Spontaneous pattern formation in a thin film of bacteriorhodopsin with mixed absorptive–dispersive nonlinearity

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We have observed the spontaneous formation of transverse spatial patterns in a thin film of bacteriorhodopsin with a feedback mirror. Bacteriorhodopsin has a mixed absorptive–dispersive nonlinearity at the wavelength used in the experiments (633 nm). Threshold values of the incident intensity for observation of pattern formation are found from a linear stability analysis of a model that describes bacteriorhodopsin as a sluggish saturable nonlinear medium with a complex Kerr coefficient. The calculated threshold intensity is in good agreement with the experimental observations, and the patterns are predicted to be frequency offset from the pump radiation.

Spontaneous formation of transverse spatial patterns owing to counterpropagating beams in a nonlinear-optical medium has been observed by use of several different geometries, including cavityless counterpropagation in a thick nonlinear medium,[1–3] a nonlinear medium placed in a Fabry–Perot cavity,[4] a ring geometry with nonlinear feedback,[5–7] and a thin nonlinear slice with a feedback mirror.[8–11] The thin nonlinear slice is an attractive system because of its relative simplicity from both an experimental and a theoretical point of view. The linear stability analysis of this geometry was performed first by Firth,[12] and hexagonal transverse patterns were predicted later on the basis of numerical simulations.[13] Experimental studies have shown that a variety of transverse patterns, hexagonal and otherwise, are accessible in this geometry.[8,9,11,14] Subsequent theoretical analysis[15] has pointed out the role of the Gaussian intensity profile in selecting the transverse pattern.

The experiments to date using the thin-slice geometry have been performed in atomic vapors,[8,11] with the pump laser tuned off resonance, or in liquid crystals.[9,10,14] In these cases the nonlinearity is essentially dispersive. We report here on the observation of transverse spatial structures by use of a thin film of bacteriorhodopsin as the nonlinear medium. Bacteriorhodopsin has attracted considerable interest recently as a nonlinear-optical medium.[16] It is characterized by a combined absorptive–dispersive nonlinearity,[17] in addition to relatively high values of the background linear absorption. The loss owing to linear absorption results in a higher threshold intensity for pattern formation than would be the case for transparent media. Furthermore, the nonlinear absorption, as described by the imaginary part of the complex n_2, results in frequency detuning of the patterns but does not otherwise affect the threshold intensity. This result is in contrast to that found in a previous analysis[12] that assumed a purely dispersive material response. The detuning predicted here is analogous to that found in photorefractive media, in which a sluggish complex nonlinearity also results in frequency detuning of the spatial sidebands.[18,19] These effects are studied on the basis of a dispersion relation that accounts analytically for both the linear and the nonlinear absorption.

The experimental arrangement is shown in Fig. 1. A 15-mW beam from a He–Ne laser with a wavelength of 633 nm is focused to a spot with a Gaussian beam radius $w \sim 26\ \mu$m, giving a peak intensity of 1400 W/cm^2. The bacteriorhodopsin in gelatin film has a thickness of $\ell = 40\ \mu$m and a refractive index of ~ 1.5. The film was prepared by use of a method similar to that described in Ref. 20, except that here a pH of 9 was used, resulting in a high saturation intensity of $\sim$900 W/cm^2. We speculate that this high saturation intensity relative to the more typical tens to hundreds of milliwatts per square centimeter[21] is related to the use of high pH values. Nonlinear saturationlike effects at intensities of several hundred watts per square centimeter have been reported previously[22] with films having a pH >9.

When a single laser beam is focused onto the nonlinear film, a characteristic concentric ring pattern is observed.[23] By counting the number of rings N (in our case $N = 7$) and using the formula[23] $N/2\pi \sim n_2' I k_0 n_0' [1 - \exp(-\alpha\ell)]/\alpha$, where $k_0 = 2\pi/\lambda$ is the vacuum wave number, $n_0' \sim 1.5$ is the background index in the film, and α is the background intensity absorption coefficient, we estimate the nonlinear refractive-index coefficient to be $n_2' \sim -1.4 \times 10^{-6}$ cm^2/W.

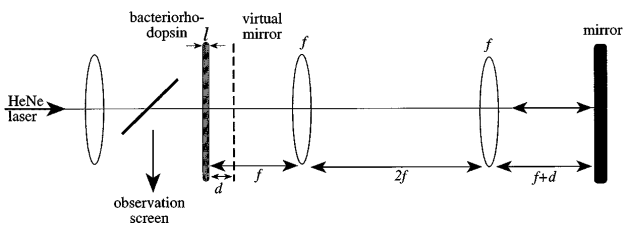

Fig. 1. Experimental setup. The mirror reflectivity, accounting for Fresnel losses at the film, was $R \sim 0.9$.

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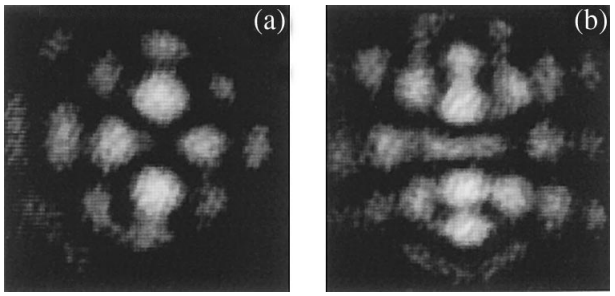

Fig. 2. Near-field image of the backward-propagating light for (a) $d = 3$ mm and (b) $d = 4$ mm.

When two counterpropagating beams are incident upon the film, the situation changes drastically. We introduced a feedback mirror, using two $f = 50$ mm lenses in a 4-f configuration.[14] An image of the nonlinear film (near field) was magnified and projected onto an observation screen and a video camera. Some typical patterns are shown in Fig. 2. The observed patterns were static, i.e., they did not move or rotate. The confocal length of the focused beam given by $k_0 w^2$ was ~6.8 mm, which is comparable with the film-to-mirror distance d. Thus the experiment was performed in a parameter regime in which the boundaries influence the pattern formation,[15] and it is not surprising that patterns with lower-than-hexagonal symmetry were observed.

We turn now to a theoretical study of the experiment. Bacteriorhodopsin may be characterized as a sluggish saturable Kerr medium with a complex index of refraction:

$$n = n_0' + in_0'' + (n_2' + in_2'')\frac{I}{1 + I/I_s}, \qquad (1)$$

where I is the optical intensity and I_s is the saturation intensity. At 633 nm, bacteriorhodopsin is a self-defocusing medium ($n_2' < 0$),[17] and nonlinear polarimetry measurements[24] yield $n_2''/n_2' \simeq -0.16$. In addition to the photochromic effect modeled by Eq. (1), a thermal nonlinearity may also contribute to n_2'. We estimate the magnitude of the thermal contribution at the end of this Letter.

To find the threshold intensity for the appearance of a transverse instability, we write the forward- and backward-propagating fields in the nonlinear film in the form of incident plane waves F_0 and B_0 plus spatial sidebands $F_0 F_{\pm 1}$ and $B_0 B_{\pm 1}$, with the sidebands detuned by $\pm\Omega$. In the limit of weak sidebands the wave equations for the slowly varying amplitudes take the form $\mathrm{d}F_{+1}/\mathrm{d}z = \mathrm{d}B_{-1}^*/\mathrm{d}z = \kappa$, $\mathrm{d}F_{-1}^*/\mathrm{d}z = \mathrm{d}B_{+1}/\mathrm{d}z = -\kappa$, where

$$\kappa = \frac{i2k_0 n_0'(n_2' + in_2'')|F_0(z)|^2}{1 + [|F_0(z)|^2/I_s](1 + q)} \frac{1}{(1 - i\Omega\tau)} \times |F_{+1} + F_{-1}^* + q(B_{+1} + B_{-1}^*)], \qquad (2)$$

$q(z) = |B_0(z)/F_0(z)|^2$, and τ is the characteristic response time of the medium. The equations have been written in the limit of a thin slice ($\ell \ll d$) so that diffraction inside the slice may be neglected. In addition, only transmission gratings have been accounted for since analysis of the reflection-grating case is complicated when the beam ratio q is a function of z.[19]

Using $q(z) = R\exp[-2\alpha_0(\ell - z)]$, where the intensity absorption coefficient is given by $\alpha_0 = 2k_0 n_0''$, we obtain the following dispersion relation for the threshold of the absolute instability:

$$1 + 4k_0 n_0'\ell(n_2' + in_2'') \times |F_0(0)|^2 \frac{1}{1 - i\Omega\tau} \sin(2k_d d)\psi = 0, \qquad (3)$$

$$\psi = \frac{1}{\alpha_0 \ell b\sqrt{1 - 4b^2\tilde{R}}} \times \operatorname{arctanh}\left\{\frac{\sqrt{1 - 4b^2\tilde{R}}[1 - \exp(\alpha_0\ell)]}{1 + 2b + (1 + 2b\tilde{R})\exp(\alpha_0\ell)}\right\} + \frac{1}{2\alpha_0\ell b}\ln\left[\frac{\exp(\alpha_0\ell) + b + bR}{1 + b + b\tilde{R}}\right], \qquad (4)$$

where $k_d = k_\perp^2/2k_0$, $\tilde{R} = R\exp(-2\alpha_0\ell)$ is the effective mirror reflectivity, and $b = |F_0(0)|^2/I_s$ is the saturation parameter at the input face. In the Kerr limit ($b = 0$) with no linear absorption ($\alpha_0 = 0$) the factor ψ reduces to R, and we recover the relation published previously[12] in the limit of no diffusion in the nonlinear medium and $\Omega = 0$. The minimum thresholds are obtained for $k_d d = (4m + 3)\pi/4$ for self-focusing media ($n_2' > 0$) and $k_d d = (4m + 1)\pi/4$ for self-defocusing media ($n_2' < 0$), where m is an arbitrary integer. Note that the lack of diffusion in the model considered here results in all the branches' having the same minimum threshold. The inclusion of a finite diffusion length, or accounting for the spatial frequency response of the nonlinear medium and its finite thickness, would select the branches with the smallest value of $k_d d$.

Setting the imaginary part of the left-hand side of Eq. (3) to zero yields immediately $\Omega\tau = -n_2''/n_2'$. The presence of nonlinear absorption implies that the sidebands are frequency detuned and that no instability is possible in the purely absorptive limit. Noting that $n_2 = n_2' + in_2''$ appears in Eq. (3) only as a factor multiplying $|F_0(0)|^2$ and replacing $\Omega\tau$ by $-n_2''/n_2'$, we find that the threshold intensity is inversely proportional to n_2' and is independent of n_2'' (and $\Omega\tau$). The effect of nonlinear absorption in the thin-slice geometry is solely that of causing a frequency shift and does not otherwise affect the threshold intensity. When the nonlinear medium is thick, both the real and the imaginary parts of n_2 enter into the dispersion relation in a complicated fashion.[18,19] The original analysis of the thin-slice model according to Firth[12] concluded that the instability threshold is an increasing function of $\Omega\tau$. The disparity between his results and those presented here can be traced to the fact that we have permitted the phase of the nonlinear response to be a function of $\Omega\tau$, as is appropriate for a sluggish medium, whereas it was assumed previously[12] that n_2 is purely real, irrespective of the detuning.

Figure 3 shows the threshold value of the dimensionless product $k_0 n_0'\ell n_2'|F_0(0)|^2$ plotted as a

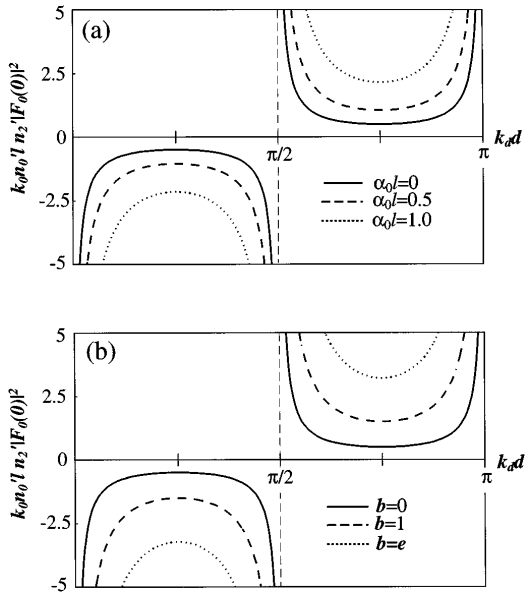

Fig. 3. Threshold intensity for various parameters: (a) variable absorption ($R = 1$, $b = 0$, and $\alpha_0 \ell = 0$, 0.5, 1) and (b) variable saturation ($R = 1$, $\alpha_0 \ell = 0$, and $b = 0$, 1, e).

function of $k_d d$ for various parameters. The experimental conditions were $n_2' \sim 1.4 \times 10^{-6}$ cm^2/W, $R \sim 0.9$, $2\alpha_0 \ell \sim 0.5$, $b \sim 1.6$, and $k_0 n_0' \ell \sim 600$. Numerical solution of Eq. (3) for these parameter values yields $I_{\text{threshold}} \sim 3000$ W/cm^2, which agrees within a factor of 2 with the experimental value of 1400 W/cm^2. It should be recalled that we calculated the threshold by neglecting the influence of reflection gratings in the bacteriorhodopsin film. On the other hand, we have performed four-wave mixing experiments that indicate that both transmission and reflection gratings are present with approximately equal strength. Analogous with the known results for a thick medium,[25] we expect that accounting for reflection gratings would tend to lower the calculated threshold by approximately a factor of 2.

The observed effects may also be due to a thermal nonlinearity. When $n_2' < 0$, the thermal contribution has the same sign and will add constructively to the total effect. A possible thermal contribution can be estimated from the formula[26] $n_{2,\text{th}}' \simeq [\alpha\lambda^2/(16\pi^2\kappa_{\text{th}})]\sin^{-2}(\theta/2)\mathrm{d}n_0'/\mathrm{d}T$, where κ_{th} is the thermal conductivity and $\theta \sim \sqrt{\lambda/d}$ is the external angle between the incident beam and the sidebands. Note that the thermal contribution increases as θ^{-2} for small θ, and θ is only $\sim$10 mrad in these experiments. Using the numerical values $\alpha = 63$ cm^{-1} (from transmission measurements at low intensity), $\theta \sim 0.014$, $\kappa_{\text{th}} \sim 0.2$ W/mK, $\mathrm{d}n/\mathrm{d}T \sim -0.0001$ K^{-1}, we find that $n_{2,\text{th}}' \sim -1.6 \times 10^{-6}$ cm^2/W, which is roughly the same as our estimate for the photochromic contribution. We conclude that the dispersive part of the nonlinearity is due to both photochromic and thermal effects.

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