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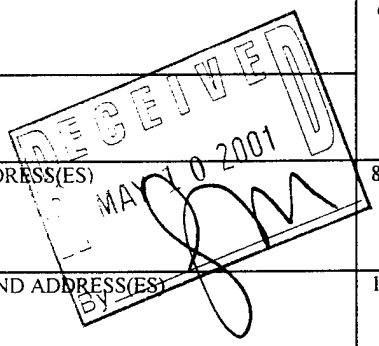

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We demonstrated for the first time a saturable polarization rotation in Bacteriorhodopsin that acts as an effective optical limiter. Using a multiple pass scheme and cross polarizes we achieved a ninety degree polarization rotation of a probe beam and a corresponding optical density of 10^{-4}.

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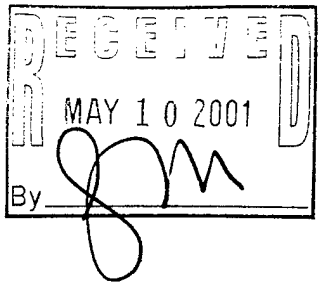

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FINAL REPORT

NONLINEAR OPTICAL PROPERTIES OF BACTERIORHODOPSIN

DAAH04-96-1-0251

JULY 1, 1996 TO JUNE 30, 1999

ABSTRACT

We demonstrated for the first time a saturable polarization rotation in Bacteriorhodopsin that acts as an effective optical limiter. Using a multiple pass scheme and cross polarizes we achieved a ninety degree polarization rotation of a probe beam and a corresponding optical density of 10^{-1}.

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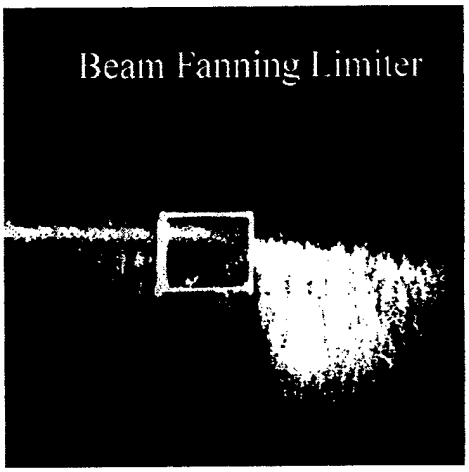
NONLINEAR OPTICAL PROPERTIES OF BACTERIORHODOPSIN

STATEMENT OF THE PROBLEM

An optical limiter is a device that limits the transmission of laser light to a safe value. For example, if a laser beam is incident on the optical limiting device, the device will switch blocking or reducing the laser beam intensity to a safe value. The ideal optical limiter will not only reduce the incident intensity, but it will do so over a broad spectral range, for a large field-of-view, for both pulsed and continuous laser sources, for laser light incident from any direction, and for both high and low intensity laser beams. Moreover, it will allow for continuous observation of outside scenes without degradation while simultaneously rejecting the potentially dangerous incident laser light.

While limiting the transmission of the incident laser light is the mission of the optical limiter, how quickly it limits the transmission is crucial to its success. If an intense laser beam is suddenly incident onto a sensor, the sensor can be protected by the addition of a neutral density filter in front of the sensor, reducing the incident intensity to a safe value. However, if this is done slowly, the sensor can be damaged before the neutral density filter is completely inserted. Therefore, it is necessary for the optical limiter to switch-in the neutral density filter fast enough to prevent damage. For example, one important sensor which has need for protection by an optical limiting device is the eye.[1,2] In this case the limiting device must switch fast enough to limit the energy density delivered on the cornea by a short laser pulse to less than $5\mu J/cm^2$.

Many techniques have been explored as an optical limiter. The majority of these exploit nonlinear optical effects, such as high intensity induced self-focusing or defocusing, two-photon absorption, excited state absorption, reverse saturable absorption and intensity-dependent scattering [3-11]. Due to the type of nonlinearity, these passive optical devices typically exhibit high transmission for low-incident intensity and low transmission for high incident intensity. Consequently, these limiters can be considered high intensity limiters. Complimenting these high intensity limiters are low intensity schemes, such as, (shown in Photograph 1 above) the photorefractive



limiter [12]. For this limiter, the incident laser light induces an index of refraction change that effectively scatters low intensity light in all directions. Our work, described in this report, is a new approach that can provide optical limiting at both low and high intensities, based on the nonlinear optical properties of bacteriorhodopsin.

SUMMARY OF THE MOST IMPORTANT RESULTS

Bacteriorhodopsine (BR) has received much attention recently due to its potential for real-time holographic recording, optical pattern recognition, and nonlinear optical effects. BR is the light harvesting protein in the purple membrane of the microorganism called halobacterium holbium, produced in salt marshes, and is closely related to the visual pigment rhodopsine. It displays a characteristic broadband evisceration profile in the visible spectral region, and when the molecule absorbs light it undergoes several structural transformations in a well-defined photocycle. One of its long list of interesting attributes is its inherent photoinducible optical anisotropy. It is this anisotropy which is explored in this letter.

The apparatus for our investigation is shown in Figure 1. The output beam from a linearly polarized argon-ion laser at 514.5 nm is split into two beams. One beam is polarized as P_1 and directed through the BR sample. The second beam is polarized at 45° relative to the first beam and is directed through the BR sample at a small crossing angle (~2°) relative to the first beam. This second beam then passes through a second polarizer P_2 which is parallel to the first. When the first (pump) beam is blocked, the second (probe) beam passes through the BR without polarization rotation, hence with minimal loss through the second polarizer (P_2). When the pump beam is unblocked the probe beam experiences a polarization rotation and is partially blocked by the second polarizer. As the intensity of the pump beam is increased, the polarization rotation of the probe beam is observed to increase and then saturate. Since the saturating effect is observed to occur at low light intensities, the rotation is in practice quickly independent of the incident

intensity of either beam and the degree of rotation is "fixed". Figure 2 shows a plot of the observed rotation versus the incident pump intenisty. For a thickness of 50 microns we observe a saturating rotation angle of 23°. The thickness of the BR sample, therefore, can be easily selected to produce a desired fixed amount of rotation for a large range of incident intensities. For a rotation of 90° , for example, a thickness of about 0.3 mm is required for the samples we have examined. For a 0.3 mm sample, used in the apparatus shown in Fig. 1, a 90 polarization rotation limits the intensity of the probe beam to at least 10^{-4} of the incident intensity depending on the quality of the polarizers, P_1 and P_2.

We have used a simple three level picture, depicted in Figure 3, to model the induced birefringence in bacteriorhodopsin. The incident light excites the trans molecular state, $|t\rangle$, to state $|1\rangle$ which immediately decays non-radiatively to the cis molecular state $|c\rangle$. The long lived cis state then rapidly accumulates a population and before long most of the molecules which are capable of absorbing the incident polarized light are in the cis molecular state. As a result, the sample becomes anisotropic. Quantitatively, $N = N_t + N_c$, where N is the total density of molecules per unit volume and N_t and N_c are the concentrations of molecules in the trans and cis states respectively. The rate of change of molecules in the cis state is then given by the rate equation:

$$\frac{dN_c}{dt} = \left(\frac{\sigma q I}{h\omega}\right)(N - N_c) - \frac{N_c}{\tau} \tag{1}$$

where I is the incident pump intensity, τ is the lifetime of the cis state, q is the quantum yield, and σ is the absorption cross-section from trans to cis states. The solution to equation (1) is:

$$N_c(t) = N\frac{I}{I + I_1}\left[1 - \exp\left[\left(-\frac{t}{\tau}\right)\left(1 + \frac{I}{I_1}\right)\right]\right] \tag{2}$$

where I_1 is equal to $h\omega/q\sigma\tau$ and is the saturation intensity or the intensity for which the population of the cis state saturates. For $\tau \rightarrow \infty$ the stationary population of the cis state therefore becomes:

$$N_c = N\frac{I}{I+I_1} \tag{3}$$

Using the Lorentz local field model where the molecule radius is much less than an optical wavelength, the susceptibility tensor can be written as the sum of an isotropic part and an anisotropic part,

$$\chi^0 + \chi^{an} \tag{4}$$

The isotropic part of the susceptibility, χ^0, is given by $\frac{N_c\alpha^0}{B}$ while the anisotropic part is given by $\frac{N_c\alpha^{an}}{B^2}$. In these expressions B is defined as $B = 1 - \frac{4\pi\alpha^0 N_c}{3}$ and α^0 is the isotropic part of the polarizability tensor while α^{an} is the anisotropic part. The dielectric permittivity tensor:

$$\varepsilon = 1 + 4\pi\chi \tag{5}$$

can also be written in terms of an isotropic and anisotropic part:

$$\varepsilon = \varepsilon^0 + \varepsilon^{an} \tag{6}$$

From the definition of isotropic and anisotropic parts of the polarizability tensor we can write

$$\varepsilon^0 = 1 + \frac{4\pi N_c\alpha^0}{B} \tag{7a}$$

$$\varepsilon^{an} = \frac{4\pi N_c\alpha^{an}}{B^2} \tag{7b}$$

Using expressions (3) and (7b) we can write

$$\varepsilon^{an} = \frac{4\pi NI\alpha^{an}}{(I+I_1)B^2} \tag{8}$$

In the laboratory coordinate system, the dielectric permittivity tensor can be found from

$$\varepsilon_L = D^{-1}\varepsilon D \tag{9}$$

where D is the Eulerian matrix transformation in three dimensional space, D = f(Θ, β, γ) in terms of Eulerian angles Θ, β, γ. For simplicity we take the Eulerian angles Θ=0, γ=0, and β = 0 to π/2. In this case the permittivity tensor becomes

$$\varepsilon_L = \begin{bmatrix} \varepsilon_0 + k\cos 2\beta & k\sin 2\beta & 0 \\ k\sin 2\beta & \varepsilon_0 - k\cos 2\beta & 0 \\ 0 & 0 & \varepsilon_0 - k \end{bmatrix} \tag{10}$$

where

$$\alpha_0 = \frac{(\alpha_{11} + \alpha_{22})}{2}$$

$$k = \frac{2\pi N_c (\alpha_{11} - \alpha_{22})}{B^2}$$

Using the tensor-vector evolution algorithim described in reference [5], we can write the electric field along the propagation direction z as:

(11)

$$\vec{E}(z) = Me^{i\Psi}\vec{E}(0)$$

where

$$M = e^{i\frac{k_o z}{2}(n_1+n_2)}\begin{bmatrix} \cos\phi + \frac{i}{\sqrt{2}}\sin\phi & 2ik\sin\phi\sin 2\beta & 0 \\ 2ik\sin\phi\sin 2\beta & \cos\phi - \frac{i}{\sqrt{2}}\sin\phi & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{12}$$

$\phi = \frac{(n_1 - n_2)k_0 z}{2}$, $k_0 = \frac{2\pi}{\lambda_0}$, λ_0 is the vacuum wavelength, $\Psi = (\varepsilon_0)^{\frac{1}{2}} k_0 z$ is the isotropic phase, and $(\varepsilon_0)^{\frac{1}{2}}$ is the isotropic index n_0. Meanwhile, n_1 and n_2 are given by

$$n_{1,2} = (\varepsilon_0)^{\frac{1}{2}}\left[1 + \frac{k_{1,2}}{\varepsilon_0}\right] \tag{13}$$

Using the expression for n_1 and n_2, ϕ can be written as

$$\phi = \frac{2\pi^2 nNzI(\alpha_{11} - \alpha_{22})}{\lambda_0 n_0 (I_1 + I) B^2} \tag{14}$$

For the case of saturation, $I \gg I_1$, the expression for ϕ becomes

$$\phi = 2\frac{\pi}{\lambda_0} z(\Delta n) \tag{15}$$

where

$$\Delta n = \frac{\pi N(\alpha_{11} - \alpha_{22}) I}{B^2 n_0 (I + I_1)} \tag{16}$$

For the apparatus shown in Fig. 1, with polarizers P_2 and P_3, the electric field seen by the detector is given by

$$\vec{E}_{out} = P_3 M e^{i\Psi} P_2 \vec{E}_{in} \tag{17}$$

where $Me^{i\Psi}$ is the transformation matrix given in equation (12) and P_2 and P_3 are the matrices for the linear polarizers, given by

$$P_2 = P_3 = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} k_{11} \tag{18}$$

In the expression for P_2 and P_3 the constant k_{11} is the transmission coefficient for incident light polarized along the polarizer axis.

Using equations (17) and (18) the electric field and incident intensity on the detector become

$$\vec{E}_{out} = k_{11} (\cos\phi) e^{i\Psi} \vec{E}_{in} \tag{19}$$

$$I_{out} = k_{11}^2 (\cos^2\phi) I_{in} \tag{20}$$

For the experiment in Figure 1, the measured ratio of I_{out}/I_{in} was 0.846. Using (20), this gives a value of 23° for ϕ. The thickness of the bacteriorhodopsine film was 50 microns. From equation (15) we calculate Δn to be 6.6×10^{-4} at $\lambda_0 = 515$ nm. Using this value of Δn, we then calculate Z =390 nm as the thickness required to produce a ϕ of 90°. We used a variation in incident intensity which ranged from 0.014 to 1.7 W/cm^2 . Our results were independent of intensity. That is, we found a 18% decrease in the transmission of I_s for $I_p = 1.7$ W/cm^2. Figure 4 shows a typical response time curve for the induced rotation. The response time was observed to vary inverselly with pump intensity and was 62 ms for an incident intensity of 0.454 W/cm^2.

In addition to our observations with P_1 parallel to P_2, we also observed the transmission behavior when P_2 is at some angle β to P_1. In this case the induced birefringence causes the transmission to rise from zero and reach a value depending on β. In particular the transmitted

intensity is given by

$$I_t = 2k^2 \sin^2 2\beta \sin^2 \phi \qquad (21)$$

As shown in Figure 5 the experimental results are in good agreement with the predictions from the three-level model.

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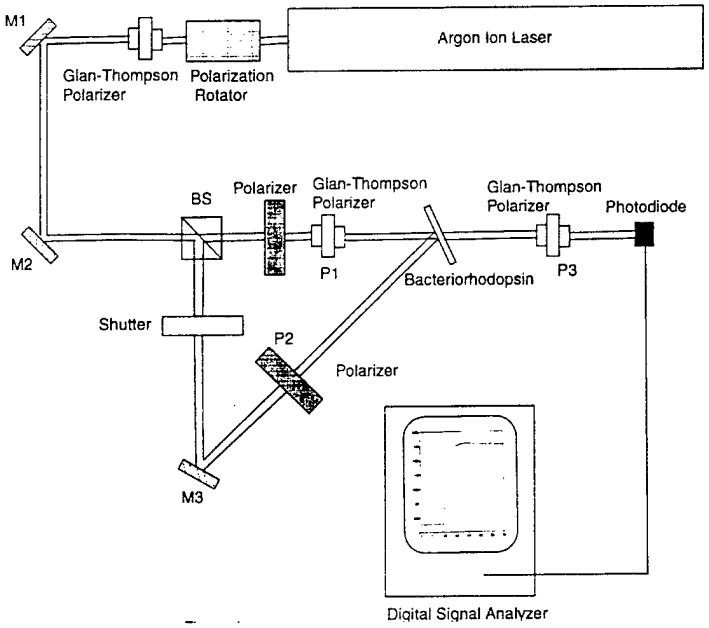

Figure 1. Apparatus

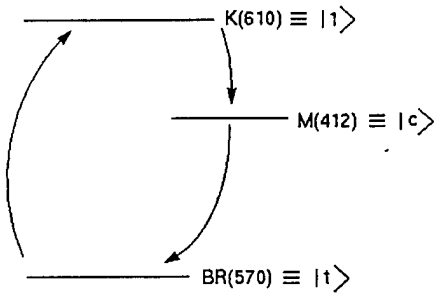

Figure 2. Three-Level System

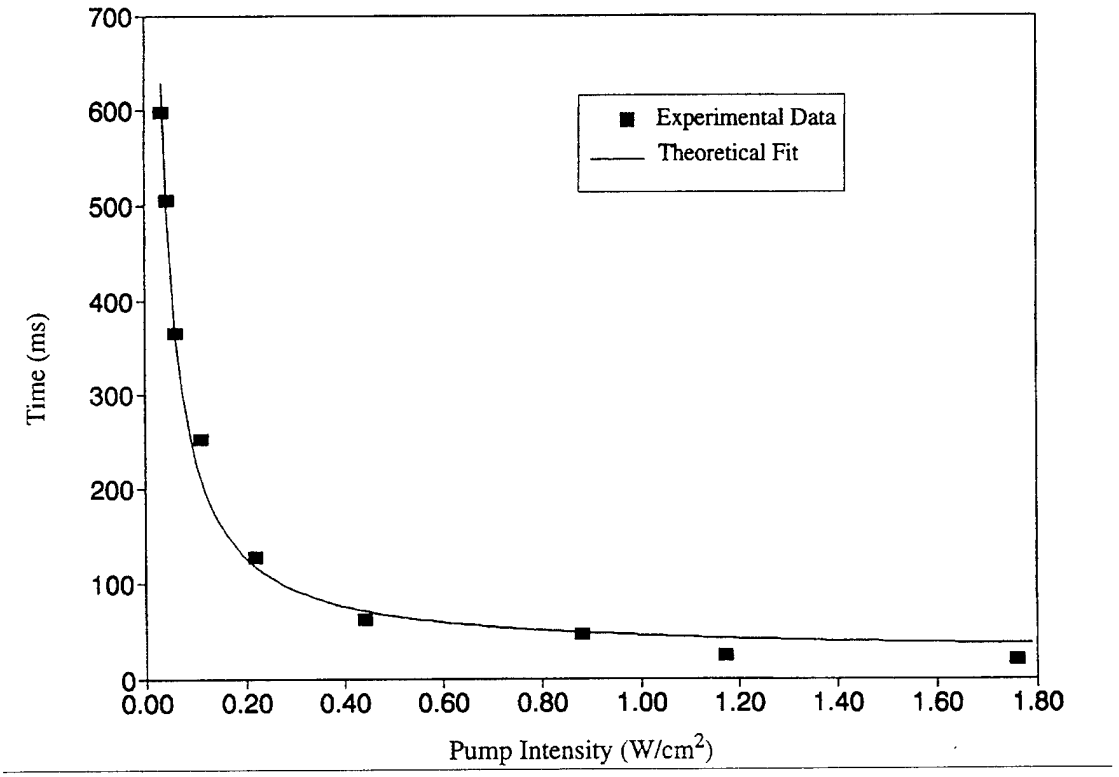

Figure 3. Response Time Verses Incident Intensity

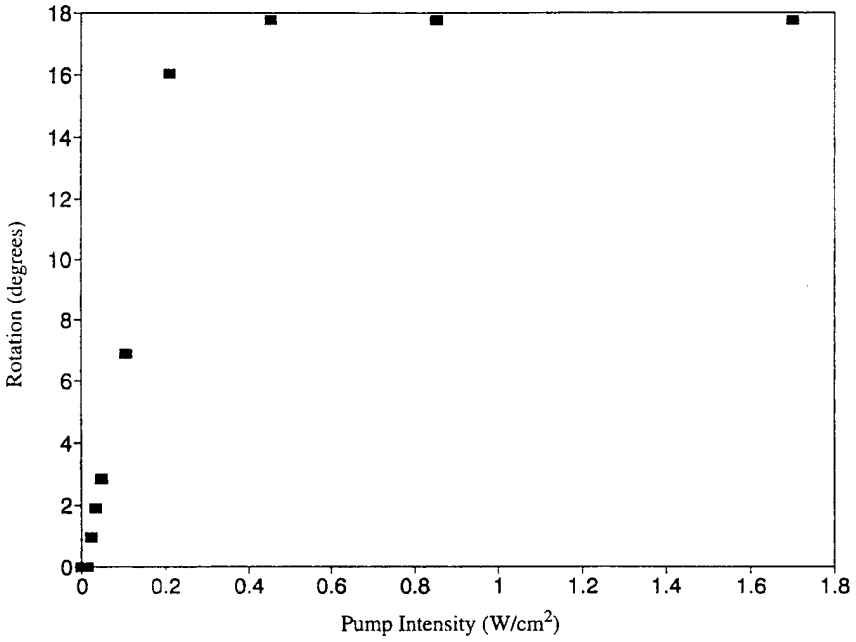

Figure 4. Saturation of Rotation with Intensity

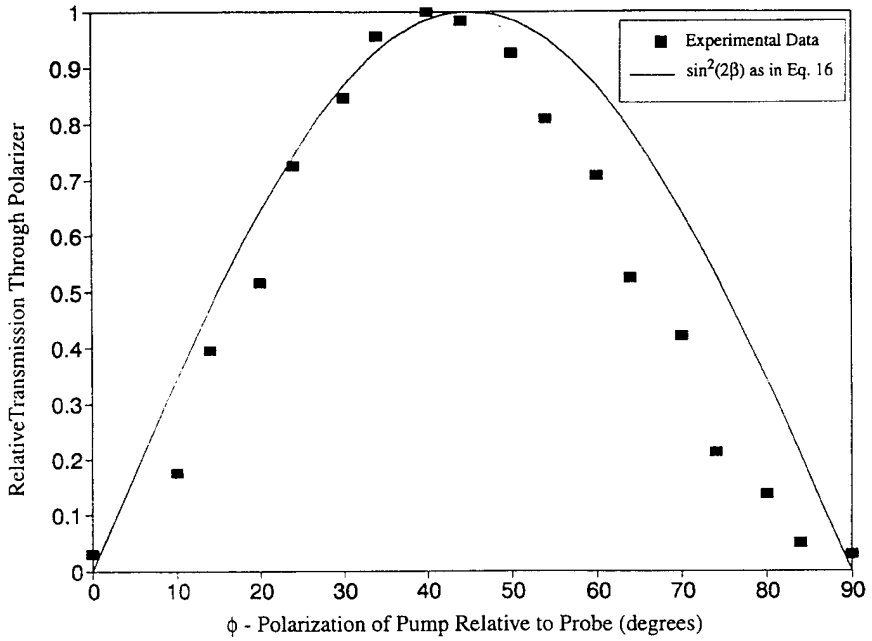

Figure 5. Induced Birefringence with incident polarization

MANUSCRIPTS

G. Dovgallenko, M. Klotz, and Greg Salamo, Optically induced birefringence in bacteriorhodopsin, Applied Physics Letters, 68, 287, 1996.

PARTICPANTS

Mr. Klotz received his Ph.D. degree at the University of Arkansas in the summer of 1999 as a result of support by this grant.

INVENTIONS

We have not reported or claim any inventions.

TECHNOLOGY TRANSFER

Although we hve not established a transfer of technology we will at the project completion.