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Solid-State Optical Maser Using Divalent Samarium in Calcium Fluoride

This Letter reports a new solid-state optical maser with an output frequency in the red portion of the visible spectrum. One of the important features of this maser is its potential for cw operation. In this respect it is similar to a previously reported optical maser using trivalent uranium in calcium fluoride.[1]

The concept of infrared and optical masers was proposed by Schawlow and Townes.[2] In their important analysis of the problems that would have to be overcome in extending maser techniques into the infrared and visible portions of the electromagnetic spectrum, they proposed the use of a Fabry-Perot interferometer as an optical "cavity." One of the principal problems in the optical region is the high density of electromagnetic modes. In the Schawlow-Townes cavity arrangement those modes whose propagation vector k is not along the axis of the interferometer will not have the high Q which characterizes those modes with propagation vector parallel to the axis. Since stimulated emission of radiation tends to occur in the highest-Q modes, an optical maser would tend to oscillate in the relatively few axially directed modes.

Both gaseous and solid state systems have been proposed as active optical maser media. In the case of a solid state optical maser a suitable crystal can be cut into the form of a cylindrical rod with ends polished and silvered. These two mirror-like surfaces form the mode-selecting Fabry-Perot interferometer. The first optical maser was reported by Maiman and used ruby (Cr^{+++} in Al_2O_3) in this configuration.[3]

General principles

The generation of coherent radiation by maser techniques in a solid requires that there be an inversion of population between two sharp atomic states, an upper metastable state (a) and a lower terminal state (b) of the optically active ion. In the case of solids this inversion is achieved by optically pumping ions from the ground state to a broad band of high energy states. The ions then undergo rapid nonradiative transitions to a metastable state (a). Under favorable conditions the accumulation of ions in state (a) can be sufficiently large to produce an inversion relative to the lower state (b). If light with a frequency corresponding to the energy difference $E_a - E_b$ passes through the medium in the absence of pumping radiation, the normal Boltzmann population distribution prevails and the light is absorbed. If an inversion of population is achieved by pumping, the light wave will gather energy by stimulating transitions from the state (a) to the lower state (b). The critical oscillation condition requires that the losses in the cavity be smaller than the energy gained by the light wave in this manner. To sum up, there is a critical excess population Δn determined primarily by the losses in the Fabry-Perot interferometer plates, and oscillation will be achieved only when the difference $n_a - n_b$ exceeds this value.

In the case of the ruby optical maser the terminal state is the ground state of the Cr^{+++} ion. To achieve inversion at least fifty percent of the total ions must be maintained in excited states, which requires very large amounts of power.

One evident way of overcoming the disadvantage inherent in ruby is to use a material in which the stimulated emission terminates on a state sufficiently far above the ground state so that n_b is sufficiently small at operating temperatures that it can be neglected. To achieve oscillation it is therefore sufficient to excite into the metastable state approximately Δn ions. This number may be several orders of magnitude smaller than the concentration of ions in the crystal. In addition, the terminal state must be sufficiently close to the ground state so that it can be rapidly emptied through nonradiative transitions to the ground state by interaction with the lattice vibrations. Energy splittings of approximately 100 to 1000 cm^{-1} are therefore desirable. Both trivalent uranium and divalent samarium in calcium fluoride have energy level structures which satisfy both of these requirements at low temperatures.

Divalent samarium in CaF_2

Figure 1 shows the energy level diagram of Sm^{++} in CaF_2, based on the work of Feofilov.[5] The principal absorption band is centered at 6320 A and is responsible for the green coloration of the crystals. There are also a few absorption bands at shorter wavelengths and fluorescence is induced by pumping in any one or all of these bands. The fluorescence appears to originate from a metastable level located 14,500 cm^{-1} (6902 A) above the $^7F_0$ ground state. In the emission spectrum there are sharp lines representing transitions from the metastable state to the $^7F_0$ ground state and to the multiplet levels $^7F_1$, $^7F_2$, $^7F_3$, ... lying above it. Of particular importance is the fact

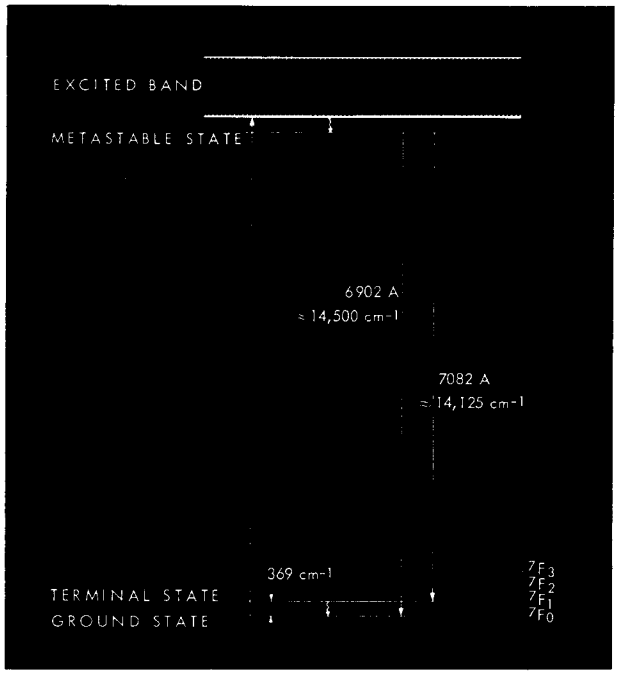

Figure 1 **Energy level diagram of divalent samarium in calcium fluoride.** *(After Feofilov, Ref. 5).*

that the intensity of the fluorescent transition to the $^7F_1$ state which lies 369 cm^{-1} above the ground state is substantially greater than the intensities of the other fluorescent transitions. This is the transition in which the maser oscillation occurs and the relative strength of the line corresponding to this transition implies that the fluorescent energy is not diverted into competing transitions. The $^7F_1$ state is spaced 369 cm^{-1} above the ground state and it is clear that this system at low temperatures is one which operates in the efficient manner described in the previous paragraph. Furthermore, since the principal band of states to which ions are pumped lies only slightly above the metastable state it is clear that a relatively small amount of energy is dissipated into lattice vibrations via the nonradiative transitions. The fluorescence lifetime was measured by Feofilov to be 1.3×10^{-6} second, and this is an indication that the divalent samarium ion is somewhat less shielded from the effects of the crystal field than the trivalent rare earth ions. An interesting feature is that there should be no Stark splitting of the terminal state $^7F_1$, for it is a general result that a state with angular momentum $J=1$ remains degenerate in a crystal field of cubic symmetry.

The crystal of CaF_2 with a 0.1 mole percent concentration of Sm^{++} was grown in the form of a boule by the Isomet Corporation under strongly reductive conditions which are necessary for the incorporation of samarium in the divalent state into the calcium fluoride lattice. Cylindrical samples approximately 20 mm long and 4 mm in diameter were cut from the boule. The ends were polished flat to one-fifth of the wavelength of sodium light and parallel to one minute of arc. One end was heavily silvered and the other end silvered until it transmitted about 1.5 percent. The maser crystal was cooled indirectly by liquid helium in a specially constructed dewar. A xenon flash lamp was used and the pumping light was focused through a window at the vertically positioned crystal. The stimulated and spontaneous emissions were passed through a monochromator and detected with a photomultiplier tube.

Figure 2 shows the photomultiplier signal as displayed on an oscilloscope with the monochromator set at 7082 A. Trace 2a is a photograph of the spontaneous emission with the pumping power below the threshold required for oscillation. Trace 2b shows the sharply nonlinear onset of oscillation when the pumping power is set somewhat above threshold. Figure 2c displays the signal at higher pump power level.

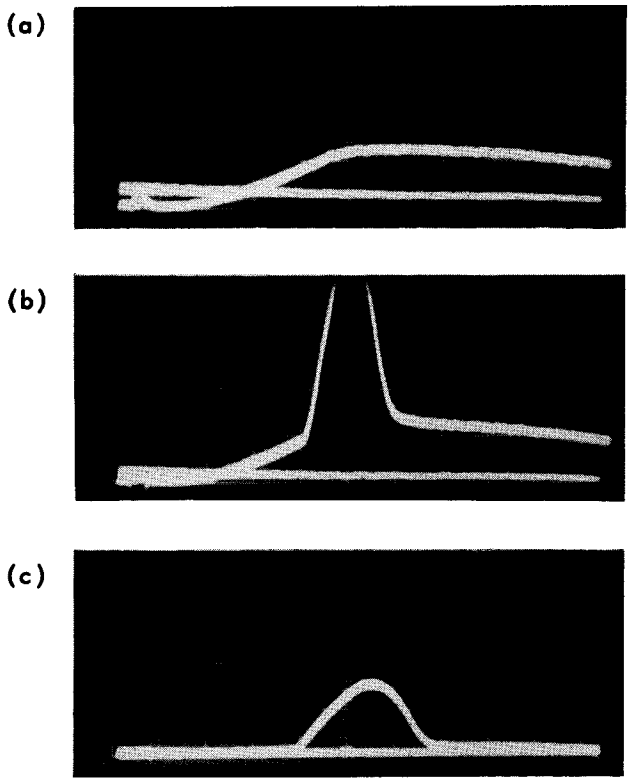

Figure 2 **Photomultiplier response vs. time.** *The sweep speed is 50 microseconds per division. (a) Trace showing spontaneous emission with pumping light below threshold. (b) Trace showing nonlinear onset of stimulated emission with pumping light adjusted very close to threshold. (c) Trace showing output with pumping light a few percent above threshold. Gain is reduced in trace (c) by a factor of 10.*

A notable feature of Fig. 2 is the absence of relaxation oscillations observed by us both in the case of ruby and trivalent uranium masers. Perhaps this is due to the small energy difference between the broad energy band and the metastable level which permits very rapid nonradiative transitions to the metastable state.

In the case of trivalent uranium, the terminal state is emptied very rapidly, as with the divalent samarium, but the relatively large energy difference between the broad pumping band and the metastable level may be the "bottleneck" in the flow of ions required to maintain sufficient excess population in the metastable state for oscillation. In the case of ruby the relaxation oscillation appears to be due to a limitation in the rate at which the ions are pumped from the ground state. This limitation exists primarily because of the large number of ions which must be excited to achieve inversion of population.

The energy level structure of divalent samarium in calcium fluoride approaches the ideal necessary to achieve cw maser operation. Reduction of losses in the interferometer mirrors by the use of multiple dielectric layers should lead to the development of a cw maser in the future.

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References

1. P. P. Sorokin and M. J. Stevenson, *Phys. Rev. Letters,* **5,** 557 (1960).
2. A. L. Schawlow and C. H. Townes, *Phys. Rev.,* **112,** 1940 (1958); A. L. Schawlow, in *Quantum Electronics,* edited by C. H. Townes, Columbia University Press, N. Y., 1960.
3. T. H. Maiman. *Nature,* **187,** 493 (1960); *British Communications and Electronics,* **7,** 674 (1960).
4. R. J. Collins, D. F. Nelson, A. L. Schawlow, W. Bond, C. G. B. Garrett, and W. Kaiser, *Phys. Rev. Letters,* **5,** 303 (1960).
5. P. P. Feofilov, *Optics and Spectroscopy,* **1,** Issue #8, 992 (1956). (In Russian.)

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