Theoretical Analysis of Photon-Mode Super-Resolution Optical Memory Using Saturable Absorption Dye

Tsuyoshi TSUJIOKA,1 Toshio HARADA,1 Minoru KUME,1 Kazuhiko KURORI1 and Masahiro IRIE2

1New Materials Research Center, SANYO Electric Co., Ltd., 1-1 Dainichi Higashimachi, Moriguchi, Osaka, 570 Japan,
2Institute of Advanced Material Study, Kyushu University, 6-1 Kasuga-koen, Kasuga, Fukuoka, 816 Japan

(Received February 16, 1995; Accepted June 8, 1995)

The possibility of a super-resolution optical memory using a saturable absorption dye mask layer is theoretically discussed. An equation which estimates the transmittance change of the mask layer has been derived. The numerical simulation shows that an efficient super-resolution is performed when the initial optical density of the mask layer is high.

Key words: optical memory, saturable absorption, super-resolution, optical density

1. Introduction

Continuous efforts have been made to increase the recording density of optical memories. One of the approaches to the high density memory is super-resolution disk technologies such as MSR (magnetically induced super-resolution).1,2) The super-resolution is based on the nonlinear response of the recording media to the readout laser light intensity.3)

Photo-bleachable organic dye mask layers are expected to show such a nonlinear response, when used as shown in Fig. 1.4,5) The transmittance of the mask layer increases with increasing readout light intensity.6) Photo-bleachable organic dye mask layers are expected to show such a nonlinear response, when used as shown in Fig. 1.4,5) The transmittance of the mask layer increases with increasing readout light intensity. The effective transmittance at the central area is higher than the transmittance of the spot edge, because of the gaussian intensity distribution of the laser light. It can be shown that an effective super-resolution spot is formed by taking the product of the readout laser intensity and the transmittance of the mask layer as shown in Fig. 2.

The aim of this paper is to derive an equation which can estimate the transmittance change of the saturable absorption dye mask layer, and predict the necessary conditions to obtain efficient super resolution.

2. Transmittance Change of Saturable Absorption Dye Mask Layer

We assumed the following simple conditions.

(1) A molecule in the ground state (molecular absorption coefficient: \( e \ M^{-1} cm^{-1} \)) is converted to the excited state by absorbing a photon.

(2) The molecule in the excited state cannot absorb the photon.

(3) The excited state has a lifetime of \( \tau s \).

We have already derived an equation for photon-mode photochromic masked super-resolution media which have an infinite lifetime.8) This equation is modified to be applicable to a system which has a finite lifetime of the photo-excited state. The readout beam is assumed to pass through the mask layer (forward), be reflected by the recording layer with reflectance \( R_{\text{rec}} \) and then be passed through the mask layer again (backward) as shown in Fig. 1. According to the above conditions, the transmittance \( T \) of the mask layer, which contains molecules in the ground state of concentration \( C_g M \), is given by the following equation,

\[
T = \exp(-2.3eCgL) ,
\]

where \( L cm \) is the thickness of the mask layer, \( eCgL \) has the time dependence and \( eCgL \) is equal to the optical density.

The total reflectance \( R \) of the medium is

\[
R = R_{\text{rec}}T^2 .
\]

The total number of molecules in a unit volume (1 liter) \( N/1 \) is constant and given by

\[
N = N_g + N_e ,
\]

where \( N_g \) and \( N_e \) are the number of molecules in the ground and in the excited state in a unit volume.

During light irradiation, the photo-exciting and deactivation processes coexist. Upon irradiation with light of wavelength \( \lambda m \) and intensity \( P W \), the number of photons absorbed by the mask layer during infinitesimal time \( dt s \) in the forward light pass and backward light pass are given by

\[
\text{forward: } dn = \frac{P}{hc} (1-T)dt ,
\]

and

\[
\text{backward: } dn = \frac{P}{hc} T \cdot R_{\text{rec}}(1-T)dt ,
\]

where \( h = 6.626 \times 10^{-34} J \cdot s \) is Planck’s constant and \( c = 3.00 \times 10^8 m/s \) is the speed of light in vacuum. Therefore, the total number of photons absorbed by the mask layer is

\[
dn + dn = \frac{P}{hc} (1-T)(1+R_{\text{rec}}T)dt .
\]

This is equal to the number of molecules that was in the ground state and is reduced by the photo-excitation. On the other hand, the number of deactivated molecules during infinitesimal time \( dt s \) is proportional to the number of excited molecules with proportional constant \( 1/\tau s^{-1} \).

\[
\frac{1}{\tau} N_e \cdot dt = \frac{1}{\tau} (N_0 - N_e)dt .
\]
Fig. 1. Structure of a super-resolution medium having an organic dye mask layer. The mask layer contains saturable dye molecules. A readout laser beam is reflected by the reflective layer after passing through the mask layer.

![Power profile of a readout laser](image)

![Transmittance profile of a mask layer](image)

Therefore, the change in the number of molecules in the ground state is given by

\[ dN_c = \frac{1}{\tau} (N_t - N_c)dt - \frac{P\lambda}{hc} (1-T)(1+R_{rec}T)dt. \]  

(8)

The number of molecules in unit volume is expressed by the molarity as shown below for the molecules in the ground state and for all molecules:

\[ N_c = C_c LS N_c \times 10^{-3}, \]  

(9)

\[ N_t = C_c LS N_t \times 10^{-3}, \]  

(10)

where \( N_c = 6.02 \times 10^{23} \text{ mol}^{-1} \) is Avogadro’s constant, \( S \text{ cm}^2 \) is the irradiation area, \( C_c \text{ M} \) is the total concentration of dye and the factor \( 10^{-3} \) is for the correction of units. From Eq. (1), the following equations are derived,

\[ \frac{dT}{dt} = -2.3eLT \frac{dC_c}{dt}, \]  

(11)

\[ C_c = -\frac{1}{2.3eL} \ln T. \]  

(12)

Initial transmittance \( T_0 \) (before irradiation) is given using total dye molarity \( C_t \),

\[ C_c = -\frac{1}{2.3eL} \ln T_0. \]  

(13)

Using Eqs. (9)-(12) and (13), Eq. (8) is transformed as follows,

\[ \frac{dT}{dt} = \frac{1}{\tau} T \ln \frac{T_0}{T} + \alpha S \lambda e T(1-T)(1+R_{rec}T), \]  

(14)

where \( \alpha = 2.3 \times 10^5/N_c hc = 1.9 \times 10^4 \text{ J m}^{-1} \text{mol}^{-1} \) is a constant.

Note that there are many independent parameters in Eq. (14) that affect the transmittance change of the mask layer, e.g., \( \tau, P/S, \lambda, \epsilon, R_{rec} \) and \( T_0 \). This equation determines the transmittance of the mask layer when irradiated with light of a uniform intensity.

Under the condition of low optical density approximation \( (A = \epsilon LC_c \leq 0.2) \), the exponential of Eq. (1) may be expanded as follows,

\[ T \sim 1 - 2.3eLC_c. \]  

(15)

Moreover, by considering the stationary state \( (\partial T/\partial t = 0) \) and a single pass through the mask layer \( (R_{rec} = 0) \), Eq. (14) can be simplified as follows,

\[ \frac{\Delta A}{A_0} = \left( \frac{1 + \frac{hcN_c S}{2.3 \times 10^5 P\lambda e T}}{1+R_{rec}} \right)^{-1}, \]  

(16)

where \( A_0 \) is the initial absorbance \( (\epsilon LC_{rec}) \) and \( \Delta A = A_0 - A \). This approximation, however, is not practical, because the initial transmittance of the mask layer must be low in order to avoid crosstalk from neighboring recording marks. Instead, we have to solve Eq. (14) numerically.

3. Several Numerical Simulations

We calculated the irradiation time dependence of \( T \) by Eq. (14) for several conditions of \( P, \epsilon, \tau \) and initial transmittance \( T_0 \). The values of \( \lambda = 680 \times 10^{-3} \text{ m}, R_{rec} = 0.8 \) and \( S = 1.0 \times 10^{-8} \text{ cm}^2 \) were kept constant. \( P = 4.0 \times 10^{-3} \text{ W}, \epsilon = 20000 \text{ M}^{-1} \text{ cm}^{-1} \), and \( T_0 = 0.2 \) were employed as standard conditions.

Figures 3 and 4 show the plots of normalized total reflectance \( R/R_{rec} \) with varying readout power \( P \) and molar extinction coefficient \( \epsilon \), respectively. When \( P \) and \( \epsilon \) are high, the stationary state with higher transmittance is readily reached. The response to the irradiation should be nonlinear in order to obtain the super-resolution effect. Ideally, at the low laser power range \( (1-2 \text{ mW}) \), the increasing speed of \( R/R_{rec} \) should be very small, and the saturated value of \( R/R_{rec} \) after more than 100 ns should be small. The curves in Fig. 3 show such a tendency.

Figure 5 shows the excited state lifetime dependence of...