Principles of Nonlinear Optics

In conventional (linear) optics, the response of a material to an applied electric field is linearly proportional to the field strength. However, when light becomes intense enough, typically from laser sources, materials begin to respond in a nonlinear fashion. This nonlinear response forms the foundation of nonlinear optics, a field with numerous applications in modern technology and fundamental research.

Linear vs. Nonlinear Optical Response

The Linear Regime

In the linear regime, the polarization $\mathbf{P}$ of a material is directly proportional to the applied electric field $\mathbf{E}$:

$$\mathbf{P} = \varepsilon_0 \chi^{(1)} \mathbf{E}$$

where ε_0 is the permittivity of free space and $\chi^{(1)}$ is the linear susceptibility tensor.

This polarization field contributes to Maxwell's equations, specifically through the electric displacement field $\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}$. Starting with Maxwell's equations in a source-free dielectric medium:

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t}$$
$$\nabla \times \mathbf{H} = \frac{\partial \mathbf{D}}{\partial t}$$

We can derive the wave equation:

$$\nabla^2 \mathbf{E} - \mu_0 \frac{\partial^2 \mathbf{D}}{\partial t^2} = 0$$

Substituting the linear polarization relation, we get:

$$\nabla^2\mathbf{E} - \mu_0\frac{\partial^2}{\partial t^2}[\varepsilon_0\mathbf{E} + \varepsilon_0\chi^{(1)}\mathbf{E}] = 0$$

$$\nabla^2\mathbf{E} - \mu_0\varepsilon_0(1+\chi^{(1)})\frac{\partial^2\mathbf{E}}{\partial t^2} = 0$$

Defining the relative permittivity $\varepsilon_r = 1 + \chi^{(1)}$ and refractive index $n = \sqrt{\varepsilon_r}$, and using $c = \frac{1}{\sqrt{\mu_0\varepsilon_0}}$ for the speed of light in vacuum, the wave equation becomes:

$$\nabla^2\mathbf{E} - \frac{n^2}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = 0$$

This transformed wave equation shows that the phase velocity in the medium is $v_p = \frac{c}{n}$, demonstrating how the material's linear response modifies the propagation of light through the medium.

The Nonlinear Regime

When the applied field becomes strong, we need to express the polarization as a power series:

$$\mathbf{P} = \varepsilon_0[\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + ...]$$

where $\chi^{(2)}$ and $\chi^{(3)}$ are the second-order and third-order nonlinear susceptibility tensors, respectively.

To understand how this nonlinear polarization affects light propagation, we can separate the polarization into linear and nonlinear parts:

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{NL}$$

where $\mathbf{P}^{(1)} = \varepsilon_0\chi^{(1)}\mathbf{E}$ is the linear polarization and $\mathbf{P}^{NL} = \varepsilon_0[\chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + ...]$ is the nonlinear polarization.

Substituting this into the wave equation derived earlier, we get:

$$\nabla^2\mathbf{E} - \mu_0\frac{\partial^2}{\partial t^2}[\varepsilon_0\mathbf{E} + \mathbf{P}^{(1)} + \mathbf{P}^{NL}] = 0$$

Rearranging terms and using the definition of the linear refractive index:

$$\nabla^2\mathbf{E} - \frac{n^2}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} = \mu_0\frac{\partial^2\mathbf{P}^{NL}}{\partial t^2}$$

This reformulation of the wave equation reveals a critical insight: the nonlinear polarization term acts as a source term in the wave equation. The left side describes wave propagation in a linear medium, while the right side represents a driving term arising from nonlinear optical processes.

The Born approximation provides a useful framework for analyzing this equation. This approximation assumes that the nonlinear effects are much weaker than the linear response, allowing us to treat the nonlinear polarization as a perturbation. Under this approximation, we first solve for the electric field considering only linear effects, then use this solution to calculate the nonlinear polarization, which in turn acts as a source for new field components.

Physically, this means that the nonlinear medium not only modifies the incident light but actually generates new electromagnetic radiation at different frequencies. For example, when the nonlinear polarization contains a term oscillating at frequency 2ω (as in second-harmonic generation), it acts as a source that radiates an electromagnetic wave at this new frequency. This perspective elegantly explains how nonlinear optical processes can create light with frequencies not present in the original input fields.

Second-Order Nonlinear Processes

Second-order nonlinear effects occur only in materials without inversion symmetry. This fundamental restriction arises from symmetry considerations: in a material with inversion symmetry, if we apply an electric field $\mathbf{E}$, the induced polarization $\mathbf{P}$ must change sign if $\mathbf{E}$ changes sign. Mathematically, if $\mathbf{P}(\mathbf{E}) = -\mathbf{P}(-\mathbf{E})$, then any even-order terms in the polarization expansion must vanish, including $\chi^{(2)}$. This explains why glasses, liquids, and many crystals (those with inversion symmetry) cannot exhibit second-order nonlinear effects, while non-centrosymmetric crystals like LiNbO , KDP, and BBO are widely used for these applications.

Second Harmonic Generation (SHG)

Using the complex representation of electric fields, we can more elegantly describe nonlinear processes. For a monochromatic wave at frequency ω, the electric field is:

$$\mathbf{E}(t) = \mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}_\omega^* e^{i\omega t}$$

where $\mathbf{E}_\omega$ is the complex amplitude and $\mathbf{E}_\omega^*$ is its complex conjugate.

The second-order polarization then becomes:

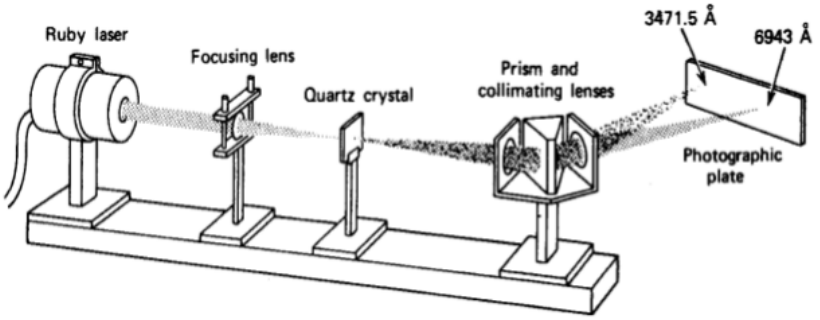

Fig: Frequency doubling of a Ruby laser: $\lambda = 694.3$ nm → $\lambda = 347.1$ nm as shown by Franken et al.[13]

Figure 1: The first experimental observation of second harmonic generation by Franken et al. (1961). This landmark experiment demonstrated frequency doubling of a ruby laser (694.3 nm) to generate ultraviolet light (347.15 nm) using a quartz crystal, establishing the field of nonlinear optics.

$$\mathbf{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \mathbf{E}^2(t) = \varepsilon_0 \chi^{(2)} (\mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}_\omega^* e^{i\omega t})^2$$

Expanding this expression:

$$\mathbf{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} [\mathbf{E}_\omega^2 e^{-2i\omega t} + (\mathbf{E}_\omega^*)^2 e^{2i\omega t} + 2|\mathbf{E}_\omega|^2]$$

This reveals three distinct components:

1. A component oscillating at 2ω (second harmonic)
2. Its complex conjugate also at 2ω
3. A DC (zero-frequency) component proportional to the intensity $|\mathbf{E}_\omega|^2$

The second harmonic term $\mathbf{E}_\omega^2 e^{-2i\omega t}$ radiates at twice the input frequency, forming the basis of SHG.

The figure illustrates how the square of the electric field leads to both the second harmonic generation (oscillating at 2) and optical rectification (the DC component) as previously derived mathematically. This visualization helps conceptualize how a single-frequency input creates multiple frequency components in the polarization response of a nonlinear medium.

Optical Rectification

Optical rectification is a second-order nonlinear process where an intense optical field generates a static (DC) polarization in a nonlinear medium. As previously noted in our discussion of SHG, when a monochromatic field is applied:

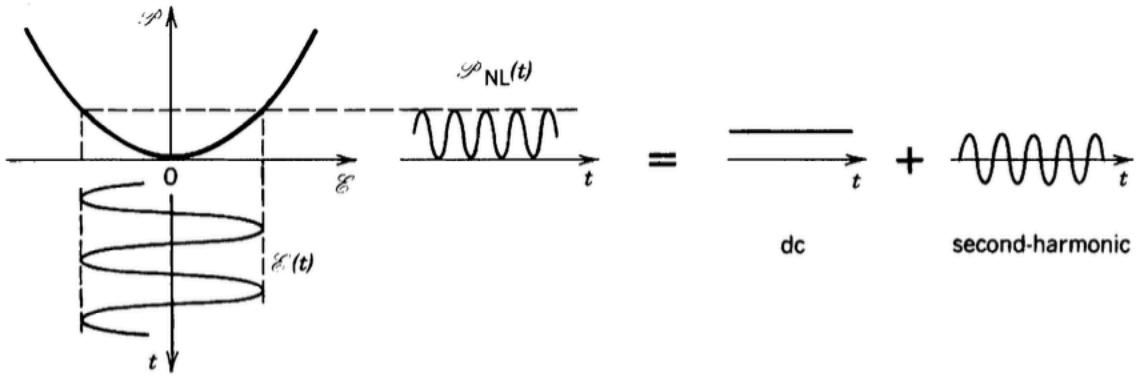

Figure 2: Second-order polarization effects. The input electric field E(t) (left) creates a nonlinear polarization P^(2)(t) (right) with two components: one oscillating at twice the input frequency (second harmonic generation) and a static component (optical rectification).

$$\mathbf{E}(t) = \mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}_\omega^* e^{i\omega t}$$

The second-order polarization includes a DC term:

$$\mathbf{P}_{DC}^{(2)} = \varepsilon_0 \chi^{(2)} \cdot 2|\mathbf{E}_\omega|^2$$

This DC polarization is proportional to the intensity of the incident light. The direction of the induced polarization $\mathbf{P}$ depends on the crystal structure through the second-order susceptibility tensor $\chi^{(2)}$, which relates the components of $\mathbf{P}$ to the square of $\mathbf{E}$. More explicitly, we can write:

$$P_i^{(2)} = \varepsilon_0 \sum_{j,k} \chi_{ijk}^{(2)} E_j E_k$$

where the indices i, j, and k represent the Cartesian components. This tensorial relationship means that the polarization direction is not necessarily parallel to the electric field, but is determined by the symmetry properties of the nonlinear medium. Physically, optical rectification can be viewed as a process that "rectifies" the oscillating electric field of light into a static electric field.

One important application of optical rectification is the generation of terahertz (THz) radiation. When ultrashort laser pulses (with broad frequency spectra) undergo optical rectification, the resulting time-varying polarization can radiate electromagnetic waves in the THz region. This

provides a convenient method for generating broadband THz radiation for spectroscopy and imaging applications.

Electro-Optic Effect

The electro-optic effect, also known as the Pockels effect, can be derived from the second-order nonlinear mixing of an optical wave and a static electric field. Consider a total electric field consisting of an optical component at frequency ω and a DC field:

$$\mathbf{E}(t) = \mathbf{E}_\omega e^{-i\omega t} + \mathbf{E}_\omega^* e^{i\omega t} + \mathbf{E}_{DC}$$

The second-order polarization in the nonlinear medium is:

$$\mathbf{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} \mathbf{E}^2(t)$$

Expanding this expression:

$$\mathbf{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)} [\mathbf{E}_\omega^2 e^{-2i\omega t} + (\mathbf{E}_\omega^*)^2 e^{2i\omega t} + \mathbf{E}_{DC}^2 + 2|\mathbf{E}_\omega|^2 + 2\mathbf{E}_\omega \mathbf{E}_{DC} e^{-i\omega t} + 2\mathbf{E}_\omega^* \mathbf{E}_{DC} e^{i\omega t}]$$

The terms oscillating at frequency ω are particularly significant as they modify the propagation of the optical wave:

$$\mathbf{P}_\omega^{(2)} = 2\varepsilon_0 \chi^{(2)} \mathbf{E}_\omega \mathbf{E}_{DC} e^{-i\omega t}$$

This adds to the linear polarization at frequency ω:

$$\mathbf{P}_\omega^{(1)} = \varepsilon_0 \chi^{(1)} \mathbf{E}_\omega e^{-i\omega t}$$

The total polarization at frequency ω becomes:

$$\mathbf{P}_\omega = \varepsilon_0 [\chi^{(1)} + 2\chi^{(2)} \mathbf{E}_{DC}] \mathbf{E}_\omega e^{-i\omega t}$$

This can be viewed as a modification of the effective susceptibility due to the applied DC field:

$$\chi^{eff} = \chi^{(1)} + 2\chi^{(2)} \mathbf{E}_{DC}$$

Since the refractive index is related to susceptibility by $n^2 = 1 + \chi$, we can directly calculate how the DC field changes the refractive index. For the case without a DC field, we have:

$$n_0^2 = 1 + \chi^{(1)}$$

When the DC field is applied, the effective refractive index becomes:

$$n^2 = 1 + \chi^{eff} = 1 + \chi^{(1)} + 2\chi^{(2)}\mathbf{E}_{DC}$$

This gives us:

$$n^2 = n_0^2 + 2\chi^{(2)}\mathbf{E}_{DC}$$

The change in refractive index is $\Delta n = n - n_0$. We can express this as:

$$n = \sqrt{n_0^2 + 2\chi^{(2)}\mathbf{E}_{DC}}$$

For typical situations where the nonlinear effect is small compared to the linear refractive index ($2\chi^{(2)}\mathbf{E}_{DC} \ll n_0^2$), we can use the binomial approximation:

$$\sqrt{a+b} \approx \sqrt{a} + \frac{b}{2\sqrt{a}} \text{ when } b \ll a$$

Applying this approximation:

$$n \approx n_0 + \frac{2\chi^{(2)}\mathbf{E}_{DC}}{2n_0} = n_0 + \frac{\chi^{(2)}\mathbf{E}_{DC}}{n_0}$$

Therefore, the change in refractive index due to the DC field is:

$$\Delta n \approx \frac{\chi^{(2)}\mathbf{E}_{DC}}{n_0}$$

This direct relationship shows how the second-order nonlinear susceptibility couples with the DC electric field to modify the refractive index of the material.

The electro-optic effect enables several important devices:

1. Electro-optic modulators: Used to modulate the amplitude, phase, or polarization of light
2. Q-switches: For generating short, intense laser pulses

3. Optical isolators: For preventing back-reflections in optical systems
4. Pockels cells: For rapidly switching polarization states

Common electro-optic materials include lithium niobate (LiNbO), potassium dihydrogen phosphate (KDP), and various organic crystals.

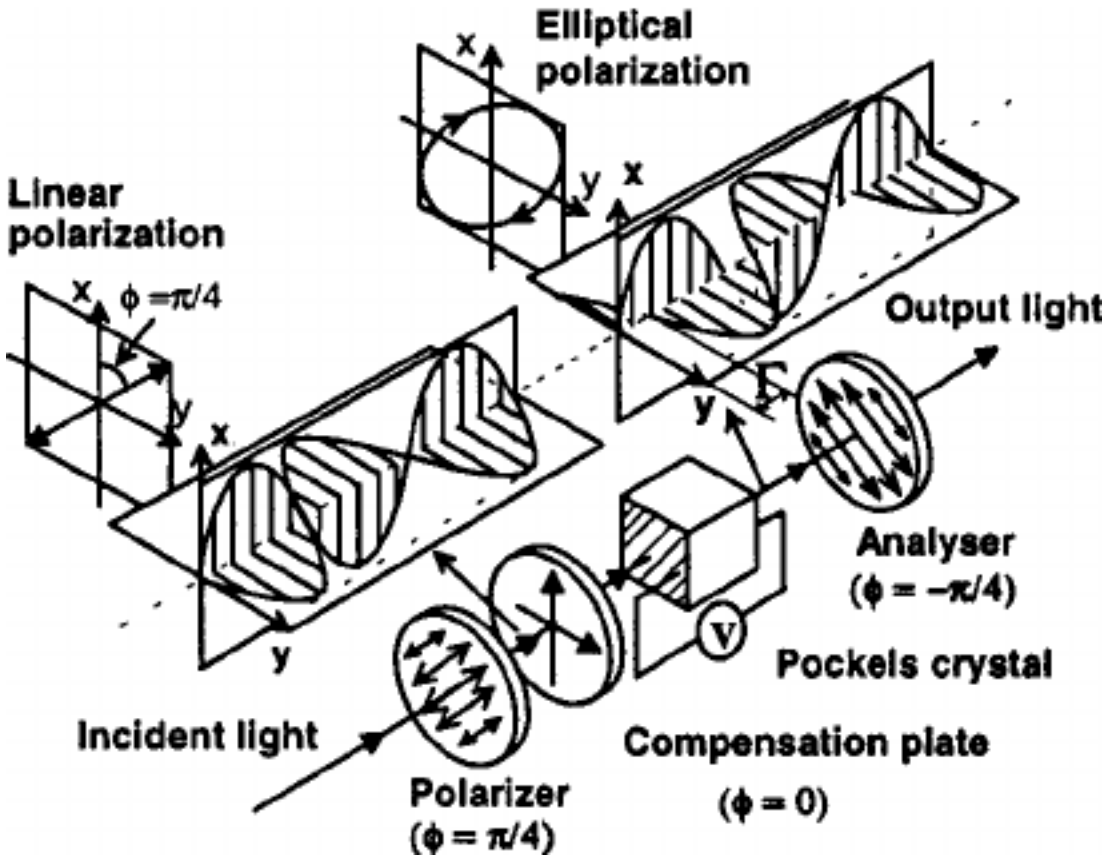

Figure 3: The principle of operation of a Pockels cell. A Pockels cell uses the electro-optic effect to modulate light by applying a voltage that changes the refractive index of the crystal. This changes the polarization state of transmitted light, which can be converted to amplitude modulation using polarizers.

Sum and Difference Frequency Generation

Sum and difference frequency generation is a natural extension of the electro-optic effect discussed earlier. While the electro-optic effect involved the interaction between a DC field and an optical wave, we now consider the interaction between two optical waves of different frequencies, allowing for the generation of new frequencies that are either the sum or difference of the input frequencies.

When two laser beams with frequencies ω_1 and ω_2 interact in a nonlinear medium, the total electric field can be written as:

$$\mathbf{E}(t) = \mathbf{E}_1 e^{-i\omega_1 t} + \mathbf{E}_2 e^{-i\omega_2 t} + c.c.$$

The second-order polarization is proportional to the square of this field. Expanding this expression:

$$\begin{aligned}
\mathbf{P}^{(2)}(t) = \varepsilon_0 \chi^{(2)}[& \quad (1)\\
\mathbf{E}_1^2 e^{-2i\omega_1 t} + \mathbf{E}_2^2 e^{-2i\omega_2 t} + & \quad (2)\\
2\mathbf{E}_1\mathbf{E}_2 e^{-i(\omega_1+\omega_2)t} + 2\mathbf{E}_1\mathbf{E}_2^* e^{-i(\omega_1-\omega_2)t} + & \quad (3)\\
c.c. + 2|\mathbf{E}_1|^2 + 2|\mathbf{E}_2|^2] & \quad (4)
\end{aligned}$$

This expression contains several important terms:

- Second harmonic terms at $2\omega_1$ and $2\omega_2$
- Sum frequency term at $\omega_1 + \omega_2$ (from $\mathbf{E}_1\mathbf{E}_2 e^{-i(\omega_1+\omega_2)t}$)
- Difference frequency term at $\omega_1 - \omega_2$ (from $\mathbf{E}_1\mathbf{E}_2^* e^{-i(\omega_1-\omega_2)t}$)
- DC terms from $2|\mathbf{E}_1|^2$ and $2|\mathbf{E}_2|^2$ (optical rectification)

While the electro-optic effect can be viewed as a special case where one of the fields is at zero frequency (DC), the sum and difference frequency generation involves two oscillating fields, creating new optical frequencies through their nonlinear interaction. This fundamental process enables frequency conversion across the electromagnetic spectrum and forms the basis for many applications, including infrared spectroscopy, tunable laser sources, and optical parametric amplifiers.

Optical Parametric Amplification and Oscillation

One of the most powerful and versatile applications of nonlinear optics is optical parametric amplification and oscillation. These processes enable the generation of tunable coherent light across spectral regions that are difficult or impossible to access with conventional lasers—a capability that has revolutionized fields from spectroscopy to quantum optics.

In optical parametric processes, a pump photon of frequency ω_p splits into signal (ω_s) and idler (ω_i) photons, where $\omega_p = \omega_s + \omega_i$. This can be viewed as the reverse of sum-frequency generation. What makes this process particularly valuable is that by adjusting phase-matching conditions through temperature tuning or crystal orientation, researchers can continuously select which frequencies are generated, effectively creating a "laser" whose output wavelength is tunable over an extremely broad range.

In the complex notation, this process is described by a polarization term:

$$\mathbf{P}^{(2)}(\omega_s) \propto \varepsilon_0 \chi^{(2)} \mathbf{E}_p \mathbf{E}_i^*$$

This term acts as a driving force that amplifies the signal field. When placed in a resonant cavity, this becomes an optical parametric oscillator (OPO), producing tunable output frequencies. Today, OPOs are indispensable tools in applications requiring mid-infrared light, including molecular spectroscopy, remote sensing, medical diagnostics, and as sources for non-destructive material testing where conventional laser sources are unavailable.

Spontaneous Parametric Downconversion for Entangled Photon Generation

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Spontaneous parametric downconversion (SPDC) represents the quantum mechanical limit of the parametric amplification process, where vacuum fluctuations stimulate the creation of photon pairs. In this process, a pump photon spontaneously converts into two lower-energy photons (signal and idler) within a nonlinear crystal, with no initial seed field required. This process is governed by the same energy conservation ($\omega_p = \omega_s + \omega_i$) and phase-matching ($\mathbf{k}_p = \mathbf{k}_s + \mathbf{k}_i$) conditions as classical parametric amplification.

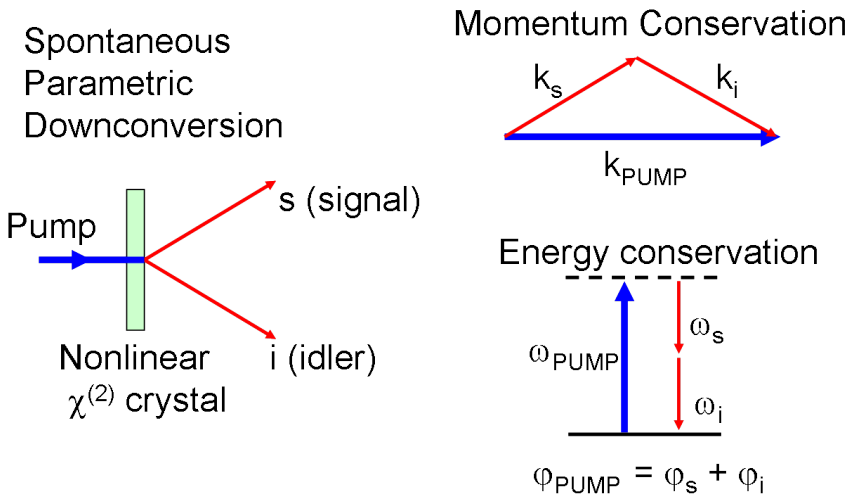

Figure 4: Spontaneous parametric downconversion (SPDC). A pump photon enters a nonlinear crystal and splits into two lower-energy photons (signal and idler) with correlated properties, forming the basis for entangled photon generation.

What makes SPDC particularly significant for quantum information science is that the generated photon pairs exhibit quantum entanglement—a non-classical correlation that Einstein famously referred to as "spooky action at a distance." The photons can be entangled in various degrees of freedom:

1. **Polarization entanglement**: The polarization states of the two photons are correlated such that measuring the polarization of one photon instantly determines the polarization of its partner, regardless of their separation distance. This can be achieved using Type-II phase matching where the signal and idler photons have orthogonal polarizations, and their emission cones overlap.

2. **Momentum entanglement**: The emission direction (wavevector) of one photon is correlated with that of its partner, constrained by the phase-matching conditions.

3. **Energy-time entanglement**: The exact time of photon pair creation is uncertain within the coherence time of the pump laser, leading to entanglement in the energy-time domain.

The quantum state of the generated photon pair can be written as:

$$|\Psi\rangle = \frac{1}{\sqrt{2}}(|H\rangle_s|V\rangle_i + e^{i\phi}|V\rangle_s|H\rangle_i)$$

for polarization-entangled photons, where $|H\rangle$ and $|V\rangle$ represent horizontal and vertical polarization states, and ϕ is a relative phase that can be controlled experimentally.

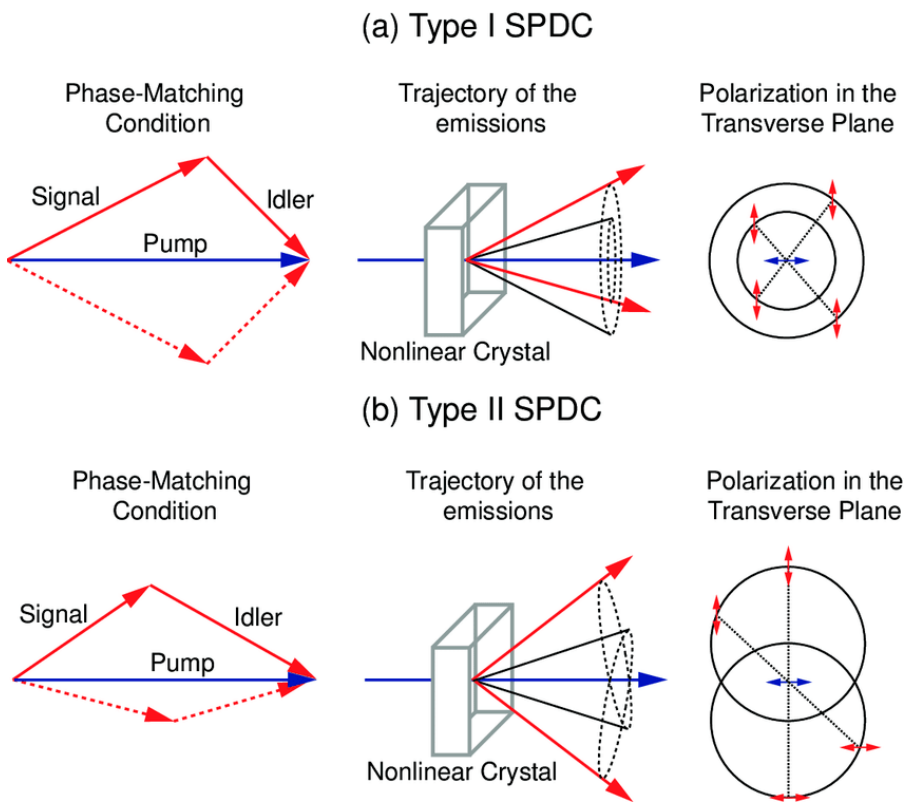

Figure 5: Type I and Type II spontaneous parametric down-conversion (SPDC) configurations. In Type I, the signal and idler photons have the same polarization, creating a single emission cone. In Type II, they have orthogonal polarizations, creating two distinct cones that intersect at two points, which provides a source of polarization-entangled photon pairs.

Entangled photon sources based on SPDC have become foundational tools for quantum information processing, including:

- Quantum key distribution for secure communication
- Bell inequality tests for verification of quantum mechanics
- Quantum teleportation protocols
- Optical quantum computing experiments
- Quantum metrology and imaging with enhanced sensitivity

A typical experimental setup for generating polarization-entangled photons consists of a UV pump laser (often 405nm) illuminating a nonlinear crystal such as beta-barium borate (BBO). The downconverted photons pass through polarization optics and narrowband filters before being collected into single-mode fibers connected to single-photon

detectors. Coincidence counting electronics then identify the correlated photon pairs, confirming their entangled nature through quantum state tomography or Bell inequality measurements.
The efficiency of SPDC is typically quite low (approximately 10^-10 to 10^-8 pair generation probability per pump photon), but recent advances in periodically poled materials, waveguide structures, and cavity enhancement have significantly improved brightness and collection efficiency, making these sources increasingly practical for quantum technologies.

Three-Wave Mixing: A General Framework

All the second-order nonlinear processes we've discussed—second harmonic generation, optical rectification, electro-optic effect, sum and difference frequency generation, and optical parametric amplification—can be understood within a unified theoretical framework called three-wave mixing. This general approach provides deeper insight into the fundamental physics underlying these phenomena and reveals the common principles that govern their behavior.

The General Three-Wave Mixing Process

In three-wave mixing, three electromagnetic waves interact through the second-order nonlinear susceptibility $\chi^{(2)}$ of a medium. These three waves, with frequencies ω_1, ω_2, and ω_3, are coupled through the nonlinear polarization. The key insight is that energy and momentum conservation require:

Energy Conservation (Photon Picture):

$$\omega_3 = \omega_1 + \omega_2$$

Momentum Conservation (Phase Matching):

$$\mathbf{k}_3 = \mathbf{k}_1 + \mathbf{k}_2$$

where $\mathbf{k}_i = n_i\omega_i/c$ are the wave vectors of the three waves.

Coupled Wave Equations

The evolution of the three interacting waves can be described by a set of coupled differential equations. Starting from Maxwell's equations with the nonlinear polarization as a source term, we can derive:

$$\frac{dA_1}{dz} = -i\frac{\omega_1 d_{\text{eff}}}{n_1 c} A_2^* A_3 e^{i\Delta k z}$$

$$\frac{dA_2}{dz} = -i\frac{\omega_2 d_{\text{eff}}}{n_2 c} A_1^* A_3 e^{i\Delta k z}$$

$$\frac{dA_3}{dz} = -i\frac{\omega_3 d_{\text{eff}}}{n_3 c} A_1 A_2 e^{-i\Delta k z}$$

where A_i are the slowly varying amplitudes of the three waves, d_{eff} is the effective nonlinear coefficient, and $\Delta k = k_3 - k_1 - k_2$ is the phase mismatch.

Derivation of the Coupled Wave Equations

To derive these coupled wave equations:

1. Start with Maxwell's wave equation including the nonlinear polarization:

$$\nabla^2 \mathbf{E} - \frac{n^2}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathbf{P}^{NL}}{\partial t^2}$$

2. Express the total electric field as a sum of three waves:

$$\mathbf{E}(z,t) = \frac{1}{2}\sum_{j=1}^{3}[A_j(z)e^{i(k_j z - \omega_j t)} + c.c.]$$

   where $A_j(z)$ are slowly varying amplitudes.

3. The second-order nonlinear polarization is:

$$\mathbf{P}^{(2)}(z,t) = \varepsilon_0 \chi^{(2)} \mathbf{E}^2(z,t)$$

4. For each frequency component ω_j, extract the relevant polarization term:

   - For ω_1: $\mathbf{P}^{(2)}(\omega_1) \propto \varepsilon_0 \chi^{(2)} A_2^* A_3 e^{i[(k_3 - k_2)z - \omega_1 t]}$
   - For ω_2: $\mathbf{P}^{(2)}(\omega_2) \propto \varepsilon_0 \chi^{(2)} A_1^* A_3 e^{i[(k_3 - k_1)z - \omega_2 t]}$
   - For ω_3: $\mathbf{P}^{(2)}(\omega_3) \propto \varepsilon_0 \chi^{(2)} A_1 A_2 e^{i[(k_1 + k_2)z - \omega_3 t]}$

5. Substitute into the wave equation and apply the slowly varying amplitude approximation:

$$\left|\frac{d^2 A_j}{dz^2}\right| \ll \left|k_j \frac{dA_j}{dz}\right|$$

6. Define $\Delta k = k_3 - k_1 - k_2$ and the effective nonlinear coefficient $d_{eff} = \frac{1}{2}\chi^{(2)}$

7. After simplification, we obtain the three coupled equations:

$$\frac{dA_1}{dz} = -i\frac{\omega_1 d_{\text{eff}}}{n_1 c} A_2^* A_3 e^{i\Delta k z}$$

$$\frac{dA_2}{dz} = -i\frac{\omega_2 d_{\text{eff}}}{n_2 c} A_1^* A_3 e^{i\Delta k z}$$

$$\frac{dA_3}{dz} = -i\frac{\omega_3 d_{\text{eff}}}{n_3 c} A_1 A_2 e^{-i\Delta k z}$$

These equations describe the energy exchange between the three waves and form the foundation for analyzing all three-wave mixing processes.

Unifying Second-Order Processes

This general framework elegantly encompasses all the second-order processes we've studied:

- **Second Harmonic Generation**: Set $\omega_1 = \omega_2 = \omega$ and $\omega_3 = 2\omega$
- **Optical Rectification**: Set $\omega_1 = \omega$, $\omega_2 = -\omega$, and $\omega_3 = 0$ (DC)
- **Electro-Optic Effect**: Set $\omega_1 = 0$ (DC field), $\omega_2 = \omega$, and $\omega_3 = \omega$
- **Sum Frequency Generation**: All three frequencies are different with $\omega_3 = \omega_1 + \omega_2$
- **Difference Frequency Generation**: Set $\omega_2 = -\omega_2'$ so that $\omega_3 = \omega_1 - \omega_2'$
- **Optical Parametric Amplification**: Pump at ω_3 generates signal at ω_1 and idler at ω_2

Energy Flow and Conservation

The coupled wave equations reveal that energy flows between the three waves in a way that conserves total energy. The Manley-Rowe relations, derived from these equations, show that:

$$\frac{1}{\omega_1}\frac{d}{dz}\left(\frac{|A_1|^2}{2}\right) + \frac{1}{\omega_3}\frac{d}{dz}\left(\frac{|A_3|^2}{2}\right) = 0$$

$$\frac{1}{\omega_2}\frac{d}{dz}\left(\frac{|A_2|^2}{2}\right) + \frac{1}{\omega_3}\frac{d}{dz}\left(\frac{|A_3|^2}{2}\right) = 0$$

These relations ensure that the number of photons is conserved in the interaction: when one photon at ω_3 is destroyed, one photon each at ω_1 and ω_2 is created, and vice versa.

The Role of Phase Matching

The exponential factor $e^{i\Delta kz}$ in the coupled wave equations reveals a critical requirement for efficient nonlinear interactions: phase matching, where $\Delta k \approx 0$. Phase matching ensures that the nonlinear polarization and the generated electromagnetic wave maintain proper phase alignment throughout the propagation distance. When perfectly phase-matched ($\Delta k = 0$), the energy transfer between interacting waves accumulates constructively along the entire crystal length, maximizing conversion efficiency. Without phase matching ($\Delta k \neq 0$), the direction of energy flow oscillates with a coherence length $L_c = \pi/|\Delta k|$, severely limiting the effective interaction length and conversion efficiency.

Several techniques have been developed to achieve phase matching:

1. Birefringent phase matching: This technique exploits the refractive index difference between orthogonal polarization states in anisotropic crystals. By carefully orienting the crystal and selecting appropriate polarizations, one can satisfy the condition $n_1(\omega_1) = n_3(\omega_3)$ for type-I phase matching or other analogous conditions for different processes. Temperature tuning can provide fine adjustment of these conditions in many materials.
2. Quasi-phase matching: When birefringent phase matching is difficult or impossible, quasi-phase matching offers an elegant alternative. Rather than maintaining $\Delta k = 0$, this approach periodically inverts the sign of the nonlinear coefficient (typically by creating periodic domain inversions in ferroelectric materials) with period $\Lambda = 2L_c$. This effectively resets the phase relationship at precisely the distance where destructive interference would begin, allowing efficient nonlinear conversion even in materials that cannot be birefringently phase-matched.
3. Modal phase matching: In waveguides and fibers, the different propagation constants of various spatial modes can be exploited to achieve phase matching between waves of different frequencies traveling in different modes.

Third-Order Nonlinear Processes

Third-order effects can occur in all materials and include:

Third Harmonic Generation

Similar to SHG, but generating light at 3ω from an input at ω.

Intensity-Dependent Refractive Index

The refractive index becomes intensity-dependent:

$$n = n_0 + n_2 I$$

where n_0 is the linear refractive index, n_2 is the nonlinear refractive index, and I is the light intensity.

Four-Wave Mixing

Four photons interact such that $\omega_1 + \omega_2 = \omega_3 + \omega_4$.

Self-Phase Modulation and Self-Focusing

Intense light can modify its own phase through the intensity-dependent refractive index, leading to spectral broadening (self-phase modulation) or focusing (self-focusing).

Applications of Nonlinear Optics

1. Frequency conversion (lasers of new colors)
2. Optical parametric oscillators for tunable light sources
3. Ultrashort pulse generation
4. Optical switching and computing
5. Optical limiting for protection against intense light
6. Imaging techniques like second-harmonic generation microscopy

Experimental Considerations

Typical Materials for Nonlinear Optics

- Lithium Niobate (LiNbO)
- Potassium Dihydrogen Phosphate (KDP)
- Beta Barium Borate (BBO)
- Gallium Arsenide (GaAs)
- Silica fibers (for third-order effects)

Typical Setup

A basic experimental setup might include:

1. A laser source (typically pulsed for high intensity)
2. Focusing optics
3. The nonlinear medium
4. Filters to select the desired output frequency
5. Detection and measurement equipment

Conclusion

Nonlinear optics represents a fascinating extension of conventional optics where the material response depends nonlinearly on the applied field. This leads to a rich variety of phenomena with numerous applications in science and technology. As laser technology continues to advance, providing ever more intense and controlled light sources, nonlinear optics remains an active and evolving field of research.