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Polarization and Power Characteristics of Pulsed Dye Lasers with Transverse Pumping

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Abstract. Starting from the classical theory on molecular luminescence in liquids, rate equation for two-polarization-mode laser oscillation under transverse pumping with intense excitation have been set forth neglecting the influence of orientational relaxation of the dye molecules. Hence, the laser output power and polarization under different configurations have been obtained. The results show that the dye laser is partially polarized even when the pump beam is unpolarized and no polarizing elements exist in the cavity. If we define a reference plane consisting of the axes of the pump beam and the dye laser, the laser system has to adopt the plane perpendicular to the reference plane as the polarization direction in order to yield a highly efficient, linearly polarized laser output. Experiments on a copper-vapor laser pumped dye laser verified these analytical results.

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Transversely pumped dye lasers and amplifiers which are pumped by pulsed YAG, N_2 molecule, excimer and copper-vapor lasers are important sources for high average power, tunable frequency, and narrow linewidth. Such sources are widely utilized in laser spectroscopy, laser isotope separation etc. [1–4]. It is necessary to understand the polarization characteristics of the laser beam and to satisfy specific requirements while investigating the interaction between laser and matter or processing materials with lasers. Organic dye solution is a typical anisotropic laser medium. Some factors such as the structures of the dye molecules and the viscosities of the solvents determine the luminescence properties, which also result in the polarization characteristics of the dye laser. Therefore, research on the dye-laser polarization properties has received much of attention. Due to the complication of the process, different analytical methods have been used in different situations. Polarization characteristics of longitudinally pumped dye-laser amplifiers have been tackled using a semiclassic theory for the density matrix [5]. The polarization properties of transversely pumped dye lasers have been studied under steady state, while using a tensor to describe the anisotropy of the dye molecules [6]. The rate equations for the population and radiation transfer in the cavity have been adopted in discussing the depolarization of cw dye laser under high saturation conditions [7]. So far as we know, however, a systematic study on the polarization characteristics of transversely pumped dye laser is missing [8].

This paper, starting from the luminescence theory of liquids under transverse pumping, considers the dye as a dichroic amplification medium under intense pumping. We calculate the output power and the degree of polarization of the laser for several kinds of device configuration using rate equation. Also, experiments on copper-vapor laser pumped a dye laser have been carried out and the results confirm our theoretical analysis.

1 Polarization of Luminescence from Dye Solution under Transverse Excitation

Suppose that partially polarized light propagates along X (Fig. 1). The intensity components along Z and Y are I_z, I_y, respectively. Assume a dye molecule that sits at the origin of the coordinates 0 jumps to the excited state after absorbing incident light and returns to the ground state while giving out the excess energy in the form of luminescence. Specify the observation along Y, I_z or I_y excites two kinds of vibrations simultaneously, their directions are parallel to and perpendicular to the exciting one, denoting a and b for their amplitudes, respectively. The polarization ratio of the incident light is

$$\beta_e = \frac{I_z}{I_y} \tag{1}$$

while the degree of polarization of the incident light is

$$P_e = \frac{I_z - I_y}{I_z + I_y} = \frac{\beta_e - 1}{\beta_e + 1}. \tag{2}$$

According to the luminescence theory of a dipole [9], the

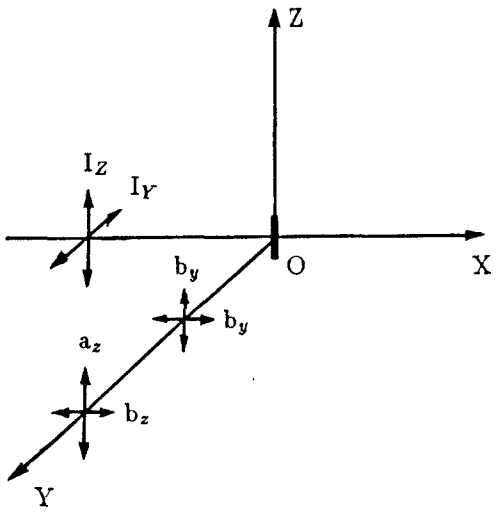

Fig. 1. Polarization of the dye molecular luminescence under external excitation

degree of polarization of the dye luminescence observed along Y is

$$P_l = \frac{(a_z^2 + b_y^2) - (b_z^2 + b_y^2)}{(a_z^2 + b_y^2) + (b_z^2 + b_y^2)}. \tag{3}$$

When $P_e = 1$, the maximum degree of polarization of the luminescence under linearly polarized light excitation is usually called the limiting degree of polarization P_p

$$P_p = \frac{a_z^2 - b_z^2}{a_z^2 + b_z^2}. \tag{4}$$

After simple algebraic manipulation, the degree of polarization of the dye luminescence under partially polarized light the transverse excitation can be expressed as follows:

$$P_l = \frac{(1 + P_e)P_p}{2 - (1 - P_e)P_p}. \tag{5}$$

In practice, laser dyes are dissolved in solvents with small viscosities, i.e. ethanol. In situation that the solvent does not freeze the emission dipoles, the dye molecules in the excited states undergo complicate movement, namely Brownian rotation, which results in the depolarization of the luminescence. P_l' denotes the degree of polarization of the dye luminescence on account of the depolarization effect of the Brownian rotational movement of the dipoles.

$$\begin{aligned} \frac{1}{P_l'} &= \frac{1}{P_l} + \left(\frac{1}{P_l} - \frac{1}{3}\right)\frac{\tau_e}{\tau_0}, \\ \tau_0 &= \frac{U\eta}{kT}, \end{aligned} \tag{6}$$

where τ_0 is the rotational time constant of a dye molecule, τ_e the lifetime of the molecular excited state, η the viscosity coefficiency of the solvent, T the temperature, k the Boltzmann constant, and U the volume of a dye molecule.

It has been verified that under ideal conditions, the limiting degree of polarization of the transverse-excited dipole luminescence P_p equals 0.5, while that for unpolarized light excitation, i.e., natural light excitation is equal to 1/3. The limiting degree of polarization depends on the material investigated and the exciting wavelength. For Rhodamine dyes, under condition of excitation in the visible region ($\geq$ 450 nm), absorption and emission oscillator behave to be roughly parallel. Due to the deviation of the absorption axis from the emission axis and other complex factors, the limiting degree of polarization is slightly less than 0.5 even for Rhodamine 6G dissolved in solvent with large viscosity. The practical value approximately equals 0.4. The viscosity is small for a Rhodamine 6G ethanol solution. The lifetime of the excited state of dye is 3 ns. It is also known that the experimental value of rotational time constant for a dipole is 0.3 ns [10]. Therefore, the observed degree of polarization of dye under weak excitation is nearly 4%.

It is necessary to point out that the lifetime of the excited state under intense excitation is determined by the spontaneous emission and stimulated emission simultaneously

$$\frac{1}{\tau_e} = \frac{1}{\tau_s} + \frac{1}{\tau^*} + \frac{1}{\tau_n}, \tag{7}$$

where τ_s is the lifetime of the spontaneous emission, $1/\tau^*$ and $1/\tau_n$ indicate the probility of the stimulated emission and the nonradiative transition, respectively. According to the Einstein's theory on radiation, (7) may be expressed as

$$\frac{1}{\tau_e} = \frac{1}{\tau_s}\left(1 + \frac{\lambda^2 g(\nu)}{8\pi n^2 h\nu} I_\nu\right) + \frac{1}{\tau_n}, \tag{8}$$

where λ, ν, n are the wavelength, frequency and the refractive index of the beam in the solution, respectively. $g(\nu)$ is the normalized lineshape function for the transition, $g(\nu) = 1/\Delta\nu$. If the intensity of the stimulated emission I_ν satisfies

$$I_\nu > \left(\frac{\tau_s}{\tau_e} - \frac{\tau_s}{\tau_n} - 1\right)\frac{8\pi n^2 h\nu}{\lambda^2 g(\nu)}, \tag{9}$$

the lifetime of the excited state is less than the rotational time constant of the dipole, then it is reasonable to neglect the depolarization of the Brownian movement, $P_l' = P_l$. For Rhodamine 6G ethanol solution, if $I_\nu > 10^3$ W/mm^2, the degree of polarization of the dye liminescence can be described by (5).

Usually, a cylindrical lens with a small focal length is used to converge pump light on the dye solution in transverse pump configuration. The cross section of the laser beam in the dye cell is less than 1 mm^2. The pulse width of the current pump source is about ten nanoseconds. The output power of the dye laser easily exceeds 1 kW once the laser builds up, thereby the depolarization of the dye molecule rotational relaxation can be neglected. Therefore, the dye solution can be regarded as a dichroic crystal: light along two different polarizing directions possesses different equivalent emission cross sections, the ratio of two equivalent cross sections equals the polarization ratio of luminescence, two polarization modes oscillate and propagate independently.

2 Rate Equations and Numerical Simulation

A dye laser can be represented by an ideal four-level system. In this paper, the triplet-state absorption and high-lying singlet-state absorption of dye molecules are neglected. The transverse pump beam and the dye oscillating beam constitute the XY plane. For the polarization direction, there are readily two modes: $\perp$ is used to denote that which oscillates along Z axis, $\parallel$ for that along X axis. From the luminescence theory mentioned above, the rate equations for a pulsed dye laser with transverse pumping are as

$$\frac{dn^{\perp}}{dt} = N\sigma_a^{\perp} I_p - \sigma_e^{\perp} \varrho^{\perp} n^{\perp} c - \frac{n^{\perp}}{\tau_s}, \tag{10}$$

$$\frac{d\varrho^{\perp}}{dt} = (\sigma_e^{\perp} n^{\perp} c - \gamma^{\perp})\varrho^{\perp}, \tag{11}$$

$$\frac{dn^{\parallel}}{dt} = N\sigma_a^{\parallel} I_p - \sigma_e^{\parallel} \varrho^{\parallel} n^{\parallel} c - \frac{n^{\parallel}}{\tau_s}, \tag{12}$$

$$\frac{d\varrho^{\parallel}}{dt} = (\sigma_e^{\parallel} n^{\parallel} c - \gamma^{\parallel})\varrho^{\parallel}, \tag{13}$$

where N stands for the concentration of dye molecules in the ground state, which at the pumping intensities used in the experiments is nearly equal to the total concentration of the dye molecules,

n is the concentration of dye molecules in the excited state,

ϱ the concentration of laser photons,

σ_a the absorption cross section of dye molecules at the pumping wavelength,

σ_e the stimulated emission cross section of dye molecules at the lasing wavelength,

τ_s the lifetime of spontaneous emission of the dye molecules,

c the speed of light

I_p the pump photon flux per unit area,

γ the loss coefficient in the cavity.

From the analyses in the preceding section, the following important relationships exist between the equivalent cross sections of two polarization modes:

$$\frac{\sigma_a^{\perp}}{\sigma_a^{\parallel}} = \frac{\sigma_e^{\perp}}{\sigma_e^{\parallel}} = \beta = \frac{1 + P_e P_p}{1 - P_p}, \tag{14}$$

$$\sigma_a^{\perp} + \sigma_a^{\parallel} = \sigma_a, \tag{15}$$

$$\sigma_e^{\perp} + \sigma_e^{\parallel} = \sigma_e. \tag{16}$$

The losses in the dye laser cavity come mainly from the transmission and absorption of both end mirrors, the losses of the inserted polarizing elements, the diffraction and scattering of laser beam in the cavity, etc. In order to facilitate a comparison with experiments, the calculations were performed on m Brewster glass plates as the polarizer in the cavity and neglected less important factors such as the diffraction and scattering of the beam. The loss in the cavity can be written as

$$\gamma^{\perp} = \frac{c}{L}\left[- m \ln T^{\perp} - \frac{1}{2}\ln(r_1 r_2)\right], \tag{17}$$

$$\gamma^{\parallel} = \frac{c}{L}\left[- m \ln T^{\parallel} - \frac{1}{2}\ln(r_1 r_2)\right], \tag{18}$$

where L is the cavity length of the dye oscillator. $T^{\perp}$ and $T^{\parallel}$ are the transmissivities of a glass plate at Brewster angle, respectively. r_1, r_2 are the reflectivities of the two end mirrors of the cavity.

There is usually no analytical solution for the rate equations. We performed our numerical simulation by using the Runge-Kutta method. Suppose that the pump beam has a Gaussian distribution in time, i.e.,

$$I_p(t) = I_{p0} \exp\left[- \left(\frac{t - t_0}{\Delta t}\right)^2 \ln 2\right]. \tag{19}$$

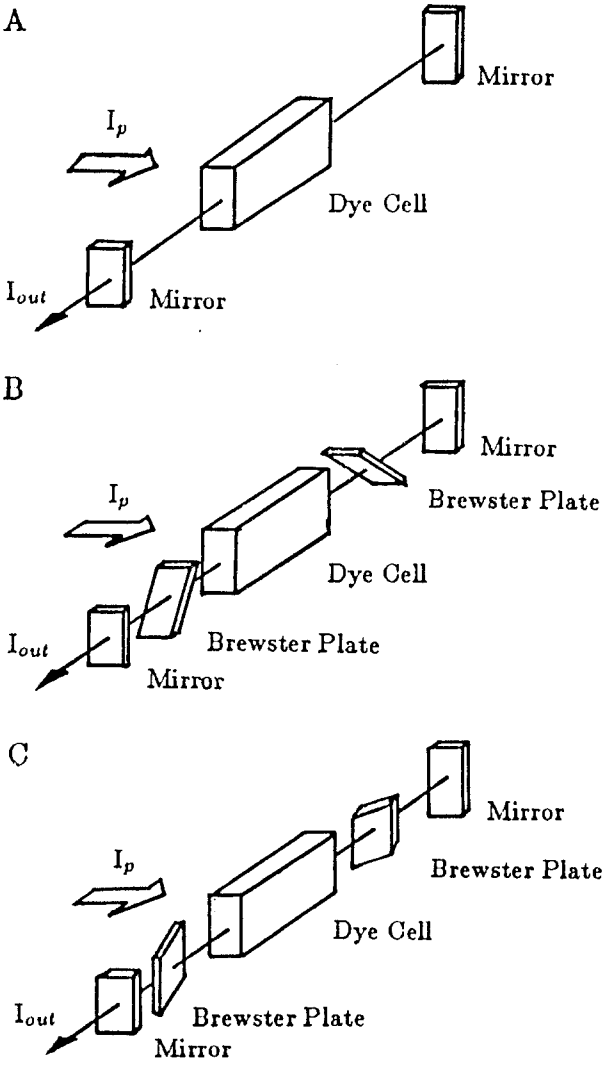

Fig. 2A–C. Three kinds of cavity configuration for a dye laser. **A** No polarizing element exists in the cavity. **B** The incident plane of the Brewster plates is perpendicular to the horizontal plane. **C** The incident plane of the Brewster plates parallels the horizontal plane

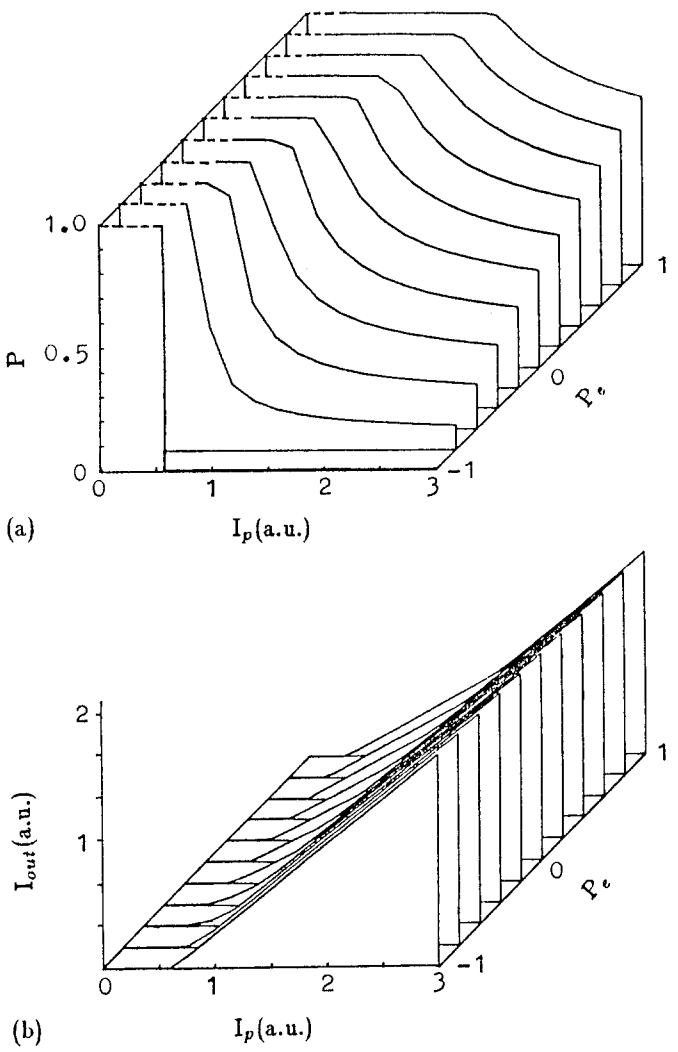

Fig. 3a, b. Calculated relationship between the degree of polarization **(a)** and the power of dye laser **(b)** vs pump power with different degrees of polarization for cavity of type A

In the above equation, I_{p0} is the maximum pump rate. Δt is the half width of the pump pulse. t_0 denotes the time when the pump beam reaches its maximum value.

The average power of the laser I is

$$I = I^{\perp} + I^{\parallel}, \tag{20}$$

$$I^{\perp} = (1 - r_2) h\nu V f \int \varrho^{\perp}(t)\, dt, \tag{21}$$

$$I^{\parallel} = (1 - r_2) h\nu V f \int \varrho^{\parallel}(t)\, dt, \tag{22}$$

where $(1 - r_2)$ is the transmissivity of the output mirror, $h\nu$ the photon energy of the dye laser, V the excited volume of the dye solution, and f the repetitive rate of the dye laser. The integrations were performed over one pulse period.

The rate equations were solved numerically with parameters corresponding to our experimental conditions: $\sigma_e = 1.6 \times 10^{-16}\,\mathrm{cm}^2$, $\sigma_a = 1.66 \times 10^{-16}\,\mathrm{cm}^2$, $N = 3 \times 10^{17}/\mathrm{cm}^3$, $L = 35\,\mathrm{cm}$, $\Delta t = 30\,\mathrm{ns}$, $r_1 = 1$, $r_2 = 0.1$. The wavelength of the pump beam is 510.6 nm, that of the dye laser is 577.5 nm. The repetitive rate f equals 6 kHz. The dimension of the dye cell V is 1.0 cm × 0.05 cm × 0.05 cm. The limiting degree of polarization of luminescence from Rhodamine 6G alcohol solution P_p is 0.4. Simple geometric calculations reveal that the transmissivity of one glass plate at Brewster angle for light polarized along two directions are 1.0 and 0.7, respectively.

For pump beams of different power and degree of polarization, we calculated the output power and degree of polarization of the dye laser under following cavity configuration: (A) No polarizing element exists in the cavity. (B) There are m Brewster plates in the cavity whose incident plane contains the Z axis, i.e., is perpendicular to the XY plane. The polarizer may be shaped like roofs. (C) There are m Brewster plates in the cavity whose incident plane is vertical to the Z axis, i.e., parallels the XY plane. The polarizer may by shaped like walls (Fig. 2).

Parts of the calculated results are shown in Figs. 3–5. For the purpose of generalization, the input pump power and the output power of the dye laser in these figures are marked in arbitrary units. Meanwhile, the degree of excess above the threshold M is introduced which is defined as the ratio of the input pump power and the threshold of the dye laser.

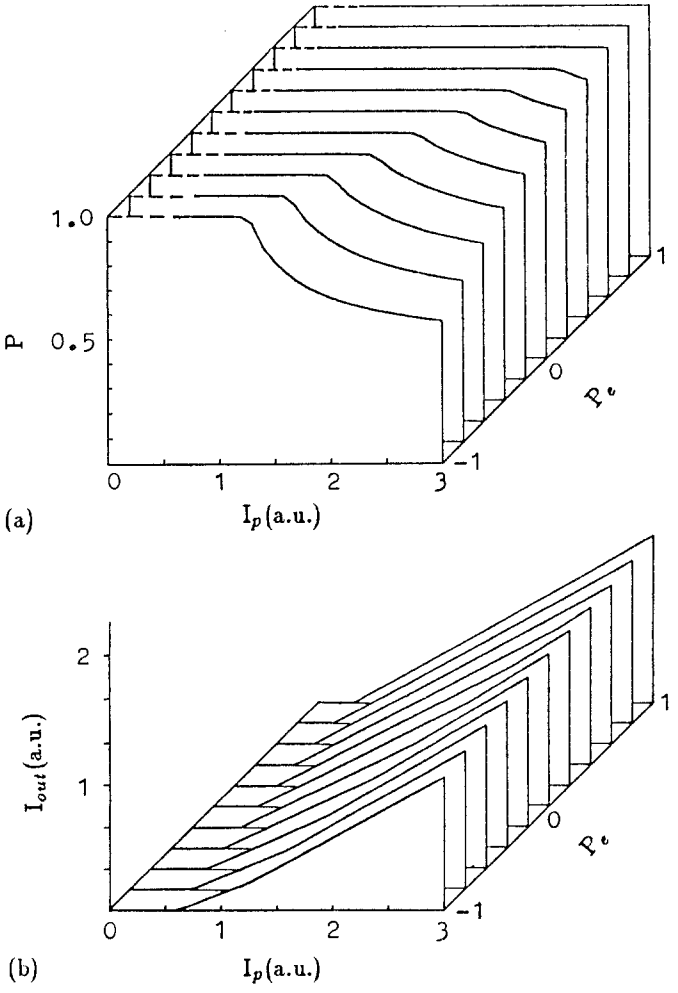

Fig. 4a, b. Calculated relationship between the degree of polarization **(a)** and the power of dye laser **(b)** vs pump power with different degrees of polarization for cavity of type B

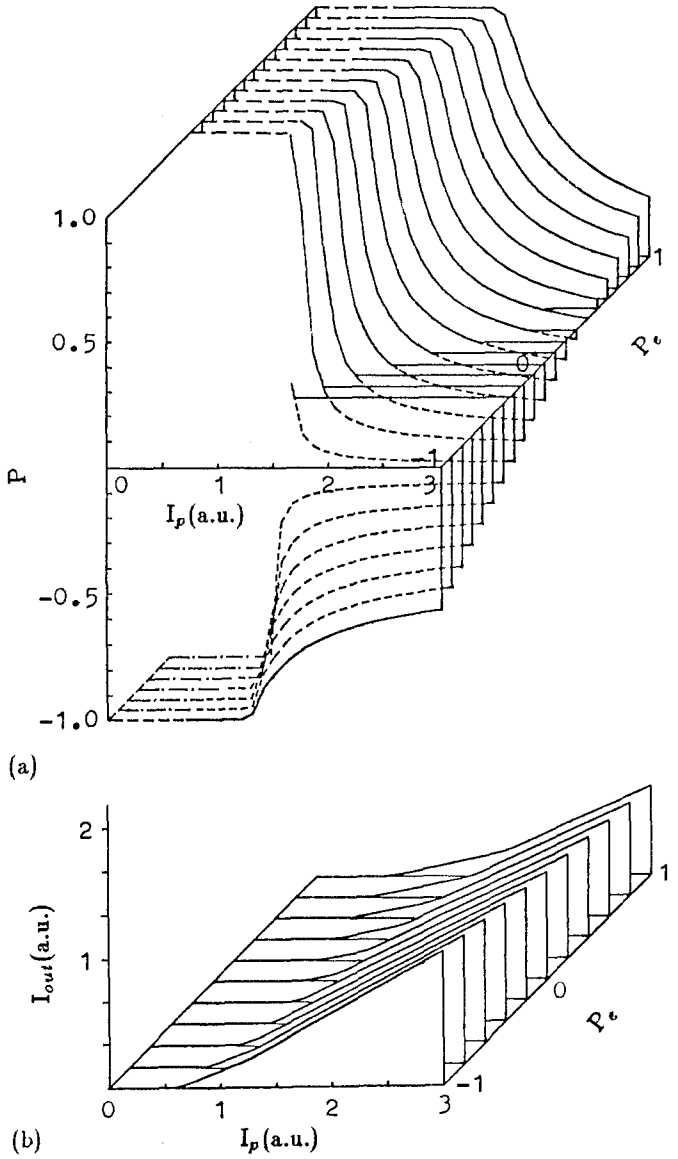

Fig. 5a, b. Calculated relationship between the degree of polarization (**a**) and the power of dye laser (**b**) vs pump power with different degrees of polarization for cavicty of type C

In a cavity without polarizing elements (Fig. 3), the dye laser is linearly polarized or partially polarized ($1 \geq P > 0$) except the pump beam is linearly polarized along Y axis ($P_e = -1$). Even when the pump beam is natural light, the dye laser is linearly polarized until the degree of excess above the threshold is about 2. With rise of the pump power, the degree of polarization of the dye laser reduces due to the increase of the output power of another polarization mode. It has been pointed out that two polarization modes have equal gain for $P_e = -1$, therefore the free-running output power is equal, i.e., the degree of polarization of the output beam equals 0.

Equation (14) indicates that the light polarized along the Z direction (i.e., $\perp$) has comparatively larger gain in the active medium for a transverse pump configuration. Therefore, in using polarizer of the same transmissivity ratio, the dye laser has larger output power and larger degree of polarization when the preferred direction of the polarizer is the same as Z direction. Figures 4 and 5 show the dye laser output characteristics with a pump beam of different power and degree of polarization while there is a pair of Brewster plates in the cavity. (The transmissivity ratio R is 1:0.49). Obviously, it is advantageous for Brewster plates as roofs.

Induced by polarizing elements whose polarizing direction is along Z, the transmissivity ratio of vertical to horizontal polarized light is greater than 1, otherwise less than 1. Figure 6 shows the degree of polarization and relative power of the dye laser as the function of transmissivity ratio of the Brewster plates when the pump power equals six times of pump threshold of the dye laser without polarizing elements under natural laser light pump. It is evident that linearly polarized light ($|P| = 1$) may be obtained as far as the transmissivity ratio R is large or small enough, but the output power will vary significantly for different configuration. For pump laser with $P_e = 1$, 0, -1 and $M = 6$, the transmissivity ratio for polarizer to be infinity (or zero) in extreme condition, the output powers are 0.81, 0.70, 0.51 (or 0.16, 0.30, 0.51) if that of no polarizing elements in the cavity equals 1. The differences among these three situations

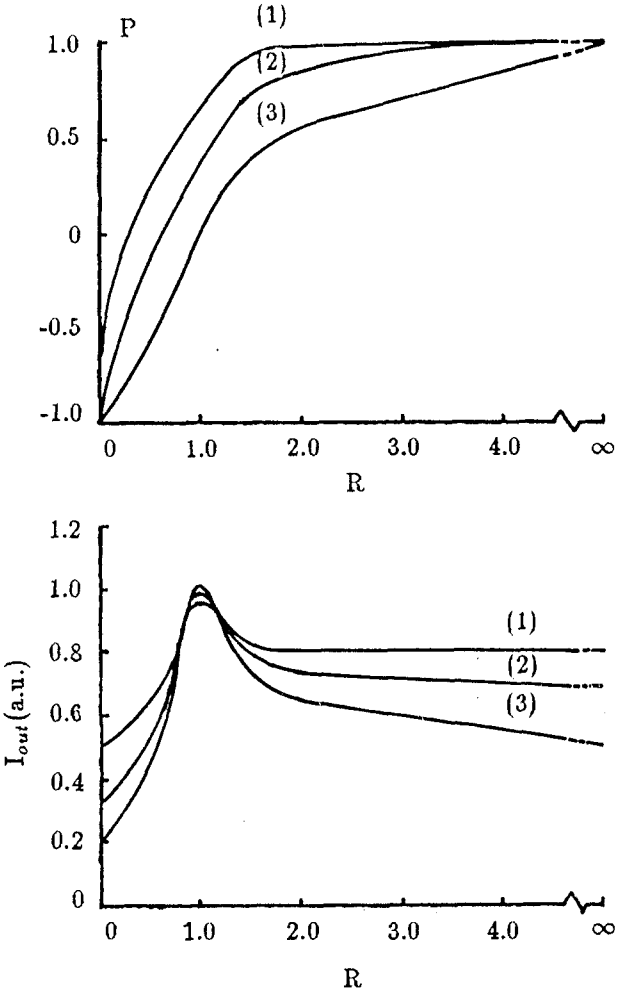

Fig. 6. The degree of polarization P and output power I_{out} of the dye laser as the function of transmissivity ratio R of the polarizer when the pump power equals six times of pump threshold of the dye laser without polarizing elements under natural laser light pump. *(1)* for $P_e = 1$; *(2)* for $P_e = 0$; *(3)* for $P_e = -1$

will diminish with the increase of the M value. Following (5) and (14), these values are 0.70, 0.625, 0.50 (or 0.30, 0.375, 0.50), respectively, as M tends to be infinity.

3 Copper-Vapor Laser Pumped Rhodamine 6G Dye Laser

To verify the theoretical analyses mentioned above, experiments have been carried out. The experimental setup is shown in Fig. 7. The pump source was a copper-vapor laser which operated at 510.6 and 578.2 nm with pulse width approximately 30 ns (FWHM) at a repetition rate of 6 kHz. The resonator of the copper-vapor laser adopted the positive-branch telescope cavity with magnification 10 to permit free oscillation of the laser. To obtain laser beams with different degree of polarization, the beam passed through polarizer which consisted of glass plates whose number and angle were adjustable. The solvent was ethanol and the concentration of Rhodamine 6G dye solution was 5×10^{-4} M. The dye was circulated during the experiment. There were three kinds of dye laser configuration corresponding to those depicted in Fig. 2. The cavity adopted was a plane-plane cavity having one of the end mirrors made of glass, the other was a total reflective one with a dielectric coating. The copper-vapor laser beam was focused by a cylindrical lens to excite the dye. The length of the dye cell was 1 cm and the length of the dye laser resonator was 35 cm. Using a Rochon prism as a polarization analyzer, the degree of polarization of the pump beam P_e and dye laser P were determined by measuring the intensities of the beams along two oscillating directions after the beam passed the polarization analyzer.

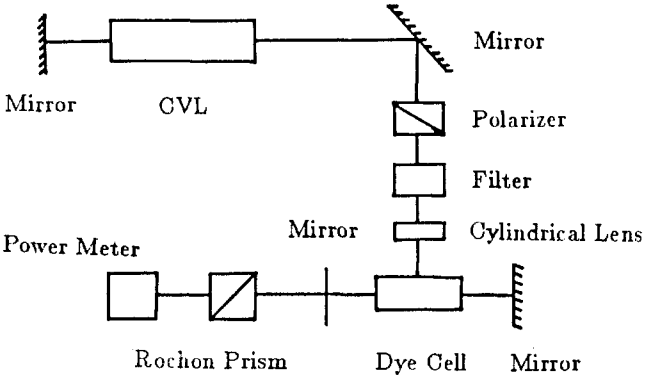

Fig. 7. Experimental setup for measuring the degree of polarization and the output power of the dye laser

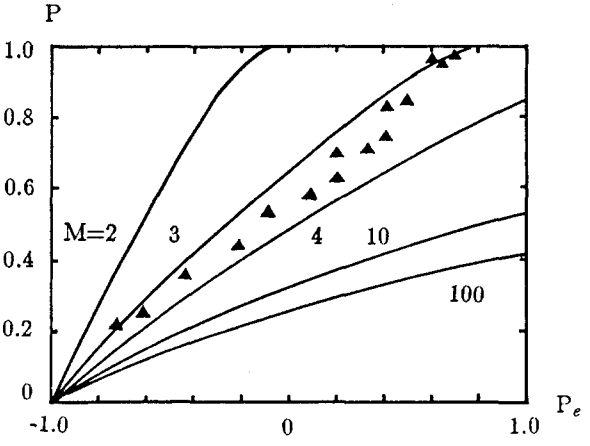

Fig. 8. The dye-laser polarization P vs pump polarization P_e. M – The degree of excess above the threshold. (–) numerical simulation; (▲) experimental results

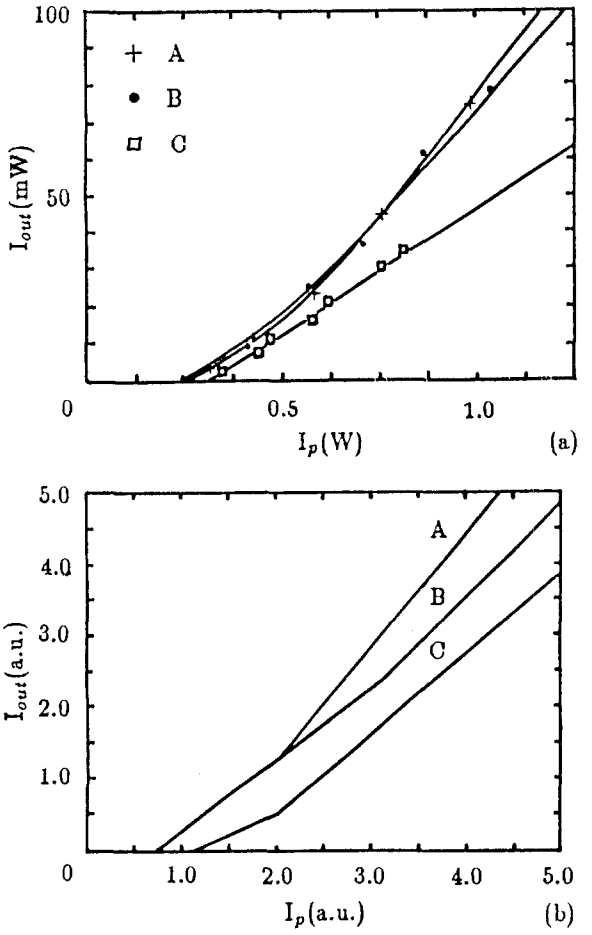

Fig. 9a, b. Dependences of the dye-laser output power I_{out} on the pump power I_p for the configuration A, B, C. **a** Experimental, **b** numerical simulation

The experimental result for the relationship between the degree of polarization of the output beam and that of pump beam are depicted in Fig. 8. The degree of excess above the threshold M was 4 in the experiment. The experimental result shows in good agreement with that of the calculation.

The relation between the output power and input power were also measured with respect to the three kinds of cavity configuration as in Fig. 2 and the pump beam was natural light (Fig. 9a). The calculated result are shown in Fig. 9b. Apparently, the agreement is quite well.

4 Conclusion

Basing on the classical theory of liquid luminescence and neglecting the molecular orientational relaxation under strong pulse excitation, this paper demonstrates that as we define a

reference coordinate plane containing the axes of the pump beam and the dye laser, transversely pumped pulsed dye may be simply regarded as a dichroic medium and the ratio of gains for two polarization directions in it depends on the polarization characteristcs of the pump beam. Furthermore, the gain for polarized light along the vertical direction is never less than the others. Therefore, the dye-laser characteristics in this configuration relate to the degree of polarization, the degree of polarization, the degree of excess above the threshold of the pumping beam and the optics of the components in the device. Even if the pump beam is natural light and no polarizing elements exist in the cavity, the dye-laser output is linearly polarized or partially polarized.

In practice, it is a necessary requirement to achieve tuning in the dye laser, some optical elements such as a Littrow grating and a beam expanding system are employed in the cavity. Usually, these elements are with reflective coatings or antireflective coatings. From Fresnel's equations, there is dichroism for the transmissivity and reflectivity of the coatings or optical surfaces. Our investigations show that the whole system must have the vertical direction as the polarization direction, otherwise unreasonable arrangements and configurations will degrade the dye-laser system. This result applies not only to the dye-laser oscillators, but also to the dye-laser amplifiers. Consequently, the incident plane of the pump laser must be chosen the vertical polarization direction if Brewster windows exist within it, too.

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