Laser-Induced Fluid Motion on a Dye/Polymer Layer for Optical Data Storage

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The interaction of highly focused light with active dye/polymer layers has received considerable attention in these days because it creates micron-size pits useful as optical data storage sites. Pit formation during laser marking takes place in a very short period of time, usually between 50 and 500 nanoseconds (ns). It consists of the following steps: the material heats, then melts, and finally flows in the lateral direction. In other words, the highly focused laser pulse results in an uneven temperature profile on the surface of an active dye/polymer layer; the variation in surface tension from one point to another creates tangential stresses on the free surface and a capillary fluid motion is therefore induced. As a result, the free surface of the pit center depresses and a bulge is formed around the pit. To be a good recording medium, an active dye/polymer layer should have high light absorption and reflectivity at the wavelength of the emitting laser beam. The greater the absorption, the faster the marking. The encoded data can be read back based on the difference in the surface reflectivity between the surroundings and the pit.

The detailed mechanism of pit formation is not fully understood yet, but it is a typical transport phenomenon problem for a chemical engineer. Surprisingly, however, almost no attention has been given to this issue by chemical engineers; most of the work has been done by optical physicists and published in physical journals. For example, Novotny and Alexandru (1979) calculated the temperature profile during the light-induced marking process and described the mechanism of structure change in dye/polymer systems. Wrobel et al. (1982) found that the surface tension forces were unimportant in rapid pit formation in thin polymer films. Broer and Vriens (1983) presented models to describe the hole-opening process, and concluded that surface tension is one dominant force during the process. Suh and coworkers investigated the writing mechanism in thin-textured inorganic films, and believed the surface tension gradient force to be one of the most effective driving forces to overcome the energy barriers for forming a pit (Suh and Craighead, 1985; Suh et al., 1985). Chung (1986) derived the governing equations for the transitional laser-induced fluid motion in extremely thin dye/polymer layers. The temperature rise in an optical layer is calculated based on the complicated Poynting vector theorem and Maxwell equations. Chung found that there are two factors controlling the pit formation, namely, the surface tension force and the gravitational force, the former being much more important than the latter.

In this paper we attempt to extend our previous work to simulate the transitional pit formation in a relatively thick optical recording layer, while the temperature profile of the recording layer is determined using a conventional chemical engineering method. Calculations will be compared to the previous experimental results to determine if the model is meaningful.

Theory

In addition to the assumptions used in the previous paper (Chung, 1986), we also assume that the optical recording layer is relatively thicker than the pit size in this work. As a result, the heat transfer and fluid motion in the lateral direction are more important than in the thickness direction. The effect of substrates on temperature is therefore neglected.

Temperature rises

The laser-induced local heating of a recording layer is a typical heat transfer problem and may be expressed as follows:

$$\frac{\partial T}{\partial t} = k\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial T}{\partial r}\right) + \frac{1}{r^2}\frac{\partial^2 T}{\partial \theta^2} + \frac{\partial^2 T}{\partial z^2}\right] \tag{1}$$

where T is the temperature rise, t is the time, k is the thermal diffusivity of a recording layer, and r and z are the lateral and thickness distances, respectively, as shown in Figure 1. The boundary condition of Eq. 1 is that the top surface is an energy ring source, which results from the illumination of a laser beam with a light intensity $I(r)$, following a Gaussian distribution

$$I(r) = Io \exp(-r^2/\delta^2) \tag{2}$$

where Io is the light intensity at the center of the beam and δ is the Gaussian beam radius at e^{-1} points. This equation indicates

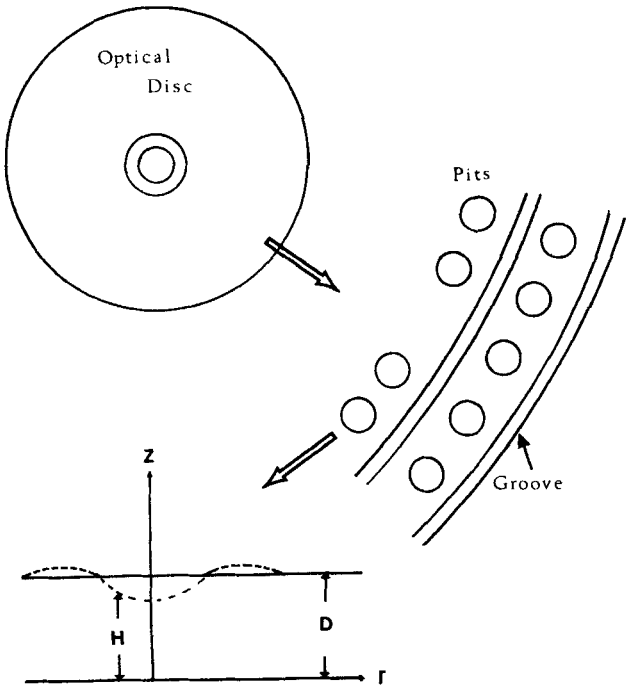

Figure 1. Optical disc, pits, and cross section of a pit.

that most of the laser energy is concentrated in the center, with the energy falling off rapidly toward the periphery. Integration of Eq. 1 with respect to r yields the total energy, Q, of the marking power, i.e., $Q = \pi\delta^2 Io$.

The temperature profile in a recording layer may be calculated using the principle of superposition, which assumes that there are infinite uniform illumination rings on the incident surface. For an instantaneous heat imposed on a single ring, the resultant temperature profile in its surroundings is (Carslaw and Jaeger, 1959)

$$T(r, z, t) = \frac{q}{8\rho c(\pi kt)^{3/2}} \cdot \exp\left(-\frac{r^2 + r'^2 + (z - H)^2}{4kt}\right) Bo\left(\frac{rr'}{2kt}\right) \quad (3)$$

where q is the heat liberated from a laser beam on an incident ring of radius r' from the pit center; its amount is equal to $2\pi r' I(r') dr' dt$. D is the thickness of the optical layer; Bo is the modified Bessel function of order zero. For infinite energy rings liberated on a recording layer for a period of time, the resultant temperature profile may be calculated by integrating Eq. 3 with respect to dr as well as dt:

$$T(r, z, t) = \frac{Io}{4\rho c}\int_0^t \frac{1}{(\pi k^3 t^3)^{1/2}} \exp\left(-\frac{r^2 + (z - H)^2}{4kt}\right) \cdot \left\{\int_0^\alpha \exp\left[-r'^2\left(\frac{1}{4kt} + \frac{1}{\delta^2}\right)\right] Bo\left(\frac{rr'}{2kt}\right) r' dr'\right\} dt \quad (4)$$

Since (Gradshteyn and Ryzhik, 1965)

$$\int_0^\alpha x e^{-\alpha x^2} Bn(\beta x)\, Jn(\gamma x)\, dx = \frac{1}{2\alpha} \cdot \exp\left(\frac{\beta^2 - \gamma^2}{4\alpha}\right) Jn\left(\frac{\beta\gamma}{2\alpha}\right) \quad (5)$$

and $Bo(0) = 1$, Eq. 4 becomes

$$T(r, z, t) = \frac{\delta^2 Io}{2\rho c(\pi k)^{1/2}}\int_0^t \frac{1}{t^{1/2}(4kt + \delta^2)} \cdot \exp\left(-\frac{(z - H)^2}{4kt} - \frac{r^2}{4kt + \delta^2}\right) dt \quad (6)$$

This is the general equation for the temperature distribution in a thick recording layer. The above derivations have been suggested by Novotny and Alexandru (1979); however, the resultant equations are slightly different. Equation 6 indicates that the temperature drops smoothly near the incident surface, while it drops dramatically with an increase in the distance away from the top surface.

Fluid motion

Considering a spatially varying temperature distribution imposed on the free surface of a thin organic film by a laser beam, the film heats and melts and then flows because of the variation of surface tension and gravitational forces. Chung (1986) has developed the governing equation for the free surface of a pit formation:

$$\frac{\partial H}{\partial t} + U_1\left(\frac{\partial \sigma}{\partial Ts}\right)\left(\frac{1}{r}\frac{\partial Ts}{\partial r} + \frac{\partial^2 Ts}{\partial r^2} + \frac{1}{U_1}\frac{\partial Ts}{\partial r}\frac{\partial U_1}{\partial r}\right) + \rho g U_2\left(\frac{1}{r}\frac{\partial H}{\partial r} + \frac{\partial^2 H}{\partial r^2} + \frac{1}{U_2}\frac{\partial H}{\partial r}\frac{\partial U_2}{\partial r}\right) = 0 \quad (7)$$

where H is the pit thickness. U_1 and U_2 are defined as

$$U_1 = \int_0^H \int_0^{z'} \frac{1}{\mu} dz' dz \quad U_2 = \int_0^H \int_0^{z'} \frac{1}{\mu}(z' - H)\, dz' dz \quad (8)$$

It may be worthwhile to point out some limitations of Eq. 7 before going into the calculation. The derivation of Eq. 7 was based on polymeric materials. It is not applicable for the hole formation for inorganic materials because:

1. Their melt viscosity is extremely low and, as a result
2. The inertial force cannot be neglected in the governing equation for this case.

Blom's (1983) model is one of the examples suitable for inorganic cases. Strictly speaking, Eq. 7 is valid only in the beginning of pit formation. Once the pit is well developed, temperature becomes extremely high and the viscosity of polymers becomes low. Therefore, the importance of inertial force has to be included in the calculation.

The initial and boundary conditions of Eq. 7 are:

1. $H = D$, at $t = 0$
2. $dH/dr = 0$, at $r = 0$, $t > 0$
3. $H = D$, at $r = \infty$, $t > 0$

Material Parameters and Numerical Schemes

A proprietary polyamide was chosen for this study. Its melt is a Newtonian fluid, and its dynamic viscosity may be expressed as μ (N · s/m^2) $= 5.8 \times 10^{-7} \exp(8{,}858/T)$. Its k is 0.88×10^{-7} m^2/s, $c\rho$ is about 2.092 kJ/kg · K. The surface tension/temperature gradient is unknown, but it is usually in the range from -0.05 to -0.15 mN/m · K. Diode laser marking was conducted and reported by Kuder and Nikles (1985) using a laser

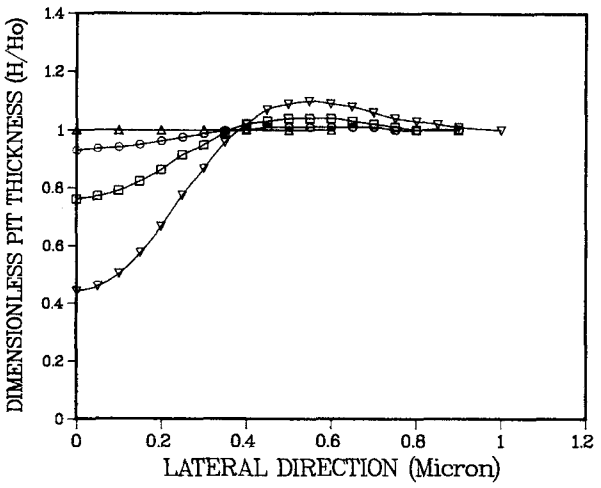

Figure 2. Pit formation as a function of time.
△ t = 0; ○ t = 200; □ t = 250; ▽ t = 300 ns

power of 8 mJ/s while varying the pulse width from 20 to 1,000 ns. The beam radius is about 1 μm. The results can be summarized as follows: Pits were created once the laser pulse was longer than 200 ns, but well-defined pits were generated after 300 ns. The playback signal-to-noise ratio remains quite high if the marking pulse is in the range of 300 to 500 ns.

Similar calculation methods (Chung, 1986) were used to obtain the transitional temperature and pit contour. A $d\sigma/dTs$ of -0.05 mN/m · K, and a layer thickness of 5 μm were used in the calculations. For simplicity, we also assume that H is approximately equivalent to D in the calculations of Eqs. 6 and 8. The laser reaches its power instantaneously when the power is turned on.

Results and Discussion

According to Eq. 7, the rate of pit thickness change is controlled by two factors, the surface tension force and the gravity. Since the film is relativity thin (<10 μm), U_2 is much smaller than U_1. Because the temperature gradient term, $(\partial\sigma/\partial Ts)(\partial Ts/\partial r)$, is much greater than the thickness variation term, $\rho g(\partial H/\partial r)$, the surface tension force dominates the fluid motion

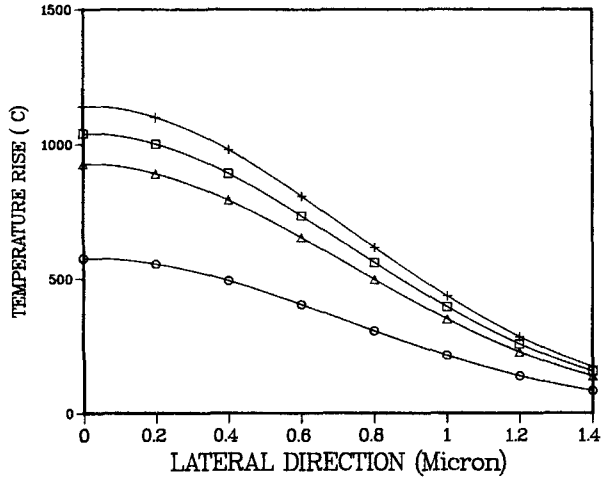

Figure 3. Lateral temperature rises with time.
○ t = 80; △ t = 200; □ t = 250; + t = 300 ns

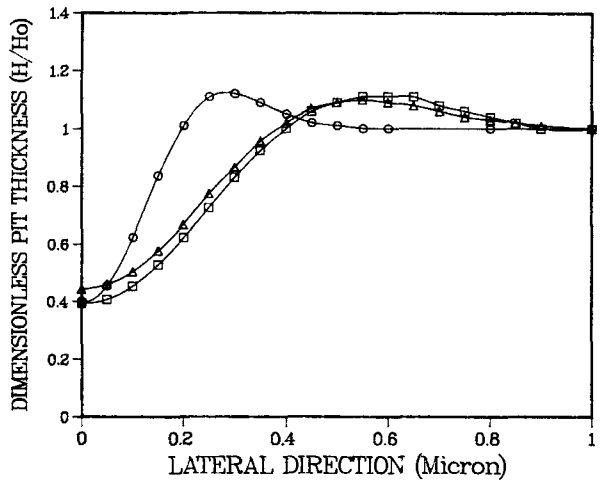

Figure 4. Effect of beam size and thermal diffusivity on pit formation.
○ δ = 0.5 μm; $k = k$; t = 60 ns
△ δ = 1.0 μm; $k = k$; t = 300 ns
□ δ = 1.0 μm; $k = 0.22 \times 10^{-7}$ m^2/s; t = 115 ns

and the effect of gravity is therefore neglected. Since both $\partial\sigma/\partial Ts$ and $\partial Ts/\partial r$ are negative, the surface tension force induces a material flow-out from a hot spot to its cool surroundings.

Figure 2 illustrates the calculated transitional free surface of a pit. The original surface is flat. Once the film is exposed to a laser beam, the temperature of the center area is increased and results in a low surface tension region. When a critical temperature is reached (after 80 ns exposure), it starts to deform and be depressed due to the drawing forces imposed on the hot spot from the cool surroundings. This creates an outflow in the upper part of the liquid layer, and a portion of the surface away from the pit center rises above its initial value and forms a bulge. Detectable pits were formed after 200 ns, while noticeable pits were developed after a 300 ns pulse. These calculated results are in agreement with previous experimental data. Therefore, the required time for the initial deformation and the deformation speed can easily be predicted using this mathematical model. Both pit width and depth increase gradually with pulse length, and these phenomena are consistent with previous data (Wrobel et al., 1982).

Figure 3 illustrates the surface temperature profile variation with lateral direction as well as time. The incident temperature profile follows a Gaussian distribution. Similar results were reported based on the complicated Poynting vector theorem and Maxwell equations (Mansuripur et al., 1982; Chung, 1986).

Comparing Figure 2 to previous data on extremely thin recording layers (Chung, 1986), we find that the thicker the layer, the faster and the easier the pit deformation. However, the degree of deformation may be reduced significantly due to continuous material transportation after the laser is shut off.

Table 1. Threshold Energy at Various Marking Powers

	Beam Power, mJ/s					
	6	8	10	12	14	16
Hole-opening time, ns	124	80	55	41	34	28
Threshold energy,nJ	0.74	0.64	0.55	0.49	0.47	0.45

This is because temperature increases faster for a thicker recording layer, while it cools down more slowly than with a thinner layer. In other words, the low conductivity of an organic layer works as a good heat insulator for temperature rises and facilitates pit formation during the heating and marking. In order to prove this, Figure 4 shows the effect of thermal diffusivity and beam size on the pit formation. Once heating is extremely localized and the heat cannot be quickly dissipated, the variation in surface tension creates tangential stresses on the free surface and a capillary fluid motion is therefore induced.

Based on the above calculations, the minimum or threshold energy E_t required to create a mark in the film may be determined. Here E_t is defined as $E_t = P\,t_i$, where t_i is considered as the initial hole-opening time when the height at pit center drops 0.1%. Table 1 summarizes the threshold energy at different laser powers and shows that it varies slightly with the marking power, but apparently levels off at high powers. This calculated phenomenon seems to be consistent with previous experimental results where E_t values are close to that of published data (Novotny and Alexandru, 1979; Broer and Vriens, 1983). At present, there are various proposals to calculate E_t, and there are many factors affecting E_t value. That is beyond the scope of this note and should be discussed in a separate paper.

Finally, it is important to point out that the relationship between $\partial\sigma/\partial Ts$ and temperature has not yet developed for most polymers; Eq. 7 would not be correct if $\partial\sigma/\partial Ts$ varies with T.

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Notation

Bn = modified Bessel function of order n
c = heat capacity, J/kg · K
D = initial thickness of optical layer, m
E_t = threshold energy, nJ
g = gravitation constant, m/s^2
H = pit thickness, m
I = light intensity, J/s · m^2
Io = light intensity at laser beam center, Eq. 2
Jn = Bessel function of order n
k = thermal diffusivity, m^2/s
K = thermal conductivity, J/m · s · K
q = heat liberated on a ring, J
Q = marking power, J/s
r = lateral distance, m
r' = ring radius, m
t = time, ns
t_i = initial hole-opening time, ns
T = temperature, K
Ts = surface temperature, K
z = thickness distance

Greek letters

δ = laser beam radius, m
ρ = density, kg/m^3
μ = viscosity, N · s/m^2
σ = surface tension, mN/m
θ = angular axis, rad

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