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End-Pumped Stimulated Emission from a Thiacarbocyanine Dye*

Using the same technique employed[1] in obtaining stimulated emission from chloro-aluminum phthalocyanine (CAP) solutions (where the beam from a giant-pulse ruby laser was caused to impinge transversely upon a cell filled with a dye solution and located within a secondary resonating cavity), we recently observed intense stimulated emission from a commercially available photosensitizing dye, 3, 3′-diethylthiatricarbocyanine iodide (hereafter, "DTTC"). Both methyl and ethyl alcohol were successfully used as solvents. This result and that of Ref. 1 combine to suggest that many other known photosensitizing dyes can be made to emit coherent beams of light similarly.

A most unusual feature of the DTTC laser is its broad spectral distribution.[2] Spectrally centered at approximately 8156 Å (in methyl alcohol solvent), the beam near threshold darkens photographic plates over a range of some 100 Å. With increasing output power the width of the spectral range quickly increases to a value of about 250 Å. Further increases in power bring only slight increases in spectral width, although the output power continues to increase linearly with applied pumping power. A broad-band laser emission of this sort provides strong basis for argument that inhomogeneous broadening prevails in symmetrical thiacarbocyanine dyes.

The center frequency of the laser beam coincides roughly with the point of maximum slope on the long wavelength side of the principal fluorescence band, which is itself Stokes-shifted by some 200 Å with respect to the principal absorption band. (Reference 3 gives the absorption spectrum for this molecule and for other members of the same vinylene-homologous series).

After the DTTC laser was successfully pumped transversely, one of us (W. H. C.) suggested end pumping. To this end a multiple dielectric mirror reflecting 40% at 0.694μ and 99% at 0.816μ was aligned in the system. This mirror served both as the output reflector for the ruby laser and as a secondary cavity reflector. The other secondary cavity reflector chosen transmits close to 25% at 0.816μ. The two were separated by 25 cm. A good-quality cylindrical cell (length, 1 in.; diameter, 0.75 in.) with antireflection coated windows was filled with the dye solution and centered in the secondary cavity. It was later found that the dye concentration had a low-level transmission at 6943 Å of roughly 45% over a 1-inch path.

When the laser system was fired a peak total output power of 0.63 MW at 0.816μ was obtained. The output mirror of the secondary cavity and the dye-filled cell were then removed and the peak total output power of the ruby laser was measured alone. This value was 4.45 MW. Hence, the efficiency in this non-optimized situation was $\sim$14%.

The far-field patterns were roughly compared by projecting the beams across the room to a target and photographing the resulting spot. With the cell and secondary cavity output mirror removed, the ruby beam illuminated a spot from which a five milliradian (half-angle) divergence was deduced. The area of the spot illuminated by the DTTC laser at the same distance was but 6% as large. Thus the actual intensity of the DTTC beam was, at an appreciable distance, some two and a half times that of the ruby beam.

References and footnotes

1. P. P. Sorokin and J. R. Lankard, "Stimulated Emission Observed from an Organic Dye, Chloro-aluminum Phthalocyanine," *IBM Journal* 10, 162 (1966).
2. The temporal behavior of the beam appeared to mirror the shape of the ruby laser giant pulse, in contrast with what is observed near threshold with the CAP laser system, pumped in the transverse geometry.
3. L. G. S. Brooker, "Absorption and Resonance in Dyes," *Rev. Mod. Phys.* 14, 275 (1942).

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