Spontaneous one- and two-dimensional optical spatial solitons supported by photoisomerization nonlinearity in a bulk polymer

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We present theoretically another type of optical spatial soliton in steady state, which is supported by photoisomerization nonlinearity in a polymer. For the photoisomerization nonlinearity with saturation, this type of one- or two-dimensional spatial soliton can be formed in a bulk polymer. The spontaneous soliton of this type can only be dark or gray. Not only a linearly polarized beam, but also a circularly polarized beam can be used to form the soliton, which is impossible for the photorefractive soliton. The effects of wavelength, polarization, and intensity of the beam on the formation of the soliton are discussed in detail.

The optical spatial soliton (SS) is one of the most intriguing phenomena of nonlinear optics. The Kerr-type SS is the first optical SS, predicted more than 30 years ago [1], yet it requires very high power and can only be observed in a one-dimensional form and in a slab waveguide owing to both catastrophic self-focusing and modulation instability. In the past decade, SSs supported by the photorefractive effect have attracted much attention from researchers [2–15]. The photorefractive effect is a saturable nonlinearity and can support one- or two-dimensional optical SSs at very low power level (microwatt) [2], however this type of SS generally requires an external electric field or an additional background beam, though some exceptions have been found [16]. Some interesting discoveries, such as interaction of the SSs [9], rotating SSs [6,17], and multiple composite SSs [18], have been reported. With the rapid development of polymer science, many researchers are dedicating themselves to forming SSs or waveguides in polymers. Sheu *et al.* [14] suggested forming the SS in photorefractive polymers, which was recently realized by Chen *et al.* [15]. Sarkisov *et al.* [19] formed dark SSs in photopolymer films by means of upconverted dye photobleaching and discussed the application of these SSs in optical interconnections. Bartuch *et al.* [20] investigated SSs in planar polymer waveguides supported by Kerr-like nonlinearity. Wright *et al.* [21] presented a two-dimensional SS in the polydiacetylene para-toluene sulfonate with a quintic nonlinearity. Shi *et al.* [22] formed the photomasked channel waveguide structure on copolyester disperse-red-19 films, and Henninot *et al.* [23] formed optical SSs in dye-doped liquid crystals. Some researchers [24,25] were interested in forming self-written waveguides caused by the photopolymerization process in photosensitive materials.

On the other hand, photoisomerization is another interesting phenomenon of nonlinear optics [26,27]. The reversible transition of *trans-* and *cis*-isomerizations of the photochromatic process in polymers can lead to photoinduced birefringence, dichroism, mass transport, and intensity-dependent refractive index change [28]. Photoisomerization nonlinearity

has been extensively applied to optical storage [26,27], recording of holographic gratings [26,27], and all-optical switching [26,27,29,30], etc. We have recently found that the photoinduced intensity-dependent refractive index change in this type of material is capable of supporting SSs and we have predicted theoretically that with the assistance of a proper background beam, a linearly polarized beam can form dark or bright one-dimensional SSs in bulk polymers with the photoisomerization nonlinearity [31].

In this paper, spontaneous one-dimensional gray and dark SSs as well as two-dimensional dark SSs, which are supported by the photoisomerization nonlinearity in the bulk polymer material, are presented, respectively. It is found that the spontaneous SSs can be only dark or gray. We show that both linearly polarized and circularly polarized beams can form this type of SSs. We also discuss the effects of the wavelength, polarization, and intensity of the beam on the formation of SSs.

For the large photoinduced change of the refractive index (see the discussion below), one can anticipate that SSs can be formed spontaneously. The reaction kinetics of the photoisomerization process in polymers induced by a linearly or circularly polarized light beam can be described by [[32], Appendix]

$$\frac{dT_l}{dt} = -q_{Ts}\sigma_T I_s T_l \cos^2\theta + q_{Cs}\sigma_C I_s(T_0 - T_l) + K_C(T_0 - T_l)$$

(1)

and

$$\frac{dT_c}{dt} = -\frac{1}{2}q_{Ts}\sigma_T I_s T_c \sin^2\theta' + q_{Cs}\sigma_C I_s(T_0 - T_c) + K_C(T_0 - T_c),$$

(2)

where T_l, (T_c), and T_0 represent the concentrations of molecules in the *trans* form under linearly (circularly) polarized illumination and in the darkness, respectively, θ is the angle between the molecular orientation and the direction of electric field of the signal beam, θ' is the angle between the molecular orientation and the direction of wave vector, q_{Ts}

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and q_{Cs} are the quantum yields of the signal beam for *trans*-to-*cis* and *cis*-to-*trans* transitions, respectively, σ_T and σ_C are the absorption cross sections of the signal beam in the *trans*-to-*cis* and *cis*-to-*trans* transitions, respectively, K_C is the thermal relaxation rate of the *cis*-to-*trans* transition, and I_s is the intensity of the light beam.

In the steady state, one can get $T=T_0(I+1)/(\beta I \cos^2 \theta + I +1)$ for the case of linear polarization, while $T=T_0(I+1)/(\beta I \sin^2 \theta'/2 + I +1)$ for the case of circular polarization, where $I=I_s/I_D$, $I_D=K_C/(q_c \sigma_C)$, and $\beta=(q_T \sigma_T)/(q_C \sigma_C)$. Then the refractive index change corresponding to the photo-isomerization is given by [32]

$$\Delta n_l = S \int (T - T_0)\cos^2 \theta d\Omega, \qquad (3)$$

$$\Delta n_c = S \int (T - T_0)\cos^2 \theta d\Omega,$$

where Ω is the solid angle, S is a positive parameter depending on material and the wavelength of the signal beam, and $\cos^2 \theta = \sin^2 \theta' \cos^2(\varphi' - \varphi)$, according to the geometric relation shown in Fig. 6. The direction of the wave vector is selected along the z axis, φ' is the angle between the x axis and the projection of the molecular orientation on the XY plane, and φ is the angle between the x axis and the electric field of the beam. Then we get

$$\Delta n_l = 4\pi ST_0 \left[\frac{I+1}{\beta I}\left(1 - \sqrt{\frac{1+I}{\beta I}} \tan^{-1}\sqrt{\frac{\beta I}{1+I}}\right) - \frac{1}{3}\right],$$

$$(4)$$

$$\Delta n_c = 4\pi ST_0 \left\{ \frac{I+1}{\beta I}\left[1 + \frac{\dfrac{I+1}{\beta I}}{\sqrt{\dfrac{2(I+1)}{\beta I}} + 1}\right. \right.$$

$$\left. \left. \times \ln \frac{\dfrac{2(I+1)}{\beta I}}{\left(\sqrt{\dfrac{2(I+1)}{\beta I}} + 1 + 1\right)^2}\right] - \frac{1}{3}\right\}.$$

The refractive index changes, Δn_l (solid lines) and Δn_c (dashed lines), are shown in Fig. 1 as functions of I for different values of β. One can see clearly in Fig. 1 that (i) the refractive index change is negative and saturable, (ii) the saturable values of $|\Delta n|$ increase with the values of β, and (iii) the values of $|\Delta n_l|$ are larger than the relative values of $|\Delta n_c|$. As the property (i) states, the refractive index change is saturable, which means that the soliton can be formed in the *bulk* material [33]; the refractive index change is always negative and decreases monotonously with the intensity of the signal beam, which means that the SSs can only be *dark or gray* [34]. From the facts that the refractive index change is induced by the light beam itself, and that the refractive index change is large enough (see the discussion below), we conclude that the SSs can be formed *spontaneously*.

The nonlinear Schrödinger equation describing the state of the beam is

$$\frac{1}{2k}\nabla_\perp^2 A + i\frac{\partial A}{\partial z} + \frac{k_0^2 \Delta(n^2)}{2k}A = 0, \qquad (5)$$

where A is the slowly varying amplitude of the electric field of the beam, $k=k_0 n_0$, $\Delta(n^2) \approx 2n_0 \Delta n$, n_0 is the unperturbed refractive index, and $k_0 = 2\pi/\lambda$, where λ is the wavelength of the beam in the vacuum.

For one-dimensional dark SSs, let $A=u(x)\sqrt{I_D}\exp(i\Gamma z)$, where Γ is the propagation constant of the SS. Then the soliton equations in dimensionless form derived from Eqs. (4) and (5) are

$$\frac{\partial^2 u(\xi)}{\partial \xi^2} = u\left\{ b - \left[\frac{u^2+1}{\beta u^2}\left(1 - \sqrt{\frac{1+u^2}{\beta u^2}}\right.\right.\right.$$

$$\left.\left.\left. \times \tan^{-1}\sqrt{\frac{\beta u^2}{1+u^2}}\right) - \frac{1}{3}\right]\right\}$$

and

$$\frac{\partial^2 u(\xi)}{\partial \xi^2} = u\left\{ b - \left[\frac{u^2+1}{\beta u^2}\left(1 + \frac{\dfrac{u^2+1}{\beta u^2}}{\sqrt{\dfrac{2(u^2+1)}{\beta u^2}} + 1}\right.\right.\right.$$

$$\left.\left.\left. \times \ln \frac{\dfrac{2(u^2+1)}{\beta u^2}}{\left(\sqrt{\dfrac{2(u^2+1)}{\beta u^2}} + 1 + 1\right)^2}\right) - \frac{1}{3}\right]\right\} \qquad (6)$$

for linearly and circularly polarized beams, respectively, where $u^2(x)=I$, $\xi=x/x_0$, $x_0=(8n_0 k_0^2 \pi ST_0)^{-1/2}$, and $b=\Gamma/(4k_0\pi ST_0)$. Let $u(\infty)=u_\infty$, and then we can get $b=(u_\infty^2 +1)/(\beta u_\infty^2)[1 - \sqrt{(u_\infty^2+1)/(\beta u_\infty^2)}\tan^{-1}\sqrt{\beta u_\infty^2/(u_\infty^2+1)}]-1/3$ for a linearly polarized beam, and

$$b = (u_\infty^2 + 1)/(\beta u_\infty^2)(1 + (u_\infty^2 + 1)/(\beta u_\infty^2)/\sqrt{2(u_\infty^2 + 1)/(\beta u_\infty^2) + 1}$$

$$\times \ln\{2(u_\infty^2 + 1)/(\beta u_\infty^2)/[\sqrt{2(u_\infty^2 + 1)/(\beta u_\infty^2) + 1} + 1]^2\}) - 1/3$$

for a circularly polarized beam. The dark SS solutions obtained are shown in Fig. 2(a) for $u_\infty = 1.0$ and $\beta = 1/2$. [The value of β depends on the wavelength and the material. As an example, the value of β will vary from about 0.3 to 1.5 with wavelength for disperse red one (DR1) doped poly-(methyl methacrylate) (PMMA) polymer [35].] The existence curves for different values of β are shown in Figs. 2(b) and 2(c).

We can see from Fig. 2 that (i) the existence curves of this type of SS are similar to those of photorefractive SS [2], (ii) the existence curves will "shift" *left* and *down* when the value of β increases, (iii) for the same intensity of the beam, when the value of β is about 2, the SSs of a linearly polarized beam and a circularly polarized beam almost have the same full width at half maximum (FWHM); and when the value of β is less than 2, the SS of a linearly polarized beam has a smaller FWHM than that of a circularly polarized beam; when the value of β is greater than 2, the FWHM of the SS of a linearly polarized beam is larger than that of a circularly polarized beam in the high-intensity range, (iv) the
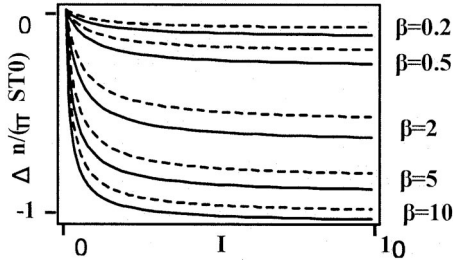
FIG. 1. The refractive index change as a function of I for different values of β, where the solid lines and the dashed lines correspond to Δn_l and Δn_c, respectively.

minimum FWHM of the soliton becomes larger when the value of β becomes smaller, for the saturated absolute value of refractive index change decreases with the value of β (see Fig. 1).

For the one-dimensional gray SS, let $A = u(\xi)\sqrt{I_D}\exp\{i[\Gamma z + \int J/u^2(\xi)d\xi]\}$, where Γ is the propagation constant of the SS, J is a positive constant, and then the soliton equations in dimensionless form derived from Eqs. (4) and (5) are
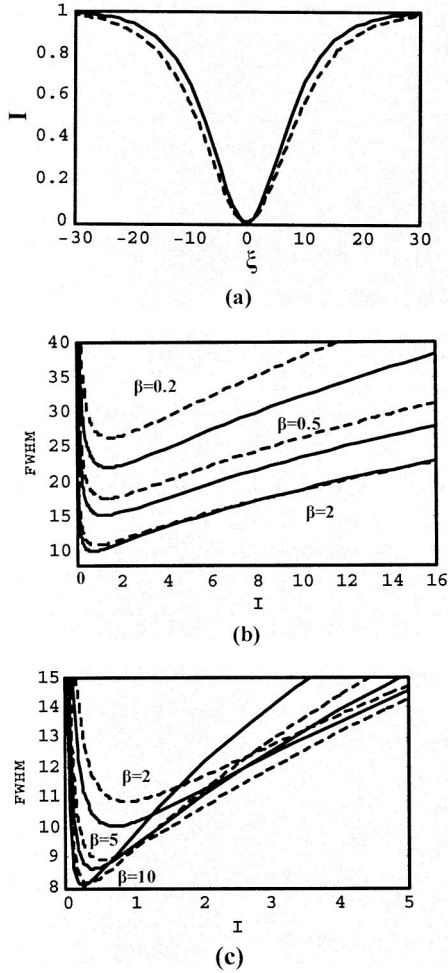

(a)

(b)

(c)

FIG. 2. The spontaneous dark spatial soliton solutions for the linearly (solid lines) and circularly (dashed lines) polarized beams. (a) The intensity profile of the solitons, where $u_\infty = 1.0$ and $\beta = 1/2$; (b) and (c) The existence curves for different values of β.

$$\frac{\partial^2 u(\xi)}{\partial\xi^2} - \frac{J^2}{u^3} = u\left\{b - \left[\frac{u^2+1}{\beta u^2}\left(1 - \sqrt{\frac{1+u^2}{\beta u^2}}\right.\right.\right.$$
$$\left.\left.\left. \times\tan^{-1}\sqrt{\frac{\beta u^2}{1+u^2}}\right) - \frac{1}{3}\right]\right\}$$

and

$$\frac{\partial^2 u(\xi)}{\partial\xi^2} - \frac{J^2}{u^3} = u\left\{b - \left[\frac{u^2+1}{\beta u^2}\left(1 + \frac{\frac{u^2+1}{\beta u^2}}{\sqrt{\frac{2(u^2+1)}{\beta u^2}+1}}\right.\right.\right.$$
$$\left.\left.\left. \times\ln\frac{\frac{2(u^2+1)}{\beta u^2}}{\left(\sqrt{\frac{2(u^2+1)}{\beta u^2}+1}+1\right)^2}\right) - \frac{1}{3}\right]\right\}$$

for linearly and circularly polarized cases, respectively. As an example, we solve the equations numerically for $u_\infty = 1.0$, $\beta = 1/2$, and $u_0 = 0.1$. The intensities and phase profiles of the gray SSs are shown in Fig. 3, where $J \approx 0.0123$ (0.0105) and $b \approx -0.0427$ (−0.0302) for the linear (circular) polarization case.

We can see from Eq. (4) that the distribution of the refractive index change only relates to the intensity distribution of the soliton beam. This means if the intensity profile of the two-dimensional beam has circular symmetry (no matter whether the polarization is linear or circular), the distribution of the refractive index change will have circular symmetry too. Recalling the saturable character of the material, we can deduce that the two-dimensional circular SS can be formed in this material. In two-dimensional case, a vortex phase profile should be introduced, and only the dark SSs can be formed. In polar coordinates, let $A(\rho, \theta) = \nu(\rho)\sqrt{I_D}\exp(im\theta)\exp(i\Gamma z)$, where $m = \pm 1, \pm 2, \ldots$, and we get the soliton equations in dimensionless form for the dark SS,

$$\frac{d^2\nu}{d\eta^2} + \frac{1}{\eta}\frac{d\nu}{d\eta} - m^2\frac{\nu}{\eta^2}$$
$$= \nu\left\{b - \left[\frac{\nu^2+1}{\beta\nu^2}\left(1 - \sqrt{\frac{1+\nu^2}{\beta\nu^2}}\tan^{-1}\sqrt{\frac{\beta\nu^2}{1+\nu^2}}\right) - \frac{1}{3}\right]\right\}$$

for the linearly polarized beam, and

$$\frac{d^2\nu}{d\eta^2} + \frac{1}{\eta}\frac{d\nu}{d\eta} - m^2\frac{\nu}{\eta^2} = \nu\left\{b - \left[\frac{\nu^2+1}{\beta\nu^2}\left(1 + \frac{\frac{\nu^2+1}{\beta\nu^2}}{\sqrt{\frac{2(\nu^2+1)}{\beta\nu^2}+1}}\right.\right.\right.$$
$$\left.\left.\left. \times\ln\frac{\frac{2(\nu^2+1)}{\beta\nu^2}}{\left(\sqrt{\frac{2(\nu^2+1)}{\beta\nu^2}+1}+1\right)^2}\right) - \frac{1}{3}\right]\right\}$$

for the circularly polarized beam, where $\eta = \rho/x_0$, $\nu^2(\rho) = I$,
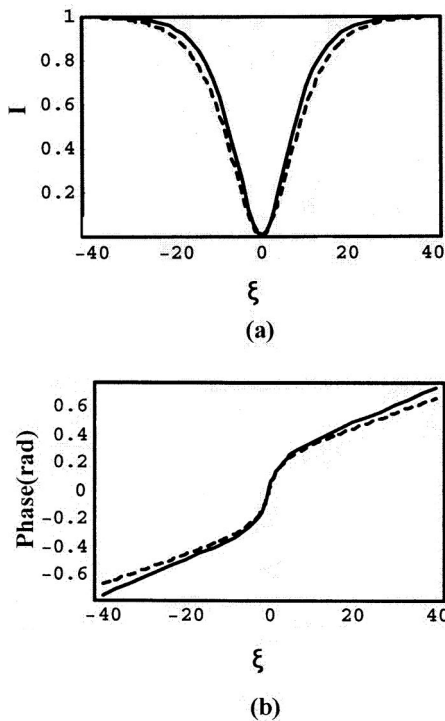
(a)

(b)

FIG. 3. The spontaneous gray spatial soliton solutions for the linearly (solid lines) and circularly (dashed lines) polarized beams, where $u_\infty = 1.0$, $u_0 = 0.1$, and $\beta = 1/2$. (a) The intensity profiles of the solitons. (b) The phase profiles of the solitons.

and m corresponds to the so-called topological charge. For $\eta \to \infty$, $\nu(\eta) \to \nu_\infty$, we get

$$b = \frac{\nu_\infty^2 + 1}{\beta \nu_\infty^2}\left(1 - \sqrt{\frac{1+\nu_\infty^2}{\beta \nu_\infty^2}}\tan^{-1}\sqrt{\frac{\beta \nu_\infty^2}{1+\nu_\infty^2}}\right) - \frac{1}{3}$$

for the linearly polarized beam, and

$$b = \frac{\nu_\infty^2 + 1}{\beta \nu_\infty^2}\left(1 + \frac{\dfrac{\nu_\infty^2+1}{\beta\nu_\infty^2}}{\sqrt{\dfrac{2(\nu_\infty^2+1)}{\beta\nu_\infty^2}+1}}\right.$$
$$\left.\times\ln\frac{\dfrac{2(\nu_\infty^2+1)}{\beta\nu_\infty^2}}{\left(\sqrt{\dfrac{2(\nu_\infty^2+1)}{\beta\nu_\infty^2}+1}+1\right)^2}\right) - \frac{1}{3}$$

for the circularly polarized signal beam.

The numerical solutions of SS's are showed in Fig. 4 for $\nu_\infty = 1.0$, $\beta = 1/2$, $m = 1, 2$, and for linearly and circularly polarized cases, respectively.

The existence curves of the vortex SSs are shown in Fig. 5 for $\beta = 1/2$. To compare, the existence curves of the one-dimensional dark SS are also given in Fig. 5. One can see that the FWHM of the two-dimensional SS is larger compared to that of the one-dimensional SS, and the FWHM of the circular-symmetry double charged vortex SS is larger than that of the single charged one for the same intensity.
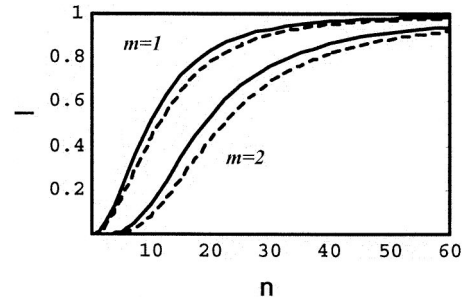

FIG. 4. The two-dimensional vortex soliton solutions for the linearly (solid lines) and circularly (dashed lines) polarized beams, where $\nu_\infty = 1.0$ and $\beta = 1/2$.

It is well known that the isomerization of the photochromatic process in polymers can lead to large intensity-dependent refractive index change [28,36]. For example, according to Table 1 in Ref. [28], when the intensity is about 10 W/cm^2 (the typical intensity for photorefractive SSs [7]), the refractive index change can be 10^{-3}–10^{-2} for 5% (concentration by weight) DR1-doped PMMA. This change is larger than that for photorefractive SSs (10^{-4}–10^{-3}) [37], which means that the *spontaneous* soliton is achievable.

The value of parameter $\beta[\beta = (q_T\sigma_T)/(q_C\sigma_C)]$ is determined by the material and the wavelength. We choose the disperse red one (DR1)-doped poly-(methyl methacrylate) (PMMA) matrix as an example here. The values of q_T and q_C (the quantum yields of the signal beam for *trans*-to-*cis* and *cis*-to-*trans* transitions, respectively) vary slightly in the visible range while σ_T and σ_C (the absorption cross section of the signal beam in the *trans*-to-*cis* and *cis*-to-*trans* transitions, respectively) will experience large variation (see Ref. [35]). In the DR1-doped PMMA material, if the wavelength of the beams is 514.5 nm (the typical wavelength for an argon laser), the value of β is about 1/2. If the wavelength of the beam is between 490 and 550 nm, the value of β can vary from about 0.3 to 1.5.

The time in which the process of isomerization reaches the steady state varies with the molecular structure, the polymeric system, the inhibiting motion of the polymer matrix, the temperature, as well as the wavelength and the intensity of the pump beam [26,27,38]. For example, in DAP-PGMA material, the formation time can reach 40 ps [39]; in the
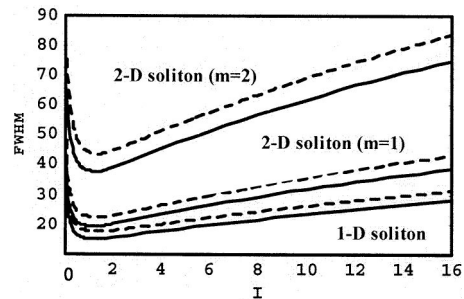

FIG. 5. The existence curves of one-dimensional soliton and two-dimensional solitons (m=1 and m=2) for $\beta = 1/2$. The solid curves correspond to the linearly polarized beam and the dashed curves correspond to the circularly polarized beam.
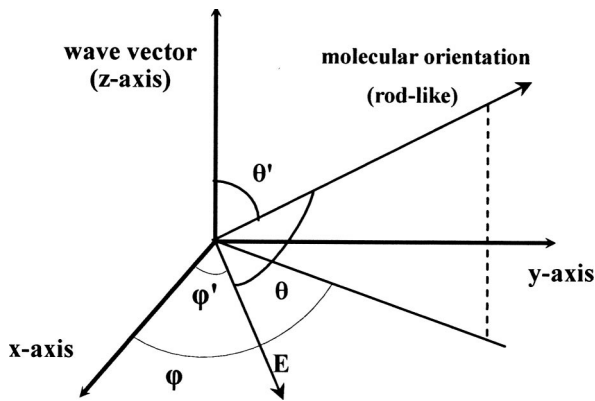
FIG. 6. Schematic of the mutual orientation of the rodlike molecule of the material, the propagation direction, and the electric field of the light beam.

DR1-doped PMMA material, the formation time can be about second, when pumped by a 514.5 nm argon laser (20 mW/cm^2) [40].

In conclusion, we have showed that the photoisomerization nonlinearity in a polymer can support one-dimensional dark and gray solitons as well as two-dimensional circularly symmetric dark solitons. This type of soliton can be spontaneous and can be formed by using not only the linearly but also the circularly polarized beams. Modification of the wavelength of the light beam can affect the existence curve of the solitons via the parameter β.

APPENDIX

From the quantum-mechanical treatment of absorption, the probability per unit time of the absorption of a photon from a monochromatic wave with an electric amplitude, $\vec{E}$, is proportional to $\Sigma_{i,j=1}^{3} p_i p_j^* E_i E_j^*$, where $\vec{p}$ is the transition dipole moment. Therefore, we can assume that the absorption cross section is $\hat{E} \cdot \vec{\sigma}_T \cdot \hat{E}$ and $\hat{E} \cdot \vec{\sigma}_C \cdot \hat{E}$ for *trans*- and *cis*-isomerizations, respectively, where $\hat{E}$ is the unit vector of the electric field of the beam,

$$\vec{\sigma}_T = \begin{pmatrix} \sigma_T^{\perp} & 0 & 0 \\ 0 & \sigma_T^{\perp} & 0 \\ 0 & 0 & \sigma_T \end{pmatrix},$$

and

$$\vec{\sigma}_C = \begin{pmatrix} \sigma_C & 0 & 0 \\ 0 & \sigma_C & 0 \\ 0 & 0 & \sigma_C \end{pmatrix}$$

in the molecular coordinate for the rodlike geometry of the *trans*-isomerization and the globular geometry of the *cis*-isomerization [26]. Generally, for the rodlike geometry of the *trans*-isomerization, $\sigma_T^{\perp}$ is much less than σ_T and can be neglected [32]. For linearly polarized light, $\hat{E} \cdot \vec{\sigma}_T \cdot \hat{E} = \sigma_T \cos^2 \theta + \sigma_T^{\perp} \sin^2 \theta \approx \sigma_T \cos^2 \theta$, $\hat{E} \cdot \vec{\sigma}_C \cdot \hat{E} = \sigma_C$, where θ is the angle between the molecular orientation and the direction of the electric field of the light beam. For circularly polarized light, the direction of the electric field of the light beam ($\hat{E}$) varies with time much faster than the process of isomerization, and then the absorption cross section of the *trans*-isomerization to the light is a time average of $\sigma_T \cos^2 \theta$, that is, $\sigma_T \sin^2 \theta'$, where θ' is the angle between the molecular orientation and the direction of the wave vector (see Fig. 6).

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