Reliable Operation of a Proustite Parametric Oscillator

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An infrared parametric oscillator using proustite and pumped at 1.065 μm has been operated with peak output powers in the kilowatt range. No significant deterioration of output power was observed after several hours of running at 2 pps. Tuning from 1.82 to 2.56 μm was obtained, limited only by the reflectivity limits of the mirrors used. With careful design, DRO proustite oscillators covering the entire range 1.2–9.5 μm now appear feasible.

Proustite (Ag_3AsS_3)[1,2] is an attractive material for parametric oscillation in the infrared since it is available as large good-quality crystals and when pumped at 1.06 μm offers a tuning range of at least 1.2–9.5 μm. The first demonstration[3] of an optical parametric oscillator (OPO) based on proustite was somewhat disappointing however, since only sporadic operation was obtained, the oscillation stopping completely after a few minutes due to crystal damage. Following a careful study[4] of laser-induced damage to proustite, we have designed an oscillator which has been run satisfactorily for many hours. The essential changes we have made from the earlier oscillator are: (i) a much-reduced pulse rate, 2 pps as opposed to 2000 pps, and (ii) single-frequency operation of the pump laser (see Ref. 5 for details of the laser used with the first oscillator). Peak output powers in excess of 1 kW have been obtained in pulses of ~25-nsec duration from our proustite OPO, as well as significant tuning around the degenerate wavelength.

Our measurements[4] have shown that surface damage of proustite occurs after 10 pulses (~20-nsec duration, 2-pps rate) at a beam-center radiation intensity ($I_{\max} = P_{pk}/\frac{1}{2}\pi W^2$, where W is the field radius of the TEM_{00} beam) equal to 25 MW/cm². In contrast, 1000 pulses at 10-MW/cm² intensity produce no damage. These values are for uncoated crystals; antireflection coatings on the crystal used in our OPO raised, very slightly, the intensity for damage.

When working with a material as susceptible to damage as proustite, the threshold intensity should be minimized rather than the power. For these conditions it can be shown[6] that the focusing parameter $\xi = l/b$ should be less than $\xi_c = \pi/4B^2$ (l is the crystal length, b is the "pump" confocal parameter within the crystal and B is the double-refraction parameter as defined in Ref. 7). At $\xi = \xi_c$, both the power and intensity thresholds are just 60% above their minimum values. $\xi < \xi_c$ results in a reduced intensity threshold. $\xi = 0.0025$ is used in our oscillator ($\xi_c = 0.006$ for a 10-mm crystal pumped at 1.065 μm). The resulting *external* confocal parameter is very large, 1.4 m, and is difficult to achieve while still maintaining the short OPO resonator needed for rapid buildup of oscillation with a brief pump pulse. Thus for our plano-concave resonator (see Fig. 1) a 30-m mirror is needed to match the beam confocal parameters ($b_p = b_s = b_i$). Estimates based on Twyman-Green interferograms of the as-grown crystal showed that the optical inhomogeneity [two fringes at 633 nm in (10 mm)³] could result in lensing equivalent to $f = \pm 5$ m. The resonator would be unstable with the negative "lens". For this reason a 2-m-curvature concave mirror was chosen giving a confocal parameter of 0.35 m. b_s and b_i are obviously no longer equal to b_p, but consideration of Asby's[8] computations for the near-field focusing limit suggest that this results in a *decrease* rather than an increase in threshold.

The proustite crystal was cut for type-I phase matching in a quadrant for which the contributions from d_{15} and d_{22} are additive.[1] An initial check of the calculated phase-matching angle was made through second-harmonic generation of 2.098-μm radiation from an Ho:YAG laser. Broadband antireflection coatings[9] deposited on the crystal give reflectivities of less than 0.5% per surface at 1.065 μm and not more than 2% over the whole of the tuning range achieved around the degenerate wavelength of 2.13 μm. A measurement using a 1.15-μm He-Ne laser of the single-pass loss of the coated crystal yielded a value of ~20%; the origin of the excess loss is unknown. The threshold power and intensity, calculated for $b_s = b_i = b_p$ and including an increase of four times due to the short duration of the pump pulse, are 10 kW and 2.5 MW/cm², respectively.

The Nd:$CaWO_4$ laser is flash-lamp pumped and water cooled to give ~250-kW 26-nsec (FWHM) pulses in the TEM_{00} mode, at a single frequency. Operation on just one longitudinal mode is ensured through the use of a two-plate resonant reflector combined with a special electro-optic Q switch.[10] To check for a single frequency the output radiation was converted to its harmonic in an ADP crystal and examined with a defocused confocal Fabry-Perot interferometer.[11]

The OPO resonator was designed to be very rigid and incorporates the necessary angular and length adjustments. In particular, the length of the resonator can be set to an accuracy of ~2 μm by a micrometer. Correct adjustment of the resonator length was found to be a critical factor in attaining reliable operation of the doubly resonant oscillator (DRO). The OPO mirrors are standard $\frac{1}{4}\lambda/4$ stocks having transmissions of 90% at 1.065 μm and reflectivities of either 98 or 95% at 2.13 μm.

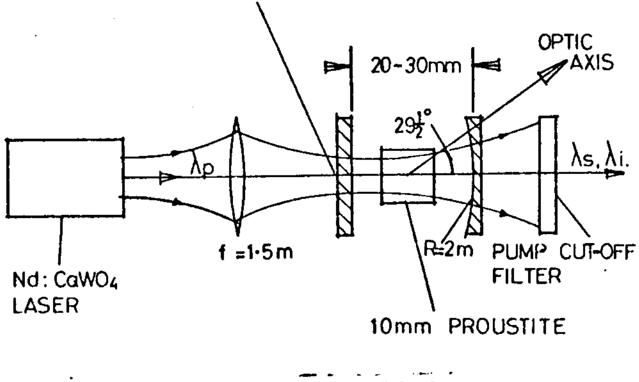

FIG. 1. Schematic diagram of OPO resonator.

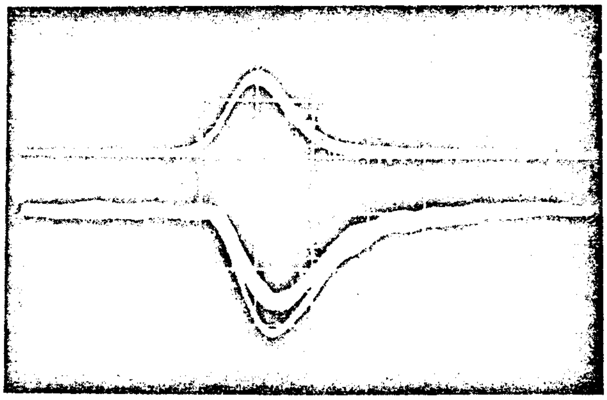

FIG. 2. Oscillator reliability. Upper trace—input pump, ~ 80-kW peak; lower trace (inverted)—OPO output, ~ 100-W peak. Time scale 20 nsec/div.

The parametric-oscillator output was filtered from the transmitted pump by a 1.06-μm mirror followed by a 2-mm slice of germanium (direct irradiation of the germanium by the laser was avoided to prevent the production of large carrier concentrations with consequent free-carrier absorption). An indium antimonide photoconductive detector with a response time of ~ 20 nsec was used to detect oscillation. The pump radiation was monitored by a F4000 vacuum planar photodiode; the response time was then limited by the oscilloscope to ~ 10 nsec.

Our first experiments were with an OPO resonator using two 98% reflectivity mirrors and 35 kW of incident pump power; this gave a safe level of 8.5-MW/cm^2 intensity at the crystal. Parametric oscillation was obtained immediately following alignment of the system using a 633-nm He-Ne laser. The peak output power was then of the order of tens of watts. The device was run in this condition for a total of about 5 h during which time optimization of the various parameters was carried out finally resulting in peak output powers of greater than 100 W. No damage occurred to the crystal during this period despite the fact that the proustite crystal had received about 36 000 pulses, in one place, at an intensity of 8.5 MW/cm^2.

For further experiments the pump power was raised to 80 kW (19 MW/cm^2 at the crystal). When set for phase matching a little off-degenerate, very reliable oscillation was obtained (see Fig. 2). Parametric oscillation occurred for every pump pulse while the laser was running on a single frequency. Operation was well above threshold, as evidenced by a delay of less than 10 nsec between the start of the pump pulse and the onset of oscillation. After 1–2 h of operation under these conditions, very slight damage was found on the exit face of the crystals; this damage appeared to have little effect, however, on the oscillation. The tuning range of the OPO was measured with a quartz-prism monochromator and the value obtained, 1.82–2.56 μm, agrees with the reflectivity limits of the 2.13-μm mirrors.

In the final experiments the output mirror was changed to one with 95% reflectivity. Peak output powers were then greater than 1 kW, corresponding to a conversion efficiency in excess of 1%. An accurate tuning curve was then determined and this is shown in Fig. 3, together with one calculated from Hobden's Sellmeier equations for proustite.[12] The degenerate phase-matching angle agreed to within experimental error with the predicted value of 29.5°. At the extremes of the tuning range the frequency reproducibility and bandwidth were better than the monochromator resolution (~ 10 cm^{-1}).

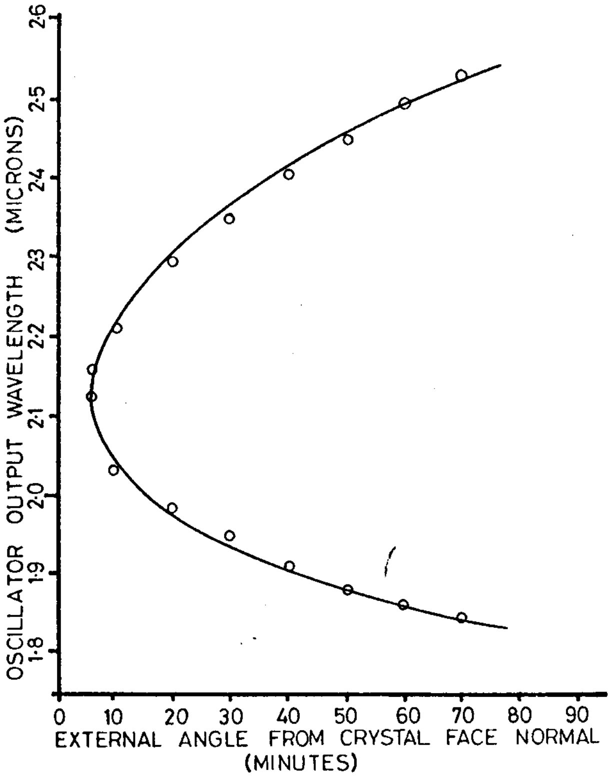

FIG. 3. Tuning curve. The solid line is calculated from refractive-index data of Ref. 11.

Our purpose here has been to show that parametric oscillators based on proustite are practical devices; at least when operated with $I_{max} < 10$ MW/cm^2 and at 2 pps. Under these conditions the devices should have long life. It may be possible to raise the pulse rate above 2 pps, but further studies of damage, etc., are needed before this can be known. We also believe that proustite oscillators tuning over substantial portions of the infrared are now feasible.

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