

Noncritically Phase Matched Mid-Infrared Generation in AgGaSe₂

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Abstract—A tunable mid-infrared optical parametric oscillator has been demonstrated using a noncritically phase matched AgGaSe₂ crystal with pump wavelength tuning. Average power experiments have generated several hundred milliwatts of 2.63 and 3.71 μm radiation using a high-repetition-rate, multiwatt 1.54 μm pump source. Interferometric measurements of the AgGaSe₂ crystal during operation indicated only minor thermal lensing.

I. INTRODUCTION

ANGLE-TUNED silver gallium selenide (AgGaSe₂) optical parametric oscillators (OPO's) have been studied in the past [1]–[6]; however, noncritically phase matched (NCPM) operation with pump wavelength tuning has not been studied in detail. Calculations based on published refractive index formulas [7] predict 2–5 μm coverage with pump wavelength tuning in the 1.45–1.58 μm range. The fact that the pump wavelength only needs to be tuned over a very narrow range within a transparent region of the crystal is especially significant, since it can potentially circumvent the severe thermal lensing limitation observed with 2- μm pumping [8]–[10].

Based on this possibility, we have identified a method of generating tunable mid-infrared radiation using two cascaded OPO's. This method uses a NCPM AgGaSe₂ OPO pumped by a second OPO with tunable pump wavelengths that results in reduced thermal loading in the AgGaSe₂ crystal, relative to the 2- μm pumping case. Furthermore, NCPM interaction would allow efficient OPO operation using multiwatt, high-repetition-rate (kHz) pump sources with low-energy (<1 mJ) pulses. Our primary goal of this investigation was to demonstrate some of the key elements of this approach.

In this paper we report, for the first time to our knowledge, a NCPM AgGaSe₂ OPO with a tuning range from 1.9–5.5 μm and negligible thermal lensing. We present the tuning data and compare them with tuning curves calculated from refractive index equations. Observations of weak thermal loading effects and average power data are described.

II. NCPM WAVELENGTH TUNING

The wavelength tuning characteristics of a NCPM AgGaSe₂-OPO were investigated first, using a tunable pump

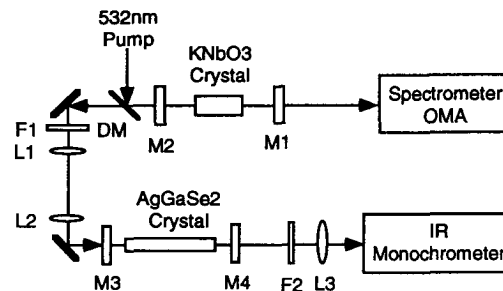


Fig. 1. Schematic diagram of the experimental arrangement used for the AgGaSe₂ NCPM wavelength tuning measurements. The tunable pump source for the AgGaSe₂ OPO was a Q-switched, Nd:YAG-pumped KNbO₃ OPO which was angle-tuned. M1: flat, 532 nm and 1.45–1.6 μm high reflector. M2: flat, 700–900 nm high reflector, 70% transmission at 532 nm, 70–80% transmission at 1.4–1.55 μm . DM: 45° incidence long-wavelength-pass dichroic mirror. F1: RG-1000 long wavelength pass glass filter. L1: 40 cm focal length lens. L2: 25 cm focal length lens. M3: 5 m concave 3.2–4 μm high reflector, 56% transmission at 1.52 μm . M4: 1 m concave 90% reflector 3.2–4 μm , 64% transmission at 1.52 μm . F2: germanium filter. L3: calcium fluoride lens.

source and a linear OPO resonator. Fig. 1 shows a schematic diagram of this experimental setup.

A potassium niobate OPO served as a tunable pump source, which generated wavelengths between 1.40–1.55 μm . The KNbO₃ OPO used a 3 × 7 × 13 mm crystal cut for type I phase matching in the bc-plane, with a face normal at 67.5° from the c-axis. The crystal faces had antireflection coatings for mid-infrared wavelengths, but not for the 532 nm pump or the signal wavelengths near 800 nm. The crystal was angle tuned inside a linear resonator comprised of two flat reflectors with a cavity length of 5 cm. One cavity mirror reflected greater than 99% from 1.45–1.6 μm and at 532 nm (M1). The other mirror (M2) reflected wavelengths between 700–900 nm and transmitted about 70% of the 532 nm pump and 70–80% of the 1.4–1.55 μm output. The pump beam was coupled into the cavity through this mirror after reflecting off a 532 nm dichroic (long-wave-pass) mirror (DM) at 45° incidence. The 1.4–1.55 μm idler output beam passed through the dichroic reflector and a long-wave-pass (RG-1000) glass filter (F1), that blocked a small amount of signal leakage through the second cavity mirror.

The OPO was pumped by a collimated 1 mm diameter beam, with pulse energies of 3–4 mJ, at a pulse repetition rate of 15 Hz. The FWHM pulse length was approximately 11 nsec. The laser was not injection seeded for this experiment. Under these operating conditions, the usable idler output energy was between 60–100 μJ per pulse over the 1.4–1.55 μm tuning range. No attempt was made to maximize

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the output energy or efficiency of the KNbO₃ OPO in this experiment.

III. AgGaSe₂ CRYSTAL

A AgGaSe₂ crystal for type I NCPM interaction ($\phi = 45^\circ; \theta = 90^\circ$) was selected at Cleveland Crystals for low absorption and scatter losses that are typical of currently available, high quality materials [11]. The crystal was cut as a rectangular bar, 33 mm in length, and the end faces were polished to yield a clear aperture of approximately 4×4 mm. A broadband antireflection coating was applied to both end faces to reduce reflection losses at wavelengths between 1.5–1.6 μm , as well as between 3–5 μm .

Prior to testing the AgGaSe₂ crystal as an OPO, we measured transmission through the crystal using the idler output beam from the KNbO₃ OPO. The beam was first demagnified and image relayed to the AgGaSe₂ crystal by a pair of lenses (see Fig. 1) with focal lengths of 40 cm (L1) and 25 cm (L2). We obtained 87% transmission at 1.523 μm .

The AgGaSe₂ crystal was extensively tested and characterized at Cleveland Crystals Inc. [12]. Those measurements indicated about 1.8% per cm absorption at 1.06 μm , and 13.5% per cm at 2.15 μm for the e-ray. The e- and o-ray absorption in the 1.45–1.6 μm region was estimated to be less than 1.5% per cm for this crystal (33 mm in length), or about 5% loss per pass. The antireflection coated witness sample indicated an average of 2.5% per surface loss at 1.5 μm and from 3–5 μm . Thus, our transmission measurement at 1.523 μm is consistent with the crystal absorption and coating reflection losses.

The AgGaSe₂ OPO tuning experiment used a linear resonator consisting of a 5 m concave high reflector (M3) and a 1 m concave 90% reflector (M4) for 3.2–4 μm . The two mirrors were separated by 4.5 cm. The idler beam from the KNbO₃ OPO pumped this OPO through the 5 m mirror, with a transmission of 56% at 1.52 μm . The 1 m mirror transmitted 64% at 1.52 μm . The transmission values varied by about $\pm 10\%$ as the input wavelength was tuned over the 1.4–1.55 μm range. The resonated radiation was coupled out through the 90% reflector, then passed through a germanium filter (F2), and focused by a calcium fluoride lens (L3) into a 0.25 m infrared monochromator equipped with a lead selenide detector at the exit slit.

OPO output was first observed with pump wavelengths between 1.464–1.54 μm , using pulse energies ranging from 95–60 μJ , respectively. The output signal/idler wavelengths tuned from 2.1/4.83 μm –2.65/3.679 μm over this pump tuning range. For pump wavelengths below 1.464 μm , the OPO oscillated without the 5 m concave input mirror. The output wavelengths tuned from 1.909/5.465 μm with 1.415- μm pump to 1.85/5.71 μm with 1.398- μm pump. At this shortest pump wavelength, which was limited by our KNbO₃ OPO tuning, the AgGaSe₂ crystal oscillated without any mirrors when the input pulse energy exceeded 50 μJ . This behavior was not entirely unexpected, since the crystal antireflection coatings did not extend to the generated wavelengths under these test conditions.

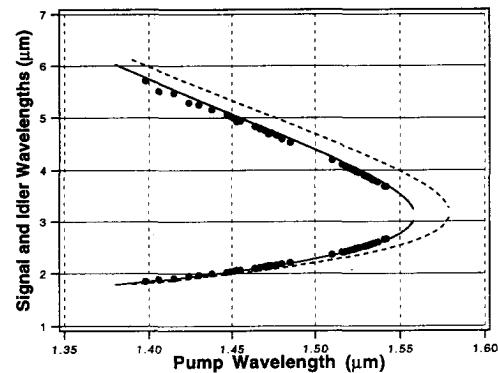


Fig. 2. OPO output wavelengths generated in a noncritically phased matched silver gallium selenide crystal, as a function of pump wavelength. Data points (\bullet) are compared with calculated tuning curves based on refractive indexes of [5] (dashed line) and [13] (solid line).

Fig. 2 shows the measured data points obtained from the tuning experiments, together with the calculated tuning curves based on two different sets of Sellmeier equations. The dashed curve represents calculated tuning based on Sellmeier equations by Kildal and Mikkelsen [7]. This was the basis of our initial prediction on NCPM tuning. A more recent set of index equations by Kato [13], however, where

$$n_o^2(\lambda) = 6.85070 + \frac{0.42970}{\lambda^2 - 0.15840} - 0.00125\lambda^2$$

and

$$n_e^2(\lambda) = 6.67920 + \frac{0.45980}{\lambda^2 - 0.21220} - 0.00126\lambda^2$$

with λ in micrometers, provides a much closer agreement with the measured values.

If the tuning curve is extrapolated further to shorter pump wavelengths down to 1.1 μm , calculations would predict a NCPM phase matching range from 1.2 μm –13 μm in a single crystal of AgGaSe₂. Although crystal absorption may limit OPO operation at these short wavelengths, pump tuned NCPM difference frequency generation in AgGaSe₂ provides a method of generating broadly tunable infrared radiation from a single crystal, in a manner similar to that demonstrated for AgGaS₂ [14].

Although the pump source for our experiment used a KNbO₃ OPO, other OPO crystals can also be considered as candidates. These include KTP, KTA (and its isomorphs), and lithium niobate, pumped by Nd:host and other lasers. Alternatively, direct laser pumping can generate a limited range of tunability. For example, a Nd:YAG laser at 1.32 μm has been used to generate NCPM OPO output at 1.6 and 6.7 μm [1].

One of the attractive features of the pump tuned NCPM AgGaSe₂ OPO is that the pump wavelengths avoid the anomalous e-ray absorption band near 2.1 μm [11]. Lack of strong absorption would reduce thermal lensing and the attendant limitation on average power scaling [8]–[10]. A critically phase matched AgGaSe₂ OPO, pumped by a 1.73 μm Er laser reported by Barnes and co-workers [3], was the first example of a pump laser whose wavelength lies far from the absorption band. Amimoto and co-workers also reported a

Raman-shifted Nd:YAG laser at $1.9 \mu\text{m}$, for pumping an angle-tuned AgGaSe₂ OPO [4]. In an effort to study average power scaling feasibility of the NCPM AgGaSe₂ OPO, we chose a demonstration experiment using a multiwatt pump source at a fixed wavelength of $1.54 \mu\text{m}$.

IV. AVERAGE POWER TEST

Average power tests were carried out on the AgGaSe₂ OPO at pump power levels of up to 3 watts. The $1.54\text{-}\mu\text{m}$ pump source was a KTP OPO, pumped by a 2.5 kHz, Q-switched, diode-pumped Nd:YLF laser, developed in our laboratory.

The Nd:YLF oscillator beam quality at $1.047 \mu\text{m}$ was quantified by measurement of the beam waist at the output coupler and in the far field. Due to differential thermal lensing along the *a*- and *c*-axes of Nd:YLF, the beam was slightly elliptical at the output coupler with a measured w_0 for the horizontal direction of $424 \mu\text{m}$, and a w_0 of $475 \mu\text{m}$ for the vertical direction. These values, combined with far field measurements, yielded a beam quality of $1.30 \times$ diffraction limited (DL) for the horizontal and $1.04 \times$ DL for the vertical direction.

The output of the oscillator was directed through a diode-pumped preamplifier and final amplifier before being mode-matched into the KTP OPO. At the nominal operating repetition rate of 2.5 kHz, the Nd:YLF master oscillator power amplifier (MOPA) produced 3-mJ pulses with 22-ns duration. Average power output exceeded 10 W at 6 kHz.

A schematic of the average power experimental arrangement is shown in Fig. 3. The output of the Nd:YLF MOPA was mode-matched to the first OPO which used two 2-cm length, x-cut KTP crystals in a linear resonator. The flat output coupler of the KTP OPO retroreflected the $1.047\text{-}\mu\text{m}$ beam to pump the crystals in both directions. An optical isolator placed at the output of the Nd:YLF oscillator rejected the retroreflected pump. A $\lambda/2$ waveplate, followed by a linear polarizer, served as a variable attenuator in the $1.047\text{-}\mu\text{m}$ beam path, so the $1.54\text{-}\mu\text{m}$ OPO output as a function of input pump energy and average power could be measured.

A second counter rotating plate attenuator, placed at the output of the KTP OPO, varied the pump power to a second OPO which used the 33-mm length AgGaSe₂ crystal. The diameter of the $1.54\text{-}\mu\text{m}$ beam at the AgGaSe₂ OPO was adjusted with mode matching lenses and monitored by a Hamamatsu IR camera. The resonated $3.71\text{-}\mu\text{m}$ signal output from the second OPO was separated from the $2.63\text{-}\mu\text{m}$ idler and $1.54\text{-}\mu\text{m}$ pump beams by the use of two short-wavelength-pass (SWP) 45° mirrors. A 1.0-m radius rear high reflector and flat output coupler completed the U-shaped OPO cavity. The $3.71\text{-}\mu\text{m}$ beam quality was quantified using an Electrophysics pyroelectric camera and beam diagnostic software.

The KTP OPO generated up to 3.0 W at $1.54 \mu\text{m}$ with 8.4 W of $1.047\text{-}\mu\text{m}$ pump power. Interferometric measurements of the KTP crystals during OPO operation indicated significant thermal lensing, caused by $3.27\text{-}\mu\text{m}$ idler absorption. It is important to note, however, that due to the end-pumped radial symmetry, the positive thermal lens did not significantly degrade the output beam quality from the OPO.

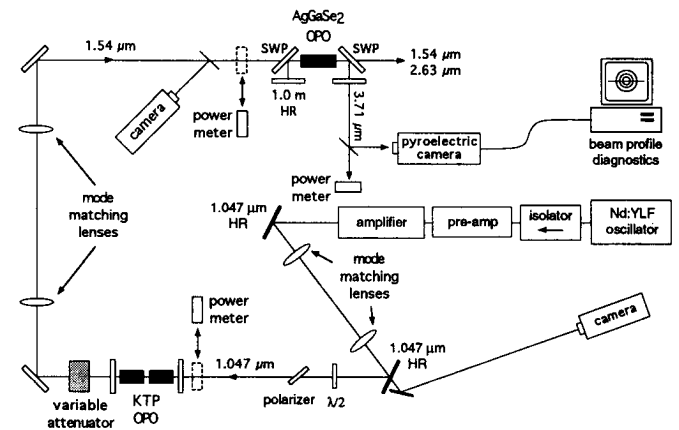


Fig. 3. Experimental arrangement for the average power test of the AgGaSe₂ OPO. $\lambda/2$: $1.047\text{-}\mu\text{m}$ half waveplate. SWP: 45° incidence short-wavelength-pass mirrors. The outputs from all three cameras were analyzed with the beam profile diagnostics.

To more accurately predict higher average power operation, we generated a quantitative model of the KTP OPO, which included diffraction, walkoff, thermal lensing, and multiple longitudinal modes. All of these phenomena were found to be necessary to explain the observed OPO performance.

Diffraction of all three waves was treated in three dimensions within the OPO cavity by propagating complex scalar fields according to the Huygens-Fresnel integral, using fast Fourier transforms. Since the Rayleigh range was much longer than the individual crystal length in the present case, diffraction was not important within the crystal itself. However, the beams were propagated between crystals and to each of the cavity mirrors where the proper phase curvature was applied. The model determined the stable mode profile which was established as a balance between diffraction, gain guiding, thermal lensing, and mirror curvature.

One effect of diffraction in the OPO is to transport signal and idler power from the center of the beam to the wings, which enhances the parametric conversion process. Another mechanism for transverse separation of the beams is walkoff, or double refraction. The dependence of optical index on angle causes the centroid of the pump and signal beams to diverge within the crystal, at an angle called the walkoff angle. In the model, walkoff is treated geometrically by translating the pump beam transversely, as it moves through the crystal. The result of walkoff is to limit the effective interaction length, as the waves become spatially separated. This effect is enhanced in the presence of thermal lensing, as the signal beam waist is a function of power loading.

Operation of KTP, at a signal wavelength of $1.54 \mu\text{m}$, placed the idler in a regime where the crystal was absorbing. At $3.27 \mu\text{m}$ the absorption in KTP is about 0.36 cm^{-1} , so that a significant fraction of the idler power was deposited as heat in the crystal. The resulting thermal lens caused the pump to be focused on each pass through the crystal. In addition, the presence of a lens in the OPO cavity reduced the signal beam radius. The model used the heat load, due to absorption of the idler, to compute the transverse temperature profile. The

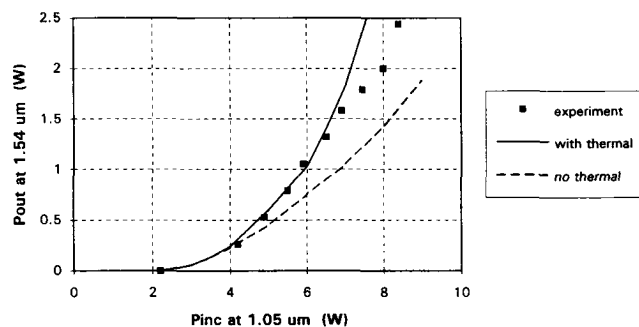


Fig. 4. Performance of the type II noncritically phase matched KTP OPO pumped by a 2.5-kHz repetition rate Nd:YLF MOPA at $1.047\ \mu\text{m}$ (■). The OPO used two 2-cm length, x-cut KTP crystals, which were pumped in both directions by the retroreflected $1.047\text{-}\mu\text{m}$ beam. KTP modeling results are shown, which include thermal effects (solid line) and without thermal effects (dashed line).

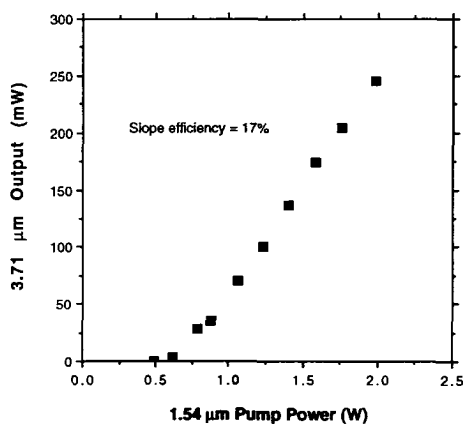


Fig. 5. Performance of the type I noncritically phase matched AgGaSe₂ OPO pumped by the $1.54\text{-}\mu\text{m}$ output of a KTP OPO operating at 2.5-kHz repetition rate. The $3.71\text{-}\mu\text{m}$ signal output is plotted as a function of $1.54\text{-}\mu\text{m}$ average pump power. Both the $3.71\text{-}\mu\text{m}$ signal and the $2.63\text{-}\mu\text{m}$ idler beams exhibited excellent beam quality.

temperature coefficient of the refractive index then determined the two-dimensional phase screen, which was applied to each beam.

The Nd:YLF pump beam had a bandwidth of about $1\ \text{cm}^{-1}$, which consisted of many longitudinal modes and results in significant pump intensity fluctuations. The effect on the KTP OPO was to lower the threshold and to increase the conversion efficiency. Bandwidth effects in the model were treated in the time domain by constructing a pump intensity waveform as the sum of many randomly phased modes. Each time slice was stepped through the gain medium and around the cavity. Synchronous enhancement was observed when the optical length of the OPO cavity was a rational fraction of the laser cavity length. The results of the model for KTP are shown in Fig. 4 as the solid (with thermal effects) and dashed (without heating) lines.

Fig. 5 shows the AgGaSe₂ OPO signal ($3.71\ \mu\text{m}$) output power as a function of input pump power. The OPO operated steadily at 0.25 W of signal output with a pump power of 2 W incident on the crystal in a $1/e^2$ pump spot radius of $393\ \mu\text{m}$ in the horizontal plane, and $327\ \mu\text{m}$ in the

vertical. The measured pump-to-signal conversion efficiency was 17% at room temperature, with no special means of cooling the AgGaSe₂ crystal. Threshold was attained at an average power of 0.56 W, which corresponds to an individual pulse energy of $224\ \mu\text{J}$ at 2.5 kHz, and a fluence of $0.055\ \text{J per cm}^2$. The maximum signal output power obtained was 0.3 W, with $\sim 2.5\ \text{W}$ of pump power (4.5 times threshold); the corresponding idler output power was 0.5 W at $2.63\ \mu\text{m}$. No output saturation was discernible at the highest pump powers used. Furthermore, the AgGaSe₂ crystal was able to withstand average pump intensities of $617\ \text{W per cm}^2$ without evidence of bulk or surface damage.

The AgGaSe₂ OPO signal temporal profile could not be measured directly due to limited time response of the PbSe detector. Based on the difference of the measured undepleted and depleted $1.54\text{-}\mu\text{m}$ pump profiles, however, we estimated the signal pulse to be $\sim 20\ \text{ns}$ FWHM.

An overall check of the internal losses in the AgGaSe₂ OPO (e.g., bulk crystal absorption of the signal and idler waves, surface scattering, mirror substrate absorption, antireflection and high reflection coatings) was carried out by comparing signal and idler output with pump depletion. We measured pump depletion by recording the temporal profile of the transmitted pump beam with and without blockage of the rear high reflector of the AgGaSe₂ OPO. Fig. 6 summarizes measurements of the pump “ \rightarrow ” signal + idler conversion efficiency by plotting the sum of the signal and idler output energies normalized to the input pump energy (open triangles) as a function of the input pump energy. The pump conversion efficiency, based on pump and signal + idler power measurements, rises to 35% at a pump energy of $330\ \mu\text{J}$. At the highest pump energies, the conversion efficiency asymptotically approached 40%. When pump depletion efficiency, defined by $[\int (\text{undepleted pump}) dt - \int (\text{depleted pump}) dt] / [\int (\text{undepleted pump}) dt]$, is plotted against input pump energy (solid triangles), good agreement is seen between the two data sets. This concurrence indicates the absence of gross loss mechanisms for the signal and idler beams in the AgGaSe₂ OPO because all the depleted pump energy is accounted for, to within the measurement uncertainties, by the sum of signal and idler energy.

The signal and idler beams had a near-Gaussian profile, and propagated with low divergence when the cavity was optimally aligned. When viewed at a distance of 1 m from the output coupler, the $3.71\text{-}\mu\text{m}$ beam diameter increased, at most 10%, throughout the $1.54\text{-}\mu\text{m}$ pump power range in which the signal beam was measurable. Taking into account the AgGaSe₂ resonator parameters, the generation of a 1–2 m focal length thermal lens at the highest pump powers would be consistent with our observations. The relatively stable far field beam diameter is in sharp contrast to the output of the KTP OPO, in which we found the $1.54\text{-}\mu\text{m}$ beam diameter to be strongly dependent on the $1.047\text{-}\mu\text{m}$ pump power.

Consistent with the far field measurements, interferometric measurements on the AgGaSe₂ crystal, using a $1.064\ \mu\text{m}$ cw Nd:YAG laser, confirmed the presence of a weak thermal lens with a focal length on the order of 1 m. Fig. 7 shows the experimental schematic for measuring optical path difference (OPD) in the OPO crystal using a Mach-Zehnder interferometer. This

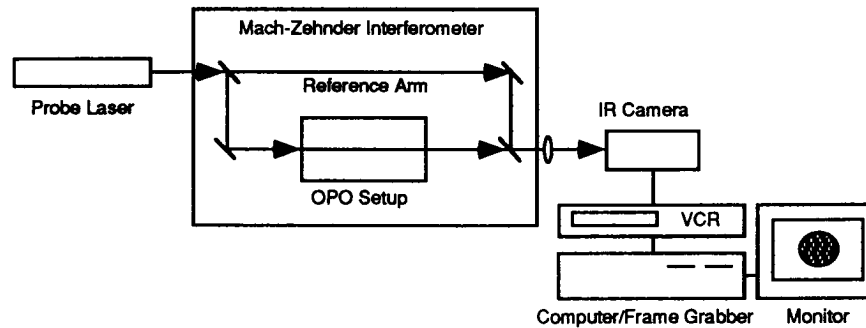


Fig. 7. Experimental arrangement for measuring the optical path difference in the AgGaSe_2 OPO using a Mach-Zehnder interferometer. The probe laser is a $1.064\text{-}\mu\text{m}$ cw Nd:YAG laser.

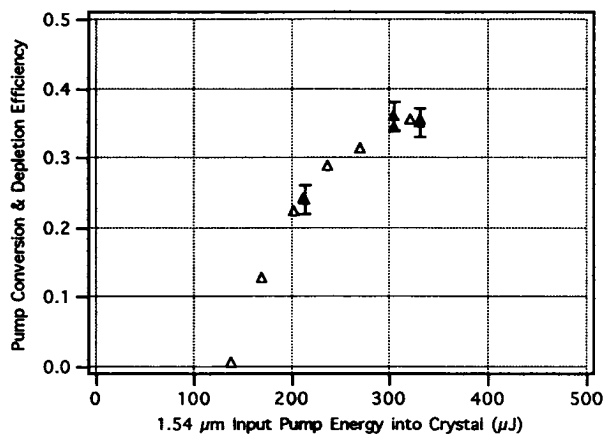


Fig. 6. AgGaSe_2 NCPM OPO pump conversion and pump depletion efficiency comparison. Open triangles (Δ): OPO pump conversion efficiency defined as $[\text{signal energy} + \text{idler energy}]/[\text{input pump energy}]$. Solid triangles (\blacktriangle): pump depletion efficiency defined as $[\int (\text{undepleted pump}) dt - \int (\text{depleted pump}) dt]/[\int (\text{undepleted pump}) dt]$. See text for discussion.

technique was previously used to investigate thermal loading effects in beta barium borate and lithium triborate crystals [15]. Fig. 8 displays AgGaSe_2 crystal interferograms at three pump power levels. The cold cavity reference interferogram, with no incident pump power, is shown in Fig. 8(a). At 1.2-W incident power, with the OPO operating steadily at ~ 100 mW of signal output, some fringe curvature becomes evident (Fig. 8(b)). Significant curvature can be seen in Fig. 8(c), with 2.25-W incident power and ~ 250 mW of signal output.

The thermal lensing was produced by contributions from both the pump and the generated radiation. We determined their relative contributions by comparison of the interferograms from the AgGaSe_2 crystal during standard OPO operation, and with the rear 1.0 m high reflector blocked to isolate the effect of the $1.54\text{-}\mu\text{m}$ pump absorption alone. Preliminary analysis indicates that the signal + idler contribution accounts for approximately 1/4 of the total lensing effect, and hence, absorption of the generated waves (in addition to pump absorption) may become a consideration at pump powers exceeding those reported here.

In conclusion, we have demonstrated a tunable, NCPM AgGaSe_2 OPO, using pump wavelength tuning. The average power tests and interferometric diagnostics showed that a



Fig. 8. Interferogram data on optical path difference measurements in the AgGaSe_2 OPO crystal at various pump power levels. Changes in the fringe pattern show weak thermal lensing with increasing pump power: (a) pump beam blocked; (b) 1.2-W incident pump power; (c) 2.25-W incident pump power.

AgGaSe_2 OPO can be operated without severe thermal lensing, when pumped by a multiwatt $1.54\text{-}\mu\text{m}$ source. Thus, a NCPM AgGaSe_2 OPO is an attractive device for generating tunable mid-infrared radiation with significant average power.

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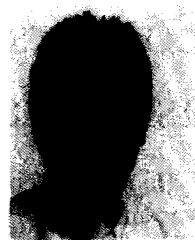
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