Spin is an intrinsic property of the photon. Poynting first identified the mechanical property of momentum of light and associated angular momentum with circular polarization. Beth presented the mechanical detection and measurement of the angular momentum of light and showed that along the propagation direction of the beam each circularly polarized photon possesses a spin angular momentum of $\hbar$. Allen et al. identified the total angular momentum of a classical light field and discussed the separation of spin and orbital angular momentum components. Barnett further derived the classical relation between spin (orbital) angular momentum flux and energy flux carried by light. Recently, Piccirillo reported an experiment in which an elliptically shaped laser beam was used to control the molecular alignment in liquid films by the transfer of orbital angular momentum of light. In this Letter we present a way of using an externally applied dc electric field to manipulate the transfer of spin angular momentum of light in an optically active medium.

To discuss this, we first consider the linear electro-optic effect in a medium possessing optical activity. Generally, optical activity and the linear electro-optic effect have been investigated individually in the past, since from a traditional point of view it may be too complicated to consider the optical activity and linear electro-optic effect at the same time. Recently, Yin et al. developed an electromagnetic theory for their electro-optic Q switch made with an optically active crystal. But their theory, including the use of refractive index ellipsoid theory, is far from general yet. Refractive index ellipsoid theory has been extensively used to analyze the linear electro-optic effect. However, the standardization of the equation of the refractive index ellipsoid is not an easy task, even using the general Jacobi method. In 2001, She and Lee presented a wave coupling theory of the linear electro-optic effect that avoids the difficulty of standardizing the equation of the refractive index ellipsoid and makes it easier to analyze the general case of the effect. Based on their theory, theories of the linear electro-optic effect in an absorbent medium and in a quasi-phase-matched medium have been developed. We find that the theory can be further generalized to the case of a medium with optical activity. In this Letter we develop a wave coupling theory of mutual action of the natural optical activity and the linear electro-optic effect and use it to discuss manipulation of the spin angular momentum of light in crystalline quartz with an externally applied dc electric field.

Spatial dispersion, which expresses the dependence of macroscopic properties of matter on the spatial inhomogeneity of the electromagnetic fields, is responsible for the phenomena of optical activity. The polarization relative to spatial dispersion can be expressed as

$$P_\mu = \varepsilon_0\chi_{\mu\rho\beta}\nabla_\rho E_\alpha = i\varepsilon_0\chi_{\mu\rho\beta}E_\alpha^*k_\beta. \quad (1)$$

In general, there are two independent polarized components in a birefringent crystal for a monochromatic plane light wave with frequency $\omega$, i.e.,

$$E(\omega) = \mathbf{E}_1(\omega) + \mathbf{E}_2(\omega) = \mathbf{E}_1(r)\exp(i\mathbf{k}_1 \cdot \mathbf{r}) + \mathbf{E}_2(r)\exp(i\mathbf{k}_2 \cdot \mathbf{r}), \quad (2)$$

where $\mathbf{E}_1(\omega)$ and $\mathbf{E}_2(\omega)$ denote two cross components of the light field when $\mathbf{k}_1 = \mathbf{k}_2$, while they denote two independent components experiencing different refractive indices when $\mathbf{k}_1 \neq \mathbf{k}_2$. The second-order polarization resulting from the spatial dispersion is

$$\mathbf{P}_1^{(2)}(\omega) = i2\varepsilon_0\kappa_1^{(2)}:\mathbf{E}_1(r)\mathbf{k}_1 \exp(ik_1r) + i2\varepsilon_0\kappa_2^{(2)}:\mathbf{E}_2(r)\mathbf{k}_2 \exp(ik_2r), \quad (3)$$

where $\kappa_1^{(2)}$ and $\kappa_2^{(2)}$ denote second-order susceptibility tensors of natural optical activity and generally $\kappa_1^{(2)} \neq \kappa_2^{(2)}$. In addition, there exists another second-order polarization responsible for the linear electro-optic effect if the optically active material is subject to an external dc electric field $\mathbf{E}(0)$. The polarization is

\begin{align*}
\mathbf{P}^{(2)}(\omega) &= i2\varepsilon_0\kappa_1^{(2)}:\mathbf{E}_1(r)\mathbf{k}_1 \exp(ik_1r) + i2\varepsilon_0\kappa_2^{(2)}:\mathbf{E}_2(r)\mathbf{k}_2 \exp(ik_2r) \\
&= i2\varepsilon_0\kappa_1^{(2)}:\mathbf{E}_1(r)\mathbf{k}_1 \exp(ik_1r) + i2\varepsilon_0\kappa_2^{(2)}:\mathbf{E}_2(r)\mathbf{k}_2 \exp(ik_2r),
\end{align*}
\[ \mathbf{P}_2^{(2)}(\omega) = 2\varepsilon_0\chi^{(2)}(\omega,0)\mathbf{E}_1(r)\mathbf{E}(0)\exp(ik_1r) + 2\varepsilon_0\chi^{(2)}(\omega,0)\mathbf{E}_2(r)\mathbf{E}(0)\exp(ik_2r). \]  

Then the total second-order polarization should be written as

\[ \mathbf{P}^{(2)}(\omega) = i2\varepsilon_0\chi^{(2)}:\mathbf{E}_1(r)\mathbf{k}_1 \exp(ik_1r) + i2\varepsilon_0\chi^{(2)}:\mathbf{E}_2(r)\mathbf{k}_2 \exp(ik_2r) + 2\varepsilon_0\chi^{(2)}(\omega,0):\mathbf{E}_1(r)\mathbf{E}(0)\exp(ik_1r) + 2\varepsilon_0\chi^{(2)}(\omega,0):\mathbf{E}_2(r)\mathbf{E}(0)\exp(ik_2r). \]  

Similarly to Ref. 11, starting from Maxwell's equations, considering only the second-order nonlinearity, and neglecting high-order nonlinearity as well as linear absorption, we can derive, under the slowly varying amplitude approximation and no walk-off approximation, the wave coupling equations for the mutual action of natural optical activity and the linear electro-optic effect, as follows:

\[ \frac{dE_1(r)}{dr} = \left( \frac{1}{n_1} f_0 - id_1 \right) E_2(r)\exp(i\Delta kr) - id_2 E_1(r), \]  

\[ \frac{dE_2(r)}{dr} = \left( -\frac{1}{n_2} f_0 - id_3 \right) E_1(r)\exp(-i\Delta kr) - id_4 E_2(r), \]

where \( \Delta k = k_2 - k_1 \) and \( d_i \) \((i = 1, 2, 3, 4)\) are the same as those in Ref. 11 and \( f_0 = -\Sigma_{jk}\chi^{(2)};\left( a_j\mathbf{a}_k \right) b_k \hat{k}_l \) \( = \Sigma_{jk}\chi^{(2)};\left( b_j\mathbf{a}_k \right) a_k \hat{k}_l \) (the second equation is the requirement of the law of conservation of energy), where \( a_j, b_k, \) and \( \hat{k}_l \) are components of three unit vectors \( \mathbf{a}, \mathbf{b}, \) and \( \mathbf{k} \) parallel to \( \mathbf{E}_1, \mathbf{E}_2, \) and \( \mathbf{k} \), respectively.

It should be noted that in the derivation of Eqs. (6a) and (6b) the relation \( k_2^{(1)} = -k_1^{(1)} \) has been used, which leads to \( \Sigma_{jk}a_j\chi^{(2)};\left( b_k \right) a_k = 0 \) and \( \Sigma_{jk}b_j\chi^{(2)};\left( a_k \right) b_k = 0 \).

For a crystal without natural optical activity \( f_0 = 0 \), Eqs. (6a) and (6b) reduce to the equations describing the linear electro-optic effect discussed in Ref. 11. And, in the absence of the external dc electric field, namely, \( d_i = 0 \) \((i = 1, 2, 3, 4)\), Eqs. (6a) and (6b) become those describing the phenomena of pure natural gyrotropy. Finding the general analytic solutions of Eqs. (6a) and (6b) is without any difficulty. But now we prefer using the theory to discuss the transfer of spin angular momentum of light in crystalline quartz controlled by an applied dc electric field. For simplicity, assuming that the light propagates along the optical axis of this crystal with an applied dc field along \( y \)-axis direction, then we have \( \hat{k} = (0,0,1), \mathbf{a} = (1,0,0), \mathbf{b} = (0,1,0), \) and \( \mathbf{c} = (0,1,0). \) In this case, \( n_1 = n_2 = n_0, \Delta k = 0, \) and \( \chi^{(2)}_1 = \chi^{(2)}_2 = \chi^{(2)}_0. \) According to the nonvanishing elements of the second-order tensor of crystalline quartz \( (\text{belonging to class } 32)\), \( k = 0 \), \( a_3 = k_0E_0/2 = d_1 = d_3, \) and \( d_2 = d_4 = 0. \) Subsequently, Eqs. (6a) and (6b) become

\[ \frac{dE_1(r)}{dr} = (f - id)E_2(r), \]

\[ \frac{dE_2(r)}{dr} = (-f - id)E_1(r), \]

where \( f = f_0/n_0 - k_0\chi^{(2)}_0. \) Assuming that the incident light beam is linearly polarized with the initial condition \( E_1(0) = E_{in}, E_2(0) = 0, \) and \( f > 0, \) then we obtain the solutions as follows:

\[ E_1(r) = E_{in}\cos(\sqrt{f^2+d^2}r), \]

\[ E_2(r) = -E_{in}\sin(\sqrt{f^2+d^2}r)\exp(i\theta), \]

where \( \theta = \arg(f + id). \) In the absence of the external electric field, \( \theta = \arg(f + id) = 0; \) Eqs. (8a) and (8b) become \( E_1(r) = E_{in}\cos(fr) \) and \( E_2(r) = -E_{in}\sin(fr), \) which are just the equations describing the phenomena of natural optical activity for dextrorotatory quartz, where the spin angular momentum remains zero throughout and \( f \) is just the known optical rotatory power.

In the following, we focus our attention on the manipulation of spin angular momentum of light in crystalline quartz with an externally applied dc electric field. In general, the light field described by Eqs. (8a) and (8b) can be expressed as the superposition of two circularly polarized lightfields: one is left-handed and the other right-handed, i.e.,

\[ E(r) = \begin{bmatrix} E_1(r) \\ E_2(r) \end{bmatrix} = E_{in} \begin{bmatrix} \alpha_L \left( 1/\sqrt{2} \right) + \alpha_R \left( 1/\sqrt{2} \right) \\ -i/\sqrt{2} \end{bmatrix}, \]

where

\[ \alpha_L = \frac{1}{\sqrt{2}} \left( \cos(\sqrt{f^2+d^2}r) - i \sin(\sqrt{f^2+d^2}r)\exp(i\theta) \right), \]

\[ \alpha_R = \frac{1}{\sqrt{2}} \left( \cos(\sqrt{f^2+d^2}r) + i \sin(\sqrt{f^2+d^2}r)\exp(i\theta) \right). \]

Suppose the input and the output surfaces of the crystalline quartz have perfect antireflection coatings. Then the numbers of left- and right-handed circularly polarized photons transmitted at the output surface per unit area per second are the respective average Poynting energy flows divided by the energy per photon, i.e.,

\[ N_L = \frac{c\varepsilon_0\alpha_L E_{in}^2}{2\hbar\omega}, \quad N_R = \frac{c\varepsilon_0\alpha_R E_{in}^2}{2\hbar\omega}. \]

The angular momentums of left- and right-handed circularly polarized photons transmitted per unit area per second are \( -N_L \hbar \) and \( N_R \hbar \), respectively. Then the total angular momentum transmitted at
It is zero at conserved. From Fig. 1(b) we find that the total spin with the applied electric field, but the total number is
circularly polarized photon number varies
lar momentum of the light in the optically active
duced by the applied electric field; i.e., the spin angu-
field. This reveals that there is a transfer of spin an-
as shown in Fig. 1.
\[ M = \frac{c\varepsilon_0 L^2}{2\omega}(|\alpha_R|^2 - |\alpha_L|^2) = \frac{c\varepsilon_0 L^2}{2\omega} \sin \theta \sin(2\sqrt{r^2 + d^2}) \]  \ hspace{1cm} (12)

Without losing generality, we take a 1 cm long (along the z axis of the crystal) and 2 mm thick (along the y axis of the crystal) quartz crystal as the working material to demonstrate the application of our theory. Assuming that the wavelength of the incident light is 550 nm, then the corresponding optical rotatory power is \( f = 4.41568 \text{ cm}^{-1} \), and the refractive indices for ordinary and extraordinary waves are \( n_o = 1.54 \) and \( n_e = 1.55 \), respectively; the nonvanishing electro-optic coefficients of crystalline quartz are \( \gamma_{11} = -\gamma_{22} = 0.2 \) and \( \gamma_{21} = -\gamma_{13} = 0.93 \) (in \( 10^{-12} \text{ m/V} \)), respectively.\(^{16}\) Using these parameters, we can get \( d = 0.097 \text{ V cm}^{-1} \), where \( V \) (kilovolts) denotes the applied dc voltage. Substituting all these parameters into Eqs. (11) and (12), we can calculate \( N_L, N_R \), and \( M \) as shown in Fig. 1.

From Fig. 1(a) we can see that the left- or right-handed circularly polarized photon number varies with the applied electric field, but the total number is conserved. From Fig. 1(b) we find that the total spin angular momentum of the photons is not conserved. It is zero at \( V = 0 \) and varies with the applied electric field. This reveals that there is a transfer of spin angular momentum from the medium to the light induced by the applied electric field; i.e., the spin angular momentum of the light in the optically active medium can be controlled through the linear electro-optic effect. According to the law of conservation of angular momentum, we know that light should transfer angular momentum to the medium in return during this process and exert a torque on it. As is well known, spin and orbital angular momentum can be transferred from a light beam to particles held within optical tweezers, forming an optical spanner.\(^{17,18}\) We have shown in the present Letter that with the aid of the external dc electric field linearly polarized light can also form an optical spanner, which can exert a torque on the medium. In addition, the transfer of spin angular momentum can be continuously manipulated by tuning the dc electric field.

In conclusion, we have shown theoretically that an externally applied dc electric field can control the transfer of spin angular momentum of light in an optically active medium and make linearly polarized light form an optical spanner.

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