Neon Gas Maser Lines at 68.329 μ and 85.047 μ*

We report here the observation of continuous optical maser oscillation at $\lambda_{vac} = 68.329\ \mu$ and 85.047 μ in a dc excited discharge of neon. These correspond to the $7p[1/2]_1 - 6d[3/2]_2^0$ and $8p[3/2]_2 - 7d[5/2]_3^0$ transitions respectively. The wavelengths, 0.068 mm and 0.085 mm, are the longest yet observed with a gas discharge interferometer maser. We have previously reported oscillation on Ne $4p$-$3d$ lines from 4 μ to 11 μ;[1] $5p$-$4d$, 13 μ–23 μ;[1] $6p$-$5d$, 20 μ–41 μ;[1,2] and $7p$-$6d$, 36 μ–57 μ.[2]

In our previous work[1-3] we found that favorable lines obey $\Delta J = \Delta k = \Delta l$; these are the lines predicted to be strong by calculations of line strengths.[3] The 68.329 μ line has the longest wavelength of the favorable $7p$-$6d$ transitions. The 85.047 μ line is the first observed of the $8p$-$7d$ set; the favorable lines of this set consist of one at 59 μ, one at 72 μ, six near 90 μ, and one at 141 μ. Optimum conditions for this line were 0.035 torr Ne, 0.7 ampere discharge current.

The maser employed opaque gold mirrors and a coupling hole[2] which is calculated to give 0.5 per cent transmission at 85 μ. It can be shown that for the Gaussian field distribution of the fundamental mode

$$\text{fractional loss} = \frac{2 \times \text{hole area}}{\text{spot area}},$$

where spot area $\equiv \pi w_s^2$ and where w_s is the spot radius defined by Boyd and Gordon.[4] The maser cavity was 3.65 meters long and 34 mm ID, giving a Fresnel number $a^2/b\lambda = 0.8$ at 100 μ. Because the atomic linewidth is comparable to and probably less than the frequency separation of TEM_{00q} modes, it was necessary to tune the cavity length to bring a resonance to line center. This was done by magnetostriction in four Invar rods driving one mirror assembly.[5]

The 68 μ and the 85 μ lines were observed both with a Golay cell and with a low temperature Ge:In bolometer.[6] The S/N was about 100 times better with the latter detector. By comparison with a calibrated Eppley thermopile we estimated the NEP to be 2×10^{-11} watt and the sensitivity to be 1.7×10^3 v/w. The time constant was 15 msec. We infer for both the 68 μ line and the 85 μ line power levels of about 7×10^{-9} w at the detector. Presumably the power emergent from the maser (before filters and spectrometer) is substantially greater.

It is quite clear that the power levels of analogous lines of the successive systems $4p$-$3d$, $5p$-$4d$, etc. fall monotonically. Under conditions near optimal for the $8p[3/2]_2$-$7d[5/2]_3^0$ line at 85.047 μ (0.05 torr Ne, 0.6 ampere discharge current), the power of the $4p[3/2]_2$-$3d[5/2]_2^0$ line at 7.4799 μ was greater by a factor of about 10^3.

R. A. McFarlane
W. L. Faust
C. K. N. Patel
C. G. B. Garrett
Bell Telephone Labs., Inc.
Murray Hill, N. J.

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Stimulated Emission in Aromatic Organic Compounds*

Most lasers made to date have used an energy level system involving those well-defined states characteristic of atoms. Organic molecules offer an attractive alternate system of molecular levels which can be used to make lasers. High power output, wide choice of output wavelength, tuning possibilities, size, simplicity and high efficiency may be some of the attractive features of organic media for use as lasers.

The first suggestion of the use of organic compounds for producing coherent stimulated emission was made by Brock.[1] Subsequently, Morantz, *et al.*,[2-4] reported the observation of phenomena in organic phosphors which they attributed to stimulated emission. However, Lempicki[5] and Wilkinson[6] have been unable to observe such effects. The purpose of this communication is to consider the various spontaneous emission processes occurring in aromatic organic compounds. In particular the relative advantages of a fluorescent as compared to a phosphorescent liquid or solid-state system for achieving laser action will be discussed.

Aromatic organic molecules have molecular energy levels which are attributable to extensive delocalization of some of the bonding electrons. The molecular energy levels which describe the allowed states of the aromatic organic molecules are rather diffuse, in contrast to the sharply defined energy levels that are characteristic of atoms. Absorption and emission between these levels result in relatively broad bands which may vary in width from less than 100 Å to over 1000 Å. The various excited states, their lifetimes, and their relationships for an aromatic hydrocarbon (where only π-π^* transitions exist) can be understood qualitatively by the modified Jablonski diagram shown in Fig. 1.

Absorption of light (transitions 1 and 2) takes the molecule from the ground singlet state to the higher singlet states. The absorption cross sections for these transitions can be very high ($\sim10^{-16}$ cm^2/molecule). If excitation is to the second excited singlet state, a nonradiative internal conversion (transition 3) places the molecule in the first excited singlet state. In this state, the molecule quickly ($\sim10^{-12}$ sec) reaches thermal equilibrium with its surroundings. The subsequent nonradiative and emission processes which will eventually take place are determined by whether the molecule *fluoresces* (transition 4)($\tau\sim10^{-8}$ sec) or undergoes an "intersystem crossing" (transition 5) to the lowest triplet state of the molecule. The triplet state is rather long lived and in fluid solvents collisional deactivation (transition 6) occurs with no significant emission being observed. In rigid media, however, a long-lived emission (10^{-4}–10^1 sec) is observed (transition 7) and this emission is termed *phosphorescence*. Further details of these transition processes can be found elsewhere.[7]

A useful criterion for selecting an appropriate 3- or 4-level fluorescent and phosphorescent organic laser system is set by the number of molecules required for the onset of oscillations in an optical cavity[8]

$$\frac{\Delta N}{V} = \frac{4\pi^2 c \Delta\tilde{\nu} \alpha \tau_{spont}}{(\pi \ln 2)^{1/2} L \mu \lambda^2_{out}} \qquad (1)$$

where $\Delta N/V$ is the population difference between the respective "lasing" levels per unit volume, τ_{spont} is the intrinsic radiative lifetime, α is the total loss per pass in the laser material and resonator (including optical scattering, reflection losses, refractive losses, absorption losses and diffraction losses), L is the length of the cavity, μ is the refractive index of the laser medium, $\Delta\tilde{\nu}$ is the fluorescence width at half maximum in wavenumbers, λ_{out} is the emission wavelength and c is the velocity of light.

A very promising candidate for a 4-level fluorescent laser system is the molecule perylene. This may be contained in a rigid or fluid solvent to utilize intermolecular radiative transfer to the emitting species for efficient utilization of available pump light. Experimental tests showed that this compound is photochemically stable under very intense optical excitation. The absorption and fluorescence spectrum for perylene in benzene at 25°C is shown in Fig. 2 depicting a characteristic mirror image symmetry. The following data have been obtained in this laboratory and elsewhere:[9]

$\lambda_{out} = 4710$ Å $\Delta\tilde{\nu} = 2.12 \times 10^4$ cm^{-1}
$\lambda_{pump} = 4100$ Å $\tau_{spont} = 6.9 \times 10^{-9}$ sec

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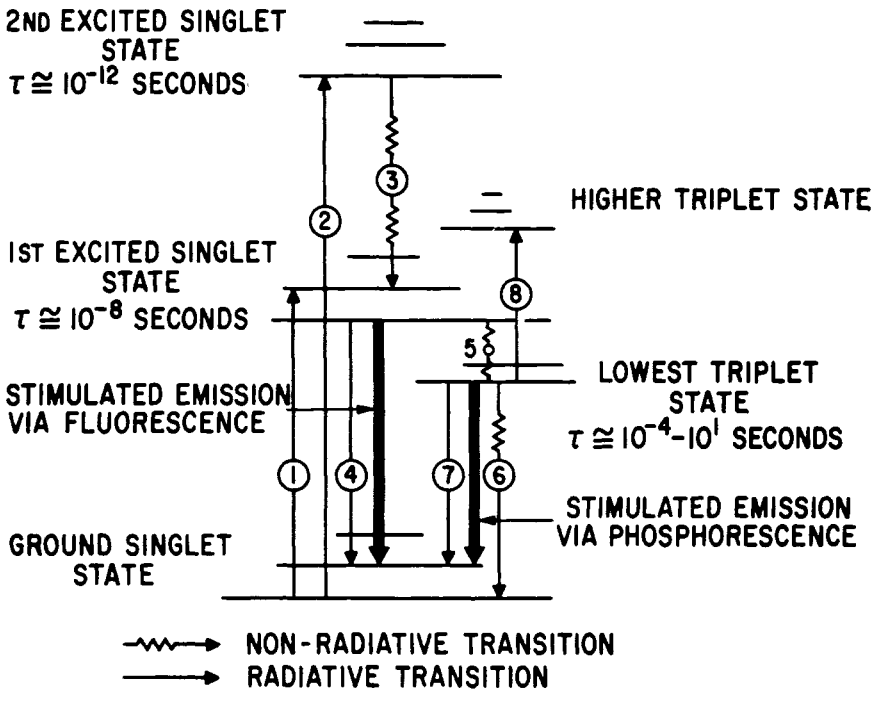

Fig. 1—A modified Jablonski diagram for an aromatic hydrocarbon.

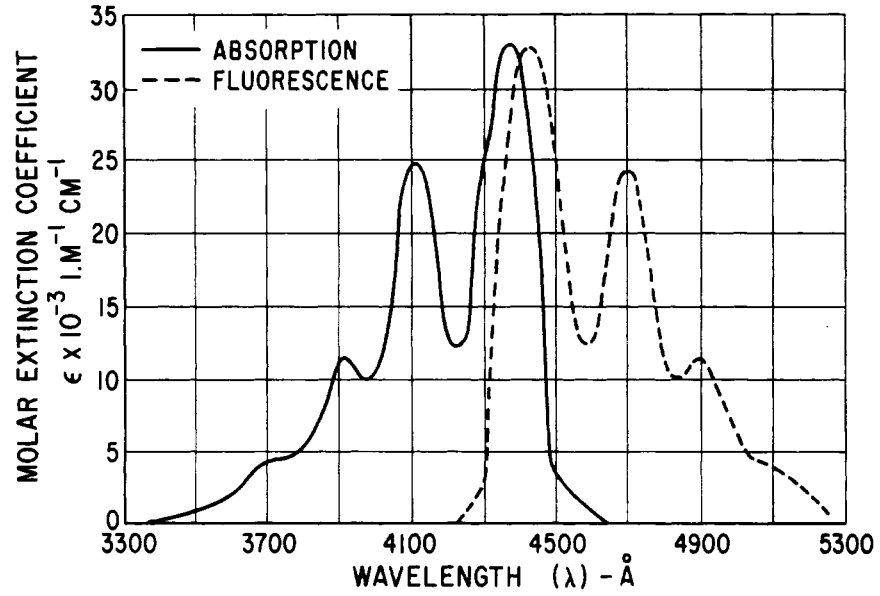

Fig. 2—Absorption and fluorescence spectrum of perylene in benzene.

(with a quantum efficiency of 0.96), and $\mu=1.5$, $L=6$ cm and $\alpha=0.18$ (assuming that scattering is the only significant contributing loss mechanism which at 1.06 microns was found to be 3 per cent/cm). Substituting into (1) the critical inversion concentration is $\Delta N/V=4.3\times10^{13}$ molecules/cm^3. This calculation assumes all radiation is in the 4710 Å band whereas only about one third is actually there (see Fig. 2). Therefore, the actual number required must be increased threefold to 1.3×10^{14} molecules/cm^3. Furthermore, a thermally depleted terminal state has been assumed while in fact the terminal-ground state separation is 1500 cm^{-1}. For a typical initial perylene concentration of 6×10^{16} molecules/cm^3, the final emitting state population has to be increased to 1.7×10^{14} molecules/cm^3. The critical or minimum pump power required for this system to oscillate is given by

$$P_{\text{critical}} = \frac{\Delta N h c}{V \lambda_{\text{pump}} \tau_{\text{spont}}} = 1.3 \times 10^{4} \text{ w/cm}^3.$$

This figure may be compared to 6×10^{2} w/cm^3 for a ruby laser.[10] Assuming a 1 per cent conversion efficiency for a broad spectral source (*e.g.*, xenon flashtube) and a coupling and absorbing efficiency of 50 per cent, the over-all electrical pump power requirement is 2.6×10^{6} w/cm^3. For a 3-cc volume, and an optical pump pulse duration of the order of 10^{-6} sec, the electrical energy input required is approximately 8 joules. Similar calculations for a potential 3-level fluorescent system show that the pump power requirements are considerably higher and probably cannot be attained with presently available optical pump sources. The threshold inverted population density for a 4-level phosphorescent compound such as benzophenone assuming no triplet-triplet absorption is estimated at 3×10^{20} molecules/cc with negligible power gain above threshold. Experiments to demonstrate laser action in fluorescent aromatic organic compounds are currently under way and will be reported later.

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D. L. STOCKMAN
W. R. MALLORY
Electronics Lab.
General Electric Co.
Syracuse, N. Y.
K. F. TITTEL
Advanced Technology Labs.
General Electric Co.
Schenectady, N. Y.

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The Dispersion Properties of Air as a Possible Limitation on the Maximum Useable Bandwidths of Coherent Optical Communication Systems*

Using an approximate analysis, this communication will suggest that an upperbound on the maximum useable bandwidth of a coherent optical communication system may exist due to the dispersion properties of the space (air) between the terminal points of this system. The maximum useable bandwidth is here defined as that bandwidth for which the distortion of the modulation of the coherent optical carrier (or the attendant loss of information accompanying this distortion) is under a specified maximum level, as set by the system designer.

Thus, imagine a coherent optical receiver-transmitter system immersed in an air path and separated by an arbitrary distance L. For space communications L can be quite large (*e.g.*, $L\approx2.10^5$ miles for earth-moon paths). If the refractive index of this path is taken as that observed for air, as given by Born and Wolf,[1] as plotted in Fig. 1, then this variation over the shown visible spectrum can be approximated by the form sug-

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