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Amplified backscattering from a rough surface through dye-doped polymer

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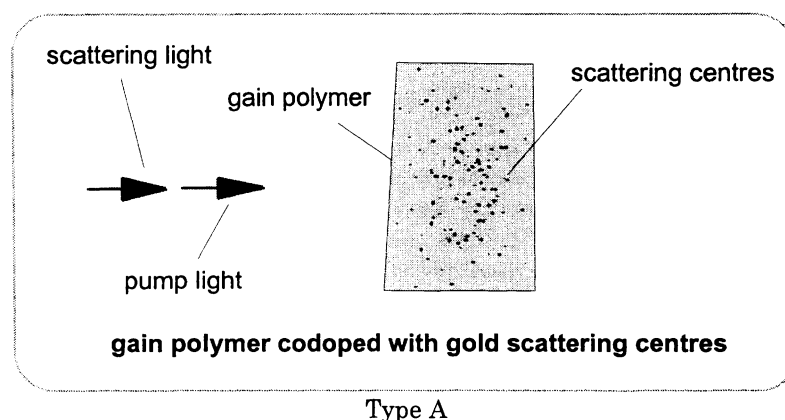
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ABSTRACT

We report the experimental study of the enhanced backscattering from a random rough surface through dye-doped polymer. The sample is a two-dimensional rough gold surface with a large slope coated with Pyrromethene-doped polymer layer. The sample is illuminated with a s-polarized He-Ne laser, which is pumped by a CW Argon Ion laser. The amplified scattering is measured. It is found that the enhanced backscattering peak is sharply increasing and the width is narrowing for a sample with low dielectric constant $|\epsilon_2|$.

I. INTRODUCTION

One of the most interesting phenomena associated with the scattering of light from a randomly rough surface is that of enhanced backscattering. This is the presence of a well-defined peak in the retroreflection direction in the angular distribution of the intensity of the incoherent component of the light scattered from such a surface, which results primarily from the coherent interference of each multiply reflected optical path with its time-reversed partner^[1-6]. Recently, theoretic study in volume and surface scattering shows that there are novel enhanced backscattering peak from amplifying random media and surface. An example of an amplifying medium is a laser material. The amplification is more pronounced for long paths which build up the structure of the enhanced backscattering peak, and qualitatively change this backscattering. In particular, the peak become sharper as the gain of the medium is increased^[7-8].



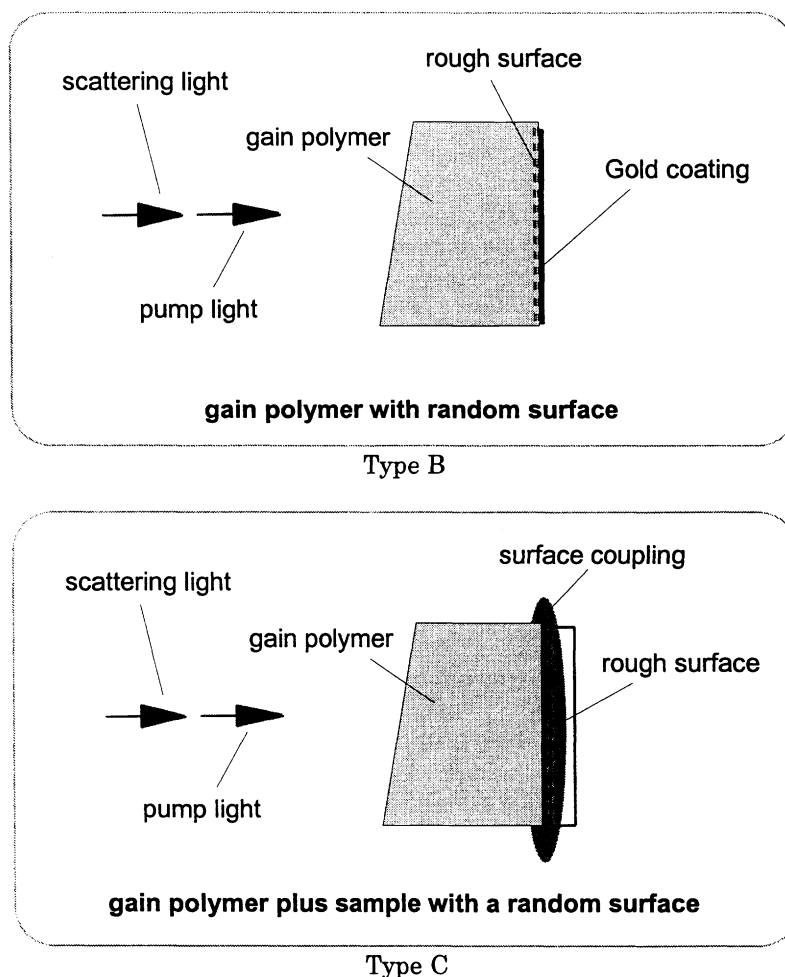


Figure 1. Three possible configurations.

In this paper we report the experimental study of the enhanced backscattering from a rough surface through dye-doped polymer. There are three possible configurations, as 1) volume effect, 2) double-passage of waves effect, and 3) surface effect. (see type A,B,C of Figure 1). We will discuss mainly the type C configuration in this paper. Section II shows gain material and sample preparation, and Section III with gain constant and fluorescence. The experimental set-up and results are in Section IV, followed by a summary in Section V.

II. GAIN MATERIAL AND SAMPLE PREPARATION

Laser dyes are highly efficient gain media either for laser sources with narrow pulse width and wide tunable range, or for optical amplifiers with high gain, high power conversion and broad spectral bandwidth. Obviously, the wide spectral range of gain of laser dyes covering the visible wavelengths are very convenient for our work here. Traditionally laser dyes are dissolved in organic solvents and thus a liquid gain medium could be easily prepared. However, the liquid gain medium is toxic and cannot be handled conveniently and safely. New techniques have been developed to make solid-state dye-doped gain media. Laser dyes captured in a solid matrix are much easier and safer to handle than their counterpart in liquid form. Moreover, the photostability of some organic laser dyes improves when molecules are trapped in solid matrices. It was reported that the photostabilities of dye molecules in solid matrices was approximately two orders of magnitude higher than in solutions^[9].

For this reason, solid-state dye gain media have attracted considerable research interest in recent years^[10,11]. There are essentially two main techniques for the preparation of solid-state dye-doped gain media. The first technique is the dye-doped organically modified silicate (ORMOSIL) ORMOSILs are synthesized using sol-gel processing at low temperature, which makes it possible to dope various laser dyes in and, at the same time, to keep them active. The second technique is impregnating laser dyes in polymeric hosts such as poly(methyl methacrylate), poly(carbonate) and poly(styrene). Much of the work are still at the demonstration stage and being carried on intensively. From recent experimental results, dye-doped polymer based on modified poly(methyl metracrylate) appears very attractive compared with solid-state dye-doped material based on ORMOSIL. It is found that, at this stage, the dye-doped polymer has better efficiency and beam quality, and superior optical homogeneity^[12].

We used the second technique to prepare our gain samples. The dye-doped samples are prepared by thermal polymerisation. The laser dyes are dissolved into the mixed monomers before the polymerisation. The monomers used include methyl methacrylate (MMA) and ethyl methacrylate (EMA) both of which have low refractive indices in the range of 1.41~1.42. Commercial monomers are inhibited for ease of transportation. The inhibitor can be removed by washing the monomers with 5% NaOH in distilled water. The washed monomers are flushed with distilled water and then the water is removed by passing the monomers through a column with activated Aluminum oxide. The monomers are then purified by means of the degassing and distillation process. After polymerisation, however, the indices increase due to the volume reduction during liquid-to-solid phase transition. The polymerised materials: Poly(MMA-EMA), have indices in the range of 1.48 ~ 1.49, depending on the polymerisation conditions.

Rhodamine 6G and rhodamine B are traditional laser dyes with high photostability and high fluorescence efficiency. In recent years, significant advances have been made in the synthesis of new dyes with higher fluorescence efficiency and also reduced triplet-to-triplet absorption. In particular, a new class of laser dyes -pyrromethene based laser dyes, have attracted considerable interest because of their superior performance over those of rhodamine or coumarin based laser dyes. It was reported that pyrromethene showed the highest fluorescence quantum efficiency (typically more than 80%) and a triplet extinction coefficient one fifth that of rhodamine 560^[13]. Hence, in addition to fabricating samples doped with conventional laser dyes such as rhodamine 6G, rhodamine B and fluorescein, we have prepared several samples doped with pyrromethene.

Table 1. Dye-doped polymer samples and parameters.

Sample No.	Gain Medium	Concentration (ppm)	Thickness (mm)
1	Rhodamine 6G	318	1, 2.6, 4.5
2	Rhodamine B	66	4.6
3	Fluorescein	440	1.6, 2.4, 4.7
4	Pyrromethene	290	1.1, 3.0, 5.3
5	Pyrromethene	8	12.0
6	Rhodamine 6G	52	6.0
7	Rhodamine B	16	6.8

All the dye-doped gain polymers are polymerized under the same thermal polymerisation process, with the process temperature progressively increased from 36°C to 98°C over a period of several days. The process was completed with heat treatment for several days at temperature between 80°C and 98°C. Finally, the dye-doped polymer samples were cut and polished to various thickness for use in

scattering experiments. The parameters of the polymer samples prepared for the experiment are summarized in Table 1. The main consideration to have a range of samples of different laser dyes, concentrations and thickness is to compare the gain effects and to obtain the optimal gain conditions.

III. GAIN CONSTANT AND FLUORESCENCE

In doping laser dyes into polymeric matrices, the thermal, chemical, mechanical and optical properties are of great importance. For the present experiment the most important factor is the gain that scattered light would have through a dye-doped polymer sample. We present a simple theoretical analysis of the gain factor based on the well-known energy level diagram of dye molecules^[14], as shown in Figure 2.

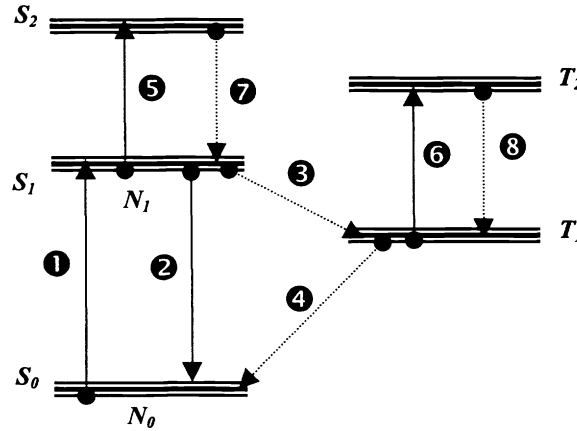


Figure 2. The schematic energy level diagram of a dye molecule.

In this simple model, the singlet states are formed by the two most energetic electrons with their spin antiparallel, resulting spin $S=0$. And the triplet states are formed when the two electrons with their spins in parallel, resulting $S=1$ which can have three possible orientations (ie. $S=-1, 0, 1$) with respect to an external magnetic field. Hence, every excited singlet state, eg. S_1, S_2 , etc., is accompanied by a triplet state, T_1, T_2 , etc., of lower energy. Many different optical processes are associated with the electron transitions between these energy levels. For example, the main absorption is normally from S_0 to S_1 (1) and the dominant spontaneous (fluorescence) or stimulated emission is from S_1 to S_0 (2). These are two processes directly related to the gain factor which we will discuss later. Other processes shown in the plot are associated with nonradiative intersystem transitions (3 and 4), excited state absorption (5 and 6) and nonradiative innersystem transitions (7 and 8).

In a gain medium, the signal intensity, $I=I(\lambda, z)$, where z is the signal propagation direction, is governed by

$$\frac{dI}{dz} = \frac{dI}{dz}\bigg|_{stim, S_1 \Rightarrow S_0} - \frac{dI}{dz}\bigg|_{absor, S_0 \Rightarrow S_1} - \frac{dI}{dz}\bigg|_{absor, T_1 \Rightarrow T_2, T_3, \text{ etc.}} \quad (1)$$

where the total signal intensity increases due to stimulated emission and decreases due to absorptions. The signal optical intensity from *stimulated* emission is well known^[14] and can be expressed as

$$\frac{dI}{dz} = \frac{\left(N_I - N_o \frac{g_2}{g_1}\right) \lambda^2 g(\nu)}{8\pi \tau n^2} I = \frac{\left(N_I - N_o \frac{g_2}{g_1}\right) \lambda^4 E(\lambda)}{8\pi \tau c n^2} I = \gamma I \quad (2)$$

where N_o , g_1 and N_I , g_2 are the populations (usually atom densities), degeneracies of S_o and S_I states, respectively. Here c , λ and ν are the velocity, wavelength and frequency of light, respectively; n is the refractive index of gain medium; τ is the fluorescence decay time for spontaneous emission; $E(\lambda)$ and $g(\nu)$ ($g(\nu) = \frac{\lambda^2}{c} E(\lambda)$) are the spontaneous emission lineshape functions that satisfy $\int_0^\infty E(\lambda) d\lambda = \int_0^\infty g(\nu) d\nu = \phi$,

with ϕ the fluorescence quantum yield; γ is defined as the gain constant due to stimulated emission:

$$\gamma = \frac{\left(N_I - N_o \frac{g_2}{g_1}\right) \lambda^4 E(\lambda)}{8\pi \tau c n^2} \quad (3)$$

To work out the gain constant, we have to evaluate the population inversion term $\Delta N = \left(N_I - N_o \frac{g_2}{g_1}\right)$.

It can be estimated from the pump laser excitation of the gain medium. In general, the pump energy E absorbed per unit volume is expressed by

$$E = \int_0^\infty \tau s(\nu) \alpha(\nu) d\nu \quad (4)$$

where $s(\nu)$ is the optical flux in watts per unit area [W/cm²/Hz], $\alpha(\nu)$ the absorption coefficient in [1/cm] and τ the fluorescence decay time for spontaneous emission in second [s]. When a single wavelength pump laser is used, the number of absorbed pump photons may be expressed as

$$N = \frac{\tau}{h\nu} s(\nu) \alpha(\nu) \quad (5)$$

If the absorption quantum efficiency is $\eta(\nu)$, then the excited molecule number per volume is

$$N_I = \frac{\tau}{h\nu} \eta(\nu) s(\nu) \alpha(\nu) \quad (6)$$

When all the pump power absorbed produces excitation of active dye molecules, eg. when the excited state absorption is neglected, we have $\eta(\nu)=1$. Of course, in most practical case, $\eta(\nu)$ is less than 1.

In practical laser and optical amplification systems, the pump wavelength is selected to be sufficiently shorter than the signal wavelength, viz. we arrange the pump at the high absorption region and, at the same time, the signal at the low absorption region. This ensures an ideal case for optimal gain: pump power efficiently excites the molecules and signal power is amplified without significant absorption. In these cases, we have simply $\Delta N = \left(N_I - N_o \frac{g_2}{g_1}\right) \approx N_I$. Therefore, the gain constant of the dye-doped polymer samples can be evaluated by

$$\gamma \approx \frac{\eta(\nu_p) s(\nu_p) \alpha(\nu_p) \lambda_s^4 E(\lambda_s)}{8\pi c n^2 h \nu_p} \quad (7)$$

As an example, we estimate the gain constant of a rhodamine 6G-doped polymer sample using Argon laser (514nm) as pump and He-Ne laser (632.8nm) as signal source. Taking typical parameters of Rhodamine 6G: $\eta(\lambda)=0.9$ and $E(\lambda) \sim 4 \times 10^4/\text{cm}$ as well as sample parameters, $\alpha(\nu) = 10\text{dB/mm}$ and $n = 1.48$, we have a gain constant about 2.3dB/cm, given that a pump power of 10W with a spot area of $0.2 \times 0.2\text{mm}^2$.

Within the gain medium, a scattering (or scattered) wave along a certain direction z can be simply represented by

$$E(z,t) = E(z)e^{j(kz - \omega t)} \quad (8)$$

where k is the propagation constant given by $k = \frac{2\pi}{\lambda} n$. The refractive index of the medium is given by

$$n = \sqrt{\epsilon} = \sqrt{\epsilon_1} + \frac{1}{2} j \frac{\epsilon_2}{\sqrt{\epsilon_1}}$$

Hence

$$e^{jkz} = e^{j \frac{2\pi}{\lambda} nz} = e^{j \frac{2\pi}{\lambda} \sqrt{\epsilon} z} = e^{j \frac{2\pi}{\lambda} \sqrt{\epsilon_1} z - \frac{\pi}{\lambda} \frac{\epsilon_2}{\sqrt{\epsilon_1}} z} = e^{jk_1 z - k_2 z}$$

Then the (intensity) gain constant defined by Equation (2) can be related to the imaginary component of dielectric constant ϵ_2 by

$$I = I_0 e^{\gamma z} = I_0 e^{-2k_2 z} = I_0 e^{-\frac{2\pi}{\lambda} \frac{\epsilon_2}{\sqrt{\epsilon_1}} z}$$

with the gain constant

$$\gamma = -2k_2 = -\frac{2\pi}{\lambda} \frac{\epsilon_2}{\sqrt{\epsilon_1}} \quad (9)$$

The fluorescence spectra of various dye-doped gain polymers have been measured. The typical fluorescence spectra are shown in Figure 2. As usual, dye molecules in polymer matrices give slightly changed and shifted fluorescence profiles compared with those from the same dye in liquid solvents. By comparing the fluorescence spectra, it is obvious that pyrromethene-doped polymer has its fluorescence peak around 650nm. This implies that pyrromethene-doped polymer would be appropriate for the current experiment using He-Ne laser at 633nm as signal source.

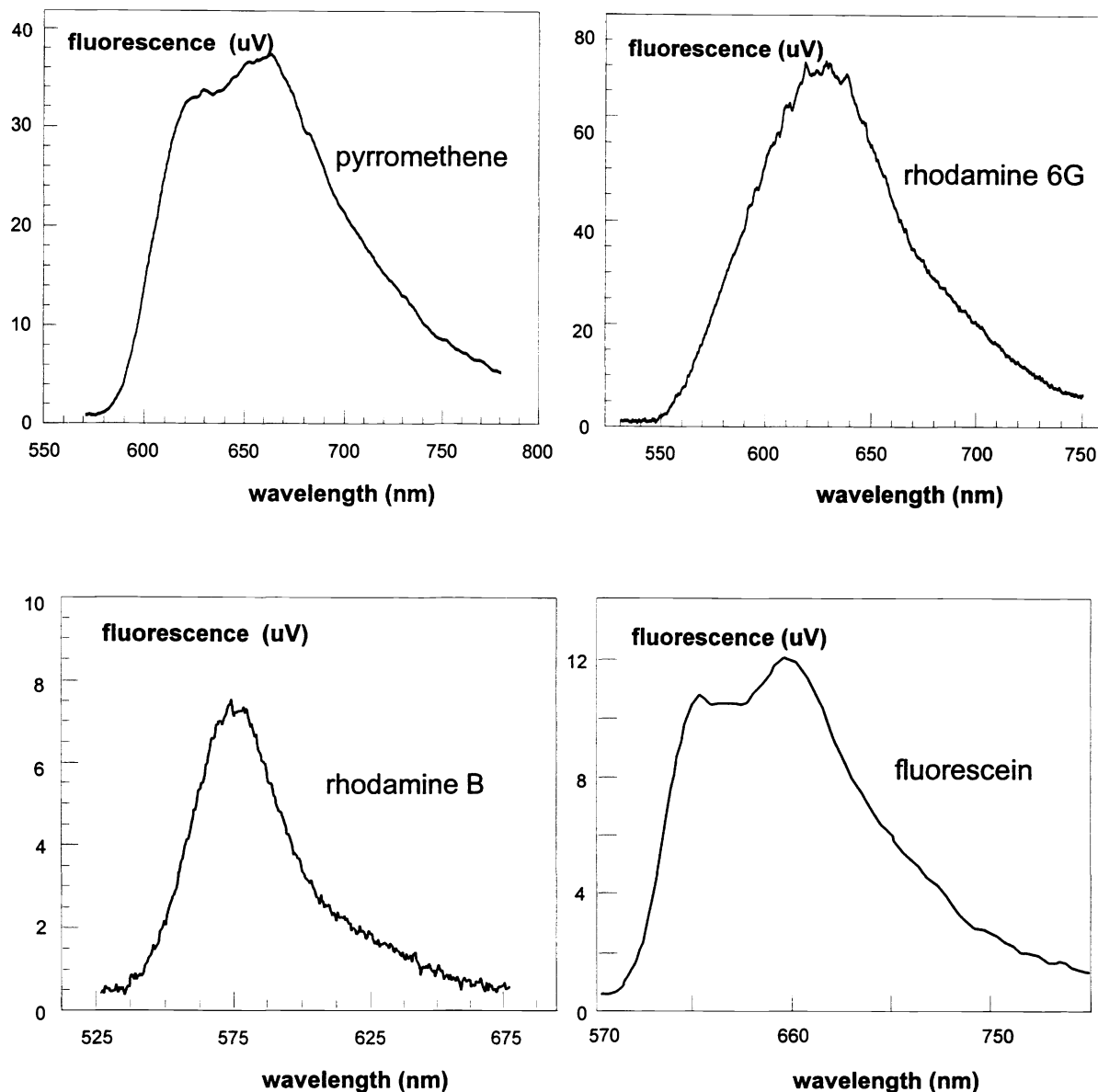


Figure 3. Typical fluorescence spectra of dye-doped polymers.

IV. EXPERIMENTAL SETUP AND RESULTS

Figure 4 is a sketch of the optical setup. Usually high pump intensity is needed. To produce optimal pump efficiency, the pump and scattered beams should be collinearly irradiating the gain layer and rough surface. Hence the scattering main beam is co-axial with the pump beam. Both pump and scattering beams would better be focussed, with pump beam size slightly larger than the scattering beam size. A narrow bandpass filter is necessary since the fluorescence from laser dyes is typically broadband and it may easily mix up to the output signal.

The wavelength of the scattered beam must be sufficiently separated from that of the pump beam to avoid high absorption around the pump wavelength. Meanwhile, it must be sufficiently close to the

fluorescence peak to ensure a significant optical gain. For the laser dye-doped polymer samples listed in Table 1, the absorption peaks are typically in the wavelength range between 470nm and 540nm, and the fluorescence peaks are typically in the wavelength range 570nm and 670nm. Hence suitable pump sources for these samples are frequency-doubled YAG lasers (wavelength 532nm) and Argon lasers (wavelength 514nm or 488nm). Based on these considerations, we have picked the He-Ne laser with the wavelength of 633nm as the scattered beam and CW Argon Ion laser with the wavelength of 514.5nm as the pump beam.

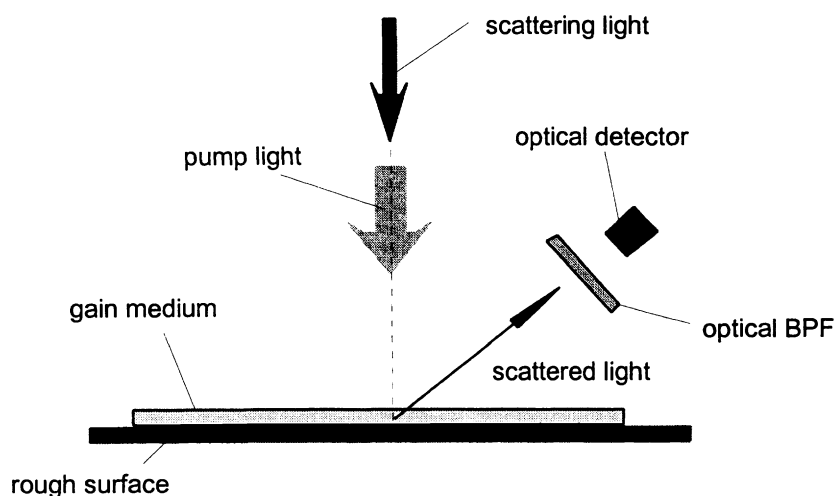


Figure 4. The sketch of optical setup.

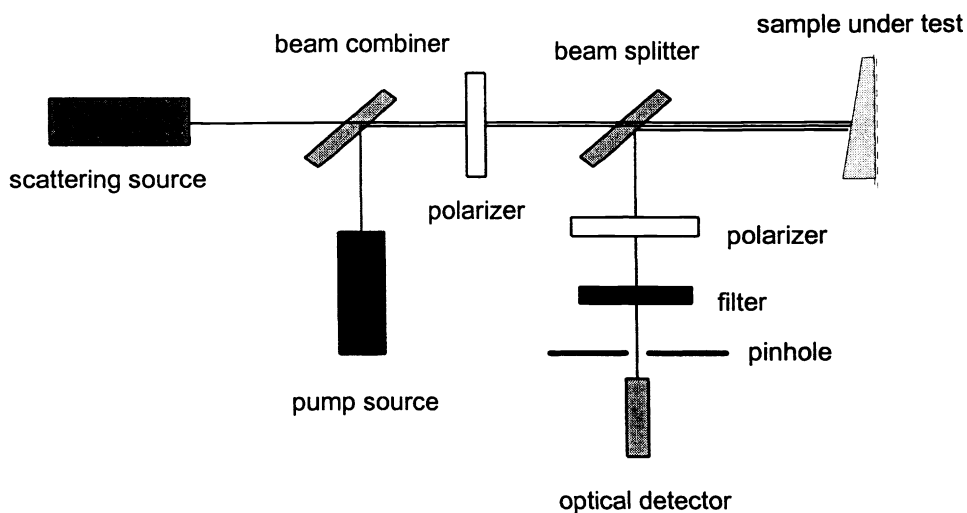


Figure 5. The schematic diagram of experimental setup.

Figure 5 is the schematic diagram of experimental set-up. A fully automated bidirectional reflectometer was used to measure the fraction of incident light reflected by the sample into incremental angles over its field of view. It uses illumination from laser sources $0.633\mu\text{m}$ as main scattering beam and this enables measurements for any combination of incident and reflected angles over the entire plane, except

for a small angle (about 0.5° away from the retroreflection direction) in which the source and detector mirrors interfere.

A laser beam passes through a polarizer and is interrupted by a chopper and a half-wavelength plate, which enables rotation of the polarization of the beam. The pump Argon ion laser joins with the He-Ne laser at the beam combiner. Then they are directed toward the sample by a folded beam system that collimates it into a parallel beam or focus to a spot. The sample is viewed by a movable off-axis paraboloid that projects the light reflected by the sample onto the detector via a polarizer and a folding mirror. Four different polarization combinations of input and receiving beams are recorded. The reference standard used for these experiments is Lab Sphere Gold, and the relative bidirectional reflectance is measured. The signal is recorded and digitized at each angular setting of interest throughout the angular range by an ITHACO lock-in amplifier and the data are stored in the memory of a personal computer (PC). The sample and the receiving telescope arm are separately mounted on two rotational stages run by two independent stepper motors that are controlled by the PC via a two-axis driver.

The main scattering source is a CW 35mw He-Ne laser while the pump source is a 10w CW Argon Ion laser. The laser beams can be focused onto an area as small as $0.1 \times 0.1 \text{ mm}^2$. We specially fabricated a sample for our experiment. The mean thickness of the designed photoresist film is about $10 \mu\text{m}$, and that of the glass is 2mm. In view of the need to accommodate 1-D or 2-D scattering theories, a 2-D very random rough surface was fabricated by use of random speckle patterns^[15]. Then a layer of Gold was deposited on the rough surface. The surface profile of the 2-D random rough surface was measured with a Dektak 3030 machine. Statistical analysis of data from 10 profiles determined that the rms roughness is $1.2 \mu\text{m}$ and the $1/e$ correlation length is $3.1 \mu\text{m}$. Finally a layer of dye-doped polymer is coupled with the Gold rough surface.

Figure 6 shows the experimental results. Figure 6 (a) shows the far-field intensity distribution of the 2-D gold rough surface coupled with Pyrromethene-doped polymer where no pump beam is used. The thickness of polymer is about 3 mm. The s-polarized He-Ne laser beam is applied with an incidental angle of -5° . An enhanced backscattering peak at the retro-reflection direction of 5° is apparent. Figure 6 (b) shows the far-field intensity distribution of the 2-D gold rough surface with Pyrromethene-doped polymer and a 2w pumping power at 514 nm. The main beam is a He-Ne laser at 633 nm with s-polarization and an incidental angle of -5° . There is a strong enhanced backscattering peak at 5° . Comparing Figure 6 (a) with 6 (b), you will find the enhanced backscattering peak has sharply increased and the width has narrowed for amplified scattering from a rough gold surface through dye-doped polymer.

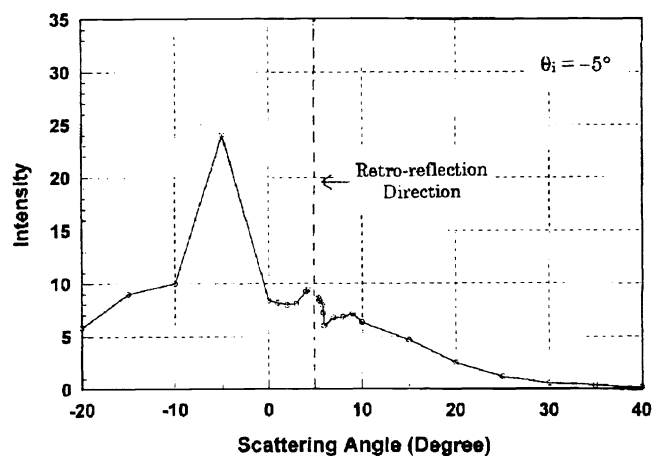
V. SUMMARY

The amplified scattering for a 2-D random rough Gold surface with dye-doped polymer is measured with an s-polarized He-Ne laser at a wavelength of 633 nm and an Argon Ion laser pumping at a wavelength of 515 nm. It is found that the enhanced backscattering peak is sharply increasing and the width is narrowing for a low dielectric constant $|\epsilon_2|$. It is also found that the accompanying background noise is because of the non-uniform distribution of the pumping power.

VI. ACKNOWLEDGEMENTS

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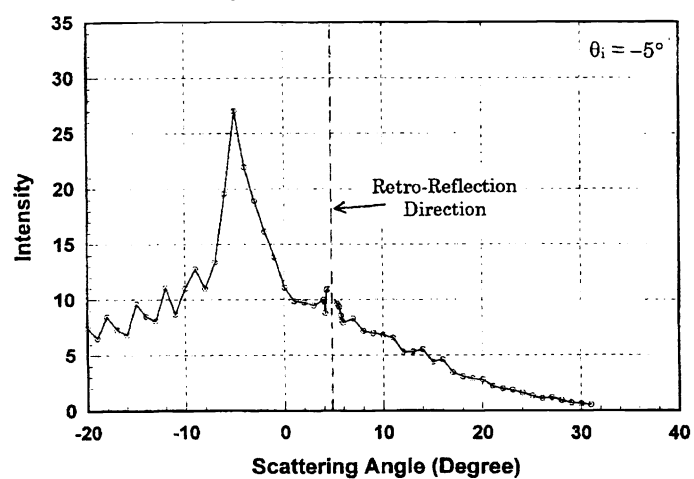
Gain Medium on 2-D Gold (No Pump Beam)



Main Beam at $0.6328 \mu\text{m}$; Pump Beam at $0.5145 \mu\text{m}$

Figure 6 (a). The intensity measurement of a dye-doped 2-D Gold rough surface with no pump beam.

Gain Medium on 2-D Gold (With Pump Beam)



Main Beam at $0.6328 \mu\text{m}$; Pump Beam at $0.5145 \mu\text{m}$

Figure 6 (b). The intensity measurement of a dye-doped 2-D Gold rough surface with pump beam.

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