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DETERMINATION OF DYE LASER INTRINSIC GAIN CHARACTERISTICS USING DIRECTLY MEASURABLE SPECTROSCOPIC QUANTITIES

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تعيين منحنيات خصائص الكسب لليزر الصبغى
بدلالة كميات طيفيه سهلة القياس

الخلاصه - يقدم هذا البحث طريقه لحساب منحنيات الكسب الذاتى لجهاز الليزر الصبغى بدلالة المقاطع العرضية للانبعاث والامتصاص للمادة النشطة .
ويرتبط مقطع الامتصاص مباشرة بمنحنيات الطيف الامتصاصى السهلة القياس . أما مقطع الانبعاث الفلورى فقد سبق اشتقاقه بدلالة الخواص الطيفيه لطيفى الامتصاص والانبعاث مع استخدام علاقة التماثل المرئى بينهما ولكن عند تطبيق هذه الصيغه المبسطه نجد أن ذيل منحنى طيف الامتصاص عند الترددات المنخفضه فى المحاليل المخففه ($\sim$ ١٠ $^{-٥}$ جزئ جرامى لكل لتر) وهى التركيزات المستخدمه فى أجهزة الليزر الصبغى - يتضاءل بسرعه كلما قل التردد .

وقد استخدمنا قانون بير - لامبرت للتغلب على هذه المشكله . وتم رسم منحنيات خصائص الكسب الذاتى مع التردد لقيم مختلفه للتعداد النسبى للحالة المثارة لصبغة الرودامين " ب " وتتفق منحنيات خصائص الكسب الذاتى للصبغة المذكورة بدرجة كبيره مع النتائج العمليه المنشورة والطريقه المذكورة فى البحث قابلة للتطبيق على الصبغات العاليه الفلوريه المستخدمة فى أنظمة الليزر ويمكن استخدام خصائص الكسب لدراسة أداء جهاز الليزر واعتماده على عديد من المتغيرات الخاصه بالمادة النشطة .

ABSTRACT

The dye laser analysis depends on the gain equation that contains cross-sections of the relevant emission and absorption molecular transitions involved in the lasing process. The absorption cross-section $\sigma_s(v)$ is directly related to the absorbance curves of the dye.

The emission cross-section $\sigma_{em}(v)$ has been derived in terms of directly measurable spectroscopic quantities. The low frequency tail of the absorption curves is minutely diminishing for very dilute solutions ($\cong 10^{-5}$ mole/lit) usually used in laser system. This low frequency tail overlaps the operating frequency band of the laser. The linear dependence of the dye absorption on the dye

concentration (Beer- Lambert law), is used to solve the low frequency tail problem. The intrinsic gain characteristics are calculated for the dye Rhodamine "B" and the results are in good agreement with the reported experimental results.

INTRODUCTION

The large number of highly fluorescent dye molecules represent a vast reservoir of potential laser sources. The vibrational and rotational states are represented as fine and hyperfine structure within each electronic state in both singlet and triplet energy ladder, Jablonski energy level diagram is shown in Fig.1.

The dye laser operation is based on the fluorescence transition that competes with the following processes:
1) Internal conversion as the nonradiative decay of the first excited singlet state S1 to the ground state So.
2) Intersystem crossing of the electrons from S1 to the first excited triplet state T1 whose lifetime is long enough to trap excited electrons out from the lasing channel.

The performance of the dye lasers as tunable coherent sources [1,2,3,4], and as wide band optical amplifiers [5,6,7], depends on the intrinsic gain characteristics expressed in terms of molecular transition cross-sections involved in the laser action [8,9,10].

In a previous work [10], we have derived a simplified expression of the intrinsic gain coefficient in terms of the absorbance and fluorescence curves. But it was not possible to use this derived gain expression for gain calculation because the low frequency tail of the absorption curve, which is the most important part of the whole absorption curve, was not available. However, the problem of low frequency tail can be solved on the basis of Beer-Lambert Law, that assumes linear dependence of the dye absorbance on its concentration. In this work, we have reviewed our previous approach, and extended it to calculate the intrinsic gain characteristics of the laser dyes.

Intrinsic Gain Analysis

The intrinsic gain coefficientis the net increase of photons number in certain mode, per unit length per photon, as a result of molecular transitions involved in the laser action. The main molecular processes involved in, or competing with, the laser action are shown in Jablonski energy level diagram in Fig. 1.

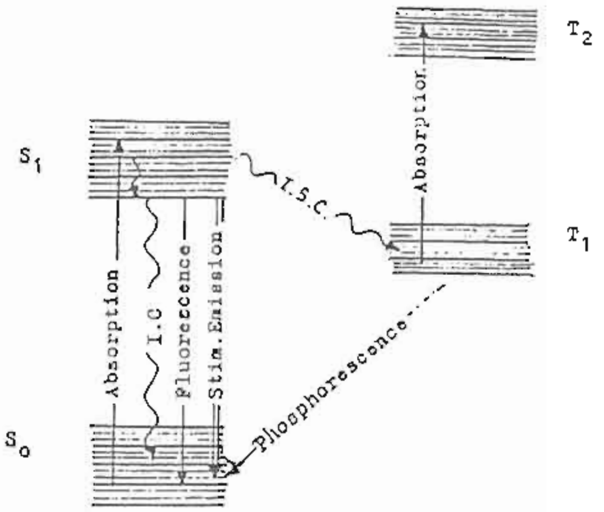

Fig.(1): Jablonski Energy Level Diagram of a Typical Organic Dye Molecule.

The intrinsic gain of the laser dye can be expressed in terms of the relevant absorption and emission cross-section, defined as the relative change in photon number per unit length per molecule in the initial state of the transition:

$$G(\bar{v},n,l).l_1 = \sigma_e(\bar{v}).N_1.l_1 - \sum_i \sigma_i(\bar{v}).N_i.l_i \qquad (1)$$

where:

$\sigma_e(\bar{v})$ = stimulated emission cross-section,

$\sigma_i(\bar{v})$ = absorption cross-section of the i^{th} lossy transition.

N_1 = population density of first excited singlet state

$\bar{v}$ = wave number

$l_i = l_1$ or l_o

l_o = total optical length of the dye.

l = fractional pumped length = l_1 / l_o

n = fractional excited state population = N_1 / N_o

The dominant lossy transitions are the single state transition ($S_o \dashrightarrow S_1$) and, to less extent, the intersystem crossing (I.S.C) with subsequent triplet absorption ($T_1 \dashrightarrow T_2$). Then the gian equation will be:

$$G(\bar{v},n,l) = \sigma_e(\bar{v}).N_1. - \sigma_s(\bar{v}).N_o/l - \sigma_T(\bar{v}).N_T. \qquad (2)$$

where $\sigma_s(\bar{v})$ is the absorption cross-section of first singlet state. However, for highly fluorescent dyes having quantum yield approaching unity, the effect of intersystem crossing is negligible. For other dyes, the quantum yield can be increased by lowering temperature, using viscous solvent, or triplet state quenching by paramagnetic additives [11], for this case the triplet state losses will be neglected and we get:

$$G(\bar{v},n,l) = \sigma_e(\bar{v}).n - \sigma_s(\bar{v})./\ l. \qquad (3)$$

This expression of intrinsic gain coefficient is to be formulated in terms of readily measurable absorbance and fluorescence of the laser dye.

A)- Singlet Absorption Cross-section $\sigma_s(\bar{v})$

From $\sigma_s(\bar{v})$ definition we get:

$$I = I_o\, e^{-(\sigma_s(\bar{v}).N_o.d)}. \qquad (4)$$

But the absorbance $Ab(\bar{v}) = \log_{10}(I_o/I)$

$$= \epsilon_s(\bar{v}).m.d \qquad (5)$$

As I_o and I are the incident and transmitted light intensity, and the absorbance $Ab(\bar{v})$ is also called the optical density

$\epsilon_s(\bar{v})$ = decadic molar extinction coefficient,
m = molar concentration of the dye solution,.
d = thickness of the dye solution,
$No = m.N_a / 1000$,
N_a = Avogadro's number.

Then we get: $\sigma_s(\bar{v}) = \ln 10.Ab(\bar{v})/(N_o.d)$ (6)

B)-Stimulated Emission Cross-section $\sigma_e(\bar{v})$:

The relative increase in stimulated photons number per unit length is:

$$\sigma_e(\bar{v}).N_1 = B_{1-o}.hv . e(\bar{v}) . N_1 .\xi/c$$

with B_{1-o} = Einstein coefficient of stimulated emission.

$e(\bar{v})$ = fluorescence shape function.
ξ = refractive index of the solvent.
h = planck's constant.
c = speed of light in vacuum.

$$\text{Then } \sigma_e(\bar{v}) = B_{1-o} .hv. e(\bar{v}).\xi/c \qquad (7)$$

$$\text{with } \int_o^\infty e(\bar{v})\, d\bar{v} = \phi \qquad (8)$$

as ϕ is the fluorescence quantum yield (≤ 1). Using Einstein relations between B_{1-o}, B_{o-1} and A for transitions between states of the same degeneracy:

$$B_{1-o} = B_{o-1} \text{ and } A = B_{1-o} . n(v) . hv \qquad (9)$$

with $n(v)$ = mode density per unit frequency = $8\pi v^2\xi^3/c^3$ (10)

Then $\sigma_e(\bar{v})$ can be expressed in fluorescence parameters as:

$$\sigma_e(\bar{v}) = (A/n(v)) . e(\bar{v})\ \xi/c \qquad (11)$$

An explicit relation between the spontaneous emission rate A and the singlet state absorption cross-section can be derived as follows.

The decrease in the radiation intensity after passing through a dye solution of concentration N_o (cm^{-3}) and thickness d(cm) is ΔI ($\bar{V}$) and equal to $I(\bar{v}).\sigma_s(\bar{v}).N_o.d$.

But from the defintion of B_{o-1} we have also for $\Delta I(\bar{v})$:

$$\Delta I(\bar{v}) = B_{o-1}.\rho(v).\ hv.\ N_o.d$$

with $\rho(v)$ = radiation energy density per unit frequency
= $I(\bar{v}).(\xi/c)/\Delta v$

The spontaneous emission rate $A(\bar{v})$ between single upper and single lower state is:

$$A(\bar{v}) = n(v).\sigma_s(\bar{v}).\Delta\bar{v}.c/\xi \qquad (12)$$

For total value of spontaneous emission rate ; eq(12) should be integrated over the whole lower states i.e.

$$A = (c/\xi)\int_0^\infty n(v).\sigma_s(\bar{v}).d\bar{v} = (8\pi\xi\ /c^2\ )^2\int_0^\infty v^2\sigma_s(\bar{v})d\bar{v} \qquad (13)$$

Using Förester approximation [12], to substitute for v its mean value v_e ;we get:

$$A = (8\pi v_e^2\xi^2/c^2)\int_0^\infty \sigma_s(\bar{v})d\bar{v} \qquad (14)$$

Equation (14) represents the fluorescence transition rate "A" in terms of singlet absorption cross-section σ_s that is directly related to the measurable dye absorbance Ab(v). The fluorescence line shape function e(v), can be related to the measured fluorescence intensity $I_f(v)$ as follows: The fluorescence intensity $I_f(v)$ of a continuous source is usually measured as power per unit steradian per unit frequency interval [13], and can be expressed as:

$$I_f(v) = K.A.h.v.e(v).N_1 d \qquad (15)$$

with K is an instrumental constant that depends on the following factors:

1. Excitation source intensity
2. Slit width
3. Absorption solid angle
4. Fluorescence solid angle,
5. Spectral ratio between source bandwidth and dye absorption bandwidth.

These factors restrict the measured fluorescence intensity use for only comparative study, For any quantitative analysis, the measured fluorescence intensity should be normalized for each dye solution. For this reason, the normalized fluorescence intensity $I_f(v)/I_{max}$ should be used in our analysis.

The normalized condition (8) can be used with eq.(15) to get:

$$\int_0^\infty (I_f(v)/v)\ dv = K.A.N_1.d.h.\phi \qquad (16)$$

and now e(v) can be expressed as:

$$e(v) = I_f(v) . \phi \, / \left\{ v \int_0^\infty (I_f(v)/v) \, dv \right\}$$

making the same approximation used in eq.(14); we get:

$$e(v) = I_f(v) \, . \, \phi(v_e/v) / \int_0^\infty I_f(v) \, dv \qquad (17)$$

The two integrals in eq.(14) and (17) represent the two areas under the two functions $\sigma_a(v)$ and $I_f(v)$ that have mirror-image symmetry about the frequency of pure electronic transition [13]. This mirror image symmetry is based on the quantum mechanical assumption that the electric dipole transition matrix element is the same for both upward and downward transitions [14]. This mirror image symmetry between $\epsilon_s(v)$ and $I_f(v)$ is shown in Fig.2. Using this symmetry; the two integrals in eqs.(14) and (17) can be written as:

$$\int \sigma_s(\bar{v}) \, .d\bar{v} = \sigma_s(\bar{v}) . \, \Delta\bar{v}_s . K_1 \qquad (18)$$

$$\int I_f(\bar{v}) \, d\bar{v} = I_{f\ max} \, . \Delta\bar{v}_e . K_1 \qquad (19)$$

Where the proportionality factor K_1 has the same value for both integrals because of the mirror image symmetry between the absorption and the floursence curves .

Where : $\Delta \bar{v}_s$ and $\Delta \bar{v}_e$ are the bandwidth of absorption and fluorescence spectra respectively. Using eqs.(18) and (19) together with (14) and (17) and substitute in eq.(10), we get the stimulated emission cross-section $\sigma_e(v)$ as:

$$\sigma_e(\bar{v}) = \sigma_{s\ max} . \, \phi(v_e/v)^3 . \, (\Delta v_s/\Delta v_e).(I_f(v)/I_{f\ max}) \qquad (20)$$

For most dyes the two band widths Δv_s and Δv_e are proved to be equal within the limits of the experimental errors [12]. This equality confirms the symmetry relation approximation used in eqs.(18) and (19). Putting $\Delta v_s = \Delta v_e$ in eq.(2), we get:

$$\sigma_e(\tilde{v}) = \sigma_{s\ max} . \phi(v_e/v)^3 \, (I_f(v)/I_{f\ max}) \qquad (21)$$

This normalized form of stimulated emission cross-section minimizes the effects of experimental errors related to the excitation sources , instrumental set-up, and detecting and recording devices. Some authors estimate these experimental errors to be ± 10% [12] , and even more (± 25 %), due to several distortion factors[15].The fluorescence intensity is usually measured in a relative manner with arbitrary scale. Therfore the importance of normalized fluorescence intensity in the expression of $\sigma_e(\bar{v})$, is confirmed.

C)- Intrinsic Gain Expression:

The absorption cross-section $\sigma_a(\bar{v})$ and the stimulated emission cross-section $\sigma_e(\bar{v})$, in eqs.(6) and (21) are expressed in terms of directly measurable quantities . Using eq.(6) and eq. (21) in eq.(3) we get :

$$G(\bar{v},n,l) = \ln 10 \left\{ Ab_{max} \phi . (v_e/v)^3 . (I_f/I_{fmax}) . (n/d) - Ab/ld \right\} \quad (22)$$

This equation represents the intrinsic gain in terms of the relative fluorescence intensity and the absorbence of the laser dye.

D) Intrinsic Gain Calculation:

To apply eq.(22) for gain calculation,we have noticed that the most important part of the absorption curve in the low frequency side(low frequency tail) is diminishingly small and is normally beyond the measurable range for low dye concentrations ($\approx 10^{-5}$mole/l). These low concentrations are usually used in dye laser systems.

In a previous work [10],we used analytic fitting function to represent the low frequency tail of absorption curve for low frequency concentrations.

The problem of low frequency tail of the absorption curves are solved here by applying Beer-Lambert Law of the linear dependence of the dye absorption on the dye concentration in the dilute solutions.

Equation (22) describes the frequency dependence of the intrinsic gain coefficient $G(\bar{v},n,l)$ and its variation with the excited state fractional population n,the pumped fractional length l,and the fluorescence quantum yield ϕ as parameters.

We have used eq.(22), together with Beer-Lambert law, to calculate the intrinsic gain characteristics for the laser dye Rhodamine "B" whose fluorescence and absorption curves are shown in Fig.2 [16,17]; in which the absorption result is plotted as molar extinction coefficient $\epsilon_g(v)$.

To express the absorbance $Ab(\bar{v})$ in terms of $\epsilon_g(\bar{v})$, eq.(22) is to be modified to get:

$$G(\bar{v},n,l) = \ln 10 \left\{ \epsilon_{smax} . \phi (v_e/v)^3 . (I_f/I_{fmax}).n.m - \epsilon_g m/l \right\} \quad (23)$$

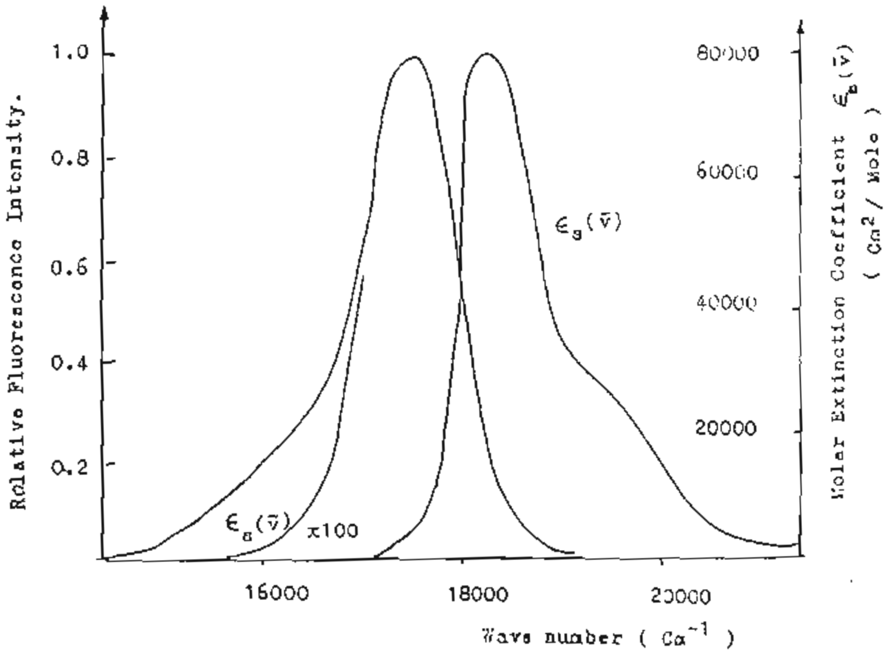

Fig. (2) Absorption and Relative Fluorescence Curves for Rhodamine B in Methanol($5x10^{-5}$ mole/lit.)

The fluorescence and absorption results in Fig.2 are for Rhodamine B molar concentration $m = 5 \times 10^{-5}$ mole/lit in methanol, with fluorescence quantum yield $\phi = 1$ [17]. With magnified low frequency tail (×100) for the absorption curve $\epsilon_a(v)$ [18]. The fractional pumped length $l = 1.0$ for axial pumping and for full length transverse pumping. We have calculated the intrinsic gain characteristics for different values of fractional excited state population n: n = 0.15, 0.1, 0.024, 0.01, 0.005, 0.0025, 0.001, and 0.0005 and the resulted gain curves are shown in Fig.3.

The calculated gain characteristics in Fig.3 are in good agreement with the reported results [16,17,19], and these curves disclose the following properties of the dye laser:

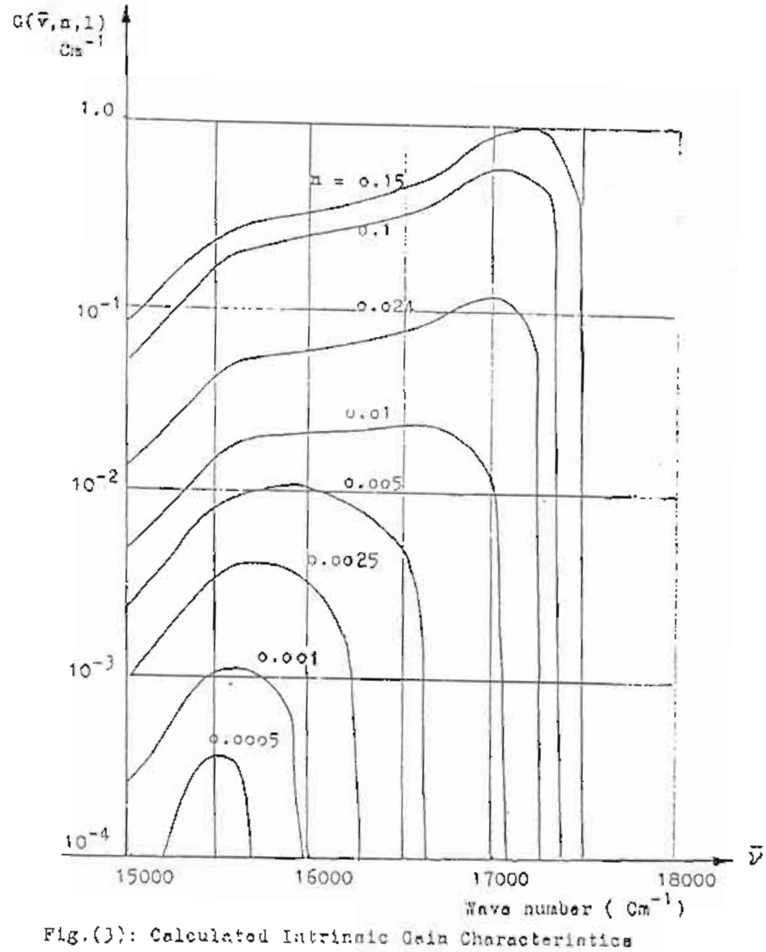

Fig.(3): Calculated Intrinsic Gain Characteristics for the laser dye Rhodamine B.

1) The dye solution exhibits positive coefficient over a wide spectral range, even for very small fractional excited state population ($n < 0.01$). Therfore the dye lasers truly typify a four-level laser scheme.
2) The dye laser gain increases and its frequency profile changes with increasing excited state fractional population which can be changed, either by varying the pumping rate or by changing dye concentration.
3) The peak of the calculated gain curves shifts to higher frequency as "n" or "l" increases, leading to laser frequency shifts to lower values by increasing the dye concentration or the dye optical length, these results are in good agreement with previously reported experimental results [20,21].

CONCLUSION

In our previous work, stimulated emission cross-section is expressed in terms ofreadily measurable spectroscopic parameters, namely normalized fluorescence intensity, frequency of maximum fluorescence, and fluorescence quantum yield . Using this expression of $\sigma_e(v)$, we have calculated the intrinsic gain coefficient for the dye Rhodamine B and plotted the frequency profile of the gain characteristics with the excited state fractional population as a parameter. These calculated gain characteristics are in good agreement with the reported results.

In this work, we have used Beer-Lambert law to calculate the low frequency tail of the absorption cross-section in the intrinsic gain expression.

This intrinsic gain expression is applicable for other fluorescent dyes and the calculated gain characteristic, can be used to study and explain the performance of the dye laser and its frequency dependence on the dye parameters.

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