Design considerations of an infrared spectrometer based on difference–frequency generation in $AgGaSe_2$

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The availability of new nonlinear optical materials such as $AgGaS_2$ and $AgGaSe_2$ and improvements in compact, tunable, pulsed and continuous-wave (cw) solid-state pump lasers now make it possible to generate tunable, infrared narrow-band coherent radiation over a wide wavelength range (4–18 μm) by means of difference–frequency generation (DFG). This article describes the wavelength and output-power characteristics of a tunable infrared source based on $AgGaSe_2$ and certain proven cw near-infrared pump sources for application to high-resolution spectroscopy.
Key words: Nonlinear optics, design of specific laser systems, infrared spectra.

Introduction

There is a need for continuing improvement of convenient laser-based sources applicable to high-resolution spectroscopy. As virtually all fundamental vibrational modes of molecules and molecular ions lie in the 2–20-μm-wavelength fingerprint region, tunable monochromatic probe lasers in this wavelength range are particularly useful for high-sensitivity, high-resolution (<100 MHz) and time-resolved molecular spectroscopy. Optimum spectral control and frequency stability are achieved with cw laser sources. Suitable cw tunable infrared-laser sources are presently limited to color-center lasers, lead salt diode lasers, sources based on sideband generation by CO and CO_2 lasers, and difference–frequency mixing sources. The color-center lasers require liquid nitrogen operation and are limited to a relatively small spectral region between 1.4 and 3.5 μm.[1] Lead salt diode lasers cover the 3–30-μm-wavelength range, but also require cryogenic operating temperatures (<100 K).[2] As the wavelength-tuning range per individual diode is limited to ~100 cm^{-1} (~400 cm^{-1} for MBE structures) and has many undesirable discontinuities, a large collection of diodes is required to cover the fingerprint-wavelength range. With continuous-tuning ranges limited to only 0.5 cm^{-1}, CO and CO_2 sideband lasers provide tunable infrared radiation between 5 and 6 μm and 9 and 11 μm, respectively.[3]

A source that is based on DFG, which was originally developed by Pine,[4] and in which an Ar^+ laser was mixed with a cw dye laser in $LiNbO_3$, has proved very useful for high-resolution infrared spectroscopy, but it is limited to wavelengths shorter than 4 μm by the infrared transmission characteristics of $LiNbO_3$. Using $LiIO_3$ as the nonlinear medium, Oka and coworkers[5] extended the long-wavelength limit for cw DFG sources to nearly 5 μm. Recently, we demonstrated the operation of a continuously tunable cw DFG spectrometer in the 4–9-μm-wavelength range based on 90° type I phase matching in $AgGaS_2$.[6] A 45-mm-long nonlinear optical crystal was pumped by two single-frequency (dye and Ti:sapphire) lasers. Using this spectroscopic source, absorption spectra of several stable molecular systems have been detected, e.g., H_2O, N_2O, NH_3, and CO. Additionally, we have observed transient spectra of vibrationally excited CO (v = 1, 2), generated by 193-nm ArF excimer laser-flash photolysis of acetone.

This paper describes the design considerations of a novel, midinfrared DFG-based laser source that is continuously tunable from 4–18 μm by mixing two cw single-frequency lasers in $AgGaSe_2$. It is based on design parameters resulting from the recent commercial availability of improved nonlinear optical crystals and new solid-state pump-laser sources.

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Characteristics of $AgGaSe_2$

A nonlinear optical material must meet several criteria to be useful in a three-wave parametric-mixing process. The material must be transparent for all three beams—pump, signal, and idler—participating in the process. Other conditions important to achieving a high nonlinear conversion efficiency are a large effective nonlinear coefficient d_{eff}, a long crystal length, a high optical-damage threshold, and phase matchability for available pump sources. The ternary chalcopyrite $AgGaSe_2$ shows a wide transparency range from 0.71 to 18 μm.[7] These crystals have recently become available commercially in large linear dimensions (> 40 mm) and with high optical quality.[8] The absorption coefficient of this material in its transmission range is comparable with that of $AgGaS_2$.[9] $AgGaSe_2$ is a birefringent uniaxial negative crystal ($n_e - n_o = -0.032$ at 5 μm) with a large nonlinear coefficient d_{36}. A value of 33 pm/V ± 10% has been determined for d_{36} from second-harmonic-generation (SHG) experiments in $AgGaSe_2$ with a Q-switched CO_2 laser at 10.6 μm as a pump source.[10] The optical-surface-damage threshold for $AgGaSe_2$ has been determined to be as high as 20–30 MW/cm^2 for 10-ns pulses at 2 μm.[9,11] The optical properties of several nonlinear optical materials useful for infrared DFG are summarized in Table 1. $ZnGeP_2$ has the highest figure of merit (d_{eff}^2/n^3) among these materials, owing to its extremely large nonlinear optical coefficient. As this material suffers from strong absorption in the 0.74–2-μm-wavelength region, the DFG process requires infrared pump sources emitting at wavelengths longer than 2 μm. However, noncritical phase matching in $ZnGeP_2$ cannot be realized for pump sources beyond 2 μm, as shown by our calculations based on the Sellmeier equations from Ref. 13, thus restricting the usefulness of $ZnGeP_2$. The figure of merit for $AgGaSe_2$, which scales linearly with the infrared DFG power, is nearly two times smaller than that for $ZnGeP_2$, but nearly six times larger than that for $AgGaS_2$.

Optimized parametric-frequency conversion requires phase matching of the three interacting waves. For a birefringent, nonlinear optical crystal this can be accomplished by means of crystal orientation, temperature, and pump-laser wavelength tunability.

Table 1. Characteristics of Important IR Nonlinear Optical Crystals

Material	Transparency Range (μm)	Absorption (% cm^{-1})[a]	Damage Threshold (MW/cm^2)	Relative Figure of Merit
$AgGaS_2$	0.5–13[b]	1.0 (1 μm)	25 (10 ns)[c]	42.3[d]
$AgGaSe_2$	0.71–18[b]	0.4 (2 μm)	20–30 (10 ns)[c]	242[d]
$ZnGeP_2$	0.74–12[b]	1.0 (5 μm)	50 (25 ns)[a]	625[d]
$LiNbO_3$	0.35–4.5[e]	0.15 (1 μm)	300 (10 ns)[a]	3.5[d]
$LiIO_3$	0.31–5.5[e]	0.08 (1 μm)	2000 (1 ns)[a]	1.5[d]

[a]Ref. 30.
[b]Ref. 7.
[c]Ref. 9.
[d]Ref. 29.

In the case of $AgGaSe_2$, the ability to tune the wavelength by varying the temperature is small.[14] Hence, we propose to use either two tunable, near-infrared cw pump sources, as in case of $AgGaS_2$,[6] or to use angle tuning.

90° Type I Phase Matching

The highest nonlinear frequency-conversion efficiency is achieved with 90° type I (noncritical) phase matching. Type II ($e \rightarrow o + e$) phase matching at $\theta = 90°$ in $AgGaSe_2$ is specifically ruled out by the combination of negative birefringence and $\bar{4}2m$ crystal symmetry. The infrared wavelength-tuning characteristics for both $AgGaS_2$ and $AgGaSe_2$ for 90° type I ($e \rightarrow o + o$) phase matching based on the Sellmeier constants from Refs. 15 and 16 are shown in Fig. 1. This comparison of phase-matching curves indicates the respective infrared-wavelength-tuning ranges for DFG in $AgGaS_2$ and $AgGaSe_2$ using several different pump-laser sources. In the case of $AgGaS_2$, single-frequency lasers in the visible spectral region can be employed as pump sources. As the transparency, and in particular the dispersion range, of $AgGaSe_2$ is shifted toward longer wavelengths as compared to $AgGaS_2$, the phase-matching conditions imply that two tunable pump sources above 1 μm are necessary

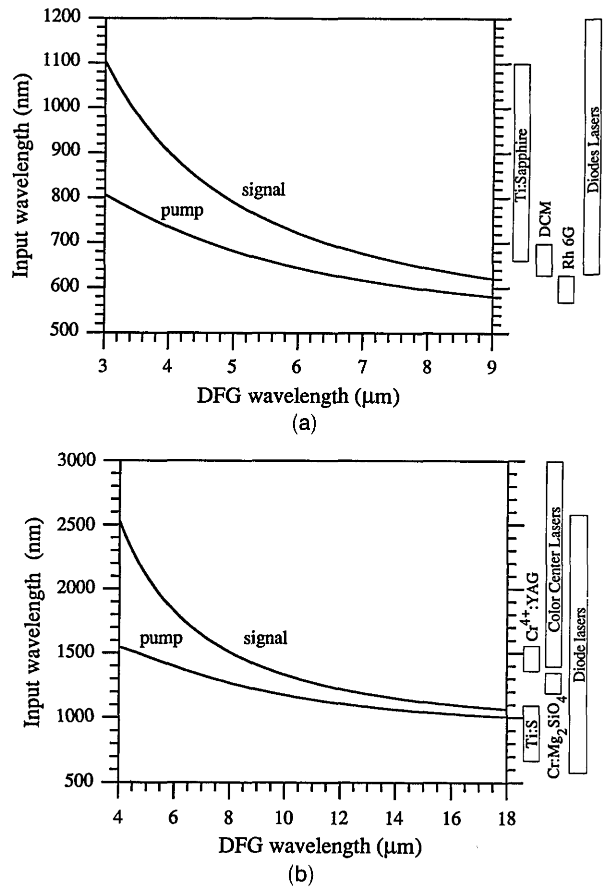

Fig. 1. Infrared-wavelength-tuning characteristics with corresponding pump sources for (a) $AgGaS_2$ and (b) $AgGaSe_2$. The 90° type I phase-matching curves of $AgGaS_2$ and $AgGaSe_2$ are based on the refractive-index data given in Refs. 15 and 16, respectively.

to generate tunable infrared radiation in the 4–18-μm-wavelength range. The lack of tunable cw pump sources in this wavelength region (especially between 900 and 1400 nm) has limited the usefulness of $AgGaSe_2$ for parametric-frequency-conversion devices in the past. The progress in the development of reliable, tunable near-infrared solid-state lasers now offers several options for cw single-frequency pump sources for DFG in $AgGaSe_2$ as indicated in Fig. 1, e.g., Ti:sapphire lasers (Ti:s), color-center lasers, forsterite lasers ($Cr^{4+}:Mg_2SiO_4$),[17] Cr^{4+}:YAG lasers,[18] and single-frequency semiconductor laser diodes. Table 2 summarizes the main characteristics of these near-infrared pump sources. The last column indicates which portion of the optical transmission range of $AgGaSe_2$ each laser can generate in the DFG mixing process (either as a signal or a pump wave) if an appropriate second pump source is used.

The use of single-frequency laser diodes[19] appears particularly attractive because these devices are able to generate the full infrared-wavelength-tuning range for DFG in $AgGaSe_2$. These lasers have been steadily improving in output power, wavelength coverage, reliability, and cost. Recently there have been important advances in spectral-control and frequency-stabilization techniques of diode lasers based on external cavities, optical feedback, injection locking, and electronic servo control.[20] Using laser diodes as master oscillators, high cw-output powers (up to several watts) have been obtained by injection-locking high-power broad-area diode lasers or diode-laser arrays and by using these devices as traveling-wave or double-path reflective amplifiers.[21] Single-frequency diode lasers, each tunable over an ~15-nm range, have recently become available commercially in the 0.63–2.6-μm spectral region, with a bandwidth of <500 kHz and output powers up to 25 mW.[22] The utility of presently available single-frequency diode lasers for nonlinear frequency-conversion experiments has been demonstrated recently. Efficient frequency doubling of AlGaAs diode laser emission using a resonator for enhancement of the fundamental light field within the nonlinear material has been reported in $KNbO_3$.[23] In addition, efficient sum-frequency generation of blue light has been accomplished by mixing a Nd:YAG laser with a single-frequency AlGaAs diode laser in a monolithic $KNbO_3$ or KTP resonator.[24,25] SFG employing a single-frequency diode laser has also been obtained without using an enhancement cavity.[24,26] Recently, we demonstrated DFG in $AgGaS_2$ utilizing cw single-frequency diode–Ti:sapphire and diode–diode pump-laser configurations.[27]

Table 2. Pump Sources in DFG Mixing with $AgGaSe_2$[a]

Laser	Tuning Range (nm)	Power (W)	Coverage Signal/Pump (%)
Ti:sapphire	660–1100	3˙	13/40
$Cr^{4+}:Mg_2SiO_4$	1200–1350	1.8[b]	19/21
Cr^{4+}:YAG	1350–1560	1[b]	14/18
Color Center	1400–3500	0.1[b]	36/14
Diodes	630–2600	3[c]	100/100

[a]Some tunable solid-state lasers that are useful as pump sources in $AgGaSe_2$-based difference-frequency generation (DFG) mixing.
[b]Requires cryogenic operating conditions.
[c]When a diode-laser amplifier is used.

The available IR power is of special interest for spectroscopic applications. To estimate the cw single-pass infrared DFG power using $AgGaSe_2$, an analysis was performed based on Refs. 28 and 29, assuming a lossless crystal and focused Gaussian input beams. In this case the DFG power can be expressed as

$$P_i = \frac{4\omega_i^2 k_s d_{\text{eff}}^2 l}{\varepsilon_0 \pi c^3 n_p n_s n_i (1 + \mu)} P_s P_p h(\mu, \xi). \qquad (1)$$

Here the subscripts p, s, and i refer to the pump, signal, and idler beams, respectively; where ω_i is the IR angular frequency; k is the wave vector, d_{eff} is the effective nonlinear coefficient which equals d_{36} for 90° type I phase matching in $AgGaS_2$ and $AgGaSe_2$; l is the crystal length, ε_0 is the free-space permittivity; c is the speed of light in vaccuo; n_j ($j = p, s$ or i) are the wavelength-dependent refractive indices; μ is the ratio of the input wave vectors ($\mu = k_s/k_p$); ξ is the ratio of crystal length l to confocal parameter b; and $h(\mu, \xi)$ is the focusing function given in Ref. 29.

The infrared DFG power calculated for mixing two cw tunable lasers (power 0.5 W each) in $AgGaS_2$ and $AgGaSe_2$ utilizing 90° Type I phase matching and optimal focusing[29] ($l = 1.125b$) is depicted in Fig. 2. As can be seen from Eq. (1), the DFG power P_i is proportional to the input-power product P_sP_p of the pump and signal sources. This relation has been verified experimentally for even unbalanced signal-to-pump power ratios of up to 10:1.[6] For different input-power levels, the DFG output power can be predicted from Fig. 2 by properly scaling the input power product P_sP_p. It should be noted that the calculated data may be higher than the experimentally achievable IR power because of limitations in input-power density caused by surface damage, crystal absorption losses, imperfect spatial overlap of the pump and signal beams inside the nonlinear crystal, or deviations of pump and signal-beam modes from an ideal Gaussian TEM_{00} mode. In the case of $AgGaS_2$,[6] our measured IR power was only approximately half of the predicted power mainly because of imperfect spatial overlap of the beam waists of the pump and signal beams inside the crystal. Unfortunately, no information is available on the cw-damage threshold of $AgGaSe_2$. However, for $AgGaS_2$, up to 3 W of cw visible laser power (Ti:sapphire and DCM dye lasers at 740 and 650 μm, respectively) was focused down to a beam waist of 100 μm inside a 20-mm-long nonlinear crystal (10 kW/cm^2) cut for 90° type I phase matching. After an exposure time of more than one hour, no damage to the front or rear crystal facet was visible with a 1-μm-resolution light microscope. Additionally, at these power levels the transmitted visible beams did not show any wavefront distortion

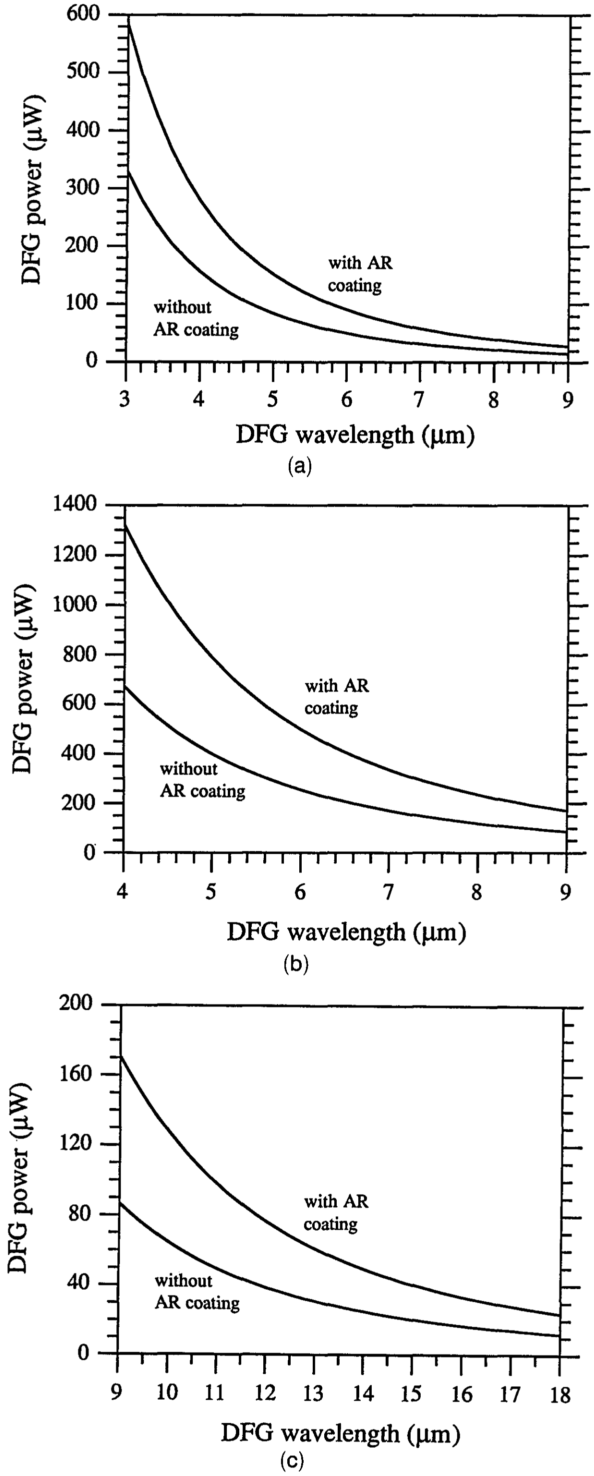

Fig. 2. Calculated DFG output power for an input power product P_pP_s of 0.25 W^2 as a function of the wavelength with (a) $AgGaS_2$ and (b, c) $AgGaSe_2$. In all cases, crystal length = 45 mm and confocal parameter = 40 mm. The case of focused Gaussian beams is considered in the calculation. Values of 12 and 33 pm/V are used for the nonlinear coefficient d_{36} of $AgGaS_2$ and $AgGaSe_2$, respectively.[10]

Table 3. Phase-Matchable DFG Wavelength Range[a]

Laser[b]	Laser Wavelength (μm)		DFG Range (μm)	Diode Range (μm)	Angle Range (Θ)
Nd:YAG	1.32	signal	10.3–18	1.17–1.23	84.7–57.7
		pump	7.32–18	1.61–1.42	82.6–54.9
Tm:YAG	2.01	signal	5.33–18	1.46–1.81	83.4–47.8
		pump	8.86–18	2.60–2.26	45.7–47.3
Tm, Ho:YAG	2.09	signal	5.07–18	1.48–1.87	83.7–48.1
		pump	10.6–18	1.60–2.35	43.9–47.4
Er:YAG	2.94	signal	3.41–18	1.58–2.53	85.7–48.3

[a]When type I angle tuning is used with tunable diode lasers (0.63–2.6 μm). The EriYAG laser at 2.94 μm is not phase matchable as a pump wave in $AgGaSe_2$.
[b]Compact, fixed-frequency, all-solid-state lasers.

behind the crystal, which indicates an absence of self-focusing effects. This lower limit for the cw-damage threshold of $AgGaS_2$ should be transferable to $AgGaSe_2$, as the damage thresholds for pulsed radiation are almost equal for both crystals. However, for high-input-power applications the surface damage threshold can be increased significantly by antireflection-coating the crystal.

Phase Matching with Angle Tuning

An alternative approach for generating tunable infrared radiation is to mix one tunable and one fixed-frequency pump laser operating at wavelengths longer than 1 μm in $AgGaSe_2$, and then to utilize angle tuning.[31] The advantage of this approach is that it allows the combination of relatively powerful fixed-frequency lasers with the radiation of weak tunable laser sources. Table 3 gives the phase-matching ranges of several compact, fixed-frequency, all-solid-state lasers that can be used in combination with tunable III-V single-frequency diode lasers for this purpose. As an example, Fig. 3 shows the phase-matching angle as a function of DFG wavelength when mixing tunable single-frequency diode lasers as the signal wave with a fixed-frequency Nd:YAG laser at 1.32 μm as the pump wave. In practice, the effect referred to as the Poynting vector walk-off[29] reduces the effective interaction length of the three waves in the crystal drastically. Owing to the birefringence of $AgGaSe_2$, the Poynting vector of the e-ray pump beam

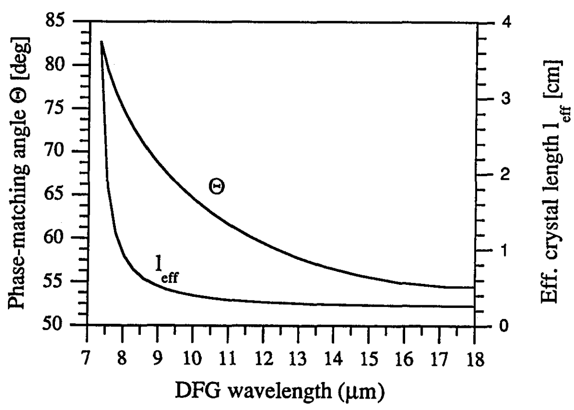

Fig. 3. Wavelength dependence of the phase-matching angle θ and effective crystal length l_{eff} for critical type I phase-matching in $AgGaSe_2$ with a Nd:YAG laser (1.32 μm) as the fixed pump input and diode-laser input as the tunable signal (1.6–1.4 μm).

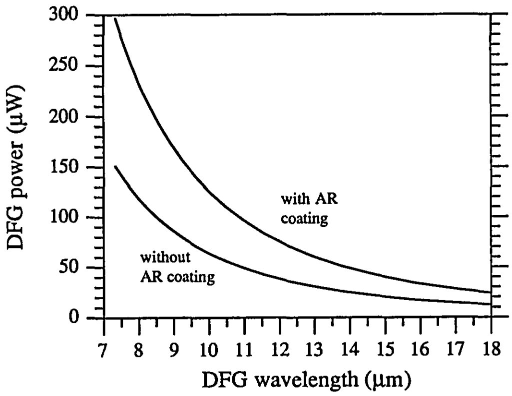

Fig. 4. Wavelength dependence of the calculated IR DFG power for critical type I phase matching in $AgGaSe_2$ with a Nd:YAG laser (1.32 μm, 500 mW) as the fixed pump and diode lasers (1.6–1.4 μm) followed by an amplifier[21] (500 mW) as the tunable signal. The conditions are an input power product $P_pP_s = 0.25$ W^2, a crystal length of 45 mm, optimal focusing conditions, and a nonlinear coefficient d_{36} of 33 pm/V.[10]

deviates from the wave vector by an angle ρ, unless the angle θ between the wave vector and the crystal c-axis equals 90°, in which case ρ = 0. The larger the deviation of θ from 90°, the larger is ρ. The length for which the birefringence gives rise to a substantial decrease in the overlap of o-ray and e-ray beams—and therefore in the nonlinear conversion efficiency—is designated by the effective length l_{eff}, which is a function of $1/\rho^2$. As can be seen from Fig. 3, the effective crystal length l_{eff} is rapidly reduced if θ deviates from 90°, and it becomes much smaller than the actual physical length l of the crystal. To estimate the infrared DFG power, Eq. (1) was modified for phase matching by angle tuning. Assuming optimal focusing and phase matching, the value of the focusing function h is given approximately by[29]

$$\mathrm{h_{mm}}(B) \approx \mathrm{h_{mm}}(0)/[1 + (4B^2/\pi)\mathrm{h_{mm}}(0)], \qquad (2)$$

with $B = 1/2\rho(lk_p)^{1/2}$; $\mathrm{h_{mm}}(0)$ the double refraction parameter and the maximum value of 1.068 for the h function at vanishing double refraction, respectively. The DFG power calculated for critical phase matching of a cw fixed-frequency Nd:YAG laser (500 mW) and an amplified diode laser (500 mW) in $AgGaSe_2$ is depicted in Fig. 4. As in the case of noncritical phase matching, for different input-power levels the DFG output power can be predicted from Fig. 4 by properly scaling the input-power product P_sP_p.

The limiting effect of l_{eff} on the DFG process, and hence the preference for 90° phase matching, is obvious. Moreover, noncritical phase matching provides other desirable features, such as a large phase-matching bandwidth and acceptance angle of pump lasers and achievement of a maximum effective nonlinear coefficient d_{eff}.

Conclusion

In summary, a widely tunable, compact, all-solid-state cw laser spectrometer in the 4–18-μm range that is suitable for high-resolution spectroscopy and kinetic spectroscopy and is based on $AgGaSe_2$ is feasible. Equally promising is the application of such lasers to methods that require a compact, portable laser-based spectrometer, such as sensitive and selective environmental monitoring. This development is possible now only because of the recent advances in new nonlinear optical crystals and solid-state, infrared, electro-optic technology. Compact, tunable, single-frequency high-power diode lasers and subsequently diode laser amplifiers appear to offer particular promise as pump sources.

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