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New Sellmeier and Thermo-Optic Dispersion Formulas for $AgGaS_2$

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Abstract: This paper reports on the new Sellmeier and thermo-optic dispersion formulas that provide a good reproduction of the temperature-dependent phase-matching conditions for second-harmonic generation (SHG) and sum-frequency generation (SFG) of a CO_2 laser and a Nd:YAG laser-pumped $KTiOPO_4$ (KTP) optical parametric oscillator (OPO) in the 0.8859–10.5910 μm range as well as those for difference-frequency generation (DFG) between the two diode lasers in the 4.9–6.5 μm range and DFG between the two periodically poled $LiNbO_3$ (PPLN) OPOs in the 5–12 μm range thus far reported in the literature.

Keywords: $AgGaS_2$; phase-matching; Sellmeier equations; thermo-optic dispersion formula; frequency conversion; mid-IR generation

1. Introduction

In previous papers [1,2] we have reported the Sellmeier equations for $AgGaS_2$ that reproduce well the phase-matching angles of a Nd:YAG laser-pumped optical parametric oscillator (OPO) in the 2.6–5.3 μm range [3] and those of difference-frequency generation (DFG) between the signal and idler outputs of a Nd:YAG laser-pumped $LiNbO_3$ OPO in the 5–12 μm range [4] at room temperature. In addition, our Sellmeier and thermo-optic dispersion formulas [1,2] have also provided a good reproduction of the temperature-tuned phase-matching conditions for DFG between the two laser diodes in the 4.9–6.5 μm range [5] and those for DFG between two Nd:YAG laser-pumped PPLN OPOs in the 5–12 μm range [6] at elevated temperatures. However, this thermo-optic dispersion formula [2] constructed from the original work of Bhar et al. [7] has given dn_o/dT and dn_e/dT three times larger than the experimental values of Aggarwal and Fan [8] at 308 K (35 °C).

In order to clarify this large discrepancy, we measured dn_o/dT and dn_e/dT at 0.6328, 1.0642, 1.1523, 2.052, and 3.3913 μm by using a prism method and found that our experimental values of $dn_o/dT = 6.048 \times 10^{-5}$ °C^{-1} and $dn_e/dT = 6.549 \times 10^{-5}$ °C^{-1} at 1.0642 μm agree well with $dn_o/dT \simeq 5.5 \times 10^{-5}$ °C^{-1} and $dn_e/dT \simeq 6.0 \times 10^{-5}$ °C^{-1} at 1.06 μm measured by Aggarwal and Fan (Figure 1 of [8]). Thus, we have used our measured dn_o/dT and dn_e/dT and have constructed the new thermo-optic dispersion formula that provides a good reproduction of the abovementioned experimental results when coupled with our new Sellmeier equations that reproduce correctly the 90° phase-matched second-harmonic generation (SHG) wavelength of λ_1 = 1.7718 μm at 20 °C.

2. Experiments and Discussion

We used three different samples in the present experiments. One sample was cut at θ = 90° and ϕ = 45°. The other two samples were cut at θ_1 = 37.2° (θ_2 = 52.8°) and ϕ = 45°, and θ_1 = 39.7° (θ_2 = 50.3°) and ϕ = 0°. They were shaped as parallelepipeds with four polished surfaces corresponding to the two directions defined by the two values of the polar angle given. The dimensions of these samples are ~10 × 10 × 10 mm^3.

These samples were mounted on a temperature-controlled copper oven, which was set on a Nikon stepmotor-driven rotation stage having an accuracy of ±0.02° to vary only the polar angle θ. The temperature stability of the oven was ±0.1 °C. By using a Nd:YAG laser-pumped $KTiOPO_4$ (KTP) OPO as a pump source, we first measured the 90° phase-matching wavelengths for type-1 SHG by heating a θ = 90° and ϕ = 45° cut crystal from 20 °C to 120 °C at 20 °C intervals. The resulting tuning points (open circles) are shown in Figure 1. As can be seen from this figure, these results give $d\lambda_1/dT$ = +0.16 nm/°C and λ_1 = 1.7718 μm at 20 °C. Since $d\lambda_1/dT$ is defined as

$$\frac{d\lambda_1}{dT} = \left(\frac{\partial n_2{}^e}{\partial T} - \frac{\partial n_1{}^o}{\partial T}\right) / \left(\frac{\partial n_1{}^o}{\partial \lambda_1} - \frac{1}{2}\frac{\partial n_2{}^e}{\partial \lambda_2}\right), \quad (1)$$

we obtain $\partial n_2{}^e/\partial T - \partial n_1{}^o/\partial T = +1.0777 \times 10^{-5}$ °C^{-1} when using our Sellmeier equations presented in [1]. This is 27% lower than $\partial n_2{}^e/\partial T - \partial n_1{}^o/\partial T = +1.4692 \times 10^{-5}$ °C^{-1} given by our thermo-optic dispersion formula (T/K) presented in [2].

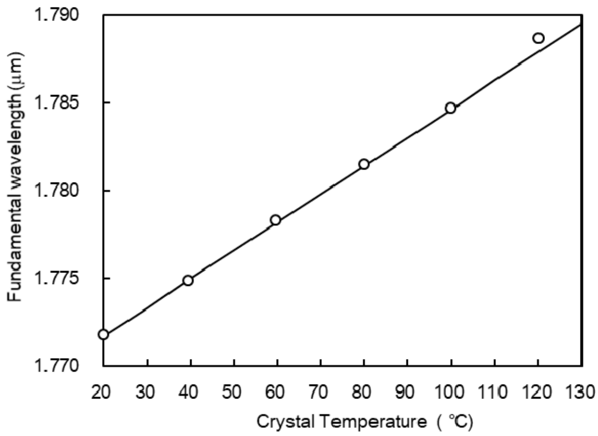

Figure 1. Temperature tuning curve for type-1 90° phase-matched second-harmonic generation (SHG) in $AgGaS_2$. The solid line is the theoretical curve calculated with the Sellmeier and thermo-optic dispersion formulas presented in this text. Open circles are our experimental points. The 90° phase-matching wavelength at 20 °C is λ_1 = 1.7718 μm with $\Delta T \cdot l$ = 7.3 °C cm.

We next measured the temperature variation of the phase-matching angles for SHG and sum-frequency generation (SFG) of a CO_2 laser (Coherent DEOS, Model EOM-10) and its SHG and third-harmonic generation as a pump source under the same experimental conditions as described in [9]. For reliable determination of a zero-angle of incidence, a reference He-Ne laser beam reflected from the entrance face of the crystal was aligned on a 0.2 mm slit located 2 m from the crystal.

The observed phase-matching angles and the temperature phase-matching bandwidths ($\Delta T \cdot l$) at full width at half-maximum (FWHM) that were determined from the temperature variation of the phase-matching angles ($\Delta\theta_{ext}/\Delta T$) and the angular acceptance angles ($\Delta\theta_{ext} \cdot l$) calculated with the following new Sellmeier equations, are tabulated in Table 1.

$$\begin{aligned} n_o{}^2 &= 5.79495 + \tfrac{0.22799}{\lambda^2 - 0.07963} - 2.4534 \times 10^{-3}\lambda^2 + 3.1814 \times 10^{-7}\lambda^4 - 9.7051 \times 10^{-9}\lambda^6, \\ n_e{}^2 &= 5.54077 + \tfrac{0.22223}{\lambda^2 - 0.09670} - 2.5240 \times 10^{-3}\lambda^2 + 2.0930 \times 10^{-6}\lambda^4 - 2.2984 \times 10^{-8}\lambda^6, \\ &(0.661 \le \lambda \le 10.5910), \end{aligned} \quad (2)$$

where λ is in micrometers. The phase-matching angles tabulated in Table 1 agree well with the values presented in [1,2]. We did not measure the temperature phase-matching bandwidth for type-1 THG of a CO_2 laser because the polarization directions of λ_1 and λ_2 were mutually orthogonal in the experimental setup, in which the 5.2955 μm input (λ_2) was generated by a type-1 SHG process. This index formula was obtained by adjusting the Sellmeier constants of Equations (1) and (2) presented in [1] to give the best fit to the type-1 90° phase-matched SHG wavelength of λ_1 = 1.7718 μm at 20 °C and the type-1 90° phase-matched DFG wavelength of λ_i = 5.333 μm generated by mixing λ_p = 661.183 nm and λ_s = 754.752 nm at 20 °C [10]. It reproduces correctly the phase-matching angles of a Nd:YAG laser-pumped OPO in the 2.6–5.3 μm range [3] and those of DFG between the signal and idler outputs of a Nd:YAG laser-pumped $LiNbO_3$ OPO in the 5–12 μm range [4] described above.

Table 1. Temperature phase-matching properties of $AgGaS_2$ for harmonic generation of a CO_2 laser at 10.5910 μm.

Wavelength (μm) [1]			Phase-Matching Angle at 20 °C θ_{pm} (deg)	Angular Acceptance $\Delta\theta_{ext}\cdot l$ (deg·cm)	$\Delta T\cdot l$ (°C·cm)	
λ_1	λ_2	λ_3			Measured	Calculated
10.5910^o	10.5910^o	5.2955^e	68.5	1.77	35.8	35.5
5.2955^o	5.2955^o	2.6478^e	32.6	0.66 (0.656)	20.6	20.8
5.2955^e	5.2955^o	2.6478^e	50.1	1.23	21.2	21.4
3.5303^o	3.5303^o	1.7652^e	33.4	0.44 (0.438)	15.2	15.2
3.5303^e	3.5303^o	1.7652^e	51.5	0.83	15.8	15.7
10.5910^o	5.2955^o	3.5303^e	43.3	0.80		28.5
10.5910^e	5.2955^o	3.5303^e	58.5	1.38	29.0	29.4

[1] $1/\lambda_1 + 1/\lambda_2 = 1/\lambda_3$. [2] The superscripts of the interacting wavelengths denote the polarization directions.

Although our Sellmeier and thermo-optic dispersion formulas presented in [2] reproduce well the temperature-dependent phase-matching conditions for the 90° phase-matched DFG between the two laser diodes in the 4.9–5.0 μm range [5] and those for the critically phase-matched DFG between the two PPLN OPOs pumped by a Nd:YAG laser in the 5–12 μm range [6], we found large differences between the temperature phase-matching bandwidths ($\Delta T\cdot l$) for SHG of a CO_2 laser and its harmonics that are tabulated in Table 1 and those listed in Table 1 of [2]. For instance, $\Delta T\cdot l$ = 35.8 °C cm observed in the present experiment at λ_2 = 5.2955 μm is a factor of ~4 smaller than the $\Delta T\cdot l$ = 139 °C cm value observed in previous experiments [2]. This may account for the systematic error in previous measurements of $\Delta\theta_{ext}/\Delta T$.

Aggarwal and Fan [8] have reported that dn_o/dT and dn_e/dT at 308 K (35 °C) measured by using the temperature-induced shift in the frequency of the interference fringes in the Fourier transform infrared spectroscopy (FTIR) transmittance spectrum of $AgGaS_2$ etalon are ~1/3 of our calculated values and those of Bhar et al. [7] and Zondy and Touahri [11]. Hence, we attempted to measure dn_o/dT and dn_e/dT at 0.6328, 1.0642, 1.1523, 2.052, and 3.3913 μm by using a minimum deviation method with a prism cut at an apex angle of 20°48′.

Using the raw data obtained by heating the prism from 20 °C to 140 °C at 20 °C intervals, we constructed a tentative thermo-optic dispersion formula to extrapolate dn_o/dT and dn_e/dT at 5.2955 and 10.5910 μm. The interpolated and extrapolated values were then iteratively adjusted to give the best fit to the measured temperature phase-matching bandwidths ($\Delta T\cdot l$) tabulated in Table 1. The newly constructed thermo-optic dispersion formula is expressed as

$$\begin{gathered}\frac{dn_o}{dT} = \left(\frac{5.4715}{\lambda^3} - \frac{13.1712}{\lambda^2} + \frac{11.1658}{\lambda} + 2.6459\right) \times 10^{-5} (^\circ\mathrm{C}^{-1}),\\ \frac{dn_e}{dT} = \left(\frac{4.8348}{\lambda^3} - \frac{11.6335}{\lambda^2} + \frac{9.8950}{\lambda} + 3.5135\right) \times 10^{-5},\\ (0.6328 \le \lambda \le 10.5910),\end{gathered} \tag{3}$$

where λ is in micrometers. Note that Equation (3) gives $\partial B/\partial T = \partial n_e/\partial T - \partial n_o/\partial T = 6.4991 \times 10^{-5} - 5.9924 \times 10^{-5} = 5.067 \times 10^{-6}$ °C^{-1} at 1.1523 μm, which is a factor of 1.7 larger than the value $\partial B/\partial T \simeq 3 \times 10^{-6}$ °C^{-1} which was reported by Suslikov et al. [12].

For reference, we have plotted dn_o/dT and dn_e/dT given by Equation (3) in Figure 2 together with the values at 20 °C (closed circles) estimated from the data points of Aggarwal and Fan at 308 K (35 °C) and 97 K (−176 °C) [8], and our experimental points (open circles). As can be seen from this figure, our calculated at 10.5910 μm are about 20% lower than the experimental values of Aggarwal and Fan [8].

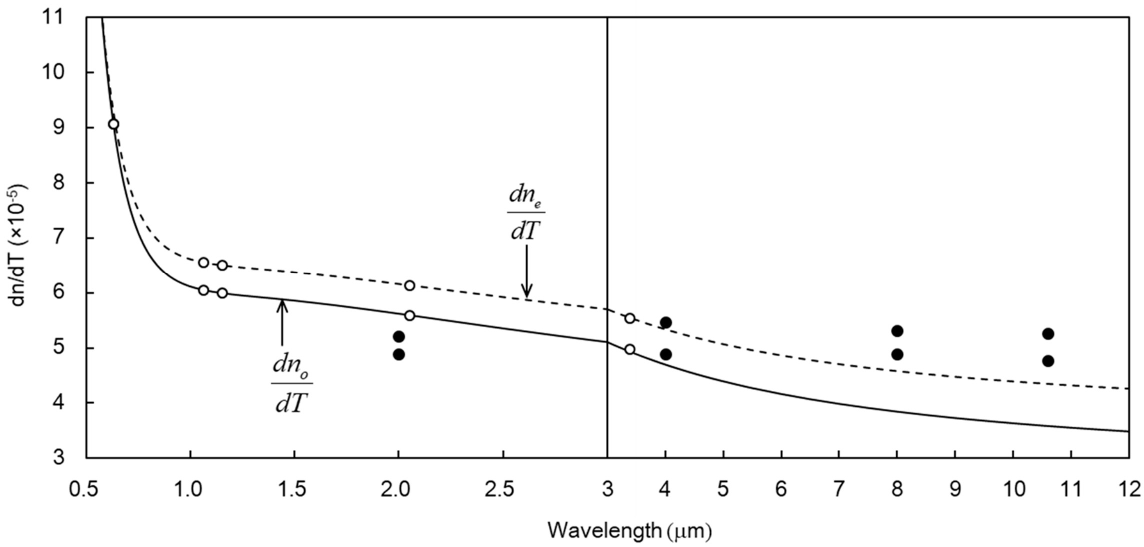

Figure 2. Thermo-optic dispersion curves of $AgGaS_2$. Closed circles are the values at 20°C estimated from the data points of Aggarwal and Fan at 308K (35 °C) and 97 K (−176 °C) [8]. Open circles are our experimental points.

In order to check the utility of Equations (2) and (3), we calculated the 90° phase-matching temperatures for type-1 DFG between the two diode lasers (λ_s = 0.791116 μm, λ_p = 0.68162–0.68290 μm). The resulting tuning curve (K) is shown in Figure 3 together with the experimental points (closed circles) of Willer et al. [5] and the tuning curves (Z/T), (H/K), and (R) that were calculated with the Sellmeier equations of Zondy and Touahri [11], Harasaki and Kato [1], and Roberts [13] coupled with our new thermo-optic dispersion formula (Equation (3)). Our tuning curve (K) reproduces the experimental points of Weller et al. [5] within an accuracy of ±5 °C except near 30 °C.

We next calculated the phase-matching temperatures at θ_{pm} = 34.30° for type-1 DFG between two Nd:YAG laser-pumped PPLN OPOs operating at λ_p = 1.60 μm and λ_s = 1.846−2.353 μm [6]. Since we found that our calculated values at θ_{pm} = 34.20° reproduce well the experimental points of Haidar et al. [6], we inserted our tuning curve (K) at θ_{pm} = 34.20° into Figure 4. It should be noted that our calculated values at θ_{pm} = 34.30° are ~12 °C larger than those of the tuning curve (K) because the phase-matching temperatures around the retracing point are strongly dependent on the phase-matching angle θ_{pm}. Also note that the dashed line (T/K) was formerly presented in Figure 3 of [6] by Haidar et al. and it agrees with fairly well with our tuning curve (K) at θ_{pm} = 34.20°. On the other hand, the Sellmeier equations of Roberts [13] give no retracing point for this process (Figure 1 of [6]). They give the phase-matching angles of θ_{pm} = 28.90°, 30.38°, and 33.24° to generate λ_i = 5.0, 8.5, and 12.0 μm at 20 °C, respectively. Hence, we did not use his index formulas for the present calculations.

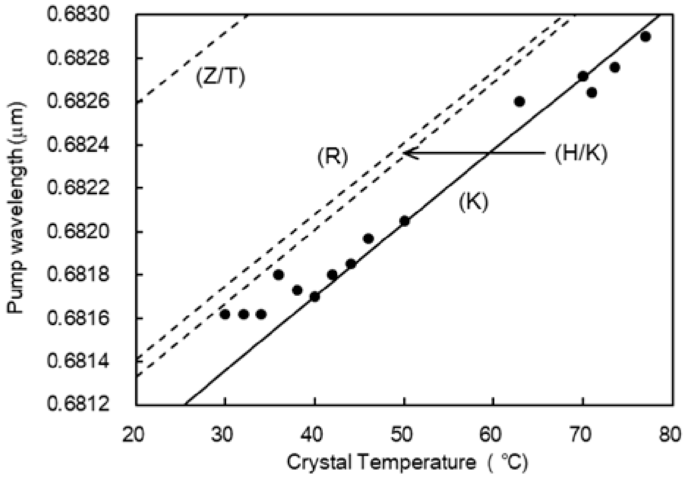

Figure 3. Temperature-tuning curves for type-1 difference-frequency generation (DFG) between the two laser diodes in $AgGaS_2$. The signal wavelength is 0.791116 μm. The solid line (K) is the theoretical curve calculated with the Sellmeier and thermo-optic dispersion formulas presented in this text. The dashed lines (Z/T), (H/K), and (R) are the theoretical curve calculated with the Sellmeier equations of Zondy and Touahri [11], Harasaki and Kato [1], and Roberts [13] coupled with our thermo-optic dispersion formula presented in this text. Closed circles are the data points taken from [5].

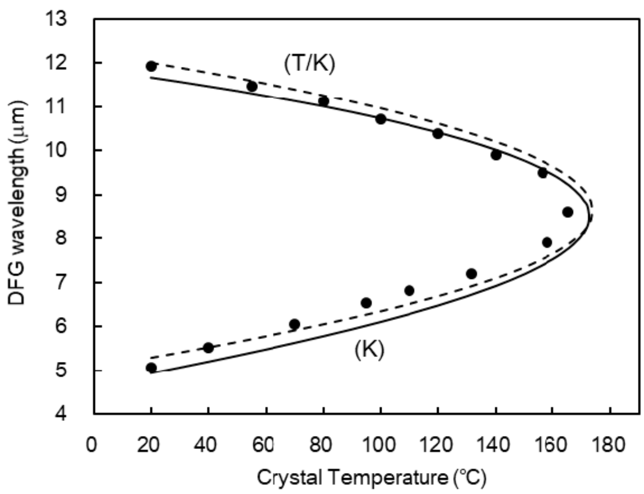

Figure 4. Temperature-tuning curves for type-1 DFG between two PPLN optical parametric oscillators (OPOs) pumped by a Nd:YAG laser in $AgGaS_2$. The solid line (K) is the theoretical curve at $\theta_{pm} = 34.20°$ calculated with the Sellmeier and thermo-optic dispersion formulas presented in this text. The dashed line (T/K) is the theoretical curve at $\theta_{pm} = 34.30°$ calculated with the Sellmeier and thermo-optic dispersion formulas presented in [2]. Closed circles are the data points taken from [6].

In order to check further the utility of our Sellmeier and thermo-optic dispersion formulas, we measured the phase-matching angles for type-2 DFG at $\lambda_i = 7.5190$ μm between the signal ($\lambda_p = 1.8645$ μm) and the idler ($\lambda_i = 2.4793$ μm) of a Nd:YAG laser-pumped $CsTiOAsO_4$ OPO [9,14] in a $\theta_1 = 39.7°$ ($\theta_2 = 50.3°$) and $\phi = 0°$ cut crystal. The measured phase-matching angles of $\theta_{pm} = 38.2 \pm 0.2°$ at 20 °C and $\theta_{pm} = 39.3 \pm 0.2°$ at 100 °C agree well the calculated values of $\theta_{pm} = 38.24°$ and 39.21°, respectively. Thus, we demonstrated the utility of Equations (2) and (3) in this wavelength range.

3. Conclusions

In this study, we have reported the new Sellmeier and thermo-optic dispersion formulas for $AgGaS_2$ that reproduce well the present experimental results as well as the temperature-dependent phase-matching conditions for type-1 DFG between the two laser diodes in the 4.9–6.5 μm range [5] and between two Nd:YAG laser-pumped PPLN OPOs in the 5–12 μm range [6]. We believe that these two formulas are highly useful for investigating the temperature-dependent phase-matching conditions for down conversion (OPO, OPA (optical parametric amplifier), and DFG) of near-IR solid state laser systems in different temporal regimes (pulse duration and pulse repetition rate) to the mid-IR spectral range.

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