Polarization singularities and orbital angular momentum sidebands from rotational symmetry broken by the Pockels effect

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The law of angular momentum conservation is naturally linked to the rotational symmetry of the involved system. Here we demonstrate theoretically how to break the rotational symmetry of a uniaxial crystal via the electro-optic Pockels effect. By numerical method based on asymptotic expansion, we discover the 3D structure of polarization singularities in terms of \textit{C} lines and \textit{L} surfaces embedded in the emerging light. We visualize the controllable dynamics evolution of polarization singularities when undergoing the Pockels effect, which behaves just like the binary fission of a prokaryotic cell, i.e., the splitting of \textit{C} points and fission of \textit{L} lines are animated in analogy with the cleavage of nucleus and division of cytoplasm. We reveal the connection of polarization singularity dynamics with the accompanying generation of orbital angular momentum sidebands. It is unexpected that although the total angular momentum of light is not conserved, the total topological index of \textit{C} points is conserved.

The study of polarization of light has a long history. Nowadays polarization has been of broad applications in many areas of science and technology, ranging from physics to biology and chemistry\textsuperscript{1}. Recent interest may be traced back to the seminal work by Nye who revealed the generic structure of polarization singularities\textsuperscript{2–4}. In the general 3D picture, there are two types of polarization singularities: lines along which the polarization is purely circular (\textit{C} lines) and surfaces on which the polarization is purely linear (\textit{L} surfaces), where the orientation and handedness of polarization ellipse are indefinite, respectively. In the context of singular optics, polarization singularities are regularly considered as the vector analog of phase singularities or optical vortices in scalar fields\textsuperscript{5}. Circular polarizations are in essence associated with spin angular momentum of photons, while optical vortices are often studied with twisted photons carrying quantized orbital angular momentum (OAM)\textsuperscript{6,7}. It has been demonstrated that manipulating optical beams with vortex lines in the forms of knots or links holds promise for future laser technology and optical trapping schemes\textsuperscript{8–10}. Beyond the uniform polarization in scalar fields, the morphology and topology of polarization singularities in vector fields are much richer and subtler, as predicted by Dennis and later verified by Flossmann \textit{et al}\textsuperscript{11,12}. Recent years have witnessed a rapidly growing interest in these amazing structures, which are found to appear in the skylight\textsuperscript{13}, isotropic microchip laser\textsuperscript{14}, near field nano-optics\textsuperscript{15}, and inhomogeneous anisotropic plates\textsuperscript{16}.

Here we report theoretically a rather fascinating phenomenon that in a uniaxial crystal when undergoing Pockels effect, the tunable evolution of polarization singularities of emerging light behaves just like the binary fission, such as in a prokaryotic cell division\textsuperscript{17}. Under the control of an externally applied electric field, the splitting of \textit{C} points and fission of \textit{L} lines can be depicted vividly in analogy with the cleavage of nucleus and division of cytoplasm. Polarization singularities in crystals have been indeed studied extensively\textsuperscript{18–21}, but apparently seldom considering the Pockels effect\textsuperscript{22,23}. Previously, we demonstrated the capability of using Pockels effect to manipulate spin and orbital angular momentum in optically active crystals or electro-optic birefrigent crystals\textsuperscript{24–27}. In contrast, we here aim to show another phenomenon of tunable polarization singularities by electro-optically breaking the rotational symmetry of a uniaxial crystal, which therefore lends itself to a flexible and real-time manipulation. Our work can also be connected to those reporting the conservation law of angular momentum related to the rotational symmetry, such as in isotropic crystals or uniaxial crystals\textsuperscript{26,28–31}. In contrast, here the rotational symmetry around the optic axis is slightly broken by the applied electric field, since the specific
second-order susceptibilities $\chi_{xyz}$ and $\chi_{yzx}$ are activated to respond for the deformation of the refractive index ellipsoid of electro-optic crystal. Furthermore, we reveal the connection of our observations with the accompanying generation of OAM sidebands. OAM sidebands have been found intrinsic to reflection due to Goos-Hanchen and Imbert-Fedorov shifts.35. But ours is resulted from transverse angular anisotropy induced by Pockels effect. Surprisingly, total angular momentum of light is not conserved, whereas total topological index of $C$ points is conserved.

Our scheme is sketched in Figure 1. We consider a $z$-cut uniaxial crystal of potassium dihydrogen phosphate (KDP), which is a typical electro-optic material belonging to class 42m. The principle refractive indices $n_0 = 1.5074$, $n_\perp = 1.4669$, and the nonvanishing electro-optic coefficients $\gamma_3 = \gamma_9 = 8, \gamma_6 = 11$ (in pm/V), respectively.35. Assume the initial light beam of wavelength $\lambda = 1.633 \mu m$ is a left-handed circularly polarized one propagating along the optic axis, namely, $E(r,z = 0) = \exp(-i(r^2/2\sigma^2))\hat{e}_z$, where $\sigma = 4.59 \mu m$ is the beam waist and $\hat{e}_z = \sqrt{1/2}[1, i]$ as well as $\hat{e}_\perp = \sqrt{1/2}[1, -i]$ forms the circular bases. The incident and exit interfaces of KDP are both coated with transparent electrode in order to apply a longitudinal electric field $E_0 = E_0\hat{e}_z$. When $E_0$ is switched on, a nonlinear polarization responsible for the Pockels effect is induced: $P^{(2)} = 2\epsilon_0\chi^{(2)}(\omega, 0): E_0$, where $\epsilon_0$ is the permittivity of free space, $\chi^{(2)}(\omega, 0)$ the second-order susceptibility tensor related to the Pockels effect, and $E$ the light field of frequency $\omega$. Starting from Maxwell equations and considering total electric displacement $D = \epsilon_0\varepsilon_0 E + P^{(2)}$, we have the following equation governing the complex amplitude of a propagating light,

$$V^2E - V(V\cdot E) + k_0^2\varepsilon_0 E + \mu_0\sigma^2 P^{(2)} = 0,$$

where $k_0 = 2\pi/\lambda, \varepsilon_0 = \text{diag}(\varepsilon_r, \varepsilon_r, \varepsilon_r)$ denotes the relative dielectric tensor, and $\mu_0$ is the magnetic susceptibility in vacuum.

In this paper, we follow Ciattoni’s angular spectrum representation method36 and our recently developed numerical method based on asymptotic expansion to calculate the complex vectorial field $E(r, z)$ in the propagating space. As $E(r, z)$ is obviously position dependent, it is naturally expected that the emerging polarization from KDP is also spatially variant. Besides, the polarization should be electrically tunable by $E_0$ via the Pockels effect. In general, the geometry of polarization ellipse can be completely described in terms of four Stokes parameters18: $S_0 = E_x E_x + E_y E_y, S_1 = 2\text{Re}(E_x E_y), S_2 = 2\text{Im}(E_x E_y)$, and $S_3 = 2\text{Im}(E_x E_y)$. Physically, the orientation of major axis and the ellipticity of polarization ellipse are characterized by $\theta = \frac{1}{2}\text{arg}(S_1 + iS_2)$ and $\varepsilon = S_3/S_0$, respectively. In very cross section, $L$ lines are those on which $S_1 = 0, C$ points are defined as the intercept of the loci $S_1 = 0$ and $S_2 = 0$. Besides, $C$ points can be classified into some basic types, such as lemon, monstar and star.36

Results

We plot in Figure 2 our numerical solution of the 3D polarization singularities embedded in the emerging light. It is found that when the KDP crystal is undergoing the Pockels effect, $C$ lines and $L$ surfaces attain different morphologies. Figure 2(a) demonstrate the simple case when $E_0$ is absent. It looks like a right circular cone with the conical surface being $L$ surface and the axis of the cone being $C$ line. As the light propagates, the $C$ line is stretching along the propagation direction coinciding with optical axis. While $E_0$ is switched on, however, the $C$ line is quickly bifurcated into two ones, appearing like a pair of compasses with both arms being left-handed circular polarization and deflecting from the optic axis. Besides, the $L$ surface is then gradually cleaved into two separate sleeves and each encircles one $C$ line. By a comparison of Figure 2(a)–2(d), we find that applying a larger $E_0$ accelerates the cleavage of $L$ surface. As $E_0$ increases from 5.31 kV/cm to 10.62 kV/cm, the cleavage point is brought forward from $z = 6000 \mu m$ to 3000 $\mu m$ or so.

Figure 2 shows only the frame of the 3D polarization structures. One can image that the volume is filled with many polarization ellipses of various shapes and orientations. For a better view, we also visualize their 2D fine structures in Figure 3, assuming the length of KDP crystal is fixed at $z = 6000 \mu m$. By tuning $E_0$, we observe a fascinating phenomenon that the dynamic evolution of polarization singularities when undergoing the Pockels effect just behaves like the binary fission of a prokaryote cell.36 In Figure 3(a), the KDP crystal is pure uniaxial without disturbance ($E_0 = 0$). So the central $C$-point is simply surrounded by one $L$ circle. Here we use “$L$ cell” to describe the region that the $L$ line embraces. As $E_0$ is increasing, the splitting of $C$ points and fission of $L$ lines can be animated in analogy with the cleavage of nucleus and division of membrane in a cell division. Specifically, the $C$ point first replicates (like a single DNA), then attaches each copy to a different part of $L$ cell. In Figure 3(b) with $E_0 = 5.31$ kV/cm, the $L$ cell begins to elongate along $x$ direction, and the original and replicate $C$ points are pulled apart to separate poles. Then the middle portion of the $L$ cell begins to sink, and a cross wall is well developed and formed at $E_0 = 6.90$ kV/cm in Figure 3(c). When $E_0 = 10.62$ kV/cm, the $L$ cell has been completely split into two daughters of identical $C$ point, shown in Figure 3(d). Obviously, present manipulation on polarization singularities could be flexible and fast, since the electro-optic Pockels effect possesses a responsible time less than one nanosecond.34

It is crucial for us to reveal the underlying reasons that support the above interesting features. By analogy between polarization and phase singularities, we attribute this to the accompanying angular momentum dynamics. In principle, we can express the transverse light field in terms of both circular polarizations and spiral harmonics,

$$E_\perp(r, z) = \sum_{l = -\infty}^{\infty} [E_l^+ (r, z)\hat{e}_+ + E_l^- (r, z)\hat{e}_-] \exp(i\ell\phi),$$

and $W_l^\pm = \frac{1}{\eta} \int_0^\infty 2\pi r|E_l^\pm (r, z)|^2 dr$ (with $\eta$ being the normalized constant) can thus be interpreted as the weight of each OAM mode. As an echo of Figure 3, we show the numerical results of $W_l^\pm$ in Figure 4. In Figure 4(a), the initial left-handed circularly polarized light with $l = 0$ is partially converted into right-handed one with $l = 2$ while acquires $2h$ OAM per photon, and therefore conserving total angular momentum. This is just the case for a pure uniaxial crystal.36 When undergoing Pockels effect, besides the energy transfer from left- to right-handed circular component, we find that the energy for each circular component is distributed over several neighboring even OAM modes, i.e., the OAM sidebands are generated due to mode coupling. For left-handed component, the main coupling is to the $L$ line $\pm 2$ modes, with a small efficiency of 1.7%, 2.7% and 4.7% in Figure 4(b), 4(c) and 4(d), respectively; while those to other higher modes are even weaker, as interfered from insets of Figure 4. For right-handed component, the significant mode coupling occurs

\[\text{Figure 1 | The theoretical scheme. (a) The proposed schematic diagram. (b) The deformed ellipse of refractive index of KDP when undergoing the Pockels effect.}\]
Figure 2 | The 3D structures of C lines (blue) and L surfaces (red). Different electric fields are applied: (a) $E_0 = 0$, (b) $E_0 = 5.31$ kV/cm, (c) $E_0 = 6.90$ kV/cm and (d) $E_0 = 10.62$ kV/cm. All coordinates are in unit of $\mu m$. See also the Supplemental information video 1.

Figure 3 | The 2D fine structure of C points (blue dots) and L lines (red lines). Green lines denote the streamlines of major axis of polarization ellipse. Under the control of $E_0$, they behave like the binary fission of a prokaryotic cell, where $E_0$ is the same as those in Figure 2. See also the supplementary information video 2.
between \( l = 0 \) and \( l = +2 \). Besides, it is electrically tunable. As has been revealed by Angelsky et al\(^{39} \), there is a relationship between topological characteristics of component vortices and polarization singularities, namely, \( C \) points locate at the vortices of the opposite circular component. In our case, the left-handed \( C \) points are coming from the vortices of right-handed component. For a pure uniaxial crystal, only \( l = +2 \) vortex exists such that only a left-handed \( C \) points emerges. It is just the superposition of \( l = 0 \) and \( l = +2 \) modes that accounts for the formation of a pair of right-handed component vortices, and therefore, the formation of a pair of left-handed \( C \) points. As \( E_0 \) is increasing, the intensity ratio of \( l = +2 \) to \( l = 0 \) decreases from 30\%, 16\% to 5\%, shown in Figure 4(b) to 4(d). As a consequence, two vortices are pushed away, so are the \( C \) points, see Figure 3(b) to 3(d).

**Discussion**

The above generation of OAM sidebands can be well understood from the transverse angular anisotropy induced by the Pockels effect. According to the refractive index ellipsoid theory\(^{33} \), we know that, with the application of electric field along \( z \) direction, the transverse isotropy of \( n_x = n_y = n \) cannot hold anymore; instead, \( n_x = n_0 - n^{\gamma_{E_0}}_x E_0/2 \) and \( n_y = n_0 + n^{\gamma_{E_0}}_y E_0/2 \). Thus we can define the quantity, \( \Delta = n_x - n_y = n^{\gamma_{E_0}}_x E_0 \), to characterize the broken degree of rotational invariance around the optic axis, which is evidently proportional to \( E_0 \). As is well known, the conservation law of angular momentum is naturally linked with the rotational symmetry. So here we expect that applying a larger \( E_0 \) will give rise to a larger nonconservative amount of angular momentum. Generally, the angular momentum per photon within emerging light can be expressed as a sum of spin and orbital parts, namely, \( J = \sum (l+1)W^+_l + \sum (l-1)W^-_l \). By calculation, we obtain the angular momentum change (after subtracting \( \hbar \) for initial left-handed light): \( \Delta J = 0 \hbar, \Delta J = -0.5794\hbar, \Delta J = -0.9015\hbar \) and \( \Delta J = -1.6008\hbar \) for Figure 4(a)–4(d), respectively, thereby confirming our prediction.

But, surprisingly enough, the total topological index of \( C \) points is preserved. In Figure 3(a), the polarization streamlines make up spiral branches. As we make a complete circuit clockwise around \( C \) point, note that the polarization ellipse rotates clockwise through a complete revolution. Consequently, the topological index is \( I_C = +1 \), and this corresponds to the double degeneracy of the central \( C \) point. While in Figure 3(b)–3(d), the signed number of turns that the streamlines makes around each \( C \) point is \(+1/2\), and the number of streamlines that terminate on the \( C \) point is 1, so each \( C \) point is a lemon type\(^{11} \). Therefore, the total topological index is preserved, namely, \( I_C = 1/2 + 1/2 = 1 \), despite that \( E_0 \) is changing. A complex Stokes field, \( \sigma = S_1 + iS_2 = \sigma_0 \exp(i2\theta) \) (\( \theta \) is the orientation of polarization ellipse), is usefully defined to study the Stokes vortex\(^{40} \). It follows that the index of \( C \) points \( I_C \) is just half the charge of the Stokes vortex. As can be seen from Figure 4, only and always the right-handed vortex of charge 2 dominates in the OAM sidebands such that \( I_C = 1 \) remain preserved in each subfigure.

In conclusion, we have discovered and visualized the interesting dynamics evolution of polarization singularities for a light field emerging from a uniaxial crystal undergoing the Pockels effect, where the splitting of \( C \) points and fission of \( L \) lines are animated in analogy with the cleavage of nucleus and division of cytoplasm in the binary fission. Because of the rotational symmetry breaking, we find that the total angular momentum of light is not conserved, but unexpectedly, the total topological index of \( C \) points is conserved. We revealed the connection of these findings with the accompanying generation of OAM sidebands, as a result of the OAM mode coupling induced by the Pockels effect. Our results may supply another perspective of angular momentum conservation law in the context of

![Figure 4](https://example.com/fig4.png)
rotational symmetry breaking, and provide a flexible and fast manipulation on the polarization singularities.

**Methods**

According to the refractive index ellipsoid theory, with the application of longitudinal electric field $E_L$, the refractive index ellipsoid of KDP is deformed as $n_z^2 = n_x^2 + b^2 n_y^2 + c^2 n_z^2 + 2a c E_L$, where $a$, $b$, and $c$ denote the crystalline axes. Considering the symmetry of $a$ and $b$ in the ellipsoid equation, we choose a new coordinate system $x$, $y$, and $z$, where $x$ and $y$ are related to $a$ and $b$ by 45° rotation while $z$ is parallel to $c$, as illustrated in Fig. 1(b). Then the equation of index ellipsoid becomes $n_z^2 = n_x^2 + n_y^2 + z^2 / n_z^2 = 1$. By considering the nonlinearity induced by the Pockels effect as a perturbation, we have introduced in Ref. [36] an accurate and numerically cheap method based on asymptotic expansion theory. Specifically, one need to calculate a typical oscillatory integral in Eq. (7), $I(z) = \int \exp(izf(x,y))dx dy$, where the function $g$ can be expanded as $f = f_1 \sim f_2$ with $f_1 \sim f_2$ with $f_1, f_2$ in one oscillating region, and making a 45° coordinate rotation back to $x$, $y$, and $z$, we finally arrive the coupling equations that the light field components satisfy,

$$\frac{\partial^2 E_z}{\partial z^2} + i k_z E_z + k_z \frac{\partial E_z}{\partial z} + \left[ k^2 (n_x^2 - n_y^2) / 2 \right] E_z + k_z^2 E_z = 0,$$

$$(4)$$

$$\frac{\partial^2 E_y}{\partial z^2} + i k_z E_y + k_z \frac{\partial E_y}{\partial z} + \left[ k^2 (n_x^2 - n_y^2) / 2 \right] E_y + k_z^2 E_y = 0,$$

$$(5)$$

$$\frac{\partial^2 E_x}{\partial z^2} + i k_z E_x + k_z \frac{\partial E_x}{\partial z} + \left[ k^2 (n_x^2 - n_y^2) / 2 \right] E_x + k_z^2 E_x = 0.$$

$$(6)$$

The above equations form a complete description of light propagation in the momentum space. Note that the coefficients of $E_z$ in Eq. (4) and $E_y$ in Eq. (5) are both electrically tunable with $E_L$, which play a key role in our flexible manipulation of polarization singularities. In our simulation, we consider the incident light is a left-handed circularly polarized Gaussian one, namely, $E_z(0, x, z, 0) = \exp(-r^2 / 2a^2) \cos(\phi)$. With the boundary condition of $E_z(0, x, z, 0) = \frac{1}{2\pi} \exp(-k_z^2 x^2 / 2a^2)$, the second-order partial differential equations (4)–(6) can be solved to obtain the solution for $E(k_x, z)$.
singularities. It is noted that our method can be extended to any crystals of arbitrary point group with the biaxial anisotropy is induced by the Pockels effect, Kerr effect or other nonlinear optical effects.

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Author contributions

L.C. conceived the theory, X.L. and L.C. performed the numerical calculations. X.L., L.C., Z.W. and W.Z. analyzed the numeric data. L.C. supervised the project. All authors discussed the results and contributed to the writing of the manuscript.

Additional information

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