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Extended Sellmeier equation for the extraordinary refractive index of 5% MgO-doped congruent LiNbO3 at high temperature

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MgO-doped congruent Lithium Niobate (MgO:CLN) is widely used in a variety of nonlinear optical processes, while its Sellmeier equation has only been derived from experiments under 200°C. Here we extend the temperature range for the Sellmeier equation study to as wide as 60 °C to 510 °C. This work is important to extend the operation temperature range of MgO:CLN crystal. [http://dx.doi.org/10.1063/1.4994104]

Lithium Niobate crystal is important as a nonlinear optical crystal because of its large nonlinear optical coefficient and wide transparent wavelength range. With periodical poling, it can be used in a variety of nonlinear optical processes for light generation from visible[1,2] to Mid-infrared regimes.[3,4] Its optical performance can be further improved by MgO-doping for lower coercive field[5] and higher photorefractive damage threshold.[6] Broad spectral tuning range has been reported using periodically-poled MgO-doped lithium niobate (PPMgOLN) crystal by temperature tuning.[7–10]

In these applications, the wavelength and temperature dependent Sellmeier equation is necessary to predict the phase matching geometry and for domain structure design of PPMgOLN crystal. The wavelength dependence has been studied in the wide spectral regimes of visible, mid-infrared (MIR), and infrared regions ranging from 0.4 μm to 5.0 μm.[11–13] However, for the temperature dependence, the reported researches[11–13] are limited below 200 °C, which limits the temperature tuning range and high temperature application of PPMgOLN.

In this paper, we derive a Sellmeier equation that is valid for a spectral range of 0.5-4 μm and temperature range of 60-510 °C. At temperature under 200 °C, this new equation predicts similar phase matching to earlier works. At temperature above 200 °C, it shows a significant improved accuracy in phase matching prediction compared with that of the previously reported Sellmeier equation.[13]This work provides accurate guidance for the applications of MgO:CLN at high temperatures.

To calculate the Sellmeier coefficients, we measure the signal and idler wavelengths of a quasi-phase-matched (QPM) optical parametric oscillator (OPO) due to its sensitivity to the refractive index of the nonlinear material.[14,15] The propagating waves (pump, signal and idler) need to satisfy both energy and momentum conservation conditions. The energy conservation condition for the three propagating waves can be expressed as:

$$\frac{1}{\lambda_p} = \frac{1}{\lambda_s} + \frac{1}{\lambda_i} \tag{1}$$

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where the λ_p, λ_s and λ_i stand for the wavelength of pump, signal and idler, respectively. In our case, the 5% MgO:CLN is periodically poled with period length Λ and the three propagating waves are extraordinary polarized (d_{ZZZ}). For collinear phase matching process, the conservation of momentum results in the phase matching condition:

$$\frac{n_e(\lambda_p, T)}{\lambda_p} - \frac{n_e(\lambda_s, T)}{\lambda_s} - \frac{n_e(\lambda_i, T)}{\lambda_i} - \frac{1}{\Lambda(T)} = \frac{\Delta k}{2\pi} \tag{2}$$

Here T is the temperature of the crystal, $n_e(\lambda, T)$ is the wavelength and temperature dependent extraordinary refractive index. Δk is the wavevector mismatch. Quasi-phase matching is achieved when $\Delta k = 0$. $\Lambda(T)$ is the poling period taking the thermal expansion into account, which can be written as:[16]

$$\Lambda(T) = \Lambda\left[1 + \alpha(\mathrm{T} - 25°\mathrm{C}) + \beta(\mathrm{T} - 25°\mathrm{C})^2\right] \tag{3}$$

where the thermal expansion coefficients are $\alpha = 1.54 \times 10^{-5}\ K^{-1}$, $\beta = 5.3 \times 10^{-9}\ K^{-2}$.

In this work, the format of Sellmeier equation $n_e(\lambda, T)$ is assumed to be similar to the one used by Jundt[12] for undoped CLN:

$$n_e^2 = a_1 + b_1 f + \frac{a_2 + b_2 f}{\lambda^2 - (a_3 + b_3 f)^2} + \frac{a_4 + b_4 f}{\lambda^2 - a_5^2} - a_6 \lambda^2 \tag{4}$$

The coefficients $\{a_j, j = 1, 2, 3, \ldots, 6\}$ account for the ultraviolet and infrared absorption pole, while $\{b_j, j = 1, 2, 3, 4\}$ represent the temperature dependence. And f is the temperature parameter. For T in degree Celsius, f is defined as:

$$f = (T - 24.5)(T + 570.28) \tag{5}$$

From equation (2), we know that for every pair of wavelength-sets $\{\lambda_p, \lambda_s, \lambda_i, \Lambda(T)\}$ measured from the OPO, we can only obtain the relative difference of the refractive indices. In order to determine the absolute value of the refractive index, we include the absolute refractive index values from previously published work of Ref. 17 in the calculation. The least-square method is applied, and the a_j and b_j coefficients are iteratively and systematically adjusted to minimize the error function defined as:

$$\chi^2 = \sum\nolimits_{n=1}^{N} (\Delta k_n)^2 + \sum\nolimits_{m} (\Delta n_{e,m})^2 \tag{6}$$

where

$$\Delta n_{e,m} = n_e^{New}(\lambda_m, T_m) - n_e^{Shen}(\lambda_m, T_m) \tag{7}$$

Δk_n is the wavevector mismatch calculated through the experimental data-sets $\{\lambda_p, \lambda_s, \lambda_i, \Lambda(T)\}$ measured in this work. $\Delta n_{e,m}$ is the absolute refractive index difference between the new Sellmeier equation and Ref. 17.

The experimental setup consists of a pump laser, a PPMgOLN crystal in a temperature controlled oven and an optical spectrum analyzer. The pump light is from a frequency-doubled Q-switch Nd:YVO_4 laser operating at 532nm, with pulse energy of 30μJ, and the pulse duration of 15ns. At repetition rate of 1kHz, the average power is 30mW. The PPMgOLN crystal comprises three sections with poling periods of 8.5 μm, 9.0 μm, 10 μm and 10.5 μm. The length of the crystal is 20mm. A home-made high-temperature oven is used to control the temperature of PPMgOLN crystal. It consists of a temperature stabilized Silicon Nitride ceramic crystal mount with build-in resistive heater and temperature sensor, which is calibrated to be of ~0.1°C accuracy. An enclosure is used to form a 20mm gap with still air around the mount and ensure its temperature uniformity, which is measured to be better than 0.2°C. The wavelength of the signal radiation from the OPO is measured by the OSA (ANDO AQ-6315A) with an accuracy of 0.05nm. As for idler, it is calculated theoretically based on the energy conservation condition equation (1). As shown in Figure 1, the output spectral tuning range could be largely extended by elevating the crystal temperature. For a given poling period of Λ = 8.5 μm, the output idler wavelength varies from 2.0 μm to 2.9 μm in the temperature range of 60 °C to 510 °C. By extending the temperature tuning range to 510 °C, the wavelength tuning range exceeds 900 nm, which is over three times large than the tuning range achieved below 200 °C.

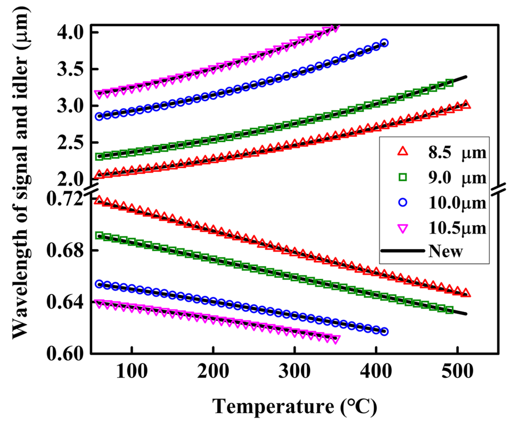

FIG. 1. Temperature dependent signal (0.6-0.72 µm) and idler (2.0-4.0 µm) wavelengths for multi-grating PPMgOLN with 532nm pump. The solid line is calculated from the new Sellmeier equation.

With the obtained wavelengths of signal and idler, and the refractive index values from Ref. 17, the coefficients in the Sellmeier equation (4) are derived by minimizing the error function χ^2 according to equation (6). Since the Sellmeier equation of MgO:CLN derived by Ref. 13 is reported to be valid in the spectral range of 0.5-4 µm, which covers our measured spectral range, the wavelength-dependent a_j coefficients from that work are set as our calculation base. The new Sellmeier equation coefficients are listed in Table I.

The new Sellmeier equation is compared with previously published works.[13,17] The quantitative results indicate that

$$\max\left|n_e^{New} - n_e^{Shen}\right|_{(\lambda_m,20°\mathrm{C})} < 4.0 \times 10^{-4}$$

$$\max\left|n_e^{New} - n_e^{Gayer}\right| < 3.7 \times 10^{-4}$$

The comparison with the absolute extraordinary refractive index from Ref. 17 is demonstrated at 20 °C for wavelengths $\lambda_m = \{0.53975\mu\mathrm{m}, 0.6328\mu\mathrm{m}, 1.0795\mu\mathrm{m}, 1.3414\mu\mathrm{m}\}$ and is shown in Figure 2. The maximum absolute value of the refractive index difference is smaller than 4.0×10^{-4}. As for the comparison with the work of Ref. 13, it is performed over the spectral range of 0.5-4µm and temperature range of 20-200 °C. The calculation based on the new Sellmeier equation is in good agreement with both previous works within the spectral and temperature range where they are reported to be valid.

When the crystal temperature is elevated from 200 °C to 510 °C, the good agreement between the new Sellmeier equation and the experimental data is maintained. While for Sellmeier equation from Ref. 13, the calculated idler wavelength starts to drift from the measured data once the temperature exceeds 200 °C, as we can see from Figure 3. The comparison of the calculated extraordinary refractive index shows the same tendency as presented in Figure 4. The absolute value of the maximum refractive index difference between the new equation and the equation from Ref. 13 varies from 3.7×10^{-4} to

TABLE I. Fitted coefficients for 5% MgO-doped congruent LiNbO3.

Coefficient	Value
a_1	5.756
a_2	0.0983
a_3	0.2020
a_4	189.32
a_5	12.52
a_6	1.32×10^{-2}
b_1	2.779×10^{-6}
b_2	5.763×10^{-8}
b_3	3.729×10^{-8}
b_4	1.415×10^{-4}

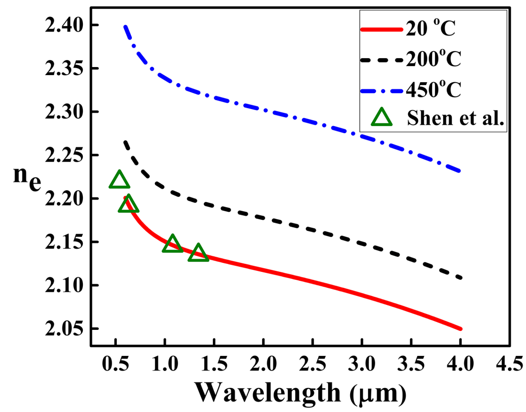

FIG. 2. Calculated extraordinary refractive index (lines) for 5% MgO-doped CLN as a function of wavelengths at different temperatures. The absolute n_e for different wavelengths at 20 °C from Ref. 17 are also plotted above (hollow triangle).

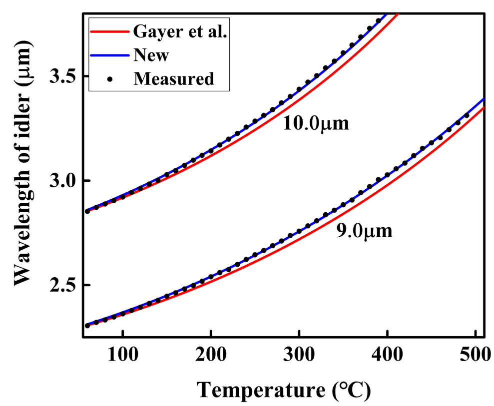

FIG. 3. Idler wavelength tuning with PPMgOLN, for Λ = 9.0μm and 10.0μm. The blue and red curves are calculated with the our Sellmeier equation and the Sellmeier equation from Ref. 13, respectively. These theoretical results are compared with the measured idler wavelengths (dot).

13.5×10^{-4} as the temperature increases from 200 °C to 510 °C. Thereby, we can see that the new equation enables more accurate prediction of the spectral and thermal characteristics of MgO:CLN at high temperatures.

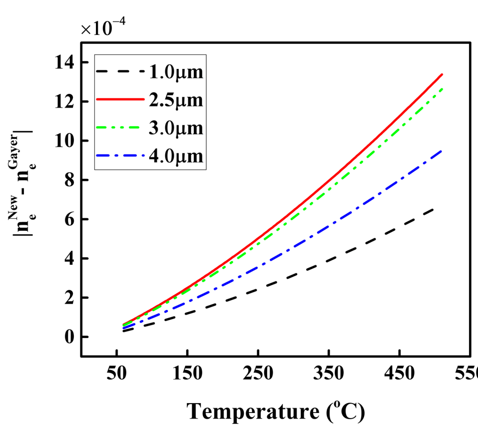

FIG. 4. The deviation of extraordinary refractive index of our experimental result and calculated result using Sellmeier equation from Ref. 13 in the temperature range of 60-510°C at wavelengths of 1.0 μm, 2.5 μm, 3.0 μm and 4.0 μm, respectively.

In summary, we present an extended Sellmeier equation study for 5% MgO-doped CLN crystal, based on an OPO experiment in the spectral range of 0.5-4 μm and ultra-wide temperature range from 60 to 510 °C. The Sellmeier coefficients are derived from the measured wavelengths of the OPO. At low temperature the new equation shows good agreement with the experimental results in previous works. At high temperatures, our results show the refractive index correction $\left|n_e^{New} - n_e^{Gayer}\right|$ of up to 13.5×10^{-4}, which is equivalent to a 53 nm improvement in phase matching calculation accuracy for idler wavelength around 2.0 μm. This work is helpful in providing accurate guidance on the applications of MgO:CLN in the high temperature range.

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