Second- and Third-Harmonic Generation in Nonlinear Optical Media

Master Student of Physics

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Abstract

This report presents an overview of second- and third-harmonic generation (SHG and THG) as key phenomena in nonlinear optics, with emphasis on theoretical foundations, phase-matching techniques, experimental configurations, and applications in spectroscopy and microscopy.

1 Introduction and Motivation

Nonlinear optical processes extend linear spectroscopy by enabling frequency conversion through intense electromagnetic fields interacting with matter. In particular, *second-harmonic generation* (SHG) and *third-harmonic generation* (THG) provide access to new spectral regions and high-resolution imaging capabilities. Applications include ultrafast laser pulse characterization, biological microscopy, and material property analysis.

2 Fundamentals of Nonlinear Polarization

The response of a dielectric medium to an applied electric field $E(t)$ can be expanded as:

$$P(t) = \varepsilon_0 \big[\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \cdots \big], \tag{1}$$

where $\chi^{(n)}$ denotes the nth-order nonlinear susceptibility tensor. The second-order term gives rise to SHG, generating polarization oscillating at 2ω, whereas the third-order term produces THG at 3ω and other phenomena such as four-wave mixing.

2.1 Energy and Momentum Conservation

Efficient harmonic generation requires satisfaction of conservation laws:

$$\text{Energy:} \quad \hbar n\omega = \sum_i \hbar\omega_i, \text{ Momentum (phase matching):} \qquad nk(\omega) = k(n\omega) + \Delta k, \quad \Delta k = 0. \tag{2}$$

Here, $k(\omega)$ is the wavevector at frequency ω and Δk quantifies the phase mismatch.

3 Second-Harmonic Generation (SHG)

SHG arises in noncentrosymmetric crystals through the second-order polarization:

$$P_i(2\omega) = \varepsilon_0 \sum_{jk} \chi^{(2)} ijk E_j(\omega) E_k(\omega). \tag{3}$$

3.1 Tensor Properties

The $\chi^{(2)} ijk$ tensor has specific nonzero components determined by crystal symmetry. For example, in a LiNbO3 crystal:

Component	Value (pm/V)
$d_{31} = \chi^{(2)}_{311}$	4.5
$d_{33} = \chi^{(2)}_{333}$	27.0

Table 1: Selected nonlinear coefficients for $LiNbO_3$.

3.2 Phase-Matching Techniques

Birefringent Phase Matching. By exploiting crystal birefringence, one can choose ordinary and extraordinary polarizations to satisfy:

$$nk_o(\omega) = k_e(2\omega). \tag{4}$$

Quasi-Phase Matching. Periodic poling in materials such as PPLN introduces a modulation of $\chi^{(2)}$ with period Λ, compensating phase mismatch via reciprocal vectors $G = 2\pi/\Lambda$.

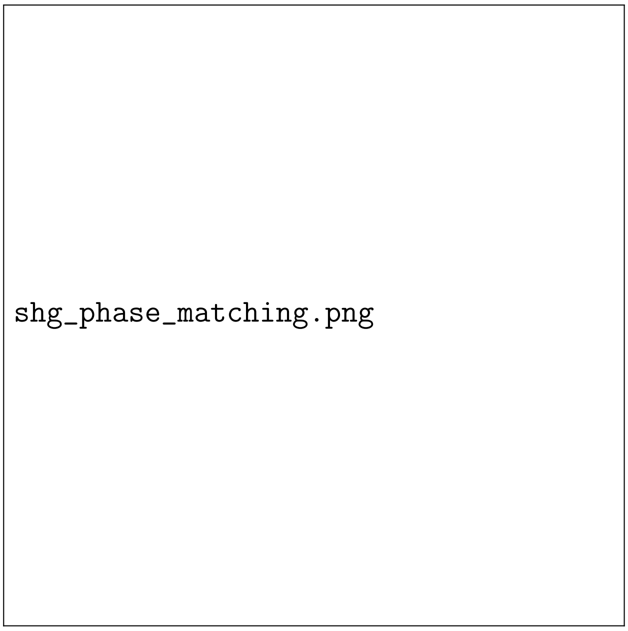

Figure 1: Schematic of quasi-phase-matched SHG in a periodically poled crystal.

4 Third-Harmonic Generation (THG)

THG is a third-order process present in both centrosymmetric and noncentrosymmetric media, described by:

$$P_i(3\omega) = \varepsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl} E_j(\omega) E_k(\omega) E_l(\omega). \tag{5}$$

4.1 Phase Matching and Cascading

Perfect phase matching for THG is challenging; one may use

- *Bulk phase matching*, adjusting dispersion via angle or temperature tuning.
- *Cascading SHG processes*, where sequential $\chi^{(2)}$ interactions ($\omega \to 2\omega$, then $\omega + 2\omega \to 3\omega$) effectively generate THG with enhanced efficiency.

5 Experimental Setup and Considerations

A typical setup for SHG/THG experiments includes:

thg_cascading.png

Figure 2: Energy-level diagram illustrating cascaded SHG and sum-frequency mixing to produce THG.

- A femtosecond pulsed laser (e.g., Ti:sapphire, 800nm, 100fs)
- Beam-shaping and focusing optics (lenses or microscope objectives)
- Nonlinear crystal mounted on a rotation/temperature-controlled stage
- Filters or dichroic mirrors to separate fundamental and harmonic beams

Key challenges include crystal damage thresholds, beam walk-off, and maintaining spatial overlap. Temperature stabilization is often required for fine-tuning phase matching.

6 Applications

6.1 SHG Microscopy

SHG provides intrinsic contrast in noncentrosymmetric biological structures (e.g., collagen), enabling label-free imaging.[Placeholder for experimental image]

6.2 Ultraviolet THG Spectroscopy

Tripling near-infrared lasers yields UV radiation for high-resolution absorption studies in materials science.

7 Summary and Outlook

We have reviewed the physical principles of SHG and THG, emphasizing the roles of $\chi^{(2)}$ and $\chi^{(3)}$ susceptibilities and phase-matching strategies. Future directions include integrated photonic waveguides with engineered dispersion and metasurfaces for enhanced harmonic conversion efficiencies.