Neon Gas Maser Lines at 68.329 μ and 85.047 μ

We report here the observation of continuous optical maser oscillation at \(\lambda_{te}=68.329\) μ and 85.047 μ in a dc excited discharge of neon. These correspond to the \(7p[1/2],-6d[3/2]\) and \(8p[3/2],-7d[3/2]\) transitions respectively. The wavelengths, 0.068 μ and 0.085 μ, are the longest yet observed in a maser employing interference bolometer. We have previously reported oscillation on Ne 4p-3d lines from 4 μ to 11 μ; 5p-4d, 13 μ-23 μ; 6p-5d, 20 μ-41 μ, and 7p-6d, 36 μ-57 μ.

In our previous work\(^1\) we found that favorable lines obey \(\Delta J=\Delta K=\Delta \ell\); these are the lines predicted to give long life calculations of line strengths.\(^2\) 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, 0.73 amper discharge current.

The maser employs gold mirrors and a coupling hole\(^3\) which is calculated to give 0.5 μ 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 \(=\pi w_0^2\) and where \(w_0\) is the spot radius defined by Boyd and Gordon.\(^4\) The maser cavity was 3.63 meters long and 34 mm ID, giving a Fresnel number \(a^2/\lambda\) = 0.8 at 100 μ. Because the atomic linewidth is comparable to and probably less than the frequency separation of TEM\(_{00}\) modes, it was necessary to tune the cavity length to bring a resonance to line center. This was done by magnetostriiction in fourteen 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 Epplpe thermouple we estimated the YEP to be \(0.068\) μ amper discharge current, the power of the 4p\([3/2]\)-3d\([3/2]\) line at 7.47 μ is greater by a factor of about 10.\(^7\)

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

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, temperature effects, 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.,\(^8\) observed the observation of phenomena in organic phosphors which they attributed to stimulated emission. However, Lempicki and Wilkinson\(^9\) 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 \(\pi^*\) 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 \((>10^{-18} \text{cm}^2/\text{molecule})\). For 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 \((\sim 10^{-13} \text{sec})\) reaches thermal equilibrium with its surroundings. The subsequent radiative and emission processes which may follow are determined by whether the molecule fluoresces \((\text{transition 4} \sim 10^{-8} \text{sec})\) or undergoes an "intersystem crossing" \((\text{transition 5})\) to the lowest triplet state of the molecule. The triplet state is rather long lived and in fluid solvents collisional deactivation \((\text{transition 6})\) occurs with no significant emission being observed. In rigid media, however, a long-lived emission \((10^{-1} - 10^{-2} \text{sec})\) is observed \((\text{transition 7})\) 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\(^6\)

\[
\Delta N = \frac{4e^2\Delta\nu_{\text{mirror}}}{(\pi \ln 2)^2/\lambda^2 n^2}
\]

where \(\Delta N/V\) is the population difference between the respective "lasing" levels per unit volume, \(\nu_{\text{mirror}}\) is the intrinsic radiative lifetime, \(z\) is the total loss per pass in the laser material and resonator \((\text{including optical scattering, reflection losses, refractive losses, absorption losses and diffraction losses})\), \(L\) is the length of the cavity, \(\mu\) is the refractive index of the laser medium, \(\Delta\nu\) is the fluorescence width at half maximum in wave numbers, \(\lambda_{\text{em}}\) 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 intense optical excitation. The absorption and fluorescence of 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:\(^9\)

\[
\lambda_{\text{em}} = 4710 \AA
\]

\[
\Delta\nu = 2.12 \times 10^4 \text{cm}^{-1}
\]

\[
\lambda_{\text{mirror}} = 4100 \AA
\]

\[
\nu_{\text{mirror}} = 6.9 \times 10^4 \text{sec}^{-1}
\]

Ground Singlet State

2nd Excited Singlet State
τ ≈ 10^{-12} SECONDS

1st Excited Singlet State
τ ≈ 10^{-8} SECONDS

Stimulated Emission via Fluorescence

Higher Triplet State

Lowest Triplet State
τ ≈ 10^{-4} - 10^{1} SECONDS

Stimulated Emission via Phosphorescence

Non-Radiative Transition

Radiative Transition

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

![Absorption and fluorescence spectrum of perylene in benzene.](image)

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 the 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 \approx 2 \times 10^{8} 

\text{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, \cite{1} as plotted in Fig. 2, then this variation over the shown visible spectrum can be approximated by the form sug-

\begin{align*}
\Delta V &= \frac{\Delta N c}{V_{\text{pump}}r_{\text{point}}} = 1.3 \times 10^{4} \text{ w/cm}^2.
\end{align*}

This figure may be compared to \(6 \times 10^{6} \text{ w/cm}^2\) for a ruby laser, \cite{13} Assuming a 1 per cent conversion efficiency for a broad spectral source (e.g., xenon flashlamps) and a coupling and absorbing efficiency of 50 percent, the over-all electrical pump power requirement is \(2.6 \times 10^{8} \text{ w/cm}^2\). For a 3-cm volume, and an optical pump pulse duration of the order of \(10^{-s} \text{ sec}\), 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 \(3 \times 10^{16} \text{ molecules/cc}\), 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.

ACKNOWLEDGMENT

The authors wish to thank H. C. Rothenberg for helpful discussions.

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.


\textsuperscript{*} Received January 20, 1964.