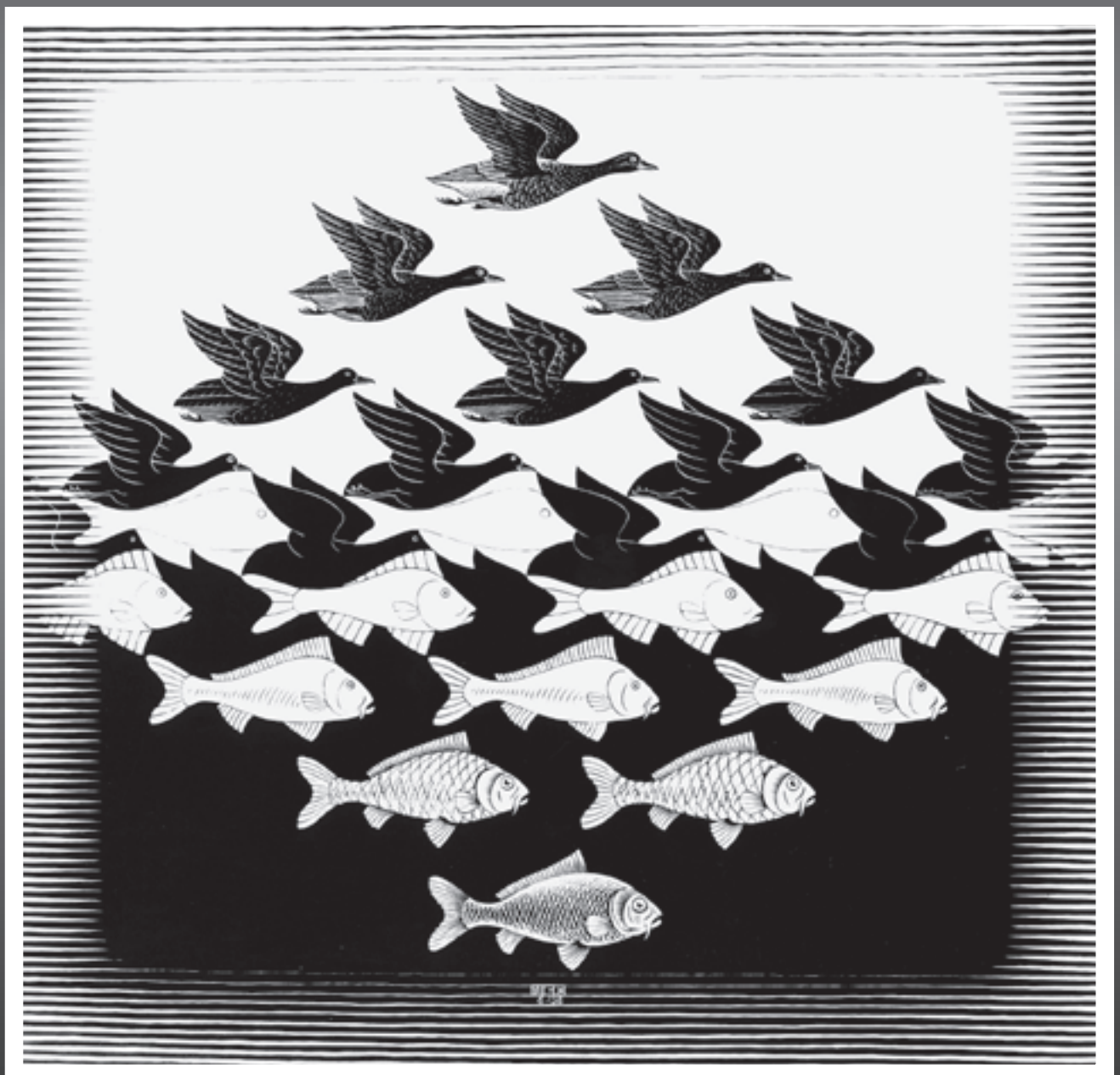
Broadband Nonlinear Frequency Conversion

Haim Suchowski, Barry D. Bruner, Ady Arie and Yaron Silberberg



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There is growing demand for coherent light in wavelength regimes that are not attainable with conventional lasers. Researchers are working to develop new techniques for nonlinear frequency conversion as well as novel crystal design structures to help meet this need. Their work is leading to applications in optical communications, metrology and other areas.

Most of the optical phenomena that we encounter in everyday life—such as refraction, absorption and dispersion—occur in the realm of linear optics. With these types of linear optical effects, the intensity or propagation direction of the light wave can change based on the interaction between light and matter. However, other properties of light—in particular, the spectrum of colors contained in the beam—remain completely unaffected.

In contrast, in the nonlinear optical regime, it becomes possible to transform this fundamental property of light beams—their color—and even to mix colors to create new ones, much like a painter combines colors on a palette. This requires high light intensities that were inaccessible before the invention of the laser.

Techniques to convert the frequency of light are currently being used in a number of different areas in optics. They have been demonstrated for a large variety of light sources, from the ultraviolet to the infrared, and for both narrowband and broadband signals.

Fundamentals of frequency conversion

At the microscopic level, we can visualize an electromagnetic wave with angular frequency ω corresponding to a specific color. As the wave passes through a dielectric material, it polarizes the atoms inside the material—that is, it induces oscillations of the electric dipoles in the atoms.

In linear optics, we assume that these dipoles all oscillate at the same angular frequency ω of the incoming beam. In this case, the induced polarization of the dipoles varies linearly with the strength of the electric field. However, if the light field is strong enough, this linearity does not hold. The polarization must be expanded as a Taylor series in powers of the electric field, where the higher order terms describe the nonlinear response. Since the nonlinearity of optical materials is relatively weak, this necessitates the use of very strong input light fields in order to observe nonlinear optical effects. The nonlinear response of the material is the source for effects such as frequency conversion.

The second term in this expansion is known as the quadratic nonlinear response—so named because its magnitude is proportional to the square of the electric field. The proportionality constant is referred to as the nonlinear susceptibility of the material $\chi^{(2)}$, which is non-zero only in a small yet important group of materials that lack inversion symmetry.

The quadratic response, which gives rise to the simplest nonlinear effects, reflects the fact that this induced polarization contains contributions from two input electric fields. Since the oscillating dipoles can be regarded as small antennas that radiate electromagnetic waves, it leads to light that is emitted at new frequencies.

If two input frequencies ω_1 and ω_2 are used, then the nonlinear response of the material can result in the generation of light at the second harmonic of each of the input frequencies (i.e., the doubled frequencies $2\omega_1$ or $2\omega_2$), as well as the sum $(\omega_1 + \omega_2)$ or difference $(\omega_1 - \omega_2)$ of the two. These frequency conversion processes could generate coherent radiation at wavelengths for which compact and efficient lasers do not exist. In this article, we shall concentrate on sum-frequency generation (SFG) as an example of a quadratic nonlinear process—but many of the conclusions can also be applied to other mixing processes.

Nonlinear interactions can occur over length scales that are considerably longer than the wavelengths of the interacting waves. The length of a typical nonlinear crystal is about 1 cm, which is roughly four orders of magnitude longer than the wavelengths of visible and near-infrared light. The distributed nature of the mixing process creates the so-called phase matching problem: Owing to chromatic dispersion, the two input light waves at frequencies ω_1 and ω_2 propagate with a different phase velocity with respect to the generated wave at $\omega_3 = \omega_1 + \omega_2$. As a result, sum-frequency-converted light that is generated in different regions in the crystal will be out of phase with the input fields, and they will destructively interfere.

(Facing page) M.C. Escher, *Sky and Water I* (1938). Escher's famous woodcut drawing helps to illustrate the concept of adiabatic (slow-varying) evolution, which can play a significant role in optical processes such as frequency conversion.

[Phase Mismatch and Quasi-Phase Matching]

The phase mismatch parameter, which originates from the chromatic dispersion of the nonlinear material, characterizes the lack of momentum conservation of the nonlinear process. For sum or difference frequency generation, it is given by

$$\Delta k_{Process} = k_1 + k_2 - k_3 = \frac{1}{c}(n_1\omega_1 + n_2\omega_2 - n_3\omega_3),$$

where k_i is the wavevector of each beam, and n_i is the refractive index of the beam in the material. Quasi-phase matching (QPM) compensates for the lack of momentum conservation by adding an artificial momentum originating from the local periodicity of the structure of the nonlinear material. The local modulation period, usually denoted as $\Lambda(z)$, provides a further contribution to the total phase mismatch:

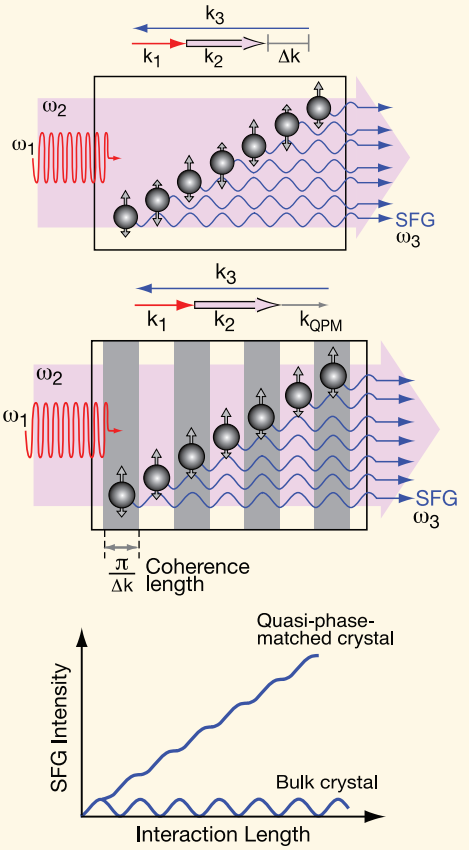
$$\Delta k_{Total} = \Delta k_{Process} + k_{QPM} = \Delta k_{Process} + \frac{2\pi}{\Lambda(z)}.$$

In a periodically poled crystal, the sign of the nonlinear susceptibility is inverted with each successive domain, with each domain width equal to the coherence length of the nonlinear process. This leads to constructive interference between the interacting beams along the entire crystal length, thereby improving upon the conversion efficiency relative to birefringent phase-matched nonlinear crystals.

Fabrication of periodically poled crystals is possible only by using certain crystal materials. Electric field poling is commonly used in ferroelectric crystals, such as $LiNbO_3$, $LiTaO_3$ and $KTiOPO_4$. Using photolithographic techniques, one can deposit a patterned metal electrode with the required modulation pattern on the top surface of the crystal, whereas the bottom surface of the crystal is coated with a planar electrode.

A high-voltage pulse induces a permanent reversal of the material dipoles in the areas with metal contact on the top surface. The typical resolution of this method is several microns for a crystal thickness of 1 mm. However, researchers have fabricated periods below 1 µm as well as thicknesses of up to 5 mm.

Another possibility for modulating the nonlinear coefficient is via selective epitaxial growth with semiconductor materials such as GaAs.



The difference between the incoming and outgoing wave vectors is usually termed the phase mismatch, denoted Δk, and the typical length over which a π phase shift is accumulated is called the coherence length. The value of the coherence length is typically just a few microns up to a few tens of microns for nonlinear interactions in the visible and near-infrared ranges of the spectrum. Thus, in bulk materials, coherence between the interacting waves is not maintained over macroscopic lengths, which results in vanishingly small frequency conversion efficiencies.

There are several possible techniques to compensate for this inherent lack of phase matching. One technique is to take advantage of the natural birefringence of the medium by choosing a specific "cut" or crystal orientation that minimizes the phase mismatch, Δk, for a specific frequency conversion process. Birefringent phase-matched crystals have been the most commonly used method for nonlinear frequency conversion over the nearly 50-year history of the field.

Growth, preparation and alignment of these crystals is straightforward. However, the applicability of simple birefringent phase-matching can be limited by a number of factors. First and foremost, since the method relies on given material properties—the dispersion and the birefringence—it is not always possible to find a crystal orientation that satisfies the phase-matching condition.

Another way to compensate for phase mismatch is to employ quasi-phase matching (QPM) techniques. In a QPM crystal, the nonlinear susceptibility is modulated spatially. In its simplest form, the nonlinear coefficient is modulated at a period equal to twice the coherence length. Hence, the phase of the newly generated waves is reversed in each domain of the crystal, resetting the phase difference between the waves and restoring constructive interference over the entire crystal length.

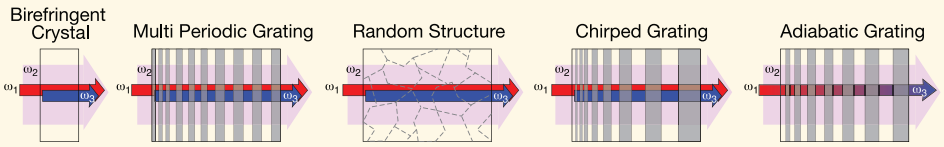
It is possible to achieve much higher conversion efficiencies with QPM crystals compared with birefringent crystals. Furthermore, this can be done for essentially any nonlinear process within the transparency window of the material. Moreover, the modulation of the nonlinear susceptibility does not have to be strictly periodic. With QPM, one can also design more sophisticated structures with almost any desired function of the phase mismatch. QPM has thus proven to be an extremely versatile technique for solving the phase-matching problem.

Nevertheless, for the most commonly used types of birefringent and QPM crystals, it is necessary to maintain a very small or vanishing Δk throughout the conversion process. This limits the robustness of frequency conversion in both types of crystals, because variations in experimental conditions that affect

QPM has proven to be an extremely versatile technique for solving the phase-matching problem.

the refractive index of the light in the material (e.g., incident angle, crystal temperature) will result in corresponding variations of the phase mismatch. The same is true for any change of the input frequencies ω_1 and ω_2, thereby limiting the conversion bandwidth for broadband or ultrafast pulses.

[A comparison of frequency-conversion methods]



	Birefringent Crystal	Multi Periodic Grating	Random Structure	Chirped Grating	Adiabatic Grating
Bandwidth	Moderate	Narrow	Very wide	Wide	Wide
Efficiency	Low	High	Very low	Low to moderate	Very high
Cost and complextiy	Very low	High	Low	High	Very high
Preservation of spectrum	No	No	Yes	Yes	Yes

Challenges in broad bandwidth frequency conversion

In order to convert broadband light, and in particular ultrafast (femtosecond and picosecond) pulses, it is necessary to overcome the effects of chromatic dispersion as well as phase mismatch. The large inherent phase mismatch for broadband signals makes it very difficult to perform frequency conversion using the methods described in the previous section, which are normally only efficient for narrow spectra. Due to dispersion, ultrashort pulses undergo temporal stretching during propagation, which results in lower pulse peak intensity and therefore lower conversion efficiency. Dispersion also causes the three interacting waves to travel at different group velocities, thereby shortening the effective nonlinear interaction region.

In birefringent phase-matched crystals, the common solution to achieving broadband conversion is to use very short crystals—less than 1 mm in length. The short propagation length lessens the effect of dispersion; however, it also considerably limits the achievable conversion efficiency.

Another approach for coping with these challenges is to use random crystal structures that circumvent the need for phase matching completely. Researchers demonstrated this in a disordered polycrystalline material that consisted of randomly distributed and oriented single crystal domains. Since the induced polarization in each domain is uncorrelated with that of its neighbors, the averaged contribution of the mixing terms of emitted frequency-converted waves from different domains is zero. In other words, there is neither constructive nor destructive interference over an extremely wide frequency range. Instead, the conversion efficiency is observed to increase linearly with the interaction length, or rather, with the number of crystal domains.

Disordered polycrystalline materials made from semiconductors such as ZnSe and GaAs, or disordered ferroelectrics such as SBN, are inexpensive and can be manufactured to very high optical quality standards. However, the conversion efficiencies that have been demonstrated to date have been low. The effectiveness of the method also depends on one's ability to manipulate the average size of the domain structures—a fact that carries implications for the applicability of this scheme to certain wavelength regimes.

Many types of QPM structures have been implemented for frequency conversion with broader bandwidths. Multi-periodic modulation patterns can be arranged in sequence along the length of the crystal, leading to phase-matching of several processes simultaneously. This is an effective method for converting a number of narrow bandwidth light sources, but not for a single broadband light source, because each distinct modulation period provides optimal phase matching only for a specific wavelength.

In a chirped QPM crystal, the modulation period is varied as a function of position along the propagation axis. Each frequency component in the pulse is phase-matched only at a particular position in the crystal. However, the sweep of the modulation period ensures that a broad phase-matching bandwidth can be supported after propagation over the entire crystal length. In addition, the spatial dependence of the conversion process means that the total group delay dispersion acquired by each distinct frequency component can be modulated, resulting in broadband frequency conversion with simultaneous temporal pulse shaping or pulse compression.

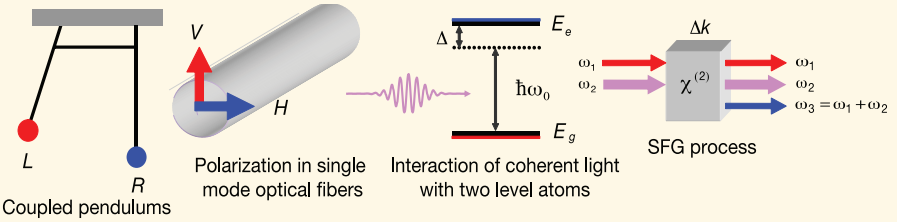
Geometric visualization of frequency conversion

For the SFG process, where $\omega_3 = \omega_1 + \omega_2$, we can use a more intuitive visual approach to understand the dynamics. Let's assume that the ω_2 wave, which we will define as the pump wave, is much more powerful than the other summed

[Two-coupled-mode systems + Bloch sphere representation]

Two-coupled-mode dynamical systems appear in many fields of physics and particularly in optics. The dynamical behavior of these systems can be fully characterized by the phase difference (mismatch) between the modes, and the strength of the coupling between them.

A formal equivalence can be shown between a number of diverse physical systems, such as (below, left to right) the difference between the lengths of the classical coupled pendulums, the phase retardation between the polarization modes in a fiber, the off-resonance detuning in the coherently excited two-level atom, and the phase mismatch Δk in the SFG process with a strong undepleted pump.

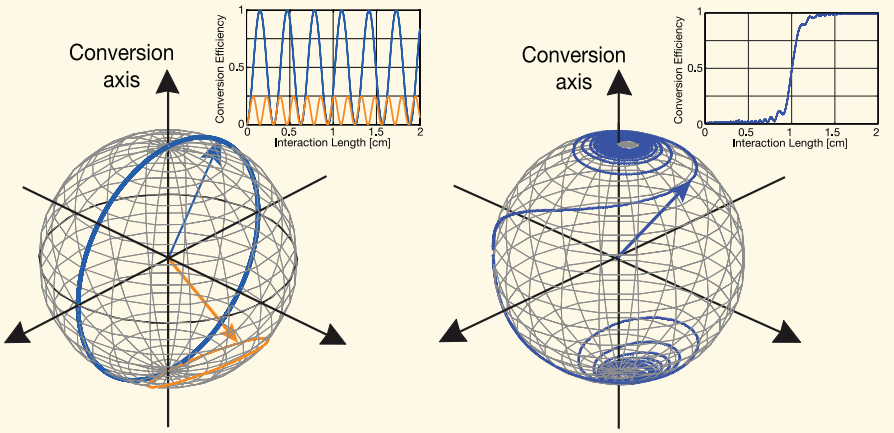


Coupled pendulums | Polarization in single mode optical fibers | Interaction of coherent light with two level atoms | SFG process

Expanding further on the likeness between the systems illustrated above, the amplitudes of the electromagnetic waves ω_1 and ω_3 in the SFG process are formally equivalent to the atomic population of the ground and excited states in the two-level atom. Thus, one can use a geometrical representation, the commonly used Bloch sphere from atomic physics (or equivalently, the Poincaré sphere in polarization optics), in order to gain physical intuition on the dynamics of the process without having to perform drawn-out analytical simulations.

The south pole of the sphere then corresponds to zero-conversion efficiency (i.e., full "population" of ω_1 without any sum frequency conversion to ω_3), and the north pole corresponds to 100 percent conversion.

When SFG process is perfectly phase matched (Δk=0), energy oscillates between ω_1 and ω_3, as the energy flows completely to ω_3 followed by a reversal of the energy flow back to ω_1 (solid blue line in the figure below and to the left). The analog of this process in a two-level atom is Rabi oscillations of the populations between the two atomic levels. For imperfect phase matching ($\Delta k \neq 0$), the maximum conversion to ω_3 reaches much lower values (orange line in figure on the left). The figure below on the right shows the dynamic trajectory for adiabatic frequency conversion. The energy is completely transferred to ω_3, and the conversion is insensitive to variations in Δk.



wave ω_1. Thus, the fractional loss of energy in ω_2 is small, and we can regard its amplitude as constant throughout the interaction.

Therefore, along the interaction length, the flow of energy occurs only between ω_1 and ω_3. In this limit, the evolution of the electric field amplitudes of ω_1 and ω_3 along the crystal can be fully described by the so-called "two coupled mode" theory. This description is analogous to a number of other well-known physical systems, such as a system of two coupled pendulums, coherent excitation of two-level atomic systems, and polarization orientation inside an optical fiber.

Adiabatic frequency conversion

Two-coupled-mode systems provide us with far more than just handy reference material for understanding frequency conversion. It is also possible to use our understanding of these analogous systems for formulating new frequency conversion schemes.

In optical physics, "rapid adiabatic passage" (RAP) is a well-known technique for performing robust and efficient population transfer in multi-level atomic and molecular systems. Adiabatic frequency conversion, which adapts the concepts of two-coupled-mode theory and adiabaticity to the context of nonlinear frequency conversion, is another approach for achieving broad bandwidth conversion with high efficiency.

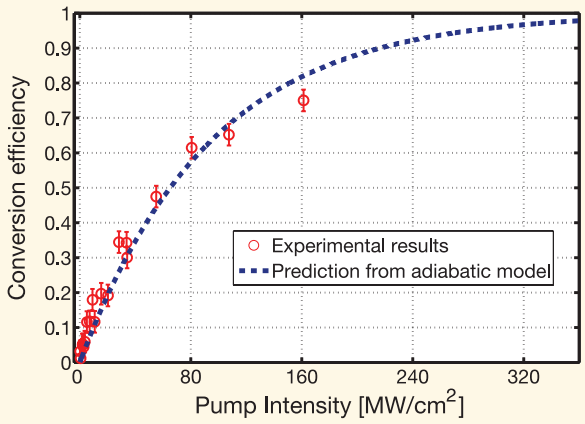
This method requires a strong pump field ω_2 and a slow (i.e., adiabatic) variation of the phase mismatch Δk during the nonlinear interaction, from large negative to large positive values. Adiabatic techniques are especially advantageous for efficient and robust frequency conversion because they are not sensitive to small changes in parameters that affect the phase mismatch, such as temperature, crystal length, acceptance angle and input wavelength.

We realized this scheme experimentally using chirped QPM crystals. An adiabatic aperiodically poled KTP crystal was realized by linearly sweeping the local poling period $\Lambda(z)$ from 14.6 to 16.2 μm along a crystal length of 20 mm. This was shown to be effective for efficient SFG of tunable narrowband pulses from a near-IR optical parametric oscillator (OPO) system (ω_1) with a strong pump field (ω_2) produced by a Q-switched Nd:YLF laser.

The sum-frequency conversion efficiency is dependent on pump intensity. The conversion efficiency increases as the pump intensity rises because the nonlinear interaction becomes increasingly adiabatic. For sufficiently large pump intensities, the conversion efficiency can reach unity.

It is possible to use our understanding of two-coupled-mode systems for formulating new frequency conversion schemes.

[Efficiency of adiabatic frequency conversion]



Efficiency of adiabatic frequency conversion for SFG of a signal beam (ω_1) centered at 1,530 nm and a strong pump beam (ω_2) centered at 1,064 nm. The maximum demonstrated conversion efficiency was 74 percent, but it can approach 100 percent using higher pump intensities. The analogy with other two-level systems enables analytic calculation of the expected conversion efficiency.

In the figure above, we observed a maximum conversion efficiency of 74 percent from ω_1 to ω_3 across a 150-nm tuning range of signal wavelengths.

The maximum available pump intensity was 160 MW/cm^2, far below the 500 MW/cm^2 damage threshold of the periodically poled crystal. Therefore, even higher efficiencies are possible. However, the trade-off for achieving large bandwidth with high efficiency is that it is necessary to use very large pump intensities and relatively long nonlinear crystals.

The wide tuning range of adiabatic frequency conversion also makes this an attractive method for the frequency conversion of broadband ultrafast pulses. In addition, the maximal conversion efficiency depends on the intensity of the strong narrowband pump and not on the peak intensity of the ultrashort pulse. This relaxes the need for using short crystals because dispersion of the ultrashort pulse does not affect the conversion. Adiabatic frequency conversion also preserves the spectral phase and amplitude, which means that there is no spectral filtering during the conversion process, so that the pulse can be compressed to the same time duration as the input seed pulse.

This implementation of broadband conversion was shown for SFG using an ultrafast Ti:sapphire laser (center wavelength near 800 nm) and a strong Q-switched Nd:YLF pump laser (center wavelength at 1,053 nm). The periodicity of the KTP crystal was varied from 5.16 to 5.58 μm along a crystal length of 3 cm. This resulted in a broadband blue pulse centered at 450 nm.

The concept of adiabatic frequency conversion can be extended to any second-order nonlinear interaction, including difference frequency generation (DFG). We also demonstrated an efficient broadband conversion into the mid-IR near 3 μm. This was done by taking the same two near-IR seed and pump sources described in the SFG implementations and mixing them in a 2-cm adiabatic SLT crystal with periodicity varying periodicity from 22.6 to 23.4 μm.

The standard method for producing intense ultrafast mid-IR pulses involves an amplified near-IR Ti:sapphire laser and two successive optical parametric amplifiers, with near-IR to mid-IR conversion efficiencies that do not exceed more than a few percent. Adiabatic DFG involves a far simpler single nonlinear conversion step, with efficiencies that could approach unity depending on the strength of the pump source.

In this article, we have attempted to provide an overview of the fundamentals of frequency conversion, especially as it pertains to the conversion of broadband light. Frequency conversion methods have matured considerably in recent years, in large part due to improvements in crystal design and quality. Solutions that can increase the bandwidth of frequency conversion are particularly exciting, as they are leading to applications for diverse broadband laser sources involving ultrafast pulses, broadband fluorescence, entangled photon pairs produced via parametric downconversion, and frequency combs. ◭

OSA Member **Haim Suchowski** (Haim.Suchowski@weizmann.ac.il), Barry D. Bruner and Yaron Silberberg are with the department of physics of complex systems at the Weizmann Institute of Science in Rehovot, Israel, Ady Arie is with the department of physical electronics at Tel-Aviv University in Tel-Aviv, Israel.

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