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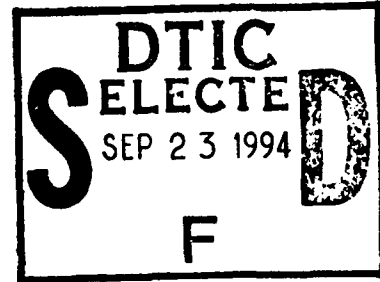
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**A AgGaSe₂ Optical Parametric Oscillator
Pumped by a Raman-Shifted YAG Laser**

2 September 1994

Prepared by

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PREFACE

We wish to thank R. C. Eckardt and R. L. Byer for useful discussions. We also wish to thank K. Schepler for the loan of a AgGaSe₂ crystal used in this work. The interest of B. K. Ching, J. C. Creswell, and M. A. Kwok are acknowledged. We wish to thank R. Welle for performing the phase-matching calculations.

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DISCUSSION

We report the operation of a silver gallium selenide (AgGaSe_2) optical parametric oscillator (OPO) pumped by a single-mode, pulsed, $1.06\text{-}\mu\text{m}$ Nd-YAG laser that is Raman-shifted using H_2 . The results reported herein are the first observations of tunable radiation produced by a $1.9\text{-}\mu\text{m}$ pump wavelength in AgGaSe_2 using short 8-ns pump pulses. Whenever pump wavelengths below $1.5\text{ }\mu\text{m}$ are used, phase matching does not allow continuous tuning over the 3- to $12\text{-}\mu\text{m}$ region. However, in this paper we show that it is possible to use a short-wavelength laser source (near $1\text{ }\mu\text{m}$) to pump AgGaSe_2 , and produce tunable wavelengths throughout the mid-infrared if one uses Raman wavelength conversion. Scaling to high average power may be possible because mature, high average power, short-wavelength lasers are commercially available, and because AgGaSe_2 has a high average power damage level. The earliest reported OPO study of AgGaSe_2 was performed by direct pumping with a Nd-YAG laser at $1.34\text{ }\mu\text{m}$ and a Holmium laser at $2.1\text{ }\mu\text{m}$.¹ More recent studies of parametric amplification² and OPO performance³ in AgGaSe_2 used an Er:YLF pump laser at $1.73\text{ }\mu\text{m}$. We measured fluence thresholds for parametric oscillation that are approximately 30% higher than the calculated theoretical fluence thresholds. They are also the lowest threshold values reported to date for AgGaSe_2 . Pump fluences up to 3 times threshold fluence levels were employed in this study. Output energies of up to 1.2 mJ/pulse were measured near $4.5\text{ }\mu\text{m}$. This corresponds to a 3.8% energy conversion efficiency when maximum pump energies of 30 mJ/pulse at $1.9\text{ }\mu\text{m}$ were used. No optical damage was observed under these conditions. The corresponding pump power and $4.5\text{-}\mu\text{m}$ output power were 2.6 MW and 500 kW respectively. Similar to the case of 2.1- and $1.73\text{-}\mu\text{m}$ pump wavelength, phase matching is possible to produce output over the 2.3- to $12\text{-}\mu\text{m}$ wavelength region using the $1.9\text{-}\mu\text{m}$ pump.

In general, good pulsed OPO performance is obtained when the threshold is low and the pump fluence is adjusted to achieve maximum conversion efficiency. This empirical approach is necessary for pulsed OPOs since theoretical solutions for pulsed OPOs are unavailable. In contrast for cw or long pulsewidth pump sources, this optimal efficiency is reached when the pump intensity is approximately 3 to 7 times the threshold value depending on the pump beam intensity profile.⁴ Often, for pulsed OPOs, the optimal fluence cannot be reached due to impurities or defects in the nonlinear material or coatings that lower the damage level of the material. Thus, a high damage threshold value is desirable in order to reach the fluence necessary to achieve maximum conversion efficiency. Several methods can be used to lower OPO thresholds while maintaining high efficiency. These include the use of a faster pump pulse with a short laser cavity length to reduce build-up time, the use of doubly resonant instead of singly resonant OPO resonator optics, and the use of pump feedback techniques. The short pump pulse and short cavity operation were explored in this study. Of course, this assumes that the damage threshold, which may be a function of the laser pulse duration, will not be appreciably lowered as well.

The pump laser for the Raman OPO pump source was a single-mode, single-frequency $1.064\text{-}\mu\text{m}$ Nd-YAG laser firing at a 10-Hz repetition frequency. The laser bandwidth is approximately 50 MHz . The laser was characterized by a near-Gaussian spatial and temporal pulse and linear polarization. Measurements of the Strehl ratio assuming a perfect-Gaussian profile yielded

results of 0.8 to 0.95. Energies per pulse of 180 to 220 mJ/pulse were typical with pulse durations of 13 ns FWHM. Single longitudinal mode operation was achieved by injection locking the laser oscillator using a seed beam from a diode laser-pumped, cw-YAG laser (Lightwave Technology).

The overall experimental scheme is shown in Fig. 1. The 1.9- μm pump beam that pumped the OPO was generated using a Raman seed generator and amplifier. The output from the YAG laser was passed through a 20% beam splitter. The weaker beam of approximately 33 mJ from the beam splitter was focused 20 to 30 cm beyond the entry window of the seed generator cell with a 1-m focal length lens. This cell was 1-m long and was filled with 230 psi of H_2 . Approximately one-half of the Raman radiation generated at 1.906 μm was produced in the forward direction and the other half was in the backward direction. The backward-scattered Stokes output was deflected to one side by a mirror coated for high reflectivity at 1.9 μm at 45 degrees incident angle. A mode-matching lens and a flat mirror redirected the backward scattered light in the forward direction through the focal region of the 1-m fl lens. This scheme produced 9 mJ in the forward direction and had the advantage of being insensitive to minor misalignments of the cell position. The Stokes output was recollimated using a single 1-m lens. Because of mirror losses and the use of uncoated optics, only 5 mJ was available to enter the amplifier. The pulse duration of the Stokes pulse was 8 ns FWHM or 13 ns at the $1/e^2$ point for a Gaussian time profile.

The remaining 80% of the 1.06- μm pump energy was sent through a delay line to equalize the paths of the Stokes seed beam and the pump beam in order to temporally overlap the beams in the Raman amplifier. Both beams were combined by a dichroic mirror. The pump beam was reduced approximately 2-to-1 by using an inverted telescope just prior to the dichroic mirror. Both beams were collimated to have approximately the same beam e^{-2} diameter of 0.2-cm within the amplifier. The Raman amplifier is a 2-m-long cell filled with 90 psi of H_2 . With 102 mJ of 1.06- μm radiation incident on the amplifier, about 40 mJ at 1.9 μm was produced. Pulse-to-pulse variation was $\pm 10\%$. The quantum conversion efficiency of the amplifier is 61%, uncorrected for Fresnel losses at the cell windows. The spectral linewidth of the Raman beam was not measured, but it is estimated to be less than 400 MHz. The injection-seeded linewidth of the YAG laser was less than 20 MHz.

The final 1.9- μm amplifier output beam was collimated using an inverted telescope. Depending on the telescope magnification, Gaussian pump beam profiles with e^{-2} diameters of 0.42, 0.54 and 0.64 cm were measured with a linear pyroelectric array detector. Beam profiles were measured over a 50-cm-long path to verify that the beam remained collimated. This is comparable to or greater than the effective parametric gain length, $\mathcal{L}$, which is defined below for the configurations used in this study. The 1.9- μm energy incident on the OPO crystal could be varied by adjusting the incident angle of a mirror coated for use at 45 degrees that would partially reflect the beam. In this way the threshold values for the OPO could be measured.

The AgGaSe_2 crystal was cut at 55 degrees for type I phase matching. The crystal dimensions were $10 \times 10 \times 20$ mm. A multilayer-dielectric, anti-reflection coating for the 3.0- to 3.6- μm region with $1\% < R < 2\%$ was applied to both end surfaces. Transmission at 1.9 μm was 85%. The OPO resonator optics consisted of two mirrors for a singly resonant cavity. A dichroic mirror of 10-m radius of curvature was used as the input mirror for the 1.9- μm pump beam. The transmission of this mirror at 1.9 μm was greater than 90%. The reflectivity for the resonant idler

beam in the 2.5- to 3.3- μm region was greater than 99% for this input mirror. The output mirror was flat with a dichroic coating. Its reflectivity for the idler wavelength region from 2.8 to 3.8 μm varied between 78 to 88% and for the signal wavelength region from 4.25 to 6.00 μm its transmission was approximately 85%. At 1.9 μm its reflectivity was 47%. The output-coupler substrate was ZnSe, and a 2-degree wedge angle between the front and the back surfaces was used to eliminate interference fringes caused by the uncoated back surface. Measurements of OPO output energies were corrected for the Fresnel loss of the back surface. Measurements of OPO spectral and temporal outputs were facilitated by a 4- to 5- μm bandpass filter and a calibrated, circularly variable filter. A fast Au-Ge infrared detector with 2-ns risetime was used to measure the temporal profiles of the Nd laser, Raman output, and the OPO output near 4 to 5 μm .

Preliminary damage measurements at 1.9 μm were performed on a $10\times 10\times 2\text{-mm}$ "companion" sample of a AgGaSe₂ crystal antireflection coated with a single layer of ThF₄. This coating was found to damage at fluences of 0.036 J/cm², provided the coating was previously exposed to laser pulses at fluences 40% below the damage level. The crystal and companion samples were both subsequently recoated using a multilayer coating design. The spectral characteristics of this new coating are described above. This recoated crystal was subjected to fluences of up to 0.18 J/cm² in an OPO cavity with no signs of damage. Given our 8-ns pulsewidth, the damage intensity measured was in excess of 22 MW/cm². This value is in agreement with the 13- to 40-MW/cm² damage levels observed earlier.¹

An angle-tuning curve was calculated using Sellmeier equations⁵ for the index of refraction. The tuning curve was experimentally verified only over the 3.8- to 4.7- μm region. Both the calculated and the theoretical curves are shown in Fig. 2. No attempt was made to operate the OPO beyond this range. It should be noted that a AgGaSe₂ OPO can be phase matched when pumped at 1.9 μm to produce output over the 2.3- to 12- μm region. To obtain agreement with the experimental measurements, it was necessary to shift the calculated curve by about 1.15 degrees to smaller internal phase-matching angles. This correction is similar to the 0.7-degree difference between calculated and measured phase-matching angles using a 2.05- μm pump wavelength in an earlier study.¹ In that study, the angular differences were attributed to small variations in the index of refraction and crystal orientation.

The FWHM pulse duration of the Nd laser of 13 ns produces a Raman output with a pulse duration of 10-ns FWHM. The OPO output is 2.5 ns for a 13-cm optical cavity length. The shortening of the OPO output pulse duration was caused in part by cavity build-up time. Fluence thresholds were measured for various pump beam diameters as shown in Fig 3. Three diameters of 0.64, 0.54 and 0.42 cm ($1/e^2$ Gaussian) were used. All three diameters gave fluence thresholds of 0.060 J/cm² for a cavity length of 17 cm. That the threshold was independent of beam diameter suggested that the OPO mode volume was smaller than that inferred from the beam diameters used. This conclusion was also verified by calculations of the beam diameters of the signal and idler beams.⁶

The energy efficiency of the OPO can be directly determined from Fig. 3. At the highest possible pump fluence of 0.18 J/cm² or 3 times the threshold fluence of 0.060 J/cm², the OPO output at 4.5 μm is 1.2 mJ for a 31-mJ pump input. The energy efficiency was 3.8% corresponding to a quantum efficiency of nearly 10%, where we define quantum efficiency as the photon conversion efficiency from the pump into either the signal or the idler photons. For

convenience, quantum efficiencies of the data of Fig. 3 are replotted in Fig. 4 to facilitate comparison with earlier work.¹ Theory predictions for pulsed quantum conversion efficiencies are not available short of numerically solving the equations in Ref. 6. A crude estimate of the quantum conversion efficiency can be made by modifying the results in Ref. 4, which apply to the case of cw pumping for a Gaussian pump beam profile in a singly resonant oscillator. The dimensionless pump power, P/P_t , in Ref. 4 was corrected for the duty cycle for the case of pulsed pumping. The relative pump powers were set equal to the average pump fluences, J/J_o , where J_o is the threshold fluence and subsequently multiplied by the ratio of the OPO output pulse duration to the pump pulse duration. For this work, the ratio of OPO to pump pulse duration is set at a fixed value of 0.20 for a cavity length of 17 cm. In Ref. 1, the pump pulse duration was 50 ns, and OPO output pulse duration varied between 10 and 30 ns as the pump fluences were varied. Both the experimental data and modified theory predictions for our data are given in units of threshold fluence as the appropriate scaling parameter. The quantum efficiency measured in this work is a very slight improvement compared to earlier work and compares well with the crude theory. The assumption of a fixed ratio of OPO to pump duration appears to allow satisfactory agreement with theory and experiment. Unfortunately, efficiency measurements were not carried out under conditions of shorter cavity length, which would serve to reduce the threshold fluence as shown below.

Thresholds for OPO operation were also measured as a function of cavity length. The optical lengths were varied from 9 to 15 cm. In Fig. 5, the observed thresholds increase monotonically with increasing cavity length. These threshold values for the AgGaSe₂ OPO are the lowest reported to date. The fluence thresholds can be calculated using the following expression developed for pulsed, singly resonant OPOs,⁶

$$J_o = \frac{2.25}{\kappa g_s L^2} \tau \left[\frac{L}{2\tau c} \ln \frac{P_n}{P_o} + 2\alpha l + \ln \frac{1}{\sqrt{R}} + \ln 2 \right]^2$$

where

$$\kappa = \frac{2\omega_s \omega_i d_{\text{eff}}^2}{n_s n_i n_p \epsilon_o c^3}$$

The n 's are the indices of refraction at the signal, idler, and pump wavelengths as identified by the subscripts. The permittivity constant is ϵ_o , and c is the velocity of light. The ratio P_n/P_o is the ratio of threshold to noise power and is set to 33 as in Ref. 6. The effective nonlinear coefficient is $d_{\text{eff}} = 32 \times 10^{-12}$ (m/v) $\sin(\Theta_m)$ for type I phase matching as recommended by Eckardt.⁷ The mode coupling coefficient is

$$g_s = \frac{w_p^2}{w_p^2 + w_s^2}$$

where w_p and w_s are the beam widths of the pump and signal respectively, and the effective parametric gain length is

$$L = l_w \operatorname{erf} \left(\frac{\sqrt{\pi} l}{2 l_w} \right)$$

where l_w is the walk-off length and l is the crystal length. The e^{-2} Gaussian half-width of the pulse duration is τ and the optical cavity length is L . The signal wavelength is $4.5 \mu\text{m}$ and the idler wavelength is $3.3 \mu\text{m}$. The pump pulsewidth, τ , was set to 4.5 ns , and reflectivity $R = 0.8$. The pump beam waist $w_p = 0.64 \text{ cm}$, $l = 2 \text{ cm}$, $g_s = 0.8$, and $\alpha = 0.02 \text{ cm}^{-1}$. The other parameters are defined in Ref. 6. Over the entire range of cavity lengths explored, the experimental thresholds exceed the calculated thresholds by 30%. However, the agreement is good when one considers the accuracy of the estimates for the input parameters. By comparison, Eckardt¹ observed a threshold of about 0.14 J/cm^2 , which is approximately 2.3 times the theoretical threshold for the pulse durations employed in their study. Some possible sources for the larger than expected experimental thresholds include variations in the beam quality, mode, and temporal pulse shapes, and differences in d_{eff} from crystal to crystal. Improvements in decreasing crystal losses and increasing the values of d_{eff} may be expected from refinements in crystal growing techniques following the work of Eckardt.

In summary, we have extended earlier AgGaSe₂ OPO operation to a new pump wavelength of $1.90 \mu\text{m}$ with significantly shorter pulse durations. The new pump beam is generated in a Raman oscillator-amplifier configuration in H₂. This will allow a short wavelength source to produce the longer wavelengths necessary to pump AgGaSe₂ in order to produce tunable, phase-matched output in the mid-infrared. The OPO tuning curve was verified over the $3.8\text{-}4.7\text{-}\mu\text{m}$ region. Energies up to 1.2 mJ at $4.5 \mu\text{m}$ and quantum conversion efficiencies up to 10% were observed. Threshold measurements are in good agreement with theory for pulsed pumping of an OPO. Significant improvements in OPO performance may be anticipated when the lower thresholds associated with shorter cavity lengths are utilized. Under these condition, it should be possible to obtain near optimized performance for this AgGaSe₂ OPO pumped by a short 5-ns, $1.9\text{-}\mu\text{m}$ Raman pulse.

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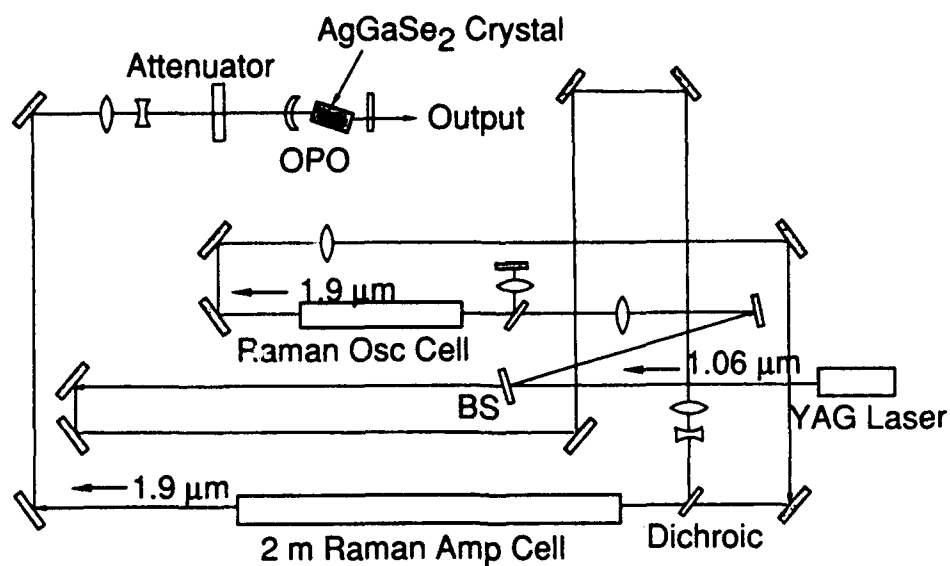


Figure 1. Experimental layout for the H₂ Raman oscillator-amplifier and the AgGaSe₂ optical parametric oscillator.

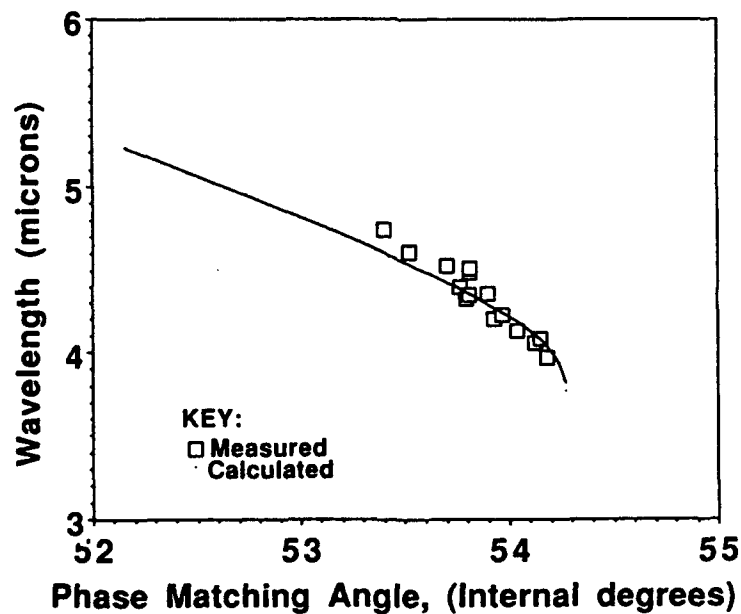


Figure 2. Experimental and calculated angle tuning curves for the AgGaSe₂ OPO pumped at 1.906 μm .

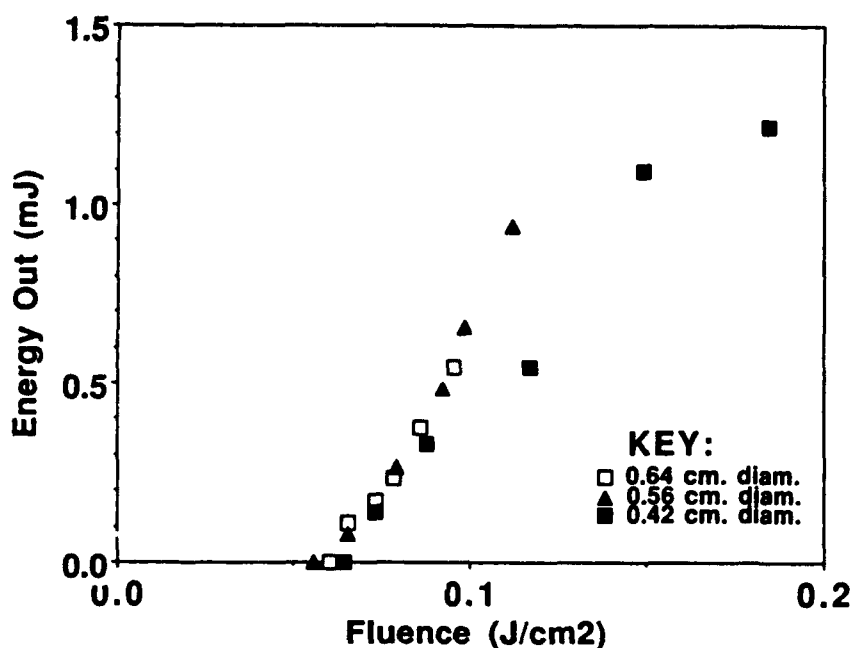


Figure 3. Energy output from the AgGaSe₂ OPO for pump beam sizes of 0.64, 0.54, and 0.42-cm, cm^{-2} diameters as a function of pump fluences.

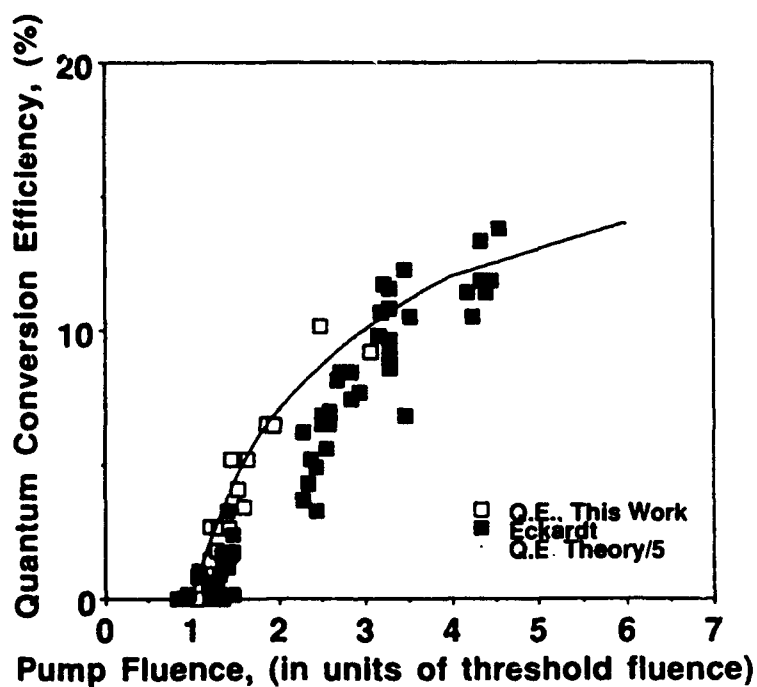


Figure 4. Photon, quantum conversion efficiencies as a function of the relative pumping parameter or fluence for the work of Eckardt (Ref. 1), the theoretical performance, and this work. The theory prediction of quantum efficiency from Ref. 4 has been multiplied by a factor of 0.2. This factor is a first order correction for the ratio of the averaged OPO output pulse to pump pulse duration.

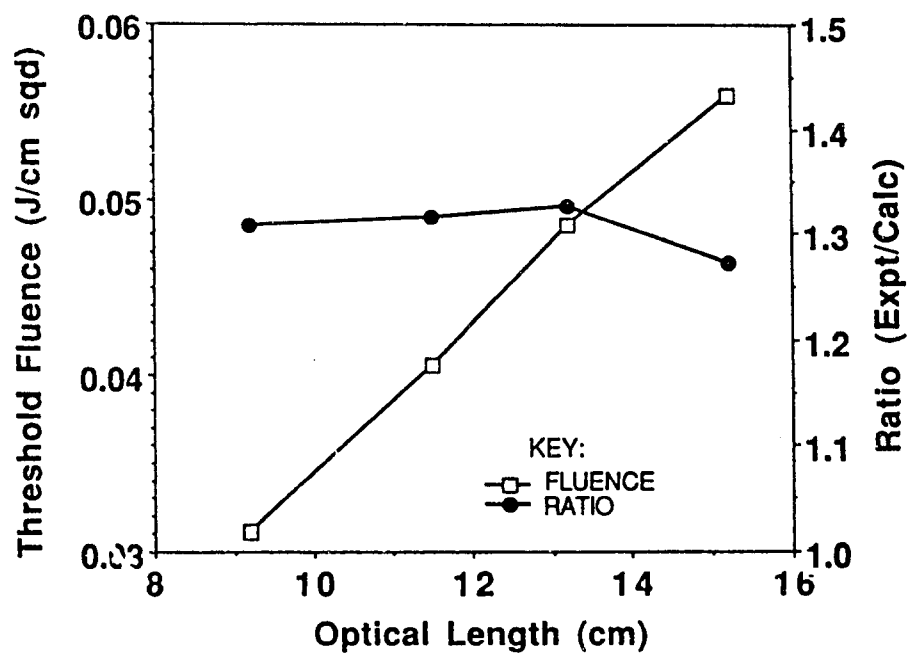


Figure 5. Experimentally measured OPO threshold fluences and theory predictions as a function of cavity length.