FLUORESCENCE EXCITATION BY THE ABSORPTION OF TWO CONSECUTIVE PHOTONS*

John F. Porter, Jr.
Radiation Laboratory, The Johns Hopkins University, Baltimore, Maryland
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Fluorescence radiation from polycrystalline La:$PrCl_3$ has been observed in this laboratory using two separate excitation sources. This fluorescence cannot be attributed to either source alone. The scheme is similar to Bloembergen's[1] proposal for a solid-state infrared quantum counter which was further elaborated upon by this author.[2] Figure 1 shows the partial energy level diagram of Pr^{+++} in a lattice of $LaCl_3$.[3] An infrared signal at 2.3 μ may induce a transition from the $^3H_4$ ground state to the $^3H_6$ state. An electron in the $^3H_6$ state may then be raised to the $^3P_2$ state by the absorption of a photon at ~558 mμ. The $^3P_2$ state is not a resonance level. Electrons in this state decay by a radiationless transition to the $^3P_1$ and the $^3P_0$ states which are resonance levels.[4] The subsequent radiative transition from the $^3P_0$ state to the $^3H_6$ state results in the emission of a photon at ~618 mμ. The electron now in the $^3H_6$ state can be re-excited to the $^3P_2$ state by the incident 558-mμ excitation, thus completing the entire cycle again. If the lattice is at a temperature T such that $kT \ll h\nu$, where $h\nu$ is the energy of the $^3H_6$ level relative to the $^3H_4$ ground state, then this level will be unpopulated and the incident radiation at 558 mμ cannot induce any transitions to the $^3P_2$ state unless there is also an incident signal at 2.3 μ to populate the $^3H_6$ level. Thus the absorption of two consecutive photons at wavelengths 2.3 μ and 558 mμ is necessary to observe fluorescence at 618 mμ.

The experimental details are essentially as given in reference 2. The infrared source was a Sylvania DLG projection lamp with Pyrex and germanium filters to give the desired bandpass. The source of the 558-mμ radiation was an AH6 mercury lamp with suitable glass and interference filters. Sharp-cutoff glass filters were used to prevent the direct excitation of the $^3P_2$ state from the $^3H_4$ ground state. An RCA 7265 photomultiplier was used to detect the 618-mμ radiation. Again interference and glass filters were used to prevent the light from the AH6 lamp from entering the

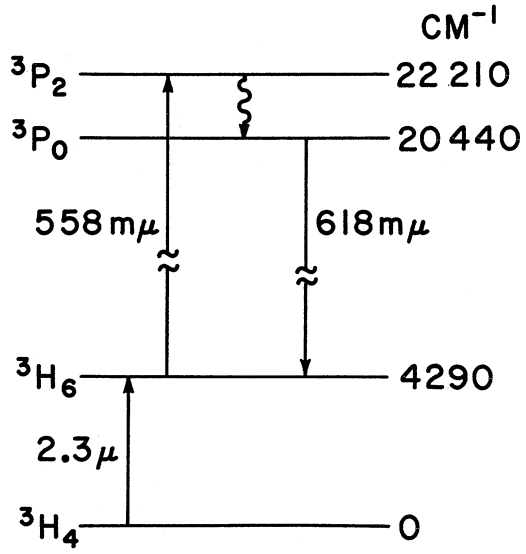

FIG. 1. Partial energy level diagram of Pr^{+++} in a host lattice of $LaCl_3$ showing the only levels involved. The energy given for the $^3P_2$ and $^3H_6$ levels are average energies of the multiplets.

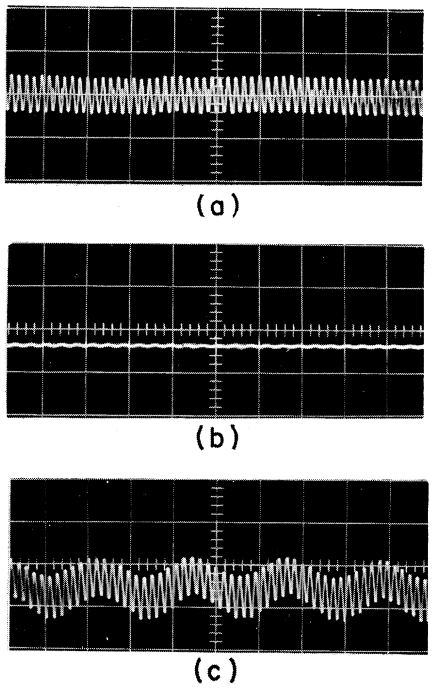

FIG. 2. Oscilloscope trace of phototube output. (a) 558-mμ source alone; (b) 2.3-μ sources alone; (c) 558-mμ and 2.3-μ sources incident on the sample showing the 10-cps modulated component of the output. This is the 619-mμ output modulated by the 10-cps 2.3-μ excitation.

photomultiplier. The infrared source was modulated at 10 cps and the output of the phototube, after amplification, was passed through a low-pass filter and presented on an oscilloscope. The sample was a polycrystalline ingot of 1% $PrCl_3$ in $LaCl_3$ which was refined by vacuum distillation. It was mounted in the tip of a liquid helium Dewar and cooled by conduction.

Figure 2(a) shows the output of the phototube with the 558-mμ excitation alone. The 120-cps signal is the "leak" of the AH6 lamp into the phototube. This is present when the sample is warmed up and is also present in a slightly diminished intensity when the sample in the Dewar tip is replaced by a glass tube similar to the Dewar tip. Figure 2(b) shows the output of the phototube when the infrared source alone is on the sample. Figure 2(c) is the output when both sources are on and clearly shows the 10-cps modulation of the 120-cps signal. This demonstrates that both excitation sources must be on the sample to observe the fluorescence. With a higher degree of isolation between the 558-mμ source and the 619-mμ detector it may be possible to use this solid-state system in the manner proposed by Bloembergen for an infrared quantum counter. This also provides an additional method for studying the lifetimes of the excited states.

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COHERENT LIGHT AMPLIFICATION IN OPTICALLY PUMPED Cs VAPOR*

S. Jacobs, G. Gould, and P. Rabinowitz
TRG, Incorporated, Syosset, New York
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An excess population density in the upper of two states connected by an optical transition makes possible light amplification by stimulated emission of radiation (LASER). This has recently been demonstrated in a gaseous system employing collisions of the second kind.[1] We have measured spatially coherent amplification in cesium vapor excited by selective optical pumping.[2]

An optical pump suitable for obtaining a population inversion in cesium is the intense He 3888A line, which overlaps the third cesium resonance line (see Fig. 1). In 1930 Boeckner reported fluorescence in cesium excited by the He 3888A line.[3] We have extended Boeckner's optical-pumping experiment by measuring the absolute intensities of Cs fluorescent lines[4] and from these measurements have computed the populations of the associated states using transition probabilities calculated by the Bates-Damgaard method.[5]

Table I shows the populations of certain energy levels obtained under optimized conditions. The population of the $8P_{1/2}$ energy level was substantially greater than that of the lower levels, $8S_{1/2}$ and $6D_{3/2}$. LASER action was therefore possible at the corresponding wavelengths, $\lambda = 7.2\ \mu$ and $3.2\ \mu$.

The amplification coefficient is related to the populations by the equation,

$$k(\nu) = (h\nu/c)(B_{21}N_2 - B_{12}N_1)S(\nu), \qquad (1)$$

where $S(\nu)$ is the normalized line-shape function $[\int S(\nu)d\nu = 1]$; B is the coefficient of induced emission (Einstein B); N_1 and N_2 are the population densities.

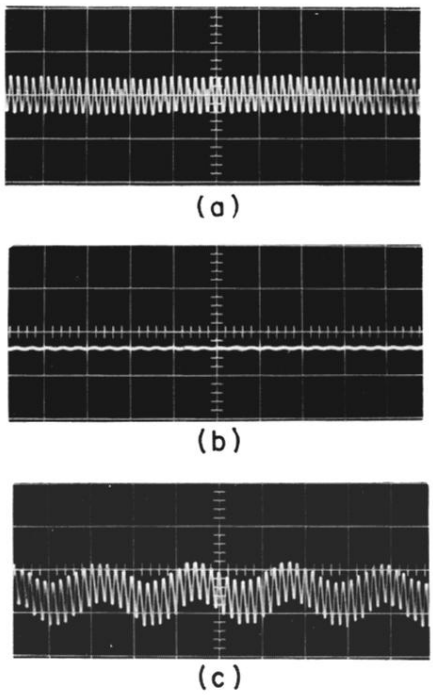
Assuming a Doppler profile, the amplification coefficient at line center reduces to

$$k(\nu_0) = \frac{1}{I}\frac{\partial I}{\partial z} = \left(\frac{\ln 2}{16\,c^2\pi^3}\right)^{1/2} A_{21}\frac{\lambda_0^4}{\Delta\lambda_d}\left(N_2 - \frac{g_2}{g_1}N_1\right), \qquad (2)$$

where A is the spontaneous emission coefficient (Einstein A), g_1 and g_2 are statistical weights, and

Table I. Populations of Cs energy levels under optimized conditions.

Cs energy levels	Population density (10^6 atoms/cm^3)
$8P_{1/2}$	100
$8P_{3/2}$	30
$8S_{1/2}$	10
$6D_{3/2}$	3
$5D_{3/2}$	200



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