ta}]}, \quad (9)$$

where [Li] and [Ta] are the concentrations of Li and Ta, respectively. It has been found that as  $x$  tends towards the stoichiometric value ( $x = 0.5$ ), the absorption edge of  $\text{LiTaO}_3$  shifts towards the ultraviolet [17–19]. The exact value of  $x$  will therefore affect the refractive index, especially in the near UV and visible regions. Additionally, the stoichiometry affects the absorption peaks of the  $\text{OH}^-$  vibration modes [20], which occur at around 2.8  $\mu\text{m}$ ; consequently  $x$  will influence the refractive index in the infrared. We do not know that exact value of  $x$  for the samples used in this work, but we assume that it is equal to or very near the values reported

**Fig. 5** Second-harmonic generation wavelength vs. temperature for four different periodicities: 1.313, 2, 3, and 4.75  $\mu\text{m}$ . The experimental data were taken from Ref. [8]



in literature,  $x = 0.485$  [17] and  $x = 0.486$  [14, 18], since the electric field that was required to produce the PPLT samples was—within experimental error—equal to the coercive field of congruently grown  $\text{LiTaO}_3$  (21–22 kV/mm).

#### 4 Conclusions

We have found a new Sellmeier equation that accurately predicts the refractive index of congruently grown lithium tantalate from 0.3 to 5  $\mu\text{m}$  and from room temperature to above 200 $^{\circ}\text{C}$ . This equation accurately predicts the quasi-phase-matched tuning curves for optical parametric generation in the infrared region ( $\sim 1.36$ – $4.83$   $\mu\text{m}$ ) in the temperature range mentioned earlier as well as the correct quasi-phase-matching conditions for frequency conversion into the ultraviolet.

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