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EFFECTIVE FLUORESCENT LIFE-TIMES IN
RUBY LASER RODS

J. A. Fleck, Jr.

April 15, 1964

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ABSTRACT

In the presence of a population inversion fluorescent life-times are reduced as a result of stimulated emission. Effective fluorescent life-times have been calculated using a Monte Carlo method for ruby rods with Brewster angle ends and roughened sides. The calculations take into account the dependence of gain and reflectivity on polarization and the dependence of gain on frequency. Results show that effective fluorescent life-times are quite insensitive to rod length but strongly dependent on rod diameter. They also indicate the possibility of oscillation in a random mode for rods of sufficiently large diameter.

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INTRODUCTION

As is well known, short duration pulses of extremely high power may be obtained from lasers through the method of Q-spoiling.¹ This method consists in first setting up as high a population inversion as possible by initially blocking the regenerative path of photons in the desired mode, then restoring the path by a rapid switching operation. Naturally, the effectiveness of this mode of operation depends on how high a population inversion can be achieved during the low-Q stage. In three-level lasers such as ruby the level of inversion is limited by two effects, the depletion of ground-state atoms capable of absorbing pump radiation and the enhanced rate of fluorescent emission, stimulated by spontaneous emission photons in their random paths through the crystal. The effect of "super-radiance", as the latter is called, has been calculated by a Monte Carlo method for ruby rods with Brewster angle ends and roughened sides using a digital computer.² The calculation involves the generation of randomly directed photon histories starting from points uniformly distributed within the rod and having frequencies selected from a Lorentz distribution. These photon histories are followed through a large number of reflections with the rod boundaries; taking into account the dependence of gain and reflectivity on polarization. The net photon statistical weight $\langle W \rangle$ per unit initial weight emerging from the rod, averaged over a large number of histories, is interpreted as a measure of super-radiance. The reciprocal of $\langle W \rangle$ is the ratio $\tau^* = \tau_{\text{eff}}/\tau$ of the effective fluorescent life-time to the fluorescent life-time in the absence of a population inversion.

Method of Calculation

One may interpret the calculation procedure as simply the summation of a large number of terms in an infinite series. Let us assume that initially a photon is generated with a statistical weight of unity. If the intensity reflection coefficient for the n th reflection is R_n and if the gain factor for the path traveled between boundary collisions n and $n-1$ is G_n , then the total photon weight W_n which emerges from the rod may be expressed as

$$W_n = (1 - R_1) G_1 + R_1 G_1 G_2 (1 - R_2) + \dots + \prod_{i=1}^{n-1} (R_i G_i) G_n (1 - R_n) + \dots \quad (1)$$

The index h in Eq. (1) expresses the fact that we are dealing with a single photon history characterized by a specific frequency, starting position, direction, etc. We are interested not only in evaluating the series (1) but also in determining an average of this series over all possible sets of initial photon data.

The series (1), however, represents a considerable simplification of the actual calculation because gain and reflectivity depend on polarization. It is therefore necessary to express the radiation amplitude in terms of two polarization components and to calculate the gain and reflectivity for each component before determining the intensity gain and reflectivity.

One can obtain qualitative information about what to expect in such a calculation by making the further simplifying assumption that the effect of averaging in Eq. (1) is to substitute suitable averages $\langle R \rangle$, $\langle G \rangle$ and $\langle RG \rangle$ for R_n , G_n and $R_i G_i$ respectively. Then $\langle W \rangle$, the average of W_n , may be expressed as

$$\langle W \rangle = \frac{[1 - \langle R \rangle] \langle G \rangle}{1 - \langle RG \rangle} \quad (2)$$

For $G = 1$ this average is unity, as it should be. As $\langle RG \rangle$ approaches 1, on the other hand, $\langle W \rangle$ approaches infinity, signifying the onset of oscillation in what

we shall call a random mode.³ This clearly represents the limit to which such calculations can or need be carried. As the conditions for such an oscillation are approached in the calculation as actually performed, the fluctuations in w_h may become rather large introducing substantial statistical errors into the calculation of $\langle W \rangle$. An additional problem which arises is that the convergence of the series (1) becomes progressively slower. One can, however, determine quite accurately the conditions under which $\langle W \rangle$ goes to infinity. One simply determines the average photon weight $\langle w_0 \rangle$ remaining in the rod after a large number of collisions for photons having the line center frequency ν . In the notation of Eq. (1) this average would be

$$\langle w_0 \rangle = \left\langle \prod_{i=1}^N \bar{R}_i G_i^0 \right\rangle \quad (3)$$

where G_i^0 is evaluated at ν_0 . By interpolation or extrapolation it is possible to determine the conditions (population inversion) such that $\langle w_0 \rangle = 1$. This obviously defines the threshold of oscillation. With this datum together with values of $\langle W \rangle$ obtained for conditions below the threshold one can thus plot a smooth curve of $\langle W \rangle^{-1} = \tau^*$ as a function of population inversion.

A very general flow diagram of the calculation procedure for a single history is presented in Fig. 1. Pertinent remarks relating to the steps indicated are made in the following sections, but many of the operations involved are quite standard. For further details regarding these the reader is referred to a standard text on Monte Carlo methods.^{4,5}

A. Initial Data

Here it is necessary to generate a set of starting coordinates x, y, z , distributed uniformly throughout the volume of the rod (see Fig. 2); a set of starting direction cosines μ_x, μ_y, μ_z , distributed uniformly in solid angle; a polarization vector making an angle δ , uniformly distributed about some reference vector in a plane perpendicular to the direction vector $\underline{\mu}$; and a dimensionless

frequency ν^* distributed according to the Lorentzian probability density function

$$f(\nu^*) = \pi/(1 + \nu^{*2}) \quad (4)$$

In Eq. (4) ν^* represents $(\nu - \nu_0)/2\Delta\nu$ where $\Delta\nu$ is the line-width. When ν^* has been determined, an absorption coefficient is determined through

$$\alpha = \alpha(0)/(1 + \nu^{*2}) \quad (5)$$

where α signifies any of the absorption coefficients which occur in the calculation. All of the aforementioned quantities are evaluated in terms of random numbers, distributed uniformly in the interval 0 to 1, which are generated by the computing machine.^{4,5}

In practice a stratified sampling procedure is necessary for generating the data. This is because a relatively large contribution to $\langle W \rangle$ is made by photons with frequencies very near the line center and initial directions close to the rod axis. Yet such photons have a low probability of being generated.⁶ The purpose of a stratified sampling procedure is to insure that the largest number of histories is generated in data regions where fluctuations are likely to be largest. In the present example the range of variation of ν^* is therefore divided into three intervals: 0 to ν_1^* , ν_1^* to ν_2^* , and ν_2^* to ∞ , where ν_1^* and ν_2^* are such that $f(\nu_1^*) = 0.99\pi$ and $f(\nu_2^*) = 0.90\pi$. Similarly the range of variation of $\mu_z = \cos \theta$ is divided into two parts such that

$$0 \leq |\mu_z| \leq \cos [\tan^{-1}(R_0/L)] , \quad (6a)$$

$$\cos [\tan^{-1}(R_0/L)] \leq |\mu_z| \leq 1 \quad (6b)$$

where R_0 is the rod radius. These domains of variation of ν^* and μ_z define in all six different data groups. An equal number of histories is then generated within each data group. But the weight of the photon histories born within a given group

must be renormalized to the statistical weight of the group. The relative weight, for example, of the highest frequency group with initial directions along the cylinder axis (μ_z - interval (6b)) is

$$\pi \left\{ 1 - \cos [\tan^{-1}(R_0/L)] \right\} \tan^{-1} v_1^* \quad (7)$$

obtained by integrating Eq. (4) from 0 to v_1^* and multiplying the result by the length of the μ_z - interval (6b). The absolute weight of a given data group is the relative weight of the group divided by the sum of the relative weights of all the groups.

B. Distance to Boundary

In this portion of the calculation the distances are calculated from the point (x, y, z) in the direction $\underline{u}$ to the planes forming the rod ends and to the cylinder which forms the remaining rod boundary. The smallest positive distance is the distance s to boundary. This operation also determines whether the photon trajectory intersects with an end, where it is specularly reflected, or with the side where it is diffusely reflected (see Sections D and E). For details on how to organize such a calculation the reader is again referred to Ref. 4.

C. Calculation of Gain

The calculation of gain is complicated, as previously mentioned, by its dependence on polarization. The polarization vector which was initially assigned randomly must be resolved into two components, one in the principal plane formed by the optic axis and the photon direction; the other perpendicular to this plane. These components correspond respectively to the extraordinary and ordinary rays. Due to the nearness of the two refractive indices in ruby the directions of the two rays may be considered coincident.

In traveling a distance s to the surface of the rod the two rays are multiplied by amplitude gain factors

$$\begin{aligned} g_e &= \exp (n \alpha_e s/2) \\ g_o &= \exp (n \alpha_o s/2) \end{aligned} \quad (8)$$

where

$$\begin{aligned} \alpha_o &= \alpha_{\perp} \\ \alpha_e &= \alpha_{\perp} \cos^2 \theta + \alpha_{\parallel} \sin^2 \theta \end{aligned} \quad (9)$$

Here n is the fractional population inversion, $\alpha_{\perp}$ and $\alpha_{\parallel}$ are frequency dependent absorption coefficients in the two polarizations, measured for light propagating perpendicular to the optic axis, and θ is the angle between the photon direction $\underline{u}$ and the direction of the optic axis $\underline{i}$.

If we consider the ruby R_1 line then

$$n = 2n_2 - n_1 = n_{21} \quad (10)$$

where n_2 is the fraction of Cr ions in the $\bar{E}$ level and n_1 is the fraction of ground state ions. The factor 2 is the statistical weight factor g_1/g_2 for ruby. A similar relationship holds for the R_2 line:

$$n = 2n_3 - n_1 = n_{31}.$$

The further relations between n_1 , n_2 , and n_3

$$n_1 + n_2 + n_3 = 1 \quad (11a)$$

$$n_3/n_2 = e^{-\Delta E/kT} = B \quad (11b)$$

enable one to express n_1 and n_{31} in terms of n_{21}

$$n_1 = \left(\frac{2}{1+B} - n_{21} \right) / \left(1 + \frac{2}{1+B} \right) \quad (12a)$$

$$n_{31} = \frac{2(B-1)}{3+B} (1 + n_{21}) + n_{21} \quad (12b)$$

In Eq. (11b) T is the temperature and ΔE is the energy separation between the $2\bar{A}$ and $\bar{E}$ levels. Since the populations of the latter two levels are locked together, as shown in Eq. (11b), and since both levels have appreciable absorption coefficients, one must calculate $\langle W \rangle$ as a weighted average for both the R_1 and R_2 transitions. To do this one first evaluates $\langle W \rangle$ for the R_1 transition and a specific value of n_{21} calling the result $\langle W \rangle_{21}$. Next, using Eq. (12b) one determines n_{31} for the same value of n_{21} and evaluates $\langle W \rangle$ for the R_2 transition, calling the result $\langle W \rangle_{31}$. The desired value of $\langle W \rangle$ is obtained through

$$\langle W \rangle = f_{21} \langle W \rangle_{21} + f_{31} \langle W \rangle_{31} . \quad (13)$$

The ratio of the weighting factors f_{21} and f_{31} must involve the Boltzmann factor B and the ratio of the spontaneous emission rates for the two transitions. In the absence of experimental information on the latter this ratio is taken to be equal to the ratio of α_L for the two transitions. Hence

$$f_{31}/f_{21} = B \alpha_{L31}/\alpha_{L21} . \quad (14)$$

D. Calculation of Reflected Amplitude

The calculation of gain requires that the field amplitude be expressed in terms of components in and perpendicular to the principal plane formed by the photon direction and the optic axis. The calculation of the reflected amplitude, on the other hand, requires that the amplitude incident on the reflecting surface be expressed in terms of components in and perpendicular to a principal plane formed by the surface normal and the photon direction. If Λ_x , Λ_y , and $\Lambda_{x''}$, $\Lambda_{y''}$ are the amplitudes in the respective coordinate systems, the transformation between the two may be very simply expressed as

$$\begin{aligned} \Lambda_{x''} &= \Lambda_x (\underline{e}_x \cdot \underline{e}_{x''}) + \Lambda_y (\underline{e}_y \cdot \underline{e}_{x''}) \\ \Lambda_{y''} &= \Lambda_x (\underline{e}_x \cdot \underline{e}_{y''}) + \Lambda_y (\underline{e}_y \cdot \underline{e}_{y''}) \end{aligned} \quad (15)$$

where $\underline{e}_x$, $\underline{e}_y$, and $\underline{e}_{x''}$, $\underline{e}_{y''}$ are orthogonal unit vectors in the two coordinate systems. One possible choice for these unit vectors is

$$\begin{aligned}\underline{e}_{x'} &= \frac{\underline{\mu} \times \underline{\tau}}{|\underline{\mu} \times \underline{\tau}|}, & \underline{e}_{y'} &= \frac{\underline{\mu} \times \underline{\tau} \times \underline{\mu}}{|\underline{\mu} \times \underline{\tau}|} \\ \underline{e}_{x''} &= \frac{\underline{\mu} \times \underline{n}}{|\underline{\mu} \times \underline{n}|}, & \underline{e}_{y''} &= \frac{\underline{\mu} \times \underline{n} \times \underline{\mu}}{|\underline{\mu} \times \underline{n}|}\end{aligned}\quad (16)$$

where $\underline{n}$ is the unit vector normal to the end plane. To evaluate the transformation coefficients in Eq. (13) all of the vectors appearing in Eq. (16) must be expressed in a common coordinate system, e.g., in the coordinate system of Fig. 1. The amplitude reflection coefficients $R_{\perp}$ and $R_{\parallel}$ are then calculated in terms of $\underline{\mu} \cdot \underline{n}$ using the Fresnel relations.⁷ The reflected amplitudes are

$$\begin{aligned}A_{x''}^{(r)} &= R_{\perp} A_{x'} \\ A_{y''}^{(r)} &= R_{\parallel} A_{y'}\end{aligned}\quad (17)$$

The contribution to $\langle W \rangle$ from the reflection will be

$$(1 - R_{\perp}^2) A_{x'}^2 + (1 - R_{\parallel}^2) A_{y'}^2 \quad (18)$$

at this point in the calculation the direction vector is transformed according to the law of specular reflection and the amplitude is transformed back to the coordinate system determined by $\underline{\tau}$ and $\underline{\mu}$.

E. Calculation of Reflected Intensity

When a photon collides with the roughened side surface of the rod it is diffusely reflected, which is to say the reflected direction $\underline{\mu}'$ is randomly distributed in azimuth about the surface normal $\underline{n}$, while the probability density for the cosine of the angle θ_0 between the reflected direction and the surface normal varies linearly with the cosine:

$$f(\cos \theta_0) = 2 \cos \theta_0 \quad (19)$$

This is also known as Lambert's law.⁸ At the same time it is assumed that the randomly oriented surface segments combine to yield an effective intensity reflection coefficient R . The reflected intensity is

$$I^{(r)} = R (A_x^2 + A_y^2) \quad (20)$$

and the contribution to $\langle W \rangle$ is

$$(1 - R)(A_x^2 + A_y^2) . \quad (21)$$

In Eqs. (18) and (19) the coefficient R is evaluated numerically using

$$R = \int_0^{\pi/2} (R_{\parallel}^2 + R_{\perp}^2) 2 \cos \theta_0 \sin \theta_0 d\theta_0 . \quad (22)$$

Numerical Results

The numerical results presented here are based on ruby rods with 0.05 wt% Cr, Brewster ends, and a 90° orientation of the optic axis with respect to the cylinder axis. The values of $\alpha_{\perp}(\nu_0)$ at the line center were taken as 0.502 cm^{-1} and 0.345 cm^{-1} for the R_1 and R_2 lines respectively.⁹ The values of $\alpha_{\parallel}(\nu_0)$ for the same transitions were 0.004 cm^{-1} and 0.0010 cm^{-1} . The corresponding weighting factors were $f_{31} = 0.375$ and $f_{21} = 0.625$. Three hundred histories were followed in each of the six data groups until the photon weight had dropped below 0.001.

Plotted in Figs. 5-6 is τ^* against $n_{21} \alpha(\nu_0)$, or simply $n \alpha$. Shown are curves based on a weighted average of the R_1 and R_2 transitions as well as curves for the R_1 transition alone. The latter provide a lower limit to τ^* in cases where the values of $\alpha_{\perp}$ and $\alpha_{\parallel}$ differ from those used here. The value $\tau^* = 0$ corresponds to the threshold for oscillation of the random modes. Also plotted is $n_1/n_1(0)$, which is proportional to the rate of absorption of pump radiation normalized to 1 at threshold, for which $n = 0$. Thus it is possible to compare the relative importance of depletion of the ground state and super-radiance as mechanisms for limiting the attainable inversion.

One may, in fact, derive the following rough estimate for the steady state pump intensity $I(n)$ required to maintain a given value of n , in terms of the threshold intensity $I(0)$

$$I(n)/I(0) \cong (1 + n) [(n_1/n_1(0))\tau^*]^{-1} . \quad (23)$$

In deriving Eq. (23) it is assumed that the rate P of pump radiation absorption, given by

$$P = I n_1 \bar{\sigma}_p \quad (24)$$

where $\bar{\sigma}_p$ is the average absorption coefficient, is equal to the rate of spontaneous emission S , which at room temperature is approximately

$$S \cong \left(\frac{n+1}{2} \right) / \tau_{\text{eff}} . \quad (25)$$

Equation (23) is plotted in Fig. 7 for rod dimensions of 1/2 in. x 6 in. and a 1/4 x 6 in. respectively.

In Table I are shown the percentage standard deviations σ in $\langle W \rangle$ for the R_1 transition in the case of Fig. 3. This may be considered to be typical of the cases exhibited in the remaining figures.

TABLE I

Table of Standard Deviations for Case of
1/2 in. by 6 in. Ruby

n, α	$\langle W \rangle$	$\sigma(\%)$
0.05	1.07	0.225
0.10	1.16	0.411
0.15	1.31	0.793
0.20	1.47	1.14
0.25	1.77	1.57
0.30	2.43	2.35
0.35	8.08	40.1

In general for values of $\langle W \rangle$ greater than -3 the "series" form of the calculation is quite unreliable. However, the value of n for which $\tau^* = 0$ can be determined to an accuracy of $\sim 1\%$.

In comparing Figs. 3 and 4 for $1/2$ in. diameters and 6 in. and 9 in. lengths, it is noted that the curves are quite insensitive to length for a given diameter.¹⁰ This can be quite generally expected as long as non-random regenerative paths do not exist. At the same time in comparing Figs. 3 and 4 for a $1/2$ in. diameter with Fig. 6 for a $1/4$ in. diameter one notices a substantial variation in the effect of super-radiance with diameter. This is explained by the cosine distribution (Eq. 19) for the scattering of radiation from the sides of the rod, which favors reflections oriented nearest to the surface normal. Thus the photons follow a random regenerative path which does not deviate greatly from the rod diameter. Photons initially directed along the rod axis will be thrown into such a random mode following the first collision with an end.¹¹ Thus as far as super-radiance is concerned length is not a problem in the construction of Brewster angle rods. Flat ended rods in contrast require anti-reflection coated ends, even for relatively modest lengths, to prevent premature oscillation.

Figures 3 and 4 for $1/2$ in. rods indicate that super-radiance can have a considerable effect in limiting inversion. The effect of water immersion for the $1/2$ in. x 6 in. ruby in Fig. 5 is striking, however. The 1.33 value of the index of refraction of water is admittedly not a very good match for the 1.78 value of ruby, but it is sufficient to reduce R from 72% to 46%. Immersion presents a further advantage, namely, a suitable absorber can be added to the immersing liquid to prevent escaping photons from re-entering the rod. But this same effect might also be created by an appropriate placement of flash lamps. (The present calculations are of course made under the assumption that no photons re-enter.) In Fig. 6 for a $1/4$ in. diameter the dominant mechanism for limiting inversion is the depletion of ground state atoms. Hence in this case the effect of immersion would be marginal.

ACKNOWLEDGMENT

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References and Footnotes

1. F. J. McClung and R. W. Hellwarth, J. Appl. Phys. 33, 828 (1962).
2. This question has also been investigated under more simplified assumption by L. Tonks, J. Appl. Phys. 35, 1134 (1964).
3. The roughened sides of the rod insure the randomness of these modes.
4. E. D. Cashwell and C. J. Everett, "A Practical Manual of the Monte Carlo Method for Random Walk Problems", (Pergamon Press, New York, 1959).
5. Another useful reference dealing with a similar type of problem is D. H. Davis, J. Appl. Phys. 31, 1169 (1960).
6. As the condition for random mode oscillation is approached, the photons near the line center grow in importance. As it turns out, however, photons with initial directions along the rod axis are not of inordinate importance and could have been eliminated from the stratified sampling.
7. See for example, M. Born and E. Wolf, Principles of Optics, (Pergamon Press, New York, London, 1959), p. 39.
8. M. Born and E. Wolf, op. cit., p. 181.
9. J. E. Swain, unpublished spectrophotometer measurements.
10. An additional calculation for a 1/2 in. x 3 in. rod, not shown, differs imperceptibly from Figs. 3 and 4.
11. Such paths may have a substantial single pass gain but in a Brewster angle system these paths have a minute statistical weight, hence their low importance.

Figure Captions

- Fig. 1: Flow diagram of Monte Carlo calculation for a single photon history.
- Fig. 2: Geometrical configuration for Brewster angle laser rod.
- Fig. 3: Effective fluorescent life-times for 1/2" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends as a function of inversion times absorption coefficient.
- Fig. 4: Effective fluorescent life-times for 1/2" x 9" 90° ruby rod, 0.05 wt% doping, and Brewster ends.
- Fig. 5: Effective fluorescent life-times for water immersed 1/2" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends.
- Fig. 6: Effective fluorescent life-times for 1/4" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends.
- Fig. 7: Normalized pump intensity as a function of inversion times absorption for 1/2" x 6" and 1/4" x 6" rods.

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Fig. 1. Flow Diagram of Monte Carlo Calculation for a Single Photon History

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Fig. 2. Geometrical Configuration for Brewster Angle Laser Rod

Fig. 3. Effective fluorescent life-times for 1/2" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends as a function of inversion times absorption coefficient.

Fig. 4. Effective fluorescent life-times for 1/2" x 9" 90° ruby rod, 0.05 wt% doping, and Brewster ends.

Fig. 5. Effective fluorescent life-times for water immersed 1/2" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends.

Fig. 6. Effective fluorescent life-times for 1/4" x 6" 90° ruby rod, 0.05 wt% doping, and Brewster ends.

Fig. 7. Normalized pump intensity as a function of inversion times absorption for $\frac{1}{2}'' \times 6''$ and $\frac{1}{4}'' \times 6''$ rods.