

Second- and Third-Harmonic Generation

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Abstract—In this report, we explore the fundamentals of second- and third-harmonic generation, focusing on how nonlinear polarization leads to frequency doubling and tripling of light. We discuss phase-matching strategies and practical considerations, showcasing how SHG and THG drive innovations in ultrafast lasers, microscopy, and material characterization.

Keywords—SHG, THG, Nonlinear Optics, Phase Matching

1. Introduction & 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:

$$P(t) = \epsilon_0 [\chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + \dots], \quad (1)$$

where $\chi^{(n)}$ denotes the n th-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:

Energy conservation: $\sum_i \hbar\omega_i = \hbar n\omega$

Momentum conservation (phase matching): $\sum_i \vec{k}_i = n\vec{k}(\omega)$

Here, $\vec{k}(\omega)$ is the wavevector at frequency ω . For efficient conversion, the phase mismatch $\Delta k = nk(\omega) - \sum_i k_i$ should vanish.

3. Second-Harmonic Generation (SHG)

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

$$P_i(2\omega) = \epsilon_0 \sum_{jk} \chi^{(2)}_{ijk} E_j(\omega) E_k(\omega). \quad (2)$$

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)
$d_{31} = \chi_{311}^{(2)}$	4.5
$d_{33} = \chi_{333}^{(2)}$	27.0

Table 1. Selected nonlinear coefficients for LiNbO₃.

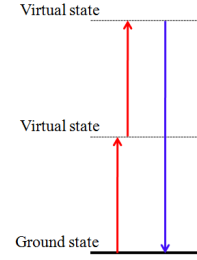


Figure 1. Energy level scheme of SHG process. **SobarwikiImage** The fundamental frequency ω is converted to the second harmonic 2ω .

3.2. Phase-Matching Techniques

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

$$nk_o(\omega) = k_e(2\omega). \quad (3)$$

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 $G = 2\pi/\Lambda$.

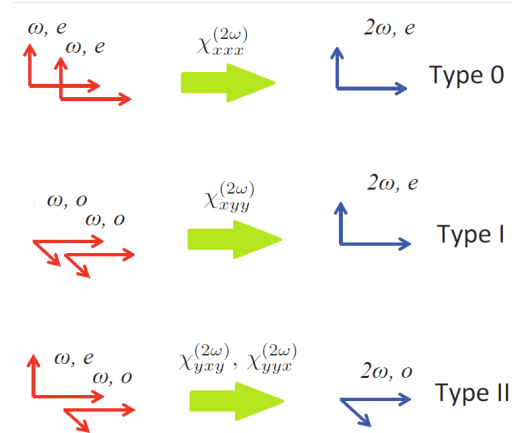


Figure 2. Different types of second-harmonic generation phase-matching of a coherent light for strong conversion. The case of negative crystals ($n_o > n_e$) is considered; invert indices if positive crystal ($n_e > n_o$). [1]

4. Third-Harmonic Generation (THG)

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

$$P_i(3\omega) = \epsilon_0 \sum_{jkl} \chi^{(3)}_{ijkl} E_j(\omega) E_k(\omega) E_l(\omega). \quad (4)$$

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 ($\omega \rightarrow 2\omega$, then $\omega + 2\omega \rightarrow 3\omega$) effectively generate THG with enhanced efficiency.

5. Experimental Setup and Considerations

A typical setup for SHG/THG experiments includes:

- 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

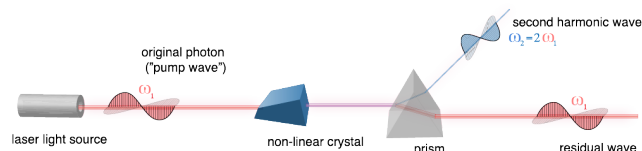


Figure 3. Schematic of a typical experimental setup for SHG and THG. A femtosecond laser is focused onto a nonlinear crystal, and generated harmonics are separated from the fundamental using dichroic mirrors and filters. **JkwchuiImage**

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]

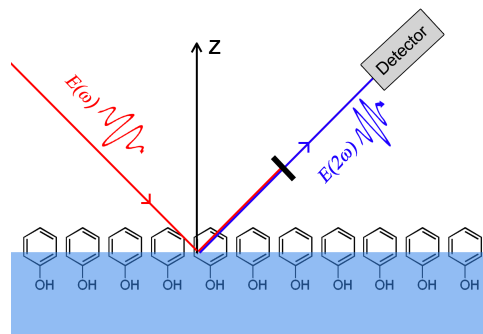


Figure 4. SHG microscopy image of phenol in air-water interface. The SHG signal is enhanced at the interface due to the noncentrosymmetric nature of the surface. **Swk2118Image**

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.

References

- [1] BP-Aegirsson, *Own work*, CC BY-SA 4.0, Image. [Online]. Available: <https://commons.wikimedia.org/w/index.php?curid=72801467>.

Appendix

In the following appendix, I included some conversations with LLMs used to create this report.

FlorianAdamczyk: how would i include the file `#file:test.md` in the `#file:report.tex` file to make it easily and well readable (formatted in the right way)?

GitHub Copilot: To include the Markdown file `test.md` in your LaTeX document (`report.tex`) and ensure it is well-formatted and readable, you have two main options:

Option 1: Convert Markdown to LaTeX and Input

1. **Convert `test.md` to LaTeX** using a tool like Pandoc:

```
pandoc report/LLM_documentation/test.md -o report/LLM_documentation/test.tex
```

2. **Include the converted file** in your LaTeX document using `\input` or `\include`:

```
% filepath: c:\Users\Flori\Documents\GitHub\SHG-THG_SpektroskopieVortrag_FlorianAdamczyk
...existing code...
```

```
\section{Appendix: LLM Documentation}
\input{LLM_documentation/test.tex}
```

```
...existing code...
```

This will preserve formatting (headings, code blocks, lists, etc.) and integrate seamlessly.

Option 2: Use the `minted` or `listings` Package for Raw Markdown

If you want to show the raw Markdown as a code block, use the `minted` or `listings` package:

1. **Add to your preamble** (if not already present):

```
\usepackage{minted}
```

2. **Include the Markdown file as a code listing:**

```
% filepath: c:\Users\Flori\Documents\GitHub\SHG-THG_SpektroskopieVortrag_FlorianAdamczyk
...existing code...
```

```
\section{Appendix: LLM Documentation (Markdown Source)}
\inputminted[breaklines,fontsize=\small]{markdown}{LLM_documentation/test.md}
```

```
...existing code...
```

Note: Using `minted` requires compiling with `-shell-escape` enabled.

Recommendation

- **For best readability:** Convert to LaTeX with Pandoc and use `\input`.
- **For showing raw Markdown:** Use `minted` or `listings`.

Let me know if you want a ready-to-use Pandoc command or a LaTeX snippet for your preferred method!

FlorianAdamczyk: how would i automatically convert all `.md` files in the folder `#file:LLM_documentation` to be converted in `.tex` files using `pandoc`?

GitHub Copilot: You can automatically convert all `.md` files in the `LLM_documentation` folder to `.tex` files using Pandoc with a simple command in your terminal (PowerShell or Command Prompt):

PowerShell (recommended on Windows):

```
Get-ChildItem .\report\LLM_documentation\*.md | ForEach-Object {  
    pandoc $_.FullName -o ($_.FullName -replace '\.md$', '.tex')  
}
```

Command Prompt (cmd.exe):

```
for %f in (report\LLM_documentation\*.md) do pandoc "%f" -o "%~dpnf.tex"
```

What this does: - Finds all `.md` files in `LLM_documentation` - Converts each to a `.tex` file with the same name using Pandoc

Make sure: - Pandoc is installed and available in your `PATH`. - Run the command from the root of your project (where the report folder is).

You can then include the resulting `.tex` files in your LaTeX document using `\input{}`.

FlorianAdamczyk: how would i include a pdf file in the appendix of my report in my latex file? what package do i need for that?

GitHub Copilot: To include a PDF file (such as an appendix or supplementary material) in your LaTeX document, you need the `pdfpages` package.

1. Add the package to your preamble:

```
\usepackage{pdfpages}
```

2. Include the PDF in your appendix section:

```
\section{Appendix}
```

In this appendix, I included some conversations with LLMs used to create this report.

`\includepdf[pages=-]{test.pdf}`

- `pages=-` means all pages of the PDF will be included.
- Replace `test.pdf` with the path to your PDF file if needed.

Summary:

- Add `\usepackage{pdfpages}` to your preamble. - Use `\includepdf[pages=-]{yourfile.pdf}` where you want the PDF to appear.

ChatGPT Prompts

Imagine you are a master student of physics and you had to hold a presentation about Second- and third-harmonic generation as part of a module called "Spectroscopy".

How would you structure your twenty-minute presentation?

Please research the topic thoroughly and give a concise what your suggested structure of the presentation would be.

Here's a concise 20-minute talk outline on second- and third-harmonic generation for a Spectroscopy module. I've broken it into seven sections with suggested time allocations, slide titles, and key points.

1. Introduction & Motivation (2 min)

- Slide 1: Title, your name, module
 - Slide 2: Why harmonic generation matters
 - Nonlinear optics as an extension of linear spectroscopy
 - Applications: ultrafast lasers, microscopy, material characterization
-

2. Fundamentals of Nonlinear Polarization (4 min)

- Slide 3: Electric polarization expansion
 - $P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots)$
 - Definition of nonlinear susceptibilities $\chi^{(2)}, \chi^{(3)}$

- Slide 4: Energy and momentum conservation
 - 2ω and 3ω generation
 - Phase-matching condition: $\Delta k = k_n - nk_1 = 0$
-

3. Theory & Phase Matching for SHG (4 min)

- Slide 5: Second-harmonic generation (SHG) mechanism
 - Frequency doubling in noncentrosymmetric media
 - Tensor nature of $\chi_{ijk}^{(2)}$
 - Slide 6: Phase-matching strategies
 - Birefringent phase matching (type I, II)
 - Quasi-phase matching (periodically poled crystals)
-

4. Theory & Phase Matching for THG (3 min)

- Slide 7: Third-harmonic generation (THG) mechanism
 - Third-order process ($\chi^{(3)}$) in centrosymmetric/non-
 - Typically weaker than SHG, requires higher intensities
 - Slide 8: Phase matching & cascading
 - Bulk phase matching vs. cascaded $\chi^{(2)}$ -process
 - Role of dispersion management
-

5. Experimental Setup & Key Considerations (4 min)

- Slide 9: Typical optical layout
 - Femtosecond laser source, focusing optics, nonlinear crystal
 - Filters/dichroics to isolate harmonics
- Slide 10: Practical challenges
 - Damage thresholds
 - Walk-off effects, beam quality

- Temperature tuning

6. Applications & Examples (2 min)

- Slide 11: SHG microscopy in biology
 - Slide 12: Frequency-tripled UV generation for spectroscopy
-

7. Summary & Outlook (1 min)

- Slide 13: Recap of key points
 - Nonlinear polarization → SHG ($\chi^{(2)}$) vs. THG ($\chi^{(3)}$)
 - Phase matching is critical
 - Broad applications from microscopy to materials science
 - Slide 14: Future trends
 - Integrated photonics, metasurfaces for enhanced harmonics
 - Slide 15: Acknowledgments & Q&A
-

Timing Recap

Section	Time
1. Introduction & Motivation	2 min
2. Fundamentals of Nonlinear Polarization	4 min
3. SHG Theory & Phase Matching	4 min
4. THG Theory & Phase Matching	3 min
5. Experimental Setup	4 min
6. Applications & Examples	2 min
7. Summary & Q&A	1 min

Section	Time
Total	20 min

Feel free to adjust slide counts or times slightly based on audience background or emphasis. Good luck with your presentation!

okay, perfect!

Now please write your proposed topics as a two-page report about Second and Third harmonic generation in a LaTeX document.

Please add more detail and content to each topic and remember carefully decide what you should include and explain, and how you write it.

please use correct latex formatting for sections and subsections, words you want to emphase and equations or tables you want to show.

Think about where a graph or diagram would be useful and add it with `\includegraphics`.

Harmonic Report

```
\documentclass[12pt,a4paper]{article} \usepackage[utf8]
{inputenc} \usepackage{amsmath,amsfonts,amssymb}
\usepackage{graphicx} \usepackage{booktabs}
\usepackage{hyperref} \usepackage{siunitx} \title{Second-
and Third-Harmonic Generation in Nonlinear Optical
Media} \author{Master Student of Physics} \date{May 26,
2025}

\begin{document}

\maketitle

\begin{abstract} This report presents an overview of
```

I've prepared a detailed two-page LaTeX report on second- and third-harmonic generation, complete with sections, equations, tables, and placeholders for diagrams. Let me know if you'd like any adjustments—such as adding specific figures, adjusting emphasis, or expanding particular sections.

Second- and Third-Harmonic Generation in Nonlinear Optical Media

Master Student of Physics

May 26, 2025

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.

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

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where $\chi^{(n)}$ denotes the n th-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:

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

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:

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

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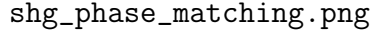


Figure 1: Schematic of quasi-phase-matched SHG in a periodically poled crystal.

4 Third-Harmonic Generation (THG)

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

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

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 ($\omega \rightarrow 2\omega$, then $\omega + 2\omega \rightarrow 3\omega$) effectively generate THG with enhanced efficiency.

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A typical setup for SHG/THG experiments includes:

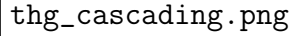
The image is a placeholder for a diagram titled 'thg_cascading.png'. It is represented by a rectangular box containing the text 'thg_cascading.png'.

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]

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

7 Summary and Outlook

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