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90° phase-matched up-conversion of CO_2 laser radiation in $AgGa_{0.86}In_{0.14}S_2$

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Abstract The CO_2 laser radiation at 10.5910–9.2714 μm was up-converted to the visible in the 90° phase-matched $AgGa_{0.86}In_{0.14}S_2$ crystal by mixing with the output of the 0.3547 μm pumped BBO optical parametric oscillator at 25–120 °C. The new Sellmeier and thermo-optic dispersion formulas that reproduce these experimental results correctly as well as the previously published data [Banerjee et al. in Appl Phys B 87:101, (2007); Opt Commun 227:202 (2007)] for difference-frequency generation at 4.05–6.98 μm and second-harmonic generation at 5.2955 μm are presented.

1 Introduction

In previous publications [1, 2], we have reported the Sellmeier and thermo-optic dispersion formulas for $AgGa_{0.86}In_{0.14}S_2$ that provide a good reproduction for difference-frequency generation (DFG) at 4.05–6.98 μm and second-harmonic generation (SHG) at 5.2955 μm. However, a somewhat large discrepancy between theory and experiment was encountered for the 90° phase-matched up-conversion of the CO_2 laser radiation achieved in this crystal. For instance, the experimentally observed OPO pump wavelengths for up-conversion of the 10.5910–9.2714 μm radiation are 14–17 nm shorter than the values given by the above-mentioned Sellmeier equations. In addition, a significant difference between theory and experiment was found for the temperature-dependent phase-matching conditions for this process. Thus, we have corrected these formulas so as to satisfy the new experimental results, and simultaneously fit the data points presented in [1, 2].

Here, we report the new experimental results on the 90° phase-matched up-conversion of the CO_2 laser radiation in $AgGa_{0.86}In_{0.14}S_2$ and the new Sellmeier and thermo-optic dispersion formulas for this crystal.

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2 Experiments and discussions

The experiments were carried out with a step-tunable CW CO_2 laser and a 0.3547 μm pumped BBO optical parametric oscillator (OPO) as the pump source. Both beams were combined with a ZnSe optical flat and collinearly incident on the refabricated, 7-mm-long, $\theta = 90°$ cut $AgGa_{0.84}In_{0.16}S_2$ crystal mounted on the temperature controlled oven.

The OPO pump power was adjusted to 5–10 mJ at 10 Hz to avoid the surface damage of the $AgGa_{0.84}In_{0.16}S_2$ crystal, and the CO_2 laser power was adjusted to less than 20 mW to avoid local heating. The unfocused beam diameter was 4 mm for the former and 2 mm for the latter.

The pump and output wavelengths were measured by a 0.5-m spectrometer with an accuracy of less than 0.1 nm.

By fixing the CO_2 laser wavelengths at 9.2714–10.5910 μm, and by tuning the BBO/OPO wavelength from 0.65 to 0.70 μm, we have measured the 90° phase-matching pump wavelengths at 25 °C. The resulting experimental points (open circles) are shown in Fig. 1 together with the theoretical curves (A) and (B) that were calculated with the Sellmeier equations of Banerjee et al. [1, 2] for this crystal and those of Badikov et al. [3] for $AgGa_{1-x}In_xS_2 (x = 0.008$ and $0.20)$, which differ significantly from the data points. The real line (C) is calculated with the following Sellmeier equations:

$$\begin{aligned} n_o^2 &= 5.8049 + \frac{0.2459}{\lambda^2 - 0.0737} - 0.00263\lambda^2 \\ n_e^2 &= 5.5738 + \frac{0.2553}{\lambda^2 - 0.0848} - 0.00273\lambda^2 \\ &\qquad (0.616 \leqq \lambda \leqq 10.591) \end{aligned} \quad (1)$$

where λ is in µm, and reproduce correctly the experimental points. This index formula was constructed by using the refractive indices previously measured by Banerjee et al. [1] at 0.6328, 1.0642, 10.5910 µm and those measured by us at 3.3913 µm, and adjusted them to give the best fit to the experimental points.

The measured acceptance angles and spectral phase-matching bandwidths at full-width at half-maximum (FWHM) are $\Delta\theta_{int}\cdot\ell^{1/2} = (2.3 \pm 0.1)$ deg cm$^{1/2}$ and $\Delta\lambda_p\cdot\ell = (0.4 \pm 0.1)$ nm cm, which agree well with the theoretical values of $\Delta\theta_{int}\cdot\ell^{1/2} = 2.35$ deg cm$^{1/2}$ and $\Delta\lambda_p\cdot\ \ell = 0.39$ nm cm for the CO_2 laser wavelength of 10.5910 µm and $\Delta\theta_{int}\cdot\ell^{1/2} = 2.34$ deg cm$^{1/2}$ and $\Delta\lambda_p\cdot\ell = 0.48$ nm cm for the CO_2 laser wavelength of 9.2714 µm.

Note that the ordinary and extraordinary refractive indices of $AgGa_{1-x}In_xS_2$ increase and the birefringence decrease as a function of In concentrations [3] as in the case of $AgGa_{1-x}In_xSe_2$ [4]. While the Sellmeier equations of Banerjee et al. [1, 2] give the ordinary refractive indices that are smaller than those of pure $AgGaS_2$ [5] at wavelengths longer than 2.42 µm; in contrast, the new Sellmeier equations give the normal dispersion [$n(AgGa_{0.86}In_{0.14}S_2) > n(AgGaS_2)$] throughout the whole spectral range. In addition, this index formula correctly reproduces the 90° phase-matched DFG between the $Ti{:}Al_2O_3$ laser at 0.84274 µm and the Nd:YAG laser at 1.0642 µm as well as the phase-matching angle of $\theta_{PM} = 80.8°$ and 73.0° for SHG of the CO_2 laser lines at 10.5910 [1, 2] and 10.2466 µm, respectively.

Next we measured the temperature-dependent phase-matching conditions by heating the crystal from 25 to 120 °C. The resulting tuning points (open circles) for the CO_2 laser wavelengths of 10.5910 and 9.5525 µm are shown in Fig. 2.

The dashed lines (A) are calculated with our new Sellmeier equations (Eq. 1) combined with the thermo-optic dispersion formula of Banerjee et al. [2], which differ again from the experimental points. The real lines (B) are calculated with our new Sellmeier equations and the thermo-optic dispersion formula given by

$$\begin{aligned} \frac{dn_o}{dT} &= \left(\frac{4.2311}{\lambda^3} - \frac{9.4687}{\lambda^2} + \frac{7.1842}{\lambda} + 5.9071\right) \times 10^{-5}\,(°C^{-1}) \\ \frac{dn_e}{dT} &= \left(\frac{4.5766}{\lambda^3} - \frac{10.4806}{\lambda^2} + \frac{8.2714}{\lambda} + 5.9655\right) \times 10^{-5}\,(°C^{-1}) \\ &(0.616 \leqq \lambda \leqq 10.591) \end{aligned} \quad (2)$$

where λ is in µm, and reproduce the experimental points correctly. The slope of the BBO/OPO pump wavelength versus crystal temperature is $d\lambda_p/dT = 0.101$ and 0.108 nm/ °C for the CO_2 laser wavelength of 10.5910 and 9.5525 µm, respectively. The temperature phase-matching bandwidth (FWHM) for this process is $\Delta T\ \ell = (3.4 \pm 0.1)$ °C cm, which agrees well with the calculated values of $\Delta T\ \ell = 3.3$ and 3.5 °C cm for the respective CO_2 laser wavelengths of 10.5910 and 9.5525 µm.

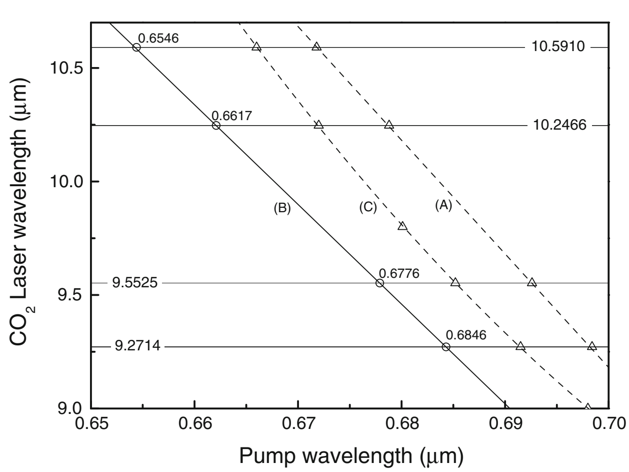

Fig. 1 90° phase-matching wavelengths for up-conversion of the CO_2 laser radiation in $AgGa_{0.86}In_{0.14}S_2$ at 25 °C. The *dashed line* (**a**) and the *real line* (**b**) are the theoretical carves calculated with the Sellmeier equations of Banerjee et al. [1, 2] and the present authors given in the text. The *dashed line* (**c**) is calculated with the Sellmeier equations of Badikov et al. [3] for $AgGa_{1-x}In_xS_2(x = 0.08$ and 0.20). *Open circles* experimental points. *Open triangle* calculated points

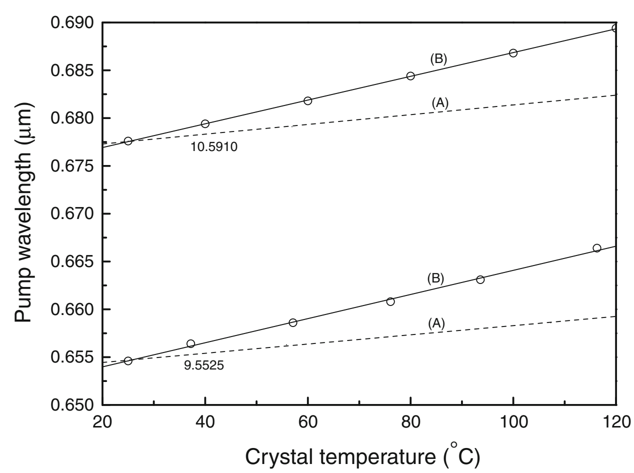

Fig. 2 Temperature-dependent 90° phase-matching wavelengths for up-conversion of the CO_2 laser radiation in $AgGa_{0.86}In_{0.14}S_2$. The *dashed lines* (**a**) are the theoretical curves calculated with the Sellmeier equations of the present authors coupled with the thermo-optic dispersion formulas of Banerjee et al. [1, 2] and the *real lines* (**b**) are the theoretical carves calculated those of the present authors presented in the text. *Open circles* experimental points

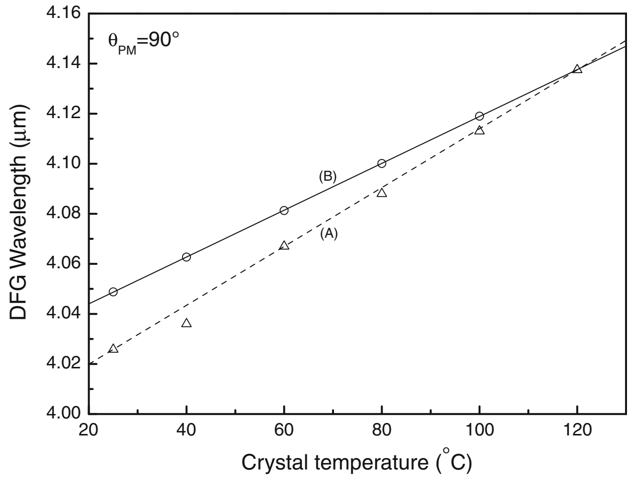

Fig. 3 Temperature-tuned 90° phase-matched difference-frequency generation between the BBO/OPO and the Nd:YAG laser in $AgGa_{0.86}In_{0.14}S_2$. The *dashed line* (**a**) and the *real line* (**b**) are the theoretical curves calculated with the Sellmeier and thermo-optic dispersion formulas of Banerjee et al. [1, 2] and those of present authors presented in this text. *Open circles* our experimental points. *Open triangle* experimental points taken from [2]

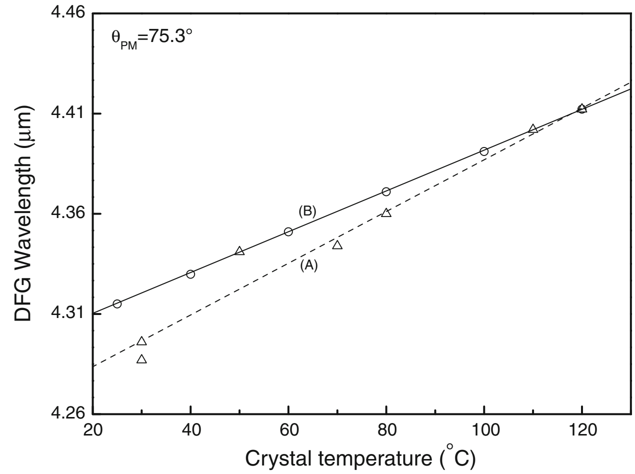

Fig. 4 Temperature-tuned difference-frequency generation between the BBO/OPO and Nd:YAG laser in $AgGa_{0.86}In_{0.14}S_2$ at $\theta_{PM} = 75.3°$. The *dashed line* (**a**) and the *real line* (**b**) are the same as noted in Fig. 3. *Open circles* our experimental points. *Open triangle* experimental points taken from [2]

In addition, we note that Eqs. (1) and (2) reproduce correctly the phase-matching points for SHG of the CO_2 laser line at 10.5910 μm given by Banerjee et al. [1, 2]. at 25–203 °C. However, because our calculated values for DFG based on the Nd:YAG laser do not fit their data points shown in Figs. 3 and 4 of [2] except at 120 °C, we have carefully checked these data points and found a distinct difference between the DFG wavelengths obtained at 25 °C by mixing the Ti:Al_2O_3 laser the Nd:YAG laser [1] and by mixing the BBO/OPO and Nd:YAG laser [2] in the same crystal. The former is 4.0497 μm as noted in the preceding, while the latter is 4.0258 μm (Fig. 3 of [2]).

In order to clarify this inconsistency, we have once again measured DFG between the BBO/OPO and the Nd:YAG laser under the identical experimental conditions described in [2]. The resulting tuning points (open circles) are shown in Figs. 3 and 4 together with the data points (triangles) taken from [2]. As can be seen from these figures, our data points agree excellently with the theoretical values calculated with Eqs. (1) and (2). Thus, the data point shown in Fig. 3 of [2] is thought to be in error.

3 Conclusion

In summary, we have reported the 90° phase-matched up-conversion of the CO_2 laser radiation at 9.2714–10.5910 μm in $AgGa_{0.86}In_{0.14}S_2$. These data were used to construct the new Sellmeier and thermo-optic dispersion formulas that provide the excellent reproduction of these experimental results as well as the previously published data [1, 2] of DFG at 4.05–6.98 μm and SHG of the CO_2 laser at 10.5910 μm. We believe that these Sellmeier and thermo-optic dispersion formulas are highly useful for predicting the temperature-dependent phase-matching conditions for frequency conversion in the $AgGa_{1-x}In_xS_2$ ($x \leqq 0.14$) crystals when combined with our index and thermo-optic dispersion formulas for pure $AgGaS_2$ [5].

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