

Setting SFG

Wednesday, February 16, 2022 1:18 AM

Setting up your own SFG system

Developing your own solid-state nonlinear SFG is stable and low maintenance, so highly preferred. Especially you want a wavelength at which there is

- no diode laser (or even there is, power output is very low --only few mW)
- Ti:sapphire laser(650-1100nm) can't be tuned to that wavelength
- While dye laser can be tuned with specific dye, it's not stable and maintenance is complex

Attributing to the quickly-developed laser technology such as fiber amplifiers and narrow-linewidth fiber lasers with quasi-phase-matched (QPM) nonlinear frequency conversion materials, research groups continue to introduce new and better performing lasers:

- **Fiber amps:** 1500nm EDFA and 1060-1080nm and YDFA -- becoming more mature, giving out dozens of W power
- **Fiber lasers:** high-quality QPM nonlinear bulk crystals (eg. PPKTP, PPLN <--which have the advantage of large effective nonlinear coefficient d_{eff}) are available at acceptable price for SHG, SFG and parametric oscillator.
 - For IR band: people prefer PPLN crystal due to higher d_{eff} , and with more developed manufacturing tech, its interaction length can be 80mm. In addition, the 5%-mol *MgO-doped* periodically poled lithium niobate (PPMgO:LN) can improve photorefractive damage as well as reduce the phase-matching temperature---very suitable for using at near room temperature.

Examples spanning 10 years:

- In 1999, Hart et al obtained 450mW of 630nm red laser output by single-pass SFG of a 1064nm Nd:YAG and 1550nm EDFA in a PPLN crystal
- In 2011, Wilson et al obtained 2W of 626nm with single-pass SFG for laser cooling Be⁺ ions/
- In 2014, Hankin et al realized 1.1W of 638nm for the Rydberg excitation of cesium atoms via single-photon transition.

Our group has investigated and built several single-pass SFG to obtain CW output at different wavelengths from different seeds+amplifiers.

PPLN Tutorials

Light sources with certain wavelengths can be made based on MgO-doped periodically poled Lithium Niobate (MgO:PPLN) = highly efficiency medium for nonlinear wavelength conversion processes.

Advantages of using this crystals: **high nonlinear coefficient, high damage threshold.**

Frequency upconversion (SHG, SFG), enabled by the **quasi-phase-matching technology**, can be used to generate otherwise hard-to-reach wavelengths in the visible range.

Combining this **QPM technology with the use of chirped PPLN structure**, we can have broadly tunable & efficient upconversion. These PPLN devices for efficient frequency conversion of lasers let us to reach wavelengths that cannot be achieved with conventional solid state lasers, diode lasers etc.

EX:

- frequency double a 1064nm laser to 532nm -- a technique used for green laser pointers
- convert 1064nm to 3um, used for gas detection or microscopy imaging techniques

Principles:

- (See my Lab Notes)

18.1 Description of nonlinear optical processes

When an external electric field is applied to a material, opposite charges inside the molecules (ie. negatively charged electrons and positively charged nucleus) are driven and displaced from the center of mass of the molecule. The non-linearity of a medium refers to the material response that varies in a non-linear manner in presence of the applied electric field. The description of nonlinear optics began with the discovery of second-harmonic generation phenomenon in 1961 by Franken *et al*, which was just shortly after Maiman demonstrated the first functional laser in 1960.

In the **linear regime** (usually when driving powers are low), each molecule in the material acts like a harmonic oscillator, and the induced dipole oscillate at the same frequency as the driving electric field. Note that at high optical frequencies, oscillations of nucleus are much weaker than electronic contribution due to the larger mass. We characterize dipole moment per volume as the **polarization** of a material $\vec{P}(t)$. The induced polarization thus depends linear with the applied electric field: $\vec{P}(t) = \epsilon_0 \chi^{(1)} \vec{E}(t)$, where ϵ_0 is permittivity of free space and $\chi^{(1)}$ is the linear polarizability.

However, in **nonlinear regime** (when applied fields are strong), oscillations become large, the dipole approximation breaks down, and material behavior deviates from harmonic oscillator. Especially when the driving field is comparable to inter-atomic field strength, the relation between applied field and material response requires quite different descriptions (eg. resonant excitation, which can lead to saturation effect).

Nonlinear behavior now comes into play, giving rise to different frequency components in the oscillations, and nuclear oscillations could be comparable to electronic. To account for this in our theoretical description, the simplest approach is to expand the polarization $\vec{P}(t)$ in power series in $\vec{E}(t)$ and include all the nonlinear terms:

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

The constants $\chi^{(2)}$, $\chi^{(3)}$ are 2nd- and 3rd- order nonlinear optical susceptibilities, respectively.

The reason why polarization is essential the description of nonlinear optics is that a time-varying polarization can act as a source of new frequency components in electric field. This is evident from the wave equation in nonlinear media:

$$\nabla^2 \vec{E}(t) - \frac{n^2}{c^2} \frac{d^2 \vec{E}(t)}{dt^2} = \frac{1}{\epsilon_0 c^2} \frac{d^2 \vec{P}_{NL}(t)}{dt^2}$$

- **Second-order nonlinear processes:** mixing of 3 EM waves in the crystal medium, where the magnitude of the crystal's nonlinear response is characterized by $\chi^{(2)}$ coefficient. --> 3 interactions: SHG, SFG, DGF
 - Frequency doubling (SHG) is the most common process that utilizes the $\chi^{(2)}$

18.1.1 SHG (Second-Harmonic Generation)

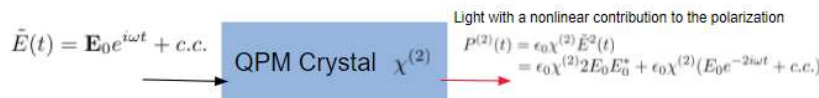
Laser beam: plane wave Gaussian

$$\vec{E}(t) = \vec{E}_0 e^{i\omega t} + c.c. \quad (5)$$

After going through crystal material with a second-order susceptibility:

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} \vec{E}^2(t) = \epsilon_0 \chi^{(2)} 2E_0 E_0^* + \epsilon_0 \chi^{(2)} (E_0 e^{-2i\omega t} + c.c.) \quad (6)$$

Additional frequencies other than ω can be seen in this second-order polarization: and one terms at 2ω frequency. According to Eqn 4, this generates radiation at 2ω frequency, while zero-frequency term does not generate radiation since it's a second derivative.



- **Sum frequency generation (SFG)** is more general: f_1, f_2 two input photons at different wavelengths are combined and through nonlinear process, generate a photon at $f_1 + f_2$

18.1.2 SFG (Sum-Frequency Generation)

Two different frequencies in optical field:

$$\vec{E}(t) = \vec{E}_1(t) + \vec{E}_2(t) = \vec{E}_1 e^{-i\omega_1 t} + \vec{E}_2 e^{-i\omega_2 t} + c.c. \quad (7)$$

After incident on a nonlinear crystal with $\chi^{(2)}$ susceptibility:

$$P^{(2)}(t) = \epsilon_0 \chi^{(2)} \vec{E}^2(t) = \epsilon_0 \chi^{(2)} [E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} + (2E_1 E_2^* e^{-i(\omega_1 - \omega_2)t})] + 2\epsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*)$$

Let's better group the complex amplitudes by frequencies:

$$\left. \begin{aligned} P_{2\omega_1} &= \epsilon_0 \chi^{(2)} E_1^2 \} \text{SHG} \\ P_{2\omega_2} &= \epsilon_0 \chi^{(2)} E_2^2 \} \end{aligned} \right\} \quad (8)$$

$$P_{\omega_1 + \omega_2} = 2\epsilon_0 \chi^{(2)} E_1 E_2 \} \text{SFG} \quad (9)$$

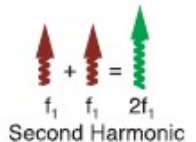
$$P_{\omega_1 - \omega_2} = 2\epsilon_0 \chi^{(2)} E_1 E_2^* \} \text{DFG} \quad (10)$$

$$P_0 = 2\epsilon_0 \chi^{(2)} (E_1 E_1^* + E_2 E_2^*) \} \text{OR} \quad (11)$$

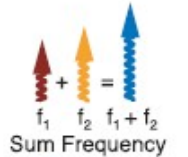
$$(12)$$

- Difference frequency generation (DFG): f_1, f_2 are combined
- (OPG)

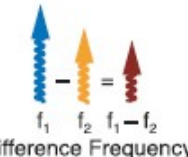
Nonlinear Effects



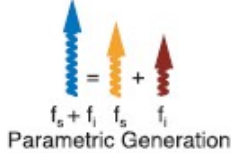
Second Harmonic



Sum Frequency

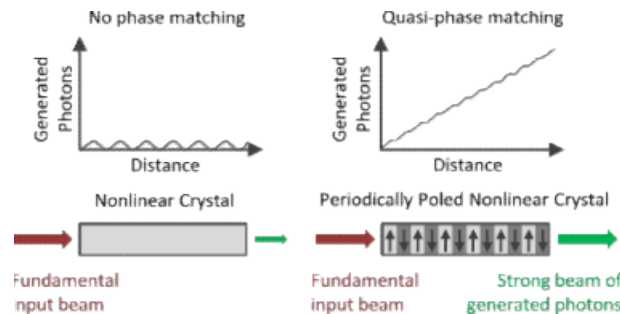


Difference Frequency



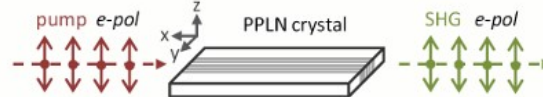
Parametric Generation

- **Phase matching:** Only when the phase of the newly generated photons matches that of previously generated photons, the combination of photons will be efficiently generated.
 - Phase matching = fixing the relative phase between two (or more) frequencies of light as the light propagates through the crystal. Because refractive index is dependent on frequency of light, so photons at different frequencies will travel at different speeds, resulting in phase delays.
 - To maintain the phase throughout the crystal, there are several ways:
 - The conventional way: **Birefringence phase matching** (eg. Frequency doubling/SHG of 1064nm in LiNbO₃), where the light is sent to propagate in a direction such that the natural birefringence cancels the chromatic dispersion and thus matches the refractive index of generated light at certain crystal temperature.
 - PPLN: **engineered quasi-phase-matched material**. The orientation of LiNbO₃ crystal is periodically poled (inverted). By inverting the crystal orientation at every peak of the sinusoidal light generation, the # of generated photons will grow as the light propagates through PPLN. Therefore, the poling period depends on the wavelength of light (obviously), and the PPLN temperature.

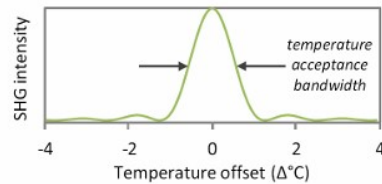


How to choose and use PPLN crystal

- **Crystal Length:** every crystal has certain associated pump acceptance bandwidth --- inversely proportional on length. This acceptance bandwidth is due to the group velocity mismatch between interacting waves/light. So longer length-->more sensitive and narrower the acceptance bandwidth.
 - For Narrowband CW source: long crystal lengths at 20-40mm give best efficiency.
 - For Pulsed sources: short lengths at 10mm (nanosecond pulse) or 0.5 to 1mm (femtosecond). For SHG of femtosecond pulses, even if the crystal is too long, causing pump bandwidth is much broader than the crystal's acceptance bandwidth, it's still possible to achieve high conversion efficiency.
- **Polarization:** to get highest nonlinear coefficient of the crystal, input light must be **e-polarized (= polarization must align with dipole moment of the crystal)**. This is done by aligning the polarization axis of light parallel to thickness of crystal. This is true of ALL nonlinear interactions!
 - This config is called Type-0 phase matching (ee-e), as all the beams have same polarization



- Type 1 phase matching (oo-e) and type 2 phase matching (eo-e) are also possible for PPLN for generation of heralded single photons.
- **Focusing and optical**
 - Typically Coevision crystals contain several grating periods. To get high conversion efficiency, the pump beam should be focused into a grating/track with the focal point on the middle of the crystal length.
 - Theory: optimum efficiency when the confocal parameters ξ ($2 \times$ rayleigh range) of fundamental beam(s): $\xi/\text{crystal length} = 2.84$.
 - General rule of thumb: **Spot size should be chosen such that Rayleigh range = 1/2 Crystal length.** The size can be reduced slightly until max efficiency is obtained. **Also for SFG, two pump/fundamental beams should be made to have the same Rayleigh range.**
- **Temperature and period:** As said above, poling period of PPLN is determined by the wavelength of the light being used. This quasi-phase-matched wavelength can be tuned slightly by changing the temperature.
 - One PPLN crystal can contain multiple different poling periods to allow different wavelength to be used at a given temperature. Use the temperature tuning curves as a guide to see the required temperature for phase-matching.
 - The temperature dependence of conversion efficiency usually follows sinc^2 function, showing a certain temperature acceptance bandwidth. Longer length-->more sensitive and narrower the acceptance bandwidth.
 - **The optimal temperature is found by heating the crystal to 20C above calculated temperature, then letting it cool down while monitoring the output optical power to see which temperature point is best.**



PPLN selection guide: <https://www.thorlabs.com/catalogpages/694.pdf>

Example: 637.5 nm generation via 1560nm + 1077nm

First, one must look at the Temperature tuning curves of the QPM crystals:

- Fundamental beam powers, theoretical optimal temperature, FWHM bandwidth of the phase-matching acceptance temperature

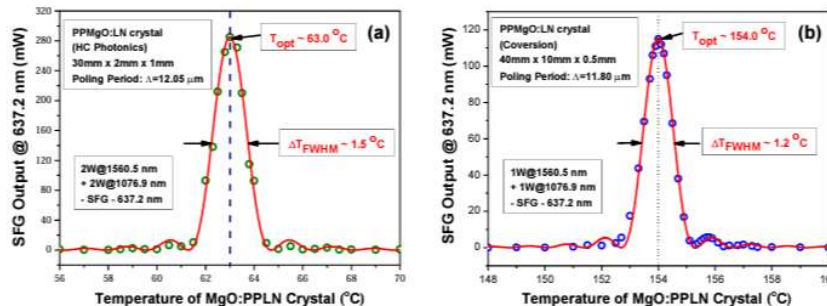
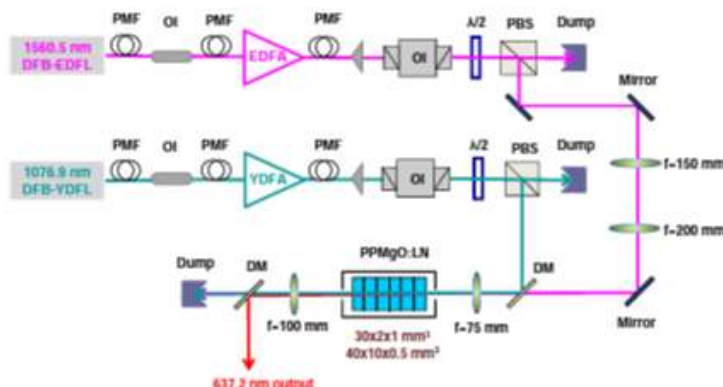


Fig. 2. The temperature tuning curves of the PPMgO:LN crystals for single-pass sum-frequency generation. Circles are the experimental data, while the solid lines are the theoretically fitted curves using sinc^2 function. (a) PPMgO:LN crystal of the dimensions 30 mm×2 mm×1 mm (Poling Period: $\Lambda=12.05 \mu\text{m}$), and the QPM optimized temperature is 63 °C with a FWHM of 1.5 °C; (b) PPMgO:LN crystal of the dimensions 40 mm×10 mm×0.5 mm (Poling Period: $\Lambda=11.80 \mu\text{m}$), and the QPM optimized temperature is 154 °C with a FWHM of 1.2 °C.

- Next, we need to plan optical arrangement (beam size, rayleigh range, focusing).
 - To get high SFG conversion, it's crucial that the two fundamental beams should have matching spatial mode (Same rayleigh range).
 - This Rayleigh range should be about 1/2 of crystal length.

With these considerations one can use telescope and lenses to **first collimate** the two pump beams coming out of fiber amplifiers, then **focus** onto the center of crystal length to appropriate beam size. Here they use final 75mm lens on each pump beam to focus into crystal with beam waist of 43μm (1560nm) and 30μm (1077nm).



- Now we must monitor and optimize the conversion output.
 - During alignment, people will use low power for the pump beams. But note down that QPM temperature must be optimized at each combination of input power.
 - So, once you optimize the conversion tweeking the alignment, focusing, etc., and temperature at low power --> take some power to **wavemeter**. Fix down power of one pump beam, also record the temp and output power values at these pump powers---->Slowly increase the power of the other pump beam, and optimize the temperature, frequency again and record! Do this until you reach the full pump beam power and optimize temperature there.
 - Finally, you should make a plot like this:

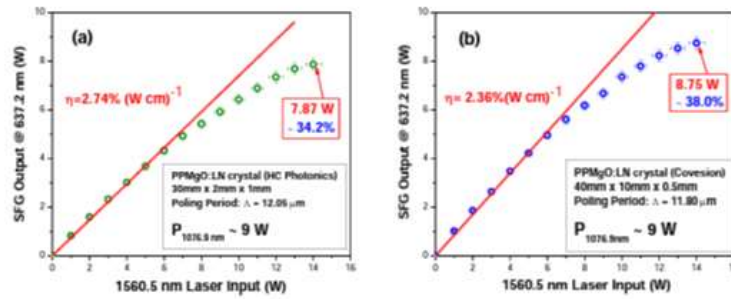


Fig. 3. SFG output power versus power of two fundamental lasers using a 75 mm focusing lens. The 1076.9 nm laser power was fixed at 9 W and the 1560.5 nm laser power varied. The error bars come from the measurement error of power meter. (a) the case of 30-mm-long PPMgO:LN crystal from HC Photonics; (b) the case of 40-mm-long PPMgO:LN crystal from Covesion.

This shows the 637.2nm SFG power as a function of one pump beam (the other pump power is kept fixed).

Reference: <https://arxiv.org/ftp/arxiv/papers/1601/1601.03579.pdf>