Continuous Operation of a Solid-State Optical Maser

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Experiments demonstrating continuous operation of a solid-state optical maser are described. A continuous coherent wave at 1.065 μ is obtained from trivalent neodymium in $CaWO_4$, operating at about 85°K. At room temperature a continuous wave at 1.058 μ is obtained. Room temperature operation is possible due to the large splitting between the terminal state of the maser transition and the ground state ($\sim$2000 cm^{-1}). Near threshold power output $\sim$10 mw is observed, using silver coatings only.

INTRODUCTION

THE optical maser characteristics of Nd^{3+} in $CaWO_4$ have been described in a recent publication.[1] Subsequently, stimulated emission has been observed in four other host crystals incorporating trivalent neodymium: CaF_2,[2] $SrMoO_4$,[3] $PbMoO_4$,[4] and $CaMoO_4$.[5] Of these, $CaWO_4$ and the molybdates (all scheelite structures) offer the most desirable crystal environments for trivalent neodymium; this follows from considerations of linewidth (3 to 7 cm^{-1} at 77°K) and fluorescence quantum efficiency. Annealing conditions for obtaining consistently clear crystals of $CaMoO_4$:Nd^{3+} have not yet been determined, and $PbMoO_4$ tends to crack under thermal shock of cooling to 77°K. It was decided, therefore, to conduct continuous operation experiments on the $CaWO_4$:Nd^{3+} and $SrMoO_4$:Nd^{3+} systems.[6]

The energy level diagram of the trivalent neodymium ion in $CaWO_4$ is shown in Fig. 1. This has been derived by combining the infrared fluorescence spectrum[1] with preliminary absorption data. Assignments are incomplete but the schematic is adequate for the discussion here. The principal pumping region for infrared fluorescence lies in the visible around 17 000 cm^{-1}. The gross features are very similar to Carlson and Dieke's detailed analysis[7] of the energy levels of trivalent neodymium in $LaCl_3$.

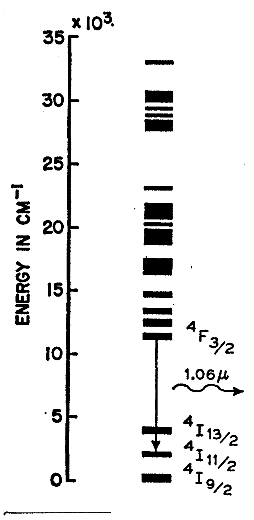

FIG. 1. Energy level diagram of Nd^{3+} in $CaWO_4$.

EXPERIMENTAL EQUIPMENT

Continuous operation of a solid state optical maser involves two major problems. The first is to obtain a pump lamp and proper housing so that there is sufficient pump light intensity incident on the crystal. The second is to provide an effective method of cooling the crystal, since a large portion of the optical energy absorbed by the crystal is necessarily released as heat.

In order to attempt to obtain continuous operation on promising crystals (promising on the basis of pulse maser threshold measurements), the apparatus shown in Fig. 2 was built. It consists of an elliptical cylinder, all of whose walls are well-polished reflecting surfaces. The flat polished top and bottom surfaces of the ellipse effectively extend its length. At the two foci of the ellipse are mounted respectively a General Electric AH6 linear high-pressure mercury lamp and the maser crystal. The lamp is water cooled, and so has the additional advantage that useless infrared radiation at wavelengths longer than 1.3 μ is eliminated. During the course of the investigation it was found very helpful to incorporate a flash tube (GE FT91) within the ellipse also, to determine (1) whether the threshold for pulsed maser operation was high or low; (2) how much the threshold for pulsed operation was reduced by steady illumination of the sample. The flash lamp is located in a nonfocusing position behind the maser crystal, where it blocks as little as possible of the light from the mercury lamp. Of course, the flash lamp is not used during continuous operation.

The form of optical wavelength resonator used is a rod of length large compared to its diameter polished with either plane parallel,[8,10] or approximately confocal[9,10] spherical ends. These surfaces are then made highly reflective with silver coatings.

[1] L. F. Johnson and K. Nassau, Proc. Inst. Radio Engrs. **49**, 1704–1706 (1961).
[2] L. F. Johnson, J. Appl. Phys. **33**, 756 (1962).
[4] L. F. Johnson and R. R. Soden, J. Appl. Phys. **33**, 757 (1962).
[5] L. F. Johnson and R. R. Soden (to be published).
[6] An account of a portion of this work is given in an earlier communication [L. F. Johnson, G. D. Boyd, K. Nassau, and R. R. Soden, Proc. Inst. Radio Engrs. **50**, 213 (1962)].
[7] E. H. Carlson and G. H. Dieke, J. Chem. Phys. **29**, 229 (1958); E. H. Carlson, Johns Hopkins University Spectroscopic Report No. 16, 1960 (unpublished).
[8] A. L. Schawlow and C. H. Townes, Phys. Rev. **112**, 1940 (1958).
[9] G. D. Boyd and J. P. Gordon, Bell System Tech. J. **40**, 489 (1961).
[10] A. G. Fox and Tingye Li, Bell System Tech. J. **40**, 453 (1961).

The method of cooling was evolved in discussions with R. J. Collins. It consists of boiling heat transfer between a bath of flowing liquid oxygen that has been precooled to liquid-nitrogen temperature in order to eliminate bubbling. For simplicity the liquid oxygen is formed by passing gasous oxygen through a copper tube immersed in liquid nitrogen. Liquid oxygen free of bubbles is thus formed which enters the Dewar. A flowing stream is necessary to prevent the crystal from being encased in a gas jacket and thus losing thermal contact with the bath. Nitrogen cooled to below its boiling point has also been used satisfactorily. It might be pointed out that on one occasion a $SrMoO_4$ crystal, exposed to the AH6 lamp driven at 2000 w, lost thermal contact with the bath of flowing liquid oxygen and melted. This required a temperature of about 1550°C.

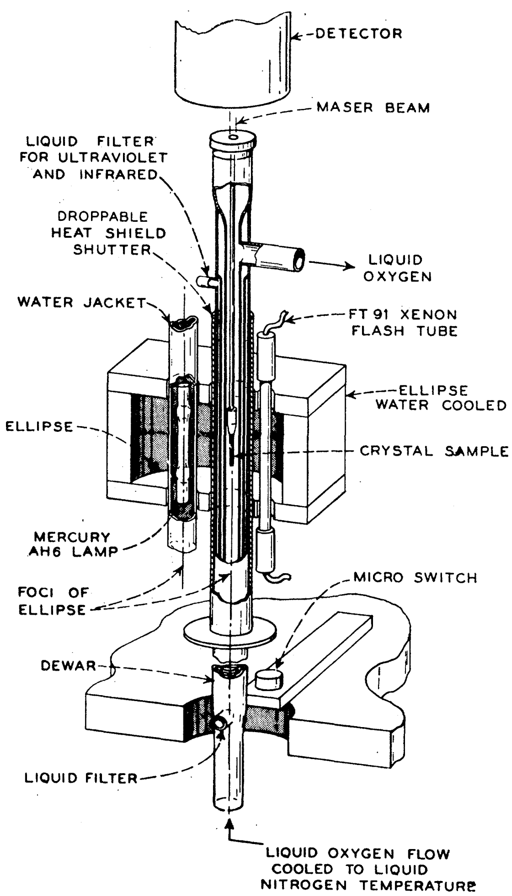

FIG. 2. Schematic of experimental equipment.

A circulating liquid filter surrounding the Dewar consists of 2 mm of sodium nitrite solution (400 g per liter of water). The filter absorbs energy below 4000 Å. Ultraviolet contributes little to the infrared fluorescence, but a considerable amount of defect production of an unknown nature seems to be caused by radiation below 4000 Å. The main symptom of these defects is to increase the threshold for stimulated emission. The effect partially anneals on warming to room temperature, analogous to an "F center" type of behavior. The filter has a negligible effect on the threshold but is a vital factor for extended continuous operation.

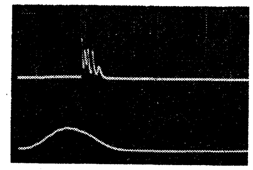

FIG. 3. Stimulated emission near threshold in $CaWO_4:Nd^{3+}$ at 1.065 μ, approximately 85°K. Pumping is achieved by a linear FT 91 xenon flash lamp in a nonfocused single ellipse configuration. Lower trace: Variation of lamp intensity with time. Upper trace: Fluorescence and stimulated emission. Time increases to the right (50 μsec per division) and the traces are triggered simultaneously.

In order to observe all the features of the stimulated emission pattern a fast response detector is required. In $CaWO_4:Nd^{3+}$ the fluorescence transition exhibiting the lowest threshold for stimulated emission lies at 1.065 μ. In $SrMoO_4:Nd^{3+}$ this line occurs at 1.064 μ. Both lie within the spectral sensitivity characteristic of an RCA 7102 infrared photomultiplier tube. Its sensitivity at 1.06 μ, however, is down by an order of magnitude from its peak. To prevent stray lamp light from over-riding the maser output, a 1-mm silicon window was placed over the photomultiplier tube to eliminate radiation below 1 μ.

$CaWO_4:Nd^{3+}$

A crystal of $CaWO_4$ containing 0.5% Nd atoms (ratio of neodymium to calcium atoms) was prepared in the confocal resonator geometry, consisting of a cylindrical rod with polished silvered spherical ends, the separation between reflecting ends being approximately equal to their common radius of curvature. The rod was 2-in. long, 0.079 in. in diameter, and one end was left partially transmitting (~1%).

The threshold for stimulated emission in the nonfocused single ellipse configuration was 6 joule of energy into the flash lamp (Fig. 3). A similar threshold is measured in a GE FT524 helical xenon flash lamp.

In the discussion of the experiment which follows, the linear xenon flash lamp is no longer used, the pumping system for continuous operation consisting solely of the linear AH6 mercury lamp at one focus of the ellipse and the maser rod at the other. In Fig. 4(a–d), the AH6 is powered by alternating current at 60 cps, producing "rectified" pump light intensity at a repetition rate of 120 cps. We shall call this "semicontinuous operation." Figure 4(a) shows the pattern of stimulated emission at about 1% above threshold for semicontinuous operation. The power input to the lamp is 920 w. The lower trace shows the variation of pump light intensity with time on a time scale of 5 msec per division. The intensity varies from a maximum to a nearly zero level of illumination. The maser output for the first six cycles of semicontinuous operation is shown in the upper trace. On each cycle normal fluorescence (spontaneous emission) increases until a critical density of inverted population is attained between the two states involved in the maser transition. A sudden burst of stimulated emission then appears, characteristic of optical maser action, continuing until the input to the lamp falls below the critical power level. In Fig. 4(b), the power input to the lamp has been raised to 1000 w (10% above threshold) and the time scale increased to 50 msec per division. Several dips in the pattern in the time interval 100–200 msec are caused by the bouncing of a metal heat shield used to trigger the operation and are to be ignored. In Fig. 4(c) the time scale has been increased to 0.5 sec per division and the vertical sensitivity reduced by a factor of 5. Note that the fluorescence level now appears as a heavy white line in the upper trace. The fluctuation in the amplitude of stimulated emission during the first 2 sec is believed to be due to the establishment of equilibrium conditions between sample temperature and heat transfer to the flowing liquid-oxygen bath. In Fig. 4(d), the time scale is 5 sec per division. It is seen that optical maser action proceeds in semi-continuous operation for 50 sec with no sign of degradation.

The technique of pumping by means of AC power applied to the AH6 mercury lamp has provided a very helpful means of identifying continuous optical maser operation and the dc power level at which it occurs. It enables us to apply dc power to the AH6 with some assurance of success. The result is shown in Fig. 5. Again, the time variation of lamp intensity is shown in the lower trace, the maser output in the upper, on a time scale of 5 sec per division. After 7 sec (to show the zero line of fluorescence and lamp intensity) the lamp is turned on and the crystal is exposed for 12 sec to illumination produced by 1300 w of dc power into the lamp. This power level is slightly above the threshold for stimulated emission and maser oscillation occurs. Note that about 5 sec is required for thermal equilibrium to be established. The power level is then lowered to 940 w for 8 sec, yielding fluorescence output only (nearly coinciding with the zero line). The power is then raised to 1600 w, well above threshold, and continuous operation is maintained for about 12 sec. Note that the dc level of maser output is now well above the fluorescence level. Finally, the lamp is turned off. The trace appears to indicate that maser output in continuous operation is not very constant. However, the trace is misleading since the "spikes" are caused by 60 cps ripple on the dc, amounting to about 20%. This is verified by Fig. 6 where the sweep speed has been increased to 5 msec per division (power level 1500 w). Note the zero line of maser output and lamp intensity. The trace clearly shows that maser output follows the 60 cps ripple on the dc, and that "spiking" is not present in continuous operation.

At the 1600-w level of pump power in Fig. 5 the maser output power transmitted through the lightly silvered end of the crystal is about 2 mw ac superimposed on a dc output of about 1 kw.

Subsequent work has resulted in continuous operation at room temperature at 1.058 μ, using water cooling of the crystal. The principal factor making this possible is the addition of monovalent sodium ions to provide local charge compensation for trivalent neodymium. A compensation ratio Na:Nd of 3:1 simplifies the fluorescence spectrum and lowers the threshold for maser oscillation by a factor of 3. At room temperature power output $\sim$10 mw has been obtained, with silvered end reflectors, at an input power to the lamp of 1.5 kw. Multiple dielectric layer reflectors should significantly

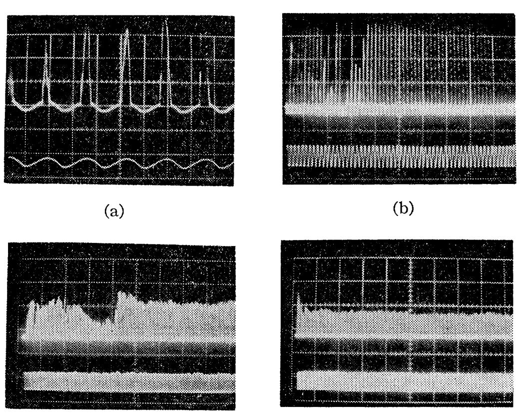

FIG. 4. Optical maser action in $CaWO_4:Nd^{3+}$ in semicontinuous operation. Pump light intensity at a repetition rate of 120 cps is provided by an AH6 mercury lamp powered by alternating current. The lower trace in each oscilloscope pattern records the variation of lamp intensity with time; optical maser output is displayed in the upper trace. (a) Power input to the lamp 920 w, or about 1% above threshold; time scale—5 msec per division. (b) Power input to the lamp 1000 w (10% above threshold); time scale—50 msec per division (c) Time scale—0.5 sec per division; vertical sensitivity of maser output has been reduced by a factor of 5. The level of fluorescence now appears as a heavy white line in the upper trace. (d) Time scale—5 sec per division. Semicontinuous operation observed for 50 sec.

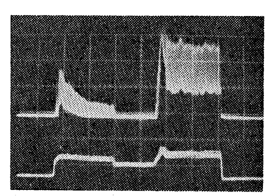

FIG. 5. Continuous operation of the $CaWO_4:Nd^{3+}$ optical maser. Lamp intensity is displayed in the lower trace, maser output in the upper trace. Time scale—5 sec per division. After 7 sec (to show zero line) the lamp is turned on and the maser crystal is exposed for 12 sec to 1300 w of dc power into the AH6 a power level slightly above the threshold for stimulated emission. The power level is then lowered to 940 w for 8 sec, yielding fluorescence output only (nearly coinciding with the zero line). The power is then raised well above threshold to 1600 w, continuous operation maintained for about 12 sec, and, finally, the lamp is turned off.

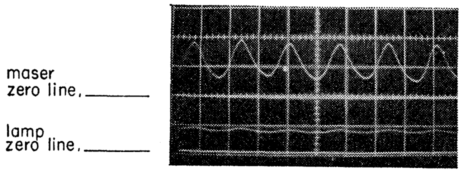

FIG. 6. Detail of continuous operation on a time scale of 5 msec per division. Power input to the lamp 1500 w (lower trace). Note that maser output (upper trace) follows the 60-cps ripple on the dc.

raise this figure, as well as lower the pump power required for continuous maser operation.

$SrMoO_4:Nd^{3+}$.

Oscilloscope traces of the $SrMoO_4:Nd^{3+}$ optical maser in semicontinuous operation are very similar to those shown for $CaWO_4:Nd^{3+}$. In the lowest threshold crystal obtained to date, the oscillations or spikes in stimulated emission in a pulse experiment are less severe (see Fig. 3). As with the tungstate, semicontinuous operation was achieved for 50 sec with no sign of deterioration. The threshold, however, was slightly higher than for the $CaWO_4:Nd^{3+}$ (10 joules as compared with 6 joules) and continuous (dc) operation could not be obtained.

CONCLUSIONS

Continuous operation of a solid-state infrared optical maser has been demonstrated at 85° and 295°K operating in an elliptical cylinder reflector. Continuous operation at room temperature is possible because of the large energy separation between the lower level of the maser transition and the ground state (approximately 2000 cm^{-1}). Power output $\sim$10 mw has been observed at room temperature. The use of multiple dielectric layer reflectors should significantly raise the resonator Q and thus decrease inversely the pump power required for continuous maser operation.

ACKNOWLEDGMENTS

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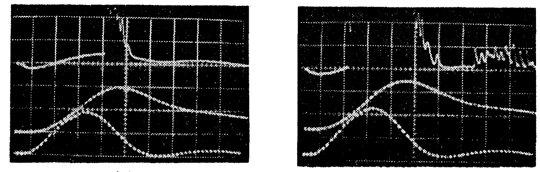
APPENDIX. REDUCTION IN THRESHOLD TECHNIQUE

An experimental procedure devised to measure how close a given crystal is to continuous operation for the available continuous light intensity is as follows. The maser crystal is mounted in the Dewar and its threshold with the flash lamp is found. A dual-beam oscilloscope is used to display the maser output and a phototube trace of the flash lamp intensity simultaneously, as in the upper and lower traces of Fig. 7(a). Also note in Fig. 7(a) the middle trace which is an electronic integration of the flash-lamp trace. The integrator has a time constant of 130 μsec. Although the fluorescence lifetime of Nd^{3+} in $CaWO_4$ has not yet been determined a lifetime of about 130 μsec was measured earlier for Nd^{3+} in $LaCl_3$, and it is not expected to be greatly different in $CaWO_4$. Therefore the height of the integrator trace at the start of maser oscillation gives a direct measure of the integrated lamp intensity required to obtain an inversion of population sufficient to achieve the maser oscillation condition.



(a) (b)

FIG. 7. Reduction of threshold experiment. (a) Maser oscillation with flash lamp only. Upper trace: maser beam; Middle trace: integration of flash lamp intensity; Lower trace: profile of flash lamp intensity vs time. (b) Maser oscillation with both continuous lamp and flash lamp on. Traces are the same as above. Sweep rate—50 μsec per division. The reduction in threshold due to illumination by the continuous lamp is given by the percentage difference in height of the integrator signal between (a) and (b) at the beginning of maser oscillation.

If now the continuous lamp is turned on and flash-lamp intensity is superimposed, a measure of the contribution of the continuous lamp is obtained from the observed reduction in threshold on the integrator trace. For convenience the AH6 lamp is operated with alternating current. A phasing circuit synchronizes the firing of the flash lamp with the peak intensity of the 120-cps continuous lamp. To prevent the maser crystal from heating up during the half minute or so stabilizing time for the continuous lamp, a metal tube is raised in position around the Dewar to shield the crystal. The heat-shield shutter is then allowed to drop and trigger a microswitch which in conjunction with a multivibrator fires the flash tube in synchronism with the peak of the light intensity from the continuous lamp. The result of such an experiment is shown in Fig. 7(b). Note that maser oscillation starts earlier in time, and that the height of the middle integrator trace at the start of maser oscillation is considerably less than in Fig. 7(a). This implies a reduction in threshold of 75%. The photos of Fig. 7 were obtained with one of our early crystals. Note that the integrator trace indicates only the flash lamp intensity since the continuous lamp merely shifts the zero line of the integrator.

The technique of measuring reduction in threshold is quite useful in evaluating the pumping efficiency of a maser material by a given lamp as well as indicating for a given type of material how close one is to continuous operation.

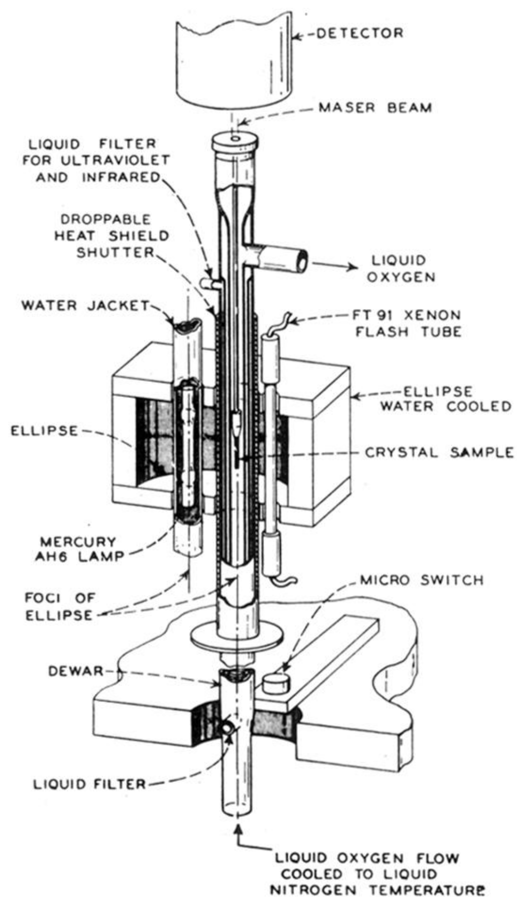
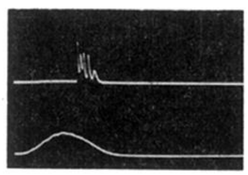


FIG. 2. Schematic of experimental equipment.



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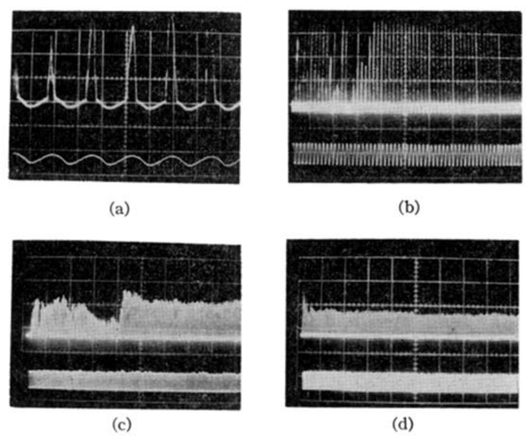

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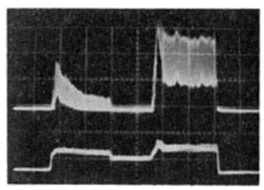

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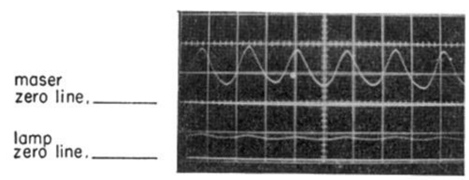

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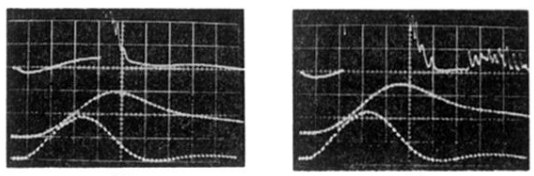

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