Applications of polarization-shaped femtosecond laser pulses

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ABSTRACT

This review article presents the generation and control of polarization-shaped femtosecond laser pulses. Some applications of these pulses to coherent control are described, including multiphoton photoionization, forced molecular rotation, optical control of lattice vibrations, nano-optics, and wavelength conversion. Theoretical expectations for enantiomeric purification and magnetization are also discussed.

1. Introduction

Quantum control concerns the active manipulation of physical and chemical processes on the atomic and molecular scales. Precise and arbitrary control of chemical reactions such as isomerization, dissociation, and ionization has been a primary goal of this field over the past two decades. Programmable pulse shaping would enable coherent control of light–matter interactions. The technology for shaping ultrashort pulses was developed experimentally by manipulating the individual frequency components of broadband femtosecond laser pulses [1]. Several review articles discuss various experimental examples of quantum control using programmable pulse shaping [2–4].
Conventional pulse-shaping technology involves modulating the spectral amplitudes and phases of linearly polarized light pulses. In such methods, the optical electric field is treated as a scalar field. However, generally, the vector components of the electric field, (i.e. the polarization state) must be considered because quantum systems are three dimensional. Numerous studies have focused on the polarization-dependent optical response due to spatial anisotropy of isolated molecules, chiral molecules, and crystalline materials. For example, multiphoton ionization, control of lattice vibrations, and conversion between enantiomers are possible using polarization-shaped pulses. One simple means of polarization control is to use a pair of linearly polarized pulses with twisted polarization directions. Changing the polarization state between the pulses in a pair enables us to impart nuclear motion [5], molecular rotation [6], and momentum of magnetization [7] in orthogonal directions.

In recent years, with advancement in techniques that generate and manipulate ultrashort laser pulses, a technique to generate vector-shaped light pulses has been developed, wherein the instantaneous polarization state dynamically changes within a single pulse envelope [8–15]. The benefit of using vector-shaped light pulses is a better understanding of anisotropic molecular and crystalline systems compared to phase- and amplitude-shaped pulses. However, shaping of the vectorial waveform is not a simple technique; thus, applications of this technique to three-dimensional quantum control are still limited. Several early applications appeared independently but simultaneously in 2004 [16–18]. Dudovich et al. used a pulse shaper capable of adjusting two of the four parameters, namely the amplitude ratio of two orthogonal polarization components and the common spectral phase, to observe quantum interference in the final state population via two-photon absorption [16]. Suzuki et al. [17] and Brixner et al. [18] showed optimal control of ionization processes in iodine molecules and potassium dimers, respectively. Since these publications appeared two decades ago, other studies have presented similar vectorial coherent control using optical pulses with a time-dependent polarization state.

Figure 1. (Left) Basic configuration of femtosecond polarization pulse shaper. (Right) Structure of the liquid–crystal spatial light modulator.
This review article presents some examples from the field of coherent control. In Section 2, we describe a basic experimental setup for a polarization pulse shaper, together with two simple but important examples of polarization-shaped pulses, namely polarization twisting and swept-polarization rotation. Section 3 discusses the applications of polarization-shaped pulses; a summary of experimental results is provided for multiphoton photoionization, forced molecular rotation, optical control of lattice vibrations, nano-optics, and wavelength conversion. Finally, theoretical expectations for the selective enantiomeric purification and magnetization are discussed.

2. Polarization pulse shaping

2.1. Basic configuration

The first computer-controlled femtosecond polarization pulse shaping was demonstrated by Brixner et al. with both the phase and polarization state of each frequency component being manipulated independently within a single pulse [8]. Figure 1 shows the basic configuration of a femtosecond polarization pulse shaper, which comprises an all-reflective zero-dispersion compressor in a folded 4f-geometry and a computer-controlled liquid crystal spatial light modulator (LC-SLM) [8,9]. The incident light polarized along the $x$ axis is spectrally dispersed by a grating and the spectral components are parallelized by a concave mirror, which is displaced by one focal length, $f$, from a grating. Lenses can also be used rather than concave mirrors, but reflective optics are preferred to avoid spectral dispersion of the lens material. Each spectral component is simultaneously focused onto the Fourier plane, which is displaced by one focal length from the mirror. This results in the Fourier transformation of the incident femtosecond pulse from the temporal to the spectral domain. In the Fourier plane, pulse shaping can be performed by phase modulation of the spectral components. Therefore, an LC-SLM with two liquid–crystal layers is placed in the Fourier plane. Each spectral component passes through a pair of liquid–crystal cells in the two successive layers of this device.

The axes of the liquid crystal molecules in the first and second layers are preferentially oriented along the orthogonal directions of $-45^\circ$ and $45^\circ$, respectively, with respect to the horizontal $x$ axis in the $x-y$ plane [1]. The liquid crystal cells independently control the phases $\phi_{-45^\circ}(\omega)$ and $\phi_{45^\circ}(\omega)$ of these two components, as expressed by

$$
\begin{bmatrix}
E_{-45^\circ}(z,t) \\
E_{+45^\circ}(z,t)
\end{bmatrix} =
\begin{bmatrix}
A_{-45^\circ}(\omega) \cos(kz - \omega t + \phi_{-45^\circ}(\omega)) \\
A_{+45^\circ}(\omega) \cos(kz - \omega t + \phi_{+45^\circ}(\omega))
\end{bmatrix}.
$$

The incident light $E^{(\omega)}(z,t) = A(\omega) \cos(kz - \omega t)$ is linearly polarized in the $x$-direction, so it is divided into two equal-amplitude polarization components
along the $\pm 45^\circ$ directions. Thus, the amplitudes of both $A_{-45^\circ}(\omega)$ and $A_{+45^\circ}(\omega)$ are $\frac{1}{\sqrt{2}}A(\omega)$.

Here, we define the phase shifts as

$$
\phi(\omega) \equiv \frac{\phi_{-45^\circ}(\omega) + \phi_{+45^\circ}(\omega)}{2}, \quad \delta(\omega) \equiv \frac{\phi_{-45^\circ}(\omega) - \phi_{+45^\circ}(\omega)}{2}.
$$

After spectral phase modulation, the horizontal $E_{x}^{(\omega)}$ and vertical $E_{y}^{(\omega)}$ electric fields are converted from $E_{-45^\circ}^{(\omega)}$ and $E_{+45^\circ}^{(\omega)}$ as follows:

$$
\begin{bmatrix}
E_{x}^{(\omega)}(z, t) \\
E_{y}^{(\omega)}(z, t)
\end{bmatrix} =
\begin{bmatrix}
\frac{1}{\sqrt{2}}E_{-45^\circ}^{(\omega)}(z, t) + \frac{1}{\sqrt{2}}E_{+45^\circ}^{(\omega)}(z, t) \\
\frac{1}{\sqrt{2}}E_{+45^\circ}^{(\omega)}(z, t) - \frac{1}{\sqrt{2}}E_{-45^\circ}^{(\omega)}(z, t)
\end{bmatrix} =
\begin{bmatrix}
\frac{1}{2}A(\omega) \left\{ \cos \left( k z - \omega t + \phi(\omega) - \delta(\omega) \right) + \cos \left( k z - \omega t + \phi(\omega) + \delta(\omega) \right) \right\} \\
\frac{1}{2}A(\omega) \left\{ \cos \left( k z - \omega t + \phi(\omega) - \delta(\omega) \right) - \cos \left( k z - \omega t + \phi(\omega) + \delta(\omega) \right) \right\}
\end{bmatrix}
$$

Therefore, the basic configuration of a polarization pulse shaper only allows the amplitude ratio $E_{y}^{(\omega)}/E_{x}^{(\omega)} = \tan \delta(\omega)$, and the common spectral phase, $\phi(\omega)$, to be adjusted. To form polarization-shaped pulses, the phase-modulated components are recombined with a time-reversed combination of a concave mirror and grating displaced by one focal length from the Fourier plane (Figure 1, left). The electric field of the resulting pulse may be described by

$$
\begin{bmatrix}
E(z, t)
\end{bmatrix} =
\begin{bmatrix}
\int_0^\infty E_{x}^{(\omega)}(z, t) d\omega \\
\int_0^\infty E_{y}^{(\omega)}(z, t) d\omega
\end{bmatrix} =
\begin{bmatrix}
\int_0^\infty A(\omega) \cos \delta(\omega) \cos \left( k z - \omega t + \phi(\omega) \right) d\omega \\
\int_0^\infty A(\omega) \sin \delta(\omega) \sin \left( k z - \omega t + \phi(\omega) \right) d\omega
\end{bmatrix}
$$

To avoid distortion of the output waveform, the spectral combining grating should differ only slightly in diffraction efficiency for the s- and p-polarizations. As dielectric mirrors introduce polarization-dependent phase retardation, metal steering mirrors are used to direct the laser beam from the pulse shaper to the sample to reduce unwanted modifications of the polarization state as low as possible.

All optical elements in this 4f-geometry are symmetrically distanced from the Fourier plane by the focal length of the concave mirrors, so a reflective pulse shaper has also been developed in which a flat mirror is placed at the Fourier plane as shown in Figure 2. This pulse shaper uses a volume-phase holographic
grating, which offers high diffraction efficiency for both the s- and p-polarizations with only a small difference between them.

2.2. Basis transformation and polarization-twisting pulses

2.2.1. Transformation into a circular polarization basis

Consider placing a quarter-wave plate (QWP) at the exit of the pulse shaper after the phase-modulated spectral components are recombined in the basic configuration. The phase retardation should be constant over the broad spectral bandwidth of the femtosecond pulses. The fast axis of the QWP is the x axis; after passing through the QWP, the two orthogonal linear polarizations are converted into right- and left-circular polarizations:

\[
\begin{bmatrix}
E_x^{(s)}(z, t) \\
E_y^{(s)}(z, t)
\end{bmatrix}
= \frac{1}{\sqrt{2}} E_{Lx}^{(s)}(z, t) + \frac{1}{\sqrt{2}} E_{Rx}^{(s)}(z, t)
= \frac{1}{2} A(\omega) \begin{bmatrix}
\cos(\delta(\omega)) & \sin(\delta(\omega))
\end{bmatrix}
\begin{bmatrix}
\cos\left(kz - \omega t + \phi(\omega) - \delta(\omega)\right) + 
\cos\left(kz - \omega t + \phi(\omega) + \delta(\omega)\right)
\end{bmatrix}
\begin{bmatrix}
\cos\left(kz - \omega t + \phi(\omega)\right)
\end{bmatrix}
\]

\[= A(\omega) \begin{bmatrix}
\cos(\delta(\omega)) & \sin(\delta(\omega))
\end{bmatrix} \cos\left(kz - \omega t + \phi(\omega)\right)
\]

The electric field of the spectrally combined output may be written as

\[
E(z, t) = \begin{bmatrix}
\int_{-\infty}^{\infty} E_x^{(s)}(z, t) d\omega \\
\int_{-\infty}^{\infty} E_y^{(s)}(z, t) d\omega
\end{bmatrix}
= \int_{-\infty}^{\infty} A(\omega) \begin{bmatrix}
\cos(\delta(\omega)) & \sin(\delta(\omega))
\end{bmatrix} \cos\left(kz - \omega t + \phi(\omega)\right) d\omega
\]

Owing to this basis transformation, the pulse shaper can adjust the azimuthal angle \(\delta(\omega)\) and common spectral phase \(\phi(\omega)\) of linearly polarized light. The QWP
linearly polarizes all spectral components, and the ellipticity of each spectral component $\delta(\omega)$ is converted into its azimuthal angle upon passing through the QWP:

$$\phi(\omega) \equiv \frac{\phi_{-45^\circ}(\omega) + \phi_{+45^\circ}(\omega)}{2}, \quad \delta(\omega) \equiv \frac{\phi_{-45^\circ}(\omega) - \phi_{+45^\circ}(\omega)}{2}.$$  

### 2.2.2. Polarization-twisting pulses

We now introduce the following spectral parameters into the pulse shaper:

$$\phi(\omega) = \frac{\beta}{2} (\omega - \omega_0)^2, \quad \delta(\omega) = \alpha (\omega - \omega_0),$$

where $\omega_0$ is the central laser frequency. The role of the spectral phase $\phi(\omega)$ is to introduce a linear chirp and stretch the light pulse in the time domain, whereas the role of $\delta(\omega)$ is to temporally shift the peak position of the pulse envelope. For simplicity, we assume a Gaussian spectral distribution $A(\omega) = e^{-\Delta^2(\omega-\omega_0)^2}$, so

$$E(z, t) = \int_{-\infty}^{\infty} e^{-\Delta^2(\omega-\omega_0)^2} \left[ \cos \delta(\omega) \sin \delta(\omega) \right] \cos \left( k_z - \omega_0 + t + \phi(\omega) \right) \, d\omega$$  

We set $|\beta|$ to be sufficiently larger than the inverse square of the bandwidth $\Delta^2$ of the laser pulse. After Fourier integration, we obtain the following formula with a phase shift $\theta$ determined by $\beta$ and $\Delta^2$:

$$E(z, t) = 2 \begin{bmatrix} \cos \left( \frac{\alpha}{\beta} t \right) \\ \sin \left( \frac{\alpha}{\beta} t \right) \end{bmatrix} \cos \left\{ k_z - \left( \omega_0 + \frac{t}{2\beta} \right) t - \left( \frac{\alpha^2}{2\beta} - \theta \right) \right\}.$$  

The output forms **polarization-twisting pulses**, in which the major-axis of polarization rotates at a fixed angular frequency $\Omega = \alpha/\beta$. The cosine and sine factors in the field amplitudes describe the rotating major axis of the optical polarization. The optical carrier frequency is the same for the two orthogonal polarization components. Note that the central optical frequency is linearly chirped as $\omega = \omega_0 + \frac{t}{2\beta}$.

Polarization-twisting pulses can be understood as a superposition of counter-rotating left- and right-circularly polarized pulses with shifted center frequencies. To observe this, we decompose Equation (7) into

$$E(z, t) = 2 \begin{bmatrix} \cos \Omega t \cos (k_z - \omega t) \\ \sin \Omega t \cos (k_z - \omega t) \end{bmatrix} \begin{bmatrix} \cos (k_z - (\omega + \Omega) t) + \cos (k_z - (\omega - \Omega) t) \\ - \sin (k_z - (\omega + \Omega) t) + \sin (k_z - (\omega - \Omega) t) \end{bmatrix}.$$  

(8)
The optical fields of the two circularly polarized pulses can now be written as

\[
\begin{align*}
E_L(t) &= \frac{1}{\sqrt{2}} \begin{bmatrix} \cos(kz - \omega_1 t) \\ \cos(kz - \omega_1 t + \pi/2) \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} \cos(kz - \omega_1 t) \\ -\sin(kz - \omega_1 t) \end{bmatrix} \quad (9) \\
E_R(t) &= \frac{1}{\sqrt{2}} \begin{bmatrix} \cos(kz - \omega_2 t) \\ \cos(kz - \omega_2 t - \pi/2) \end{bmatrix} = \frac{1}{\sqrt{2}} \begin{bmatrix} \cos(kz - \omega_2 t) \\ \sin(kz - \omega_2 t) \end{bmatrix}. \quad (10)
\end{align*}
\]

Here, \(\omega_1 = \omega + \Omega\) and \(\omega_2 = \omega - \Omega\) denote the central frequencies of the left- and right-circularly polarized pulses. Superimposing these two pulses creates a pulse with a central frequency of \(\omega = \frac{\omega_1 + \omega_2}{2}\) and have a twisting envelope at a rotating rate of \(\Omega = \frac{|\omega_1 - \omega_2|}{2}\).

### 2.2.3. Swept polarization rotation

Another important example of polarization shaping is polarization-twisting pulses, wherein the angular frequency of the rotating major axis changes linearly in time. For this purpose, the following spectral parameters should be introduced into the pulse shaper:

\[
\phi(\omega) = 0, \quad \delta(\omega) = -\frac{\beta}{2}(\omega - \omega_0)^2,
\]

where \(\omega_0\) is the central laser frequency. The spectral phase \(\delta(\omega)\) serves to introduce a linear chirp. For simplicity, we again assume the Gaussian spectral distribution \(A(\omega) = e^{-\Delta^2(\omega-\omega_0)^2}\), which leads to

\[
E(z, t) = \int_{-\infty}^{\infty} e^{-\Delta^2(\omega-\omega_0)^2} \begin{bmatrix} \cos \delta(\omega) \\ \sin \delta(\omega) \end{bmatrix} \cos(kz - \omega t + \phi(\omega)) d\omega \quad (11)
\]

Again, we set \(|\beta|\) to be sufficiently larger than the inverse square of the bandwidth \(\Delta^2\) of the laser pulse. After Fourier integration, we obtain

\[
E(z, t) = 2 \begin{bmatrix} \cos \left(\frac{t}{2\beta} t + \theta\right) \\ \sin \left(\frac{t}{2\beta} t + \theta\right) \end{bmatrix} \cos(kz - \omega_0 t). \quad (12)
\]

### 2.3. Complete vector-field control

The basic configuration first introduced by Brixner and Gerber does not offer arbitrary control of the spectral polarization. The available polarization states are limited to elliptical polarizations with their major axes fixed along the \(x\) or \(y\) directions. Full control over the electric fields of ultrashort pulses requires four independent degrees of freedom, namely the spectral phases and spectral amplitudes of the two orthogonal polarization components. Two different approaches are possible for achieving complete vector-field control.
The first method for complete arbitrary control of the vector field is to use a four-layer SLM. Two layers of the SLM are aligned with their optical axes directed at \( \pm 45^\circ \) relative to the incident linearly polarized light for amplitude and phase shaping. The combination of the third liquid crystal layer aligned with its axis directed at +45° followed by the fourth with its axis directed at 0°, enables us to exercise arbitrary control over the polarization state [15]. When the total energy of the shaped pulse is maintained during the control experiments, adding a third SLM to the basic configuration is sufficient [10]. A set of two layers enables shaping of elliptical polarizations with a fixed major axis. The following single-array at the appropriate orientation is capable of shifting the relative phase between the \( x \)- or \( y \)-polarization components. This setup using three consecutive SLMs can adjust three of the four parameters: amplitude ratio, common phase, and phase difference.

The second method is to modulate the phase and amplitude of the two orthogonal polarization components in two separate arms of an interferometer, and then recombine the components into a single laser pulse [11–14,19]. Thermal deformation or vibration of optical elements causes drifts and fluctuations in the relative path length of the interferometer. In order to avoid serious distortions of shaped vectorial waveforms, it is crucial to maintain a stable relative phase between the polarization components. In most cases, the two perpendicularly polarized pulses are simultaneously introduced into a single 4\( f \) setup in the frequency domain by allocating the two polarization components to different groups of pixels within the same SLM. This single-shaper setup for both paths of the interferometer results in more stability than two independent 4\( f \)-shaper layouts [11]. For further phase stabilization, common path setting [12,13,19] or an active feedback mechanism [14] should be used to reduce the phase drift between the two orthogonal polarization components.

In the common path-setting approach [12,13,19], the input laser pulse is split into two orthogonal polarizations with a Wollaston prism. Figure 3 shows a schematic of the setup. The two orthogonally polarized pulses are imaged onto the input plane of a 4\( f \) zero-dispersion stretcher by a concave mirror. The angles of incidence on the grating are adjusted to map the spectrum from each polarization onto roughly one half of the spatial region on the SLM. After the flat mirror set to a retro condition at the SLM, the two beams retrace their incoming optical paths through the pulse shaper and are recombined by the prism into a single polarization-shaped field. In this way, the amplitude and phase can be manipulated for both polarization components separately and independently, realizing full control over all four degrees of freedom of the vectorial electric fields.

In comparison, the present authors reliably and stably generated polarization-shaped pulses by incorporating an active stabilization mechanism of the relative phase between the two orthogonal polarization components in the interferometric pulse shaper [14]. As a reference signal for the feedback, the interference
Figure 3. Common path setup of the shaper. Reproduced with permission from Ref. [12].

Figure 4. Schematic diagram of Mach–Zehnder pulse shaper and stabilization mechanism. BS: beam splitter; SLM: spatial light modulator; PBS: polarizing beam splitter; PZT: piezoelectric actuator. Reproduced with permission from Ref. [14].

The intensity of continuous wave light from an external laser diode was used, as shown in Figure 4. The interferometer was stabilized by adjusting a mirror in the interferometer using a piezoelectric actuator to fix the interference intensity.

The relative phase stability was evaluated with and without stabilization as a function of time in the long- and short-terms. Without stabilization, the error reached $300^\circ$ after one hour. On the other hand, significant improvement was observed with stabilization; the standard deviations of the errors of the stabilized relative phase were $2.3^\circ$ in the long term and $4^\circ$ in the short term, respectively.

2.4. Characterization of polarization-shaped pulses

The vectorial waveforms of polarization-shaped pulses are usually measured either by use of time-resolved ellipsometry [11] or dual-channel spectral interferometry [20] methods. Time-resolved ellipsometry, also referred as polarization-resolved intensity cross-correlation, directly provides the temporal intensities of the angle of the major principal axis (major-axis orientation), and the ratio of the axis of the elliptical polarization (ellipticity). A small portion of the unshaped pulse is selected by a beam sampler as a reference pulse for cross-correlation measurement. The polarization of the shaped pulse is first rotated through a half-wave plate, and then the polarization component parallel to the reference pulse is extracted by a polarizer. The sum-frequency signal intensity between the
shaped and reference pulses in a $\beta$-barium borate (BBO) crystal is recorded as a function of the reference pulse delay. Correlation intensity profiles are obtained for every step of rotation angle of the wave plate. In correlation measurements of temporally shaped pulses, all parameters are functions of the delay. Using the measured intensity profiles, one can calculate the instantaneous absolute intensity, the instantaneous orientation of the major axis, and the instantaneous ellipticity. For the details of the calculation procedure, see references [11,14].

The above ellipsometric cross-correlation method requires the rotation of wave plates and polarizers, together with the scanning of a time delay for each orientation of these components. Moreover, from such ellipsometric measurements, only the temporal evolution of the phase difference $\phi_{x\text{pulse}}(t) - \phi_{y\text{pulse}}(t)$ can be obtained, not the individual phases $\phi_{x\text{pulse}}(t)$ and $\phi_{y\text{pulse}}(t)$. One alternative method shown in Figure 5 is dual-channel spectral interferometry with an adequately characterized reference pulse [20]. This technique requires neither the scanning of the time delay nor the rotation of the optical axis of the wave-plate. The reference pulse linearly polarized at $45^\circ$ with respect to the $x$—axis is divided into two equal $x$ and $y$ components. The reference and the shaped pulses are combined collinearly with a fixed time delay between the two pulses. A spectral interferometric measurement is then performed on the $x$ and $y$ components by separately measuring the spectral fringes of each polarization component with a spectrometer. In this way, both the amplitudes and phases of the two orthogonal components of a polarization-shaped pulse can be temporally characterized. This technique is linear and is therefore more sensitive than time-resolved correlation techniques using a nonlinear process. One important requirement is a fully characterized reference pulse, which can be obtained using a nonlinear optical measurement such as frequency-resolved optical gating (FROG) [21].

Spectral interferometry is a very sensitive method, however, as it requires interferometric stability between the shaped and reference pulses to obtain a
good fringe resolution. Since the path length in the polarization shaper is long, the requisite interferometric stability is not expected to be high. Hence, some other works on polarization-pulse shaping [12,22,23] employ a combination of the cross-correlation measurement and phase-retrieval techniques for the x and y components; i.e. polarization-sensitive cross-correlation frequency-resolved optical gating (XFROG) [24].

3. Applications

3.1. Multiphoton absorption and photoionization

The polarization-shaped femtosecond laser pulses that have been discussed so far are used to actively control the quantum natures of materials. In particular, the final state of multiphoton interactions is governed by the polarization order of the sequentially interacting photons. One simple example is that of a two-photon absorption process through degenerate intermediate states. Dudovich et al. showed the interference at a particular final state between paths through intermediate states with different angular momenta [16]; these authors detected the fluorescence from Rb vapor atoms via two-photon absorption excited with a photon pair of parallel or perpendicular polarizations. The relative phase between the two different intermediate paths was found to determine the final-state population. For this reason, Dudovich et al. rotated the polarization from the x axis to the y axis in a prescribed region of the broadband pulse spectrum using the basic configuration of the pulse shaper.

Multistage-photoionization processes constitute another example of multiphoton interactions. The ionization yield of such photoionization in diatomic molecules depends upon the polarization state of the incident photons because the ionization pathway is also polarization dependent, as per the selection rules of the electronic transitions involved. The demonstration of quantum control using the vector-field shaping was done by Suzuki et al. and Brixner et al. who experimentally maximized the yield of multiphoton ionization of iodine molecules [17] and potassium dimers [18], respectively.

A pulsed supersonic beam of diatomic molecules was introduced into a time-of-flight (TOF) mass spectrometer. The molecular beam was parallel to the TOF axis and the pump-probe laser beams were focused from the crossed direction. The basic configuration of the polarization pulse shaper, which was capable of adjusting two of the four parameters, was used to generate tailored femtosecond pulses. Ion yields were measured in a TOF mass spectrometer and used as feedback signals for learning-loop optimal quantum control. The crucial concept of adaptive feedback control is an experimental procedure employing closed-loop optimization guided by learning algorithm to identify the optimal laser pulse shapes for steering the system toward the desired result of measurements [2,25]. Suzuki et al. and Brixner et al. started with an optical pulse with an appropriate vector waveform and then determined the fitness of the vector waveform by
measuring the ionization yield. Next, the incident vector waveform was repeatedly modified to optimize the fitness. An evolutionary learning algorithm in which the above iteration was automatically conducted by computer-controlled instruments was used in both works to obtain the best vector waveform for the maximum reaction yield of multiphoton ionization. Figure 6 shows that vector-field shaping produced a twofold-increase in ionization yield compared with the unshaped optical pulse.

3.2. Forced molecular rotation

Previous theoretical work has proposed the use of a strong, non-resonant field to exert large optical torques on anisotropic molecules [26]. Using such optical torques, diatomic molecules can be rotationally accelerated to the point where the molecular bond is broken by the centrifugal force. The scheme also acts as an optical centrifuge for distinguishing different molecules based on their moments of inertia. Slowly rotating the polarization of the infrared field drives the molecule to rotate with the same angular frequency of the polarization rotation. By accelerating the rotation of the polarization, the molecule must undergo several hundred successive Raman transitions with continuously changing Raman transition energies. Thus, the excitation laser frequency must be constantly swept to match the changing transition energy.

Villeneuve et al. demonstrated that Cl₂ molecules are rotationally accelerated until the molecular bond is broken by the centrifugal force [27]. Such an optical centrifuge requires pulses with linear polarization rotating at an angular frequency $\Omega$, which may be generated by combining two counter-rotating circularly polarized fields with shifted central frequencies. To sweep $\Omega$ over time, they prepared two circularly polarized pulses with opposite frequency chirps (see Section 2.2.3).

To accomplish this in practice, broadband laser pulses from a Ti:sapphire chirped-pulse amplifier (10 mJ, 35 fs, 1 kHz) were dispersed by a diffraction
grating and split in the center of the spectrum at the Fourier plane of the focusing lens into long- and short-wavelength components. The frequency chirp was individually controlled by separate grating stretchers. The chirp of the long-wavelength component was reversed and its polarization was rotated by $90^\circ$, whereas the short-wavelength component was left unchanged. The two oppositely chirped, orthogonally polarized components were combined and passed through a QWP to transform the basis, as discussed in Section 2.2.3.

In a vacuum chamber, the two combined counter-rotating beams were focused into a beam of sample gas along with two diagnostic pulses. The two diagnostic pulses comprised a 400-nm pulse (20 µJ, 2 ps) used to dissociate the molecules.
and an 800-nm ionizing pulse (15 µJ, 50 fs) that served to ionize any atoms or molecules such that they could be detected by a TOF measurement. In the experiments, the supersonic molecular beam of chlorine mixed with neon buffer gas emerged from a pulsed valve at an estimated rotational temperature of 10 K. The TOF mass spectra of the spinning chlorine molecules in the kinetic energy region of Cl\(^+\) were then acquired. The translational energy of the fragments was determined from the arrival time, as shown in Figure 7. Figure 7(a) shows that the signal around 1 eV, which represents Cl\(^1\)\(^+\) dissociation, drastically increased with increasing energy of the centrifuge laser pulse. The maximum spinning frequency, \(\Omega_{\text{max}}\), had to exceed a particular threshold to break to the bond. Reducing \(\Omega_{\text{max}}\) suppressed the rotational dissociation into Cl\(^1\)\(^+\), as shown in Figure 7(b).

Following this stimulating work, several papers were published on rotational spectroscopy of molecular beams using an optical centrifuge prepared by swept polarization rotation (see, for example, Ref. [28]). For example, the gas of rotating molecules was experimentally observed to become optically birefringent in the presence of an external magnetic field [29]. An electronic spin was proposed to play a mediating role for such magneto-rotational birefringence.

3.3. Coherent Raman scattering process

A difference frequency version of two-photon process involves the generation of a coherent vibration due to stimulated Raman excitation. Especially, multiplex measurement of coherent Raman spectra uses a broad-band pulse for impulsive excitation of the Raman levels followed by a narrow-band probe pulse. Incorporation of polarization selection rules has been presented by femtosecond phase and polarization pulse shaping in single-pulse coherent anti-Stokes Raman scattering (CARS) [30]. By rotating the polarization of the prescribed spectral region in the excitation pulses from the x- to y-polarization, one can selectively measure a nonlinear susceptibility component such as \(|\chi_{\text{yxxy}} - \chi_{\text{yyxx}}|^2\) with a single pulse.

A major drawback in CARS spectroscopy is non-resonant background due to electronic response in a four-wave mixing process, which limits the signal-to-noise ratio of CARS measurements and distorts the spectral profile of resonant vibrational spectrum. The polarization-sensitive CARS is one of the most commonly used techniques for overcoming the problem of the non-resonant background. In this context, the combination of both phase and polarization control of femtosecond pulses provides a powerful method for effective suppression of the non-resonant background signal. Figure 8 shows how the phase and polarization-shaped pulses yields background-free CARS spectra.

3.4. Optical control of lattice vibrations

Coherent superposition of two degenerate states with different polarization selection rules is a suitable demonstration of polarization pulse-shaping. The relative phase between the doubly degenerate E-symmetry Raman-active vibrational
modes of $\alpha$-quartz along two orthogonal coordinates was shown to depend upon the timing between two excitation pulses with different polarizations [5]. This yields optical control over two-dimensional vibrational trajectories along degenerate lattice vibrational modes. This was first demonstrated using pairs of pulses with different polarizations, however, the Fourier-transformed spectrum of a pair of pulsed electric fields has a beating structure, which means the frequency selectivity is imperfect due to overtone resonance. Lattice vibrations of the same symmetry are excited simultaneously at different mode frequencies. Employing polarization-shaping techniques, for which polarization states can be arbitrarily adjusted as a function of time within a light pulse, allows full control of two-dimensional vibrational trajectories by means of both symmetry and frequency [31].

The present authors selectively drove the desired pseudorotational motion of a single vibrational mode in an $\alpha$-quartz crystal using polarization-twisting pulses, as described in Section 2.2.2 [32]. Impulsive stimulated Raman scattering (ISRS) was used to study time domain mode selection using polarization-shaped pulses. Pump-induced spectral phase modulation was detected as a means of measuring the transients. The $\alpha$-quartz crystal has a $C_3$ axis of symmetry that yields three normal modes of oscillation. One is the breathing mode with $A$ symmetry and the other two are degenerate $E$ modes.

Using Fourier-transform-limited pulses, $A$ and $E$ modes were simultaneously excited and four distinct peaks were observed, as previously reported, owing to the broad bandwidth of the pump pulse. The measured frequencies of 128,
Figure 9. Left- and right-circularly rotating trajectories of the degenerate E-mode vibration.

203, 357, and 465 cm\(^{-1}\) were consistent with the reported values of 128, 207, 356, and 464 cm\(^{-1}\) [33]. The degenerate E modes were discriminated by tuning the twist frequency. The two-dimensional pseudorotation of the E mode was unidirectionally enhanced in either the clockwise or counter-clockwise direction, whereas the A mode was strongly suppressed.

The frequency response of the dynamical displacement \(q_v(\omega)\), which is the Fourier transform of the nuclear dynamical displacement \(q_v(t)\), can be represented as the product of the Raman tensor components and the Fourier transform of the second-order optical electric field, \(E_i(t)E_j^*(t)\). If the quartz crystal is excited with polarization-twisting pulses traveling along the C\(_3\) axis, the frequency response of the A mode \(q_A(\omega)\) and E modes \(q_{Ea}(\omega)\) and \(q_{Eb}(\omega)\) can be reduced to the following simple cosine and sine forms, owing to the threefold crystal symmetry of \(\alpha\)-quartz [31]:

\[
q_A(\omega) = \chi_A(\omega) \int_{-\infty}^{\infty} I(t')e^{+i\omega t'} dt' 
\]

\[
q_{Ea}(\omega) = \chi_{Ea}(\omega) \int_{-\infty}^{\infty} I(t') \cos(-2\Omega t')e^{+i\omega t'} dt' 
\]

\[
q_{Eb}(\omega) = \chi_{Eb}(\omega) \int_{-\infty}^{\infty} I(t') \sin(-2\Omega t')e^{+i\omega t'} dt' 
\]
Here, \( I(t) \) and \( \Omega \) denote the intensity envelope of the excitation pulse and the frequency of rotation of the polarization’s azimuthal angle, respectively. Notice that the E-mode vibration circularly rotates at two times the frequency of the polarization rotation \((-2\Omega)\). The direction of rotation of the linear polarization determines the direction of vibrational pseudorotation, which opposes the polarization rotation.

Figure 9 shows a set of ISRS transients with several probe-polarization angles relative to the horizontal direction. The \( \alpha \)-quartz crystal was excited by polarization-twisting pulses with a rotation frequency resonant with the 128 cm\(^{-1}\) E mode. The amplitude of the oscillatory structure of the ISRS transients was proportional to the nuclear displacements in the direction of the probe polarization. The oscillation period was 258 fs, which corresponds to a vibrational frequency of 128 cm\(^{-1}\). The oscillation phase gradually shifted as the direction of the probe polarization rotated. The data from various probe-polarization directions were combined to construct the trajectories of the degenerate E-mode vibration (bottom of Figure 9).

3.5. Selective production of enantiomers

One highly notable example of coherent control by vector-shaped pulses is the selective production of enantiomers from chiral molecules. The preparation of molecules with selective chiralities is an attractive topic in organic chemistry and biochemistry. Chiral molecules can form enantiomers (i.e. isomers without mirror image symmetry), which may have D- or L-forms. Selective chiral synthesis requires breaking of the mirror symmetry in the reaction environment, which may originated from chiral reactants or chiral crystalline surfaces. Although the cause has not yet been elucidated, proteins in living organisms are composed only of L-form amino acids. Molecular interactions differ depending on whether the reactant is D- or L-form. The particular L-form amino acids in living organisms gradually denature into D-forms via mirror image isomerization, which can be a cause of disease and aging. Thus, manipulating enantiomers is important in chemistry, biology, and pharmaceutical science. Vectorial coherent control technology has the potential for wide applications if enantioselective isomerization can be induced as desired.

Shapiro and Brumer first demonstrated the theory that a suitable set of plane-polarized light triggers photodissociation of a prochiral molecule containing two enantiomers showing different cross sections for the production of D- and L-form products [34]. The model system was a molecule of the form \( BAB' \), which includes enantiomers \( B \) and \( B' \) but is symmetric with respect to the reflection plane. Active control over photodissociation resulted in selective production of the photofragments \( B + AB' \) or \( BA + B' \), either of which can have D- or L- form. The first theoretical proposal was based on angular momentum selection of photofragments using quantum interference induced by linearly polarized light. Photoisomerization starting with racemic mixture is vastly preferable because
photodissociation may yield a by-product as fragments. Brumer et al. also discussed enantioselective optical pumping from one enantiomer to the other in a polarized racemic mixture through two rovibrational intermediate states, which were symmetric and antisymmetric with respect to reflection. Unidirectional pumping was shown to be possible with an additional control light, which coherently coupled these two intermediate levels [35].

Fujimura et al. made pioneering contributions to the theory of vectorial coherent control of molecular vibrations [36]. The structure of the target molecule is shown near the center in the left panel of Figure 10. This structure switches between D- and L-forms as the upper-right H atom in the molecular structure rotates around the z axis. The potential energy curve as a function of the angle, $\phi$, of rotation of the H about the z axis has two symmetric minima. Starting with an equal distribution of the D- and L-form chiral H$_2$POSH molecules, the vibrational mode coupled to the isomerization between enantiomers was controlled by vector-shaped far-infrared (THz) pulses through dipolar interactions. The theory indicates that the enantiomeric ratio (D- to L-form) should reach almost unity.

Figure 11 shows the optimal laser waveform for selectively producing L-form H$_2$POSH. The optimal laser pulse with electric field components $E_x$ (top) and $E_y$ (bottom) are shown as functions of time. The inset shows an enlarged view of the electric field components $E_x$ (dotted line) and $E_y$ (solid line). The enlarged inset in Figure 11 reveals how the amplitude, frequency, and relative phase of the optical fields $E_x$ and $E_y$ vary with time. The optimal electric field forms elliptically, rather than circularly polarized light. Moreover, the major axis of the elliptical polarization switches over the duration of the entire pulse. During the first three subpulses, the magnitude of the $y$ component of the elliptically polarized light is greater than that of the $x$ component. However, the magnitude of the $x$ component of the fourth subpulse is greater than that of the $y$ component, as observed in Figure 11.

The above control scheme assumed initially oriented molecules as the starting material. However, it is important to obtain an excess amount of enantiomers from a randomly oriented racemic mixture. For this purpose, several control
scenarios have been proposed for enantiomeric purification using the $x$-, $y$-, $z$- components of the electric field simultaneously or sequentially [37]. First, strong IR fields, $E_z(t)$, force the molecules to align via interactions between the electronic dipole moment and the polarizability. Subsequently, the ultraviolet (UV) $E_x(t)$ and $E_y(t)$ laser pulses discriminate either one of the enantiomers in the racemic mixture. A highly purified yield is expected by the polarization shaping of UV pulses adjusted to the directions of the molecular dipole moment.

Despite the above-mentioned theoretical work and several other theoretical publications [38–40], coherent control of the chiral molecules has not yet been experimentally verified. The major target of the present author’s research is to control this chirality-related photoreaction by means of vector-field pulse shaping. Realization of polarization-shaped terahertz pulses [41], allows us to
conduct these experiments. Thus, in the near future, we expect experimental demonstration of enantioselective generation of chiral molecules.

### 3.6. Nano-optics

Polarization pulse-shaping has been mainly applied to control of the physical and chemical properties of atomic or molecular systems. It is still challenging to incorporate polarization pulse-shaping into the field of solid-state physics. However, one of the successful demonstration in solid-state physics has been ultrafast optical near-field control in the vicinity of a metal nanostructure [42–44]. Subwavelength dynamic localization of electromagnetic fields on the nanometer scale is expected to contribute to the optimization of near-field imaging techniques and tip-enhanced Raman spectroscopy. Controlling the evolution of the local vectorial electric field on the nanometer scale can be also achieved by polarization-shaped pulses. A transient spatial distribution of photoexcited electrons in a nanostructure is strongly dependent upon the directions of the electric fields.

Aeschlimann et al. demonstrated spatial control of the excitation pattern by tailoring the optical near field in the vicinity of silver nanostructures through adaptive polarization shaping [43,44]. The lateral field distribution and its transient response were monitored via time-resolved two-photon photoemission electron microscopy. Photoemission patterns were recorded with a spatial resolution better than 50 nm by a photoemission electron microscope (PEEM). The cross-correlation of the excitation created by a polarization-shaped pump pulse was also measured with a bandwidth-limited probe pulse.
The sample nanostructure was circular Ag disks arranged in a star-like shape. The nanostructure positions were marked by white circles. Figure 12(a) shows the experimental photoelectron distribution for $p$-polarized excitation. The two-photon PEEM pattern exhibits three maxima near the gap of the dimers. The evolution of the photoemission contrast in the regions of interest A and B as a function of generation number is shown in Figure 12(b) for adaptive maximization and minimization. The experimental PEEM images after adaptive A/B maximization and minimization show predominant emission from the upper (Figure 12(c)) and lower (Figure 12(d)) regions under respective excitation by the optimally polarization-shaped pulses (Figure 12(e) and 12(f)).

### 3.7. Magnetization

Ultrafast control of magnetization becomes possible with ultrashort laser pulses, and the vectorial properties of magnetization can be controlled by adjusting the phase and amplitude of two degenerate modes. Kanda et al. manipulated the two-dimensional trajectory of magnetic oscillations in an antiferromagnetic NiO crystal, but the excitation was limited to a pair of linearly polarized laser pulses with different polarization angles [7]. The control scheme was almost the same as that used in the selective manipulation of two degenerate lattice-vibrational modes [5].

On the other hand, magnetization in quantum antiferromagnets is an interesting phenomenon in terms of the quantum coherence of many-body systems. In general quantum magnets, a proper unitary transformation was theoretically...
proposed to map a rotating magnetic field of circularly polarized light into an effective static magnetic field [45]. The photon energy in the terahertz range was comparable with the energy scale of the spin systems. An optical frequency of 1 THz was calculated to be equivalent to an effective field as strong as 40 T [46]. Polarization control in the THz range is indispensable for this purpose; in particular, an upchirped circularly polarized THz laser pulse is required in order to realize adiabatic full magnetization up to saturation. However, previous demonstrations of THz polarization shaping lack the flexibility and arbitrariness of polarization-shaped waveforms. We expect to experimentally demonstrate this unitary transformation from a rotating THz field to achieve an effective static magnetic field using our recently developed THz polarization pulse shaping with arbitrary field control [41].

3.8. Wavelength conversion using nonlinear optical processes

Femtosecond polarization pulse shaping has, in most cases, been restricted to the output from a Ti:sapphire laser oscillator or amplifier in the near infrared (NIR). Tailored polarization-shaped pulses in the UV or mid-infrared (MIR) are very useful for various applications. For example, polarization-shaped pulses in the UV are especially suitable for near-field plasmonic control in metal nanostructures [43], whereas applications of shaped MIR pulses range from quantum control of molecular vibrations to multidimensional infrared spectroscopy. There are two approaches to shaping the phase and amplitude of wavelength-converted pulses: UV or MIR pulses are shaped directly, and alternatively a modulated NIR laser pulse can be frequency-converted to the desired wavelength by nonlinear optical processes. Polarization control is based on the fact that polarization-shaped pulses can be expressed as linear superpositions of two electric fields with orthogonal linear polarization. The common method of achieving this indirect polarization pulse shaping uses two orthogonally oriented nonlinear crystals to generate these orthogonal components in the desired wavelengths. Seidel et al. demonstrated femtosecond polarization shaping in the MIR tuned around 3.5 μm using two potassium niobate (KNB) crystals, both cut at the type-I phase-matching angle but oriented perpendicularly to each other (Figure 13) [22]. Selle et al. generated polarization-shaped pulses at a central wavelength of 400 nm with two perpendicularly oriented BBO crystals [47].

The present authors demonstrated a method for generating predesigned THz polarization-shaped waveforms by optical rectification of laser pulses [41]. The instantaneous polarization state and intensity of the input laser pulses were controlled by an optical pulse shaper. Wavelength conversion of light to the terahertz frequency range was achieved by exploiting the simple polarization selection rules in the optical rectification process for pulses propagating along the threefold axes of a nonlinear optical crystal.

We used a Ti:sapphire regenerative amplifier, which delivered amplified pulses at a repetition rate of 120 kHz and which had a central wavelength of 803 nm
and a pulse duration of 165 fs. We used GaP as the nonlinear optical material due to its large nonlinear optical coefficient, which satisfied the phase-matching condition of optical rectification.

On the basis of a given terahertz polarization waveform, we developed a deterministic algorithm to find a set of shaper parameters for near-infrared pulses. By using a laser pulse shaped by the parameter set thus obtained, we generated the target polarization waveforms of terahertz radiation. Figure 14 shows an example of the experimentally obtained electric field trajectory of the terahertz polarization waveforms (Figure 14(a)) and the generated targeted terahertz polarization waveform (Figure 14(b)). At the beginning of the waveform, the electric field vector rotated counterclockwise with time; by the end of the waveform, the vector field rotated in the clockwise direction. In addition to controlling the direction of rotation of the electric field vector, the amplitude also matched the designed value. Figure 14 shows how the designed waveform (Figure 14(a)) matched the experimentally obtained waveform (Figure 14(b)).

Next, we controlled the circularly polarized terahertz radiation by independently adjusting the bandwidth and central frequency. Circularly polarized terahertz pulses are important for terahertz spectroscopy and coherent control. We modified the parameters \( \alpha \) and \( \beta \) of the polarization-twisting pulse described in Section 2.2.2. The bandwidth of the resulting terahertz wave was inversely proportional to the duration of the incident laser pulse, which was modified by adjusting the chirp parameter \( \beta \). Furthermore, the angular velocity \( \alpha / \beta \) of the laser’s azimuthal angle determined the central frequency \( \Omega_{\text{THz}} = 2 \alpha / \beta \) of the terahertz radiation. Figure 15(a) shows the instantaneous electric field of the laser pulses adjusted for various pulse durations with constant \( \alpha / \beta \). When the pulse duration increased, the spectrum of the terahertz wave narrowed (Figure 15(b)). The bandwidth is inversely proportional to the pulse duration, and varied from 0.3 (bottom) to 1.0 THz (top). While maintaining a constant pulse duration, the
Figure 15. (a) Experimentally obtained instantaneous electric fields of the shaped laser pulses with various durations; (b) Intensity spectra of the terahertz waves generated by the laser pulses in (a). (c) Instantaneous electric fields of the laser pulses with various angular velocities of the azimuthal angle. (d) Intensity spectra of the terahertz waves generated by these laser pulses. Reproduced from Ref. [41].

The central frequency of the terahertz wave could be tuned (Figure 15(d)) by varying the azimuthal angular velocity of the laser pulse (Figure 15(c)). The central frequency was adjusted from 1.0 (bottom) to 3.0 THz (top), while maintaining the bandwidth at 0.3 THz.

4. Conclusion and outlook

In summary, this study provides a concise overview on the field of coherent control using polarization-shaped femtosecond laser pulses. This paper aims to provide a perspective on a steady advance in the development of polarization-shaped pulses and their application to spectroscopy. The underlying concept, from the author’s point of view, is the incorporation of the polarization selection rules, involved in the light–matter interaction of interest, as a function of time. An anisotropic mode selection in both the time and frequency domains can be achieved using polarization pulse shaping. Furthermore, a spatial distribution of the excited species in materials is favorably determined by the transient directions of the optical electric fields, which can also be adjusted by this shaping.

In future, it is likely that the pulse-shaping technology will be tested by a wide range of end users and not only by the experts in optics. The author may anticipate realization of new applications of polarization pulse shaping in the fields of condensed matter physics and chemistry. Selective production of enantiomers may be a challenging target of the polarization coherent control, particularly, in complex biological systems.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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