Internal reflection from an amplifying layer

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The reflectance of a homogeneous amplifying layer between two transparent regions is determined theoretically with the help of the Fresnel formulas. The transparent region containing the incident beam has a higher refractive index, corresponding to an internal reflection configuration. In the limit of infinite layer thickness, the reflectance is a continuous monotonic function of incident angle, greater than unity for all angles. For certain finite values of layer thickness, the reflectance has a singular point. The results explain the reported observation of a reflectance of 1000 from an excited laser dye in contact with a quartz prism.

Total internal reflection can occur at the interface between two transparent regions when light is incident within the region having higher refractive index. If the region with lower refractive index has gain (i.e., a negative absorption coefficient), then it is possible for the reflectance to be greater than unity. This situation is referred to as enhanced internal reflection.

Romanov and Shakhidzhanov[1] (hereafter denoted R & S) present a treatment of this problem based on the Fresnel reflection coefficient for light incident at a single interface. Their result is that the reflectance is discontinuous at the critical angle of total internal reflection. They predict that enhanced internal reflection occurs only for angles greater than the critical angle with a maximum value occurring at the critical angle. Lebedev, Volkov, and Kogan[2] show that the largest possible value for the reflectance is 5.83, based on the theory of R & S. They also report the experimental observation of a reflectance of 1000. They attribute the large reflectance to a gradient in the index of refraction of the amplifying region, due either to a spacial variation of the temperature or the gain coefficient.

In Sec. I we treat the problem of the reflectance from a single homogeneous layer with gain. We demonstrate that in the limit as the thickness of the layer becomes infinite, the reflectance does not behave as predicted by R & S. Enhanced internal reflection is possible for angles of incidence less than the critical angle for both finite and infinite values of the layer thickness.

In Sec. II the application of the theoretical treatment to the experimental results of Lebedev *et al.* is discussed.

I. THEORY

Consider the case of a homogeneous amplifying layer with thickness d between two optically transparent regions with refractive indices n and 1 as shown in Fig. 1. The index of the layer is $1 - i\gamma$, where positive values of the gain coefficient γ correspond to amplification in the layer.[3]

Assume $n > 1$ and let a wave be incident from the high-index side. The numbered arrows in Fig. 1 represent the wave vectors of the various waves involved. The incident wave is labeled 1 and θ is the angle of incidence. Wave 2 is the refracted wave within the layer and ϕ is

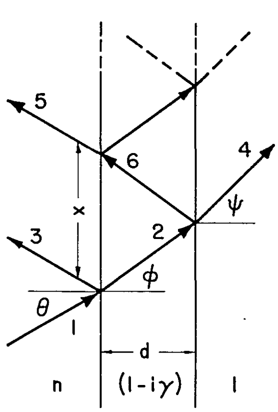

FIG. 1. Multiple reflections and propagation across an amplifying layer with thickness d. The numbered arrows represent the various waves and are defined in the text. The refractive indices are given at the bottom. The displacement between two successive reflected waves is denoted x.

the angle of refraction. The other waves in Fig. 1 are identified later as they are mentioned.

Before proceeding, it is useful to determine the amplitude of the first reflected wave, labeled 3 in Fig. 1. This corresponds to the reflectance of the first interface by itself and should give the result for $d=\infty$. For s polarization (electric field perpendicular to the plane of incidence) the amplitude reflectance given by the Fresnel formula is

$$r_1=\frac{n\cos\theta-(1-i\gamma)\cos\phi}{n\cos\theta+(1-i\gamma)\cos\phi}, \tag{1}$$

where

$$(1-i\gamma)\cos\phi=(1-\gamma^2-n^2\sin^2\theta-2i\gamma)^{1/2}. \tag{2}$$

With the convention that γ is positive, the solution of Eq. (2) may lie in either the second or fourth quadrant of the complex plane. Because of the relationship between $(1-i\gamma)\cos\phi$ and the wave vector of the refracted wave, the solution in the fourth quadrant corresponds to a refracted wave propagating away from the interface with an exponentially increasing amplitude. The root in the second quadrant corresponds to a refracted wave propagating toward the interface having an amplitude which decreases exponentially with distance from the interface. We refer to these two waves as the growing and decaying waves, respectively.

Figure 2 is a graph of the corresponding reflectance $|r_1|^2$ versus angle of incidence for these two solutions. The curves are labeled D and G corresponding to the cases of the decaying and growing refracted waves, respectively. For this graph we have used $n=1.0001523$ and $\gamma=0.00015$. These values are chosen to correspond to the experiments done by Lebedev *et al.*[2,4] discussed in Sec. II. Also, the choice $\gamma\simeq n-1$ gives the optimum reflectance according to R & S.

When these two solutions to the problem of reflectance from a single interface occur, the choice is usually made in favor of the decaying wave to avoid an infinite amplitude at infinite distance from the interface. However, in this case, where the second region has a positive gain coefficient, a growing wave seems appropriate, especially since near normal incidence the decaying wave results in a reflectance of approximately 10^8 for the parameters we have chosen.

This discussion of the single interface case parallels that of R & S. They resolve the above question by using the growing wave for angles of incidence less than the critical angle and the decaying wave for angles greater than the critical angle. The resulting reflectance is a discontinuous function of angle, corresponding to the solid curves in Fig. 2. They claim that this form of the solution results from treating the case of a layer with gain which has finite thickness and allowing the thickness to become infinite.

The reflectance of a layer with finite thickness is well known[5] and may be written in the form

$$r=r_1+(1-r_1^2)r_2e^{2i\delta}\sum_{m=0}^{\infty}(-v)^m, \tag{3}$$

where r_1 is the amplitude reflectance at the first interface and is given by Eqs. (1) and (2) above. Similarly, r_2 is the reflectance at the second interface and is

$$r_2=\frac{(1-i\gamma)\cos\phi-\cos\psi}{(1-i\gamma)\cos\phi+\cos\psi}. \tag{4}$$

The angle ψ is the angle of refraction of the wave transmitted by the layer (labeled 4 in Fig. 1) and is given by

$$\cos\psi=(1-n^2\sin^2\theta)^{1/2}. \tag{5}$$

The parameter δ is given by

$$\delta=(2\pi/\lambda)d(1-i\gamma)\cos\phi, \tag{6}$$

where λ is the wavelength of the light in the low-index region. Further, we define v as

$$v=r_1r_2e^{2i\delta}. \tag{7}$$

Equation (3) is derived by summing the series of reflected waves resulting from the multiple reflections of the wave within the layer. (The first of these waves is labeled 5 in Fig. 1.) The parameter v represents the relative amplitude and phase of two successive waves in this series.

Assuming for the moment that the series in Eq. (3) converges, we can rewrite the result in the form

$$r=(r_1+v/r_1)/(1+v). \tag{8}$$

The sign of the root in Eq. (5) can be chosen in the usual manner because the transmitted wave is in a transparent region. The critical angle θ_c for the system is

$$\theta_c=\arcsin(1/n), \tag{9}$$

and $\cos\psi$ is positive real for $\theta<\theta_c$, corresponding to a transmitted wave which is a homogeneous plane wave propagating away from the layer. For the case $\theta>\theta_c$, $\cos\psi$ is positive imaginary, corresponding to a decaying or evanescent transmitted wave.

It should be pointed out that an absorbing medium is used for the transmitted wave by R & S. This is an alternate method of obtaining the correct signs for $\cos\psi$. The choice of a transparent region simplifies the computation and does not affect the result in the limit as the thickness of the layer becomes large.

To compute r using the above equations, it is necessary to select one or the other of the roots of Eq. (2) to use for the calculations. However, as might be ex-

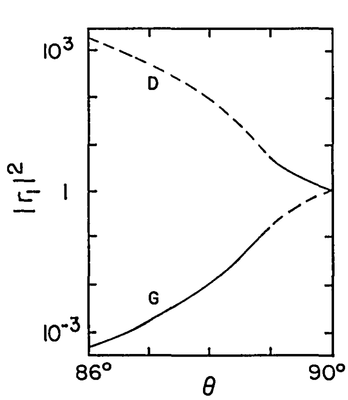

FIG. 2. A graph of reflectance versus incident angle for the two possible roots of Eq. (2) with $\theta_c=89°$ and $\gamma=0.00015$. The solid curve corresponds to the theory of R & S.

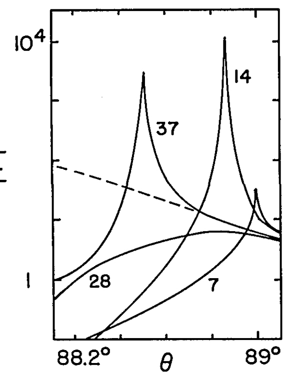

FIG. 3. A graph of reflectance versus incident angle for various layer thicknesses. The number by each curve is the corresponding value for d/λ. The dashed curve is for $d=\infty$. For all curves $\theta_c = 89°$ and $\gamma = 0.00015$.

pected, the result is independent of which root is chosen.

A graph of the reflectance $|r|^2$ versus the angle of incidence is presented in Fig. 3 for several values of the layer thickness d. The refractive index and gain coefficient are the same as in Fig. 2. The critical angle for this case is 89°. The curves for $d/\lambda = 14$ and 37 have extremely sharp peaks. These values of d are close to thicknesses for which the reflectance curve has a singular point. The location of the singular points and the corresponding values of the thickness are determined below. For values of d/λ between 14 and 37, the peak is not as pronounced and, as can be seen from Fig. 3, for $d=28$ wavelengths there is no peak at all.

There remains the question as to the convergence of the series in Eq. (3). The necessary requirement is that $|v| < 1$. There are two possible values for v, depending on which root is chosen for $(1-i\gamma)\cos\phi$. These two values are reciprocals of each other and thus it is possible to choose the root so that the series converges. The choice of root also determines whether the refracted wave is the growing or decaying wave since $(1-i\gamma)\cos\phi$ is proportional to the component of the wave vector in the direction perpendicular to the interface.

To aid with the discussion, we define an angle θ_s for a particular layer thickness d as the angle of incidence for which

$$|v| = 1. \tag{10}$$

Since $|v|$ is a monotonic function of angle, this equation is satisfied by at most one angle.

For $\theta < \theta_s$, the fourth-quadrant root of Eq. (2) results in $|v| < 1$. In this case, the refracted wave corresponds to the growing wave. Conversely, for $\theta > \theta_s$, the second-quadrant root is required for $|v| < 1$, and the refracted wave is the decaying wave.

The identification of the refracted wave as the growing or decaying wave is perhaps a semantic point since both waves are present in the layer (e.g., waves 2 and 6 in Fig. 1). In fact the whole question of convergence can be side-stepped by solving Maxwell's equations directly with appropriate boundary conditions. The result is Eq. (8), which is independent of the root chosen for Eq. (2). It is still true that the total wave within the layer is predominantly growing for θ sufficiently less than θ_s and decaying for θ sufficiently greater than θ_s.

This result is similar in some ways to the prediction made by R & S: Near normal incidence, the refracted wave is growing, and near grazing incidence, it is decaying. However, we see that the form of the refracted wave does not switch at the critical angle as they predict, but rather at θ_s as defined above.

The value of θ_s depends on the thickness of the layer, and a graph of θ_s versus d/λ is presented in Fig. 4. The refractive index and gain coefficient are the same as for the other figures. The values are obtained by solving Eq. (10) for d/λ using Eqs. (6) and (7). The result is

$$\frac{d}{\lambda} = \frac{\ln|r_1| + \ln|r_2|}{4\pi\,\mathrm{Im}[(1-i\gamma)\cos\phi]}, \tag{11}$$

where Im [ ] signifies "the imaginary part of." Equations (1), (2), (4), and (5) are used to express the right hand side in terms of θ which in this case is equal to θ_s.

Notice that θ_s decreases monotonically as d increases. There is a value of d for which $\theta_s = 0$. For this value of d, and for all larger values, the refracted wave will be the decaying wave for all angles of incidence. In the limit as d tends to infinity, the total wave in the layer has an amplitude which decays exponentially with distance from the first interface. Referring to Fig. 2, we conclude that the curve labeled D is the correct limit for the case $d \to \infty$.

If γ is now allowed to approach 0, the resulting curve is not the expected reflectance at an interface between regions with indices n and 1, but rather its reciprocal. To obtain consistent results, one must take the limit as $\gamma \to 0$ with d fixed at a finite value and then let $d \to \infty$.

Of particular interest are the singular points of Eq. (8). These occur when $v = -1$. Referring to Eq. (10), we see that a singularity occurs at θ_s when d is chosen such that arg(v) is an odd multiple of π. Several of these values of d are listed in Table I to several significant figures. The values are numbered in order of increasing d. This number m is also the order number of interference for the reflected waves. It corresponds to the number of wavelengths of optical path difference between two successive reflected waves.

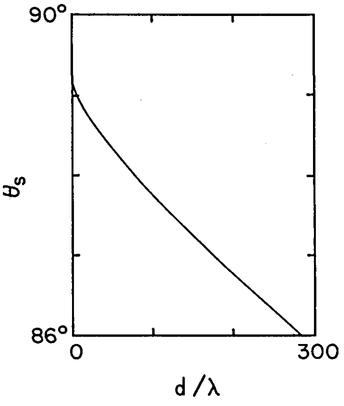

FIG. 4. A graph of θ_s versus layer thickness with $\theta_c = 89°$ and $\gamma = 0.00015$.

TABLE I. Singular points of the reflectance for $\theta_c = 89°$ and $\gamma = 0.00015$.

m	d/λ	θ_s (deg)
1	13.942	88.868
2	36.798	88.508
3	52.002	88.312
4	64.742	88.158
5	76.042	88.028
6	86.353	87.912
7	95.922	87.807
8	104.906	87.710
9	113.413	87.620
10	121.519	87.536

II. APPLICATION AND DISCUSSION

The foregoing analysis predicts that enhanced internal reflection is possible from an amplifying region. However, certain implicit assumptions limit its application to practical situations. For example, the assumption that γ is uniform over a well defined layer and zero beyond is difficult to satisfy experimentally. Also, edge effects are not taken into account. These limitations can be understood more clearly in relation to the experiment reported by Lebedev *et al.*

The experiment[2,4] involves a $10^{-3}M$ solution of rhodamine 6G in contact with a quartz prism. The critical angle is approximately 89°. The dye solution is excited by a frequency-doubled neodymium laser beam focused to a line at the surface of the prism. The excited region of the dye solution constitutes the layer with gain. A reflectance of 1000 is measured for light incident on the interface at the position of the excited region. The angular width of the reflected beam is roughly 10^{-2} rad.

In order to discuss these results, an estimate of γ for the experiment is needed. An upper limit is obtained by assuming complete population inversion and ignoring quenching by the triplet state. In this case, the gain coefficient may be written[6]

$$\gamma = \lambda\sigma_e N/4\pi. \tag{12}$$

The wavelength λ is taken to be 0.4×10^{-6} for yellow light inside a solution of refractive index 1.49 (quartz). The emission cross section of the dye σ_e is 9×10^{-21} m^2 from Ref. 6. The molecular concentration N is $6\times10^{23}/$m^3 for a $10^{-3}M$ solution. The resulting upper limit on γ is 0.00017. This is close to the value used for the figures in the previous section.

Because the dye solution in the experiment is pumped optically, the population inversion, and thus the value of γ, decreases with distance from the interface. Thus γ is not uniform over a finite layer. If the pump beam does not saturate the absorption of the dye, the population inversion decreases exponentially with distance from the interface. The lower limit d_e for the $1/e$ length of this exponential is given by

$$d_e = 1/\sigma_p N. \tag{13}$$

Here σ_p is the absorption cross section of the dye, having approximately the value 1.6×10^{-20} m^2 found in Ref. 6, and N is the molecular concentration as above. The resulting value for d_e is 100 μm. In terms of the wavelength of yellow light used above, this becomes $d_e/\lambda \simeq 250$.

The assumptions leading to these values of γ and d_e cannot both obtain simultaneously. However, in the absence of more information regarding the size, shape, and power of the pump beam used in the experiment, a more detailed analysis seems unwarranted. Thus the experimental situation is modeled by a homogeneous layer with $\gamma = 0.00015$ and d/λ approximately 250.

Figure 5 is a graph of $|r|^2$ vs θ as given by the square of Eq. (8) for $d/\lambda = 257.178$ and with $\gamma = 0.00015$ and $\theta_c = 89°$. For this value of d, θ_s is 86.232° and there is a singularity at this angle with $m = 33$.

Additional secondary maxima are present in Fig. 5. They result from constructive interference of reflected waves arising from multiple reflections of the beam within the region with gain. They occur at angles for which v is a negative real number close to -1. The minima correspond to destructive interference and occur when v is positive real on the order of $+1$.

The value of d for Fig. 5 is chosen specifically to illustrate a curve with a singularity. However, for nearby values of d the general features of the reflectance curves are similar except that the singularity is absent. For example, for $d/\lambda = 254.787$, v is $+1$ at $\theta_s = 86.25°$. This corresponds to a local minimum of the reflectance at this angle. This minimum is flanked on either side by peaks with maximum values on the order of 10^5, and subsidiary maxima of similar magnitudes and spacing as shown in Fig. 5.

The curve for the case $d = \infty$ is also included in Fig. 5 as a dashed line. For angles larger than 87°, the two curves are nearly identical. For angles between 87° and θ_s, it appears that the curve for finite d oscillates about the curve for $d = \infty$. For angles less than θ_s, the reflectance for finite d drops to very small values.

Thus we infer a further interpretation of the relationship between d and θ_s. For angles of incidence sufficiently larger than θ_s, the refracted wave is attenuated sufficiently at a depth d from the first interface that multiple reflections do not contribute to the reflectance.

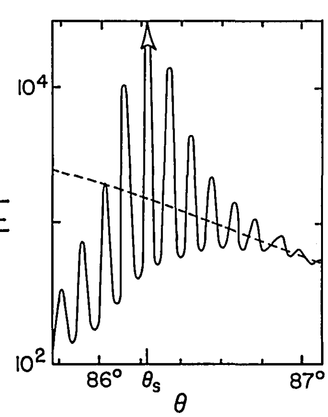

FIG. 5. A graph of reflectance versus incident angle. The solid curve is for $d/\lambda = 257.178$. The dashed curve is for $d = \infty$. For both curves $\theta_c = 89°$ and $\gamma = 0.00015$.

For these angles, the reflectance is determined by the first reflected wave and thus corresponds to the case with $d = \infty$.

For angles sufficiently smaller than θ_s, the (growing) refracted wave is not amplified sufficiently in a distance d to overcome the low reflectance at the interfaces. In this case the reflectance of the system drops to a low value typical of an interface between the two transparent regions without the layer with gain.

For angles in the neighborhood of θ_s, the additional reflected waves arising from multiple reflections are significant. They give rise to the oscillations in the reflectance curve as discussed above.

In the discussion leading to Eq. (8), an infinite number of reflected waves are assumed to be present. Each reflected wave is displaced from the preceding one by a distance x along the interface as shown in Fig. 1. Thus the region with gain must have infinite length along the interface.

In the experiment the length of the region with gain corresponds to the width of the pump laser beam. Thus the pumped region does not have infinite length and the multiply reflected wave leaves this region after a finite number of reflections. This situation is sometimes referred to as beam walkoff.

Without more accurate information regarding γ, d, and θ_c, it is difficult to obtain a meaningful value for x appropriate to the experiment. Also, the size of the pump beam is not reported so the length of the region with gain is not known. Thus it is difficult to estimate the number of reflected waves present in the experiment. However, the description of the reflected beam seems to imply that multiple reflections do not contribute to the large reflectance observed.

Two or three reflected waves should be sufficient to produce a visible modulation of the reflected beam in the neighborhood of θ_s. The angular separation of the maxima as shown in Fig. 5 is 0.1° or about 0.002 rad. This is large enough to be observable. However, the reflected beam is described as a "spot" in Ref. 2. This indicates that the secondary maxima were not observed experimentally.

In addition, the angular widths of the singular peak and the secondary maxima in Fig. 5 are less than 0.001 rad. This is an order of magnitude less than the beam divergence as given in Ref. 4. In fact, the reported beam divergence of 0.01 rad corresponds to the width of the broad features of Fig. 5 without the peaks, i.e., to a smoothed version of the curve. In particular the curve for $d = \infty$, taken only over the range $\theta_s < \theta < 90^\circ$, has a width at half-maximum of 0.5°, or nearly 0.01 rad.

Thus it appears that multiple reflections do not play a role, and that the large reflectance observed constitutes a direct measurement of $|r_1|^2$ at an angle of incidence less than the critical angle.

It should be pointed out that the curves presented in Figs. 2–5 are all for the same values of γ and θ_c and that a change in either of these values produces a marked change in the curves. In particular, the value of γ appropriate to the experiment may be lower than 0.00015. This lower value of γ results in a value of θ_s closer to θ_c with a corresponding narrowing of the peaks.

In addition, the fact that the layer with gain is not really homogeneous should be taken into account. We are presently studying the reflectance from a region with an exponentially decreasing gain coefficient. Preliminary results indicate that enhanced internal reflection is possible for this case. The results are similar to those presented in Figs. 3 and 5.

III. SUMMARY

In Sec. I the reflectance of a homogeneous layer with gain is determined. As the thickness of the layer tends to infinity, the reflectance is found to be a continuous function of angle. In addition, large values of reflectance are possible for certain finite as well as infinite values of the layer thickness. We conclude that the theory of R & S is incorrect.

In Sec. II we discuss the reflectance from a layer with gain coefficient and thickness appropriate to the experiment of Lebedev *et al.* The large reflectance which they report seems to correspond to a smoothed version of the calculated reflectance curve, indicating that multiple reflections do not play a significant role. The experiment confirms our result in Sec. I that large values of the reflectance are possible.

The discussion in Sec. II suggests further work in several areas: (1) experimental verification of the structure in the reflectance curves caused by interference of multiply reflected waves, (2) proper theoretical treatment of the case for a finite number of multiple reflections corresponding to a pumped region of finite length, and (3) careful analysis of cases in which the gain coefficient is an exponential or other function of distance from the interface.

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[2]S. A. Lebedev, V. M. Volkov, and B. Ya. Kogan, Opt. Spectrosc. 35, 565 (1973).

[3]The sign convention regarding γ assumes waves of the form $\exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$.

[4]B. Ya. Kogan, V. M. Volkov, and S. A. Lebedev, ZhETF Pis. Red. 16, 144–147 (1972) [JETP Lett. 16, 100 (1972)].

[5]O. S. Heavens, *Optical Properties of Thin Solid Films* (Butterworths, London, 1955), p. 57. Note: The sign of δ differs from ours due to the different form of the waves used. See Ref. 3 above.

[6]S. A. Tuccio and F. C. Strome, Jr., Appl. Opt. 11, 64 (1972).